MEETING THE CHALLENGES OF CONTINENTAL POLLUTANT PATHWAYS:
Volume II- Case Studies

prepared by Members of
The North American Expert Advisory Panel
on Continental Pollutant Pathways

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CASE STUDIES

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Case Study

Mercury

Authors

Wilfred Pilgrim- N.B. Ministry of Environment, Fredericton, Canada
Marc Lucotte- GEOTOP, Université du Québec à Montréal, Canada
Shelagh Montgomery- GEOTOP, Université du Québec à Montréal, Canada
Carlos Santos-Burgoa- Instituto de Salud, Ambiente y Trabajo, Mexico City, Mexico
Maria Isbel Ibarrola Uriarte - Instituto de Salud, Ambiente y Trabajo, Mexico City, Mexico
Francisco Abascal-Garrido- Instituto Nacional de Ecologia, Mexico City, Mexico
Margaret Round- NESCAUM, Boston, USA
Don Porcella- Electric Power Research Institute, Palo Alto, USA

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MERCURY CASE STUDY

Executive Summary *(draft, Hg Case Study March 24, 1997)*

The international science community has recognized Hg as a serious pollutant, and international protocols are being developed to control emissions. Scientists have shown from the analyses of lake sediment cores that in industrialised regions of the world (primary sources of mercury are Waste Incineration, Coal Combustion and Smelting), mercury deposition has increased on average 2-3 fold from the beginning of the industrial period, approximately 100 years ago, to present day. It is through fish consumption that mercury exposure in humans and in fish-eating birds and mammals takes place. Therefore, the key to understanding risk from mercury contamination is the linkage of mercury inputs to aquatic environments and accumulation in fish, a link dominated by methylation.

Some uncertainties with respect to biogeochemical cycling and potential chronic effects of mercury on humans and ecosystems have not permitted a smooth transition between science and policy decisions on the mercury issue. In particular, the noted delay in release of the US EPA Mercury Report to Congress. With respect to human health the uncertainty is in "the effects of low level exposure to methylmercury", Hg environmental policy is based on the fact that deleterious health effects related to methylmercury tend to be viewed solely in terms of clinical signs of intoxication. The critical uncertainty is identifying the level at which sensitive neurobehavorial consequences of prenatal exposure to methylmercury occur, and the extent to which the public, especially sensitive populations (e.g. pregnant women), are exposed to methylmercury from fish consumption. There is also a misconception that lots of accurate mercury data have been gathered since the 1950s. Much of the older mercury data are inaccurate due to analytical difficulties and sample contamination, Hg concentrations in the atmosphere and water tend to lie in the ppt (parts per trillion) to low ppb range. It has only been in the past 5 years that technical advances have permitted researchers to produce reliable mercury results for air and water.

To reduce risk to humans and ecosystem health, Canada, Mexico and the United States need to develop cooperative programs on a variety of environmental issues. However, because of the scientific evidence of mercury contamination, and considering it's very volatile and highly toxic nature, and inter-media distribution, this is an excellent pollutant to develop a holistic scientific approach between the NAFTA countries. With the paucity of information available in the literature concerning the status of Hg in the Mexican environment and, in contrast, the tremendous number of studies ongoing in Canada and the United States, trilateral cooperation could only yield positive results.

▪ ACKNOWLEDGEMENTS

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Panel Members

Santos-Burgoa, Carlos
Director
Instituto de Salud, Ambiente y Trabajo
Coapa 160, Torellio Guena
Mexico, DF 14050
Mexico

Round, Margaret
Program Analyst
Northeast States for Coordinated Air
Use Management (NESCAUM)
129 Portland Street
Boston, MA 02114
USA

Pilgrim, Wilfred
Mercury Coordinator
Ecological Monitoring & Assessment Network
NB Environment
P.O. Box 6000 - 364 Argyle Street
Fredericton, New Brunswick E3B 5H1
Canada

Porcella, Donald
Project Manager
Electric Power Research Institute
P.O. Box 10412, 3412 Hillview
Palo Alto, California 94304
USA

Montgomery, Shelagh
Researcher
Université du Québec à Montréal
C.P. 8888 Suc. Centre Ville
Montréal, Québec M3C 3P8
Canada

Lucotte, Marc
GEOTOP
Université du Québec à Montréal
C.P. 8888, succursale Centre-Ville
Montréal (Québec)
H3C 3P8, Canada

Abascal Garrido, Francisco (member)
Director de Laboratorio Central de Calidad Ambiental
Instituto Nacional de Ecología
Av. Revolución 1425
Mexico, DF
Mexico

Ibarrola-Uriarte, Maria Isbel
Toxicologist
Instituto de Salud, Ambiente y Trabajo
Coapa 160, Torellio Guena
Mexico, DF 14050
Mexico

■ INTRODUCTION

▸ Background

A great amount of peer-reviewed information is now available about mercury, much of which has been presented in international conferences held in Gavle, Sweden (Lindqvist et al., 1991a, 1991b), Monterey, California USA (Watras and Huckabee, 1994), Whistler British Columbia, Canada (Porcella et al., 1995), and more recently in Hamburg Germany in 1996. Elemental mercury (Hg^0) is a highly volatile metal, and the methylated form (CH_3-Hg) is a potent neurotoxin which bioaccumulates through the biotic food chain to levels that are thousands of times greater than that found in water.

▸ **Relative changes in levels of Hg from pre-industrial times compared to present day, (background, local, and regional emissions) (Figure 1).**

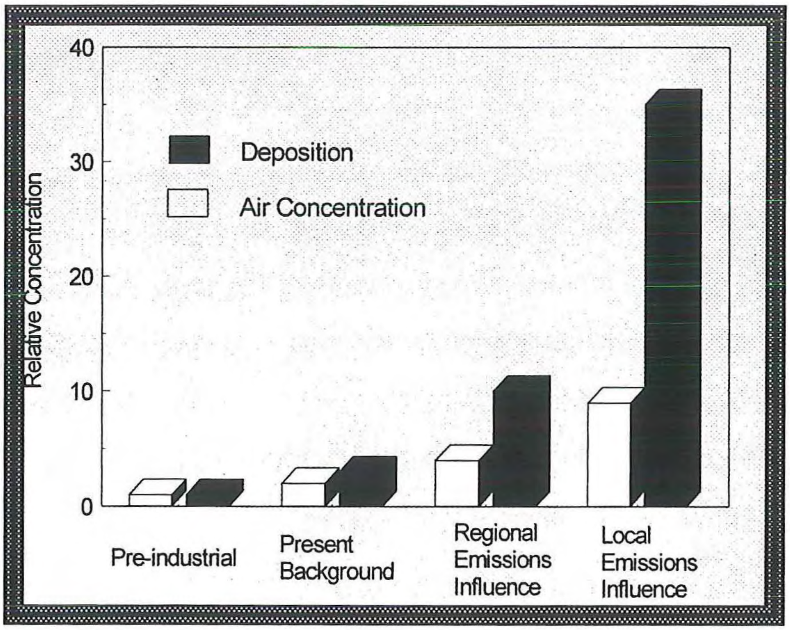

Figure 1. Relative past and present influences of emission sources (Expert Mercury Panel, 1994).

- ***Historic Records of Increased Mercury Deposition***

- **Scientists from around the world have shown from the analyses of lake sediment cores, that since the beginning of the industrial period, approximately 100 years ago, mercury deposition has increased between 2-5X in regions of eastern Canada and the northeast United States, and Sweden (Figure 2 and 3).**

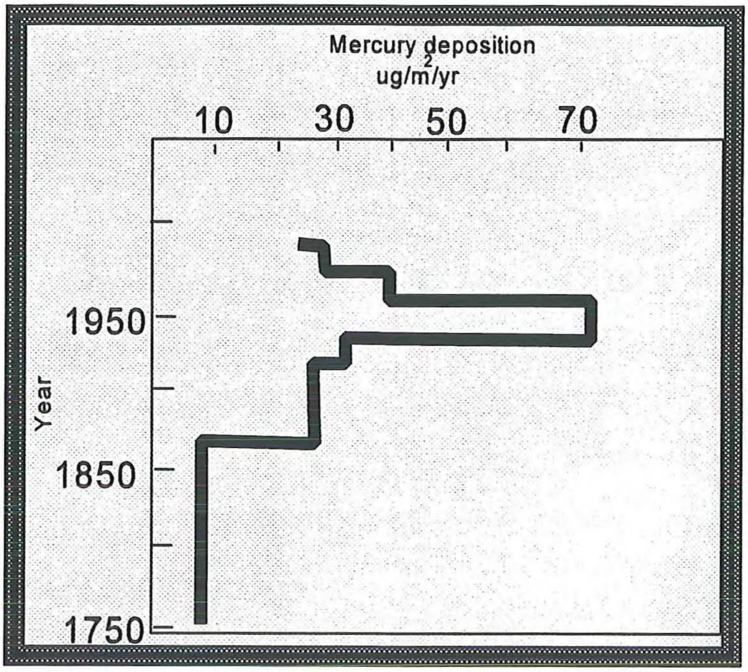

Figure 2. Historic profile of mercury deposition as recorded in Arlberg bog, Minnesota (Benoit et al., 1994).

Within the past decade a tremendous amount of research has been dedicated to the quantification of the historical record of Hg emissions. While global estimates will not be well known until a more detailed inventory of the sources of this heavy metal is produced, the general consensus among internationally recognized scientists is that, as a direct result of increased industrial activity within the last century, Hg emissions have increased by two to five times, with anthropogenic emissions now surpassing natural emissions (Johansson, 1985; Verta et al., 1986; Wiener, 1987; Nriagu and Pacyna, 1988; Fitzgerald 1986, 1989; Nriagu, 1989; Rada et al., 1989; Iverfeldt, 1991; Lindqvist, 1991; Nater and Grigal, 1992; Slemr and Langer, 1992; Swain et al., 1992; Gobeil and Cossa, 1993; Engstrom et al., 1994; Benoit, 1994; Mason et al., 1994; Landers et al., 1995; Lucotte et al. 1995; Munthe et al., 1995).

Engstrom and Swain (In Press) showed data for lake cores in Alaska with a continued increase and no peak which suggest that the global background is increasing (Alaskan cores) while some regional sources have decreased since the mid 1950s, but deposition is still 3 to 4 times greater than pre-industrial times. A working group of scientific experts on the collection and analysis of Hg in lake, peat and other archive cores prepared a protocol that will make it easier for scientists around the globe to obtain, analyze, and interpret these patterns of Hg deposition (Protocol, 1996). The protocol is available to any scientist with an interest in making historic deposition measurements. A meeting to discuss results of globally distributed coring operations will be held in 1999, around the time of the Fifth International Conference on Mercury as a Global Pollutant scheduled to be held in Rio de Janeiro, May 1999.

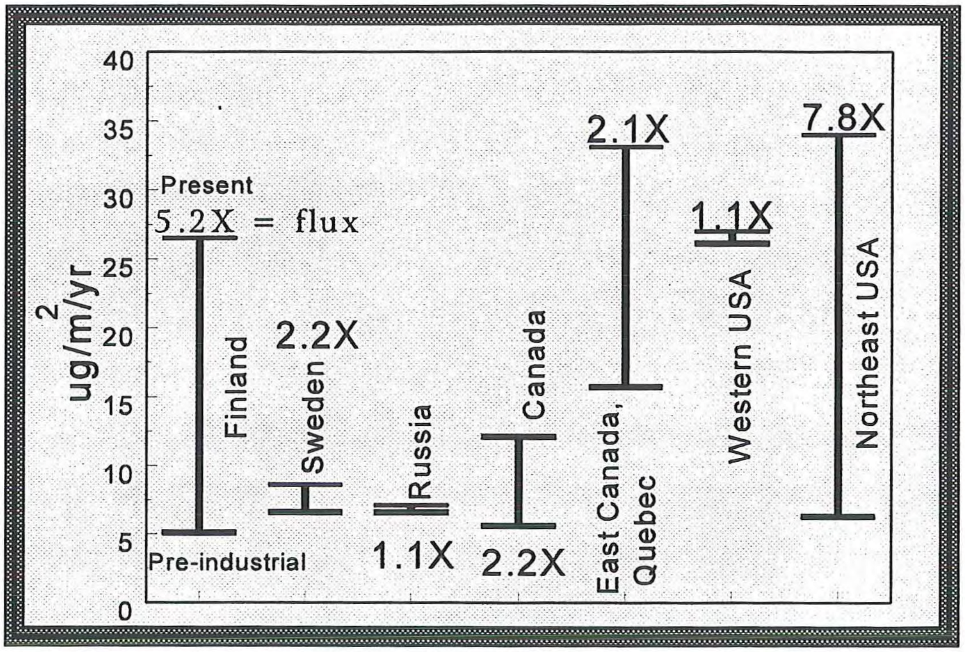

Figure 3. Average change from pre-industrial to present in mercury accumulation in lake sediments from different regions of the world (adapted from Landers et. al, 1997).

In their review article, Mason et al. (1994) re-evaluated the global Hg cycle and concluded that presently about 70% of the annual Hg flux to the atmosphere is attributable to anthropogenic sources. They calculated, furthermore, that 60% of the anthropogenic emissions are transported by long range atmospheric processes, while the remaining 40% are deposited locally. Since the conversion of simple concentration profiles to fluxes is more indicative of atmospheric Hg deposition, Table 1 shows both pre- and post-industrial fluxes determined using sediment cores collected from Arctic and boreal lakes (adapted from Landers et al., 1997). Based on these data from widely distributed sites, the researchers concluded that there has been a three fold increase in Hg flux since the onset of the industrial era. Similarly, in an earlier independent study, Engstrom et al. (1994), working on headwater lakes in Minnesota and Wisconsin, found similar variation between modern and pre-industrial fluxes.

- **Mercury emissions, transport and deposition are not as well known in Mexico and Canada, as in Europe or the United States.**

Mercury concentrations surpassing the World Health Organization guideline of 0.5 ppm now appear to be more prevalent in fish from many remote lakes, which also have elevated Hg levels in the sediments. Five Canadian provinces and 38 US states have issued health advisories for pregnant women and children to limit the consumption of freshwater fish due to Hg contamination. Fish collected in many sport fishing waters surpass the WHO guideline. Furthermore, recent data have shown that these sensitive populations are not adhering to the health advisory restrictions implemented by health authorities (LaRue et al., 1996). The risk to reproductive and behavioural health of fish eating birds and mammals are also of concern. The 1996 version of the US EPA Mercury Report to Congress is a comprehensive document that outlines the significance of the Hg issue and summarizes the current scientific knowledge. To date, this report exists only in a draft format thus hampering its availability and use as a reference document. The international science community has recognized Hg as a serious global pollutant and some countries such as Sweden have banned the use or recycling of Hg. With the paucity of information available in the literature concerning the status of Hg in the Mexican environment and, in contrast, the tremendous number of studies which have been documented for Canada and the United States, trilateral cooperation could only yield positive results.

Location	Source	Lake Name	Post Hg Flux ($ug/m^2/yr$)	Pre Hg Flux ($ug/m^2/yr$)
Finland	Verta	Iso-Iehmatampl	28.7	3.2
Finland	Verta	Valkea Kotinen	49.5	15.3
Finland	Verta	Sonnanen	42.1	2.7
Finland	Verta	Slikajarvi	39.6	4.5
Finland	Verta	Kangasjarvi	39.5	6.5
Finland	Verta	Valkeinen	4.8	4.0
Finland	Verta	Suo-Valkeinen	8.9	2.0
Finland	Verta	Ahveroinen	2.3	1.9
Finland	Verta	Vaslkkajarvi	22.9	6.0
Sweden	Johansson	Tussjon	17.0	2.5
Sweden	Johansson	Skarvsjon	14.3	1.9
Sweden	Johansson	Bjorken	9.0	4.0
Sweden	Johansson	Uggsjon	11.0	4.8
Sweden	Johansson	Karsvattnet	6.7	2.3
Sweden	Johansson	Vaster-Tackelsjon	20.7	17.2
Sweden	Johansson	Rensjon	19.5	9.4
Sweden	Johansson	Ellsjaur	8.5	3.3
Sweden	Johansson	Nattajarvl	9.9	5.4
Sweden	Johansson	Ala Makkarijarvl	12.6	6.5
Sweden	Johansson	Tunturljarvl	8.6	6.6
Russia	Landers	Nyagome	30.2	23.3
Russia	Landers	Khutudaturka	7.1	6.6
W. Canada	Lockhart	ELA	21.3	7.4
W. Canada	Lockhart	Kusawa	8.5	5.3
W. Canada	Lockhart	Laberge	17.7	14.5
W. Canada	Lockhart	Fox	8.1	5.3
W. Canada	Lockhart	Hawk	5.0	0.7
W. Canada	Lockhart	Far	7.7	2.3
W. Canada	Lockhart	Belot	3.8	3.5
W. Canada	Lockhart	Colville	8.1	4.2
W. Canada	Lockhart	Amituk	28.4	7.0
Québec	Lucotte	Jobert	33.9	18.9
Québec	Lucotte	La Cabane	30.1	11.1
Québec	Lucotte	des Voeux	5.3	2.6
Québec	Lucotte	Cabonga Reservoir	37.6	19.6
Québec	Lucotte	Duncan	51.6	24.1
Québec	Lucotte	Laporte	40.0	18.1
USA	Landers	Wonder	3.3	2.5
USA	Landers	Chandler	38.4	39.9
USA	Landers	Feniak	40.4	54.4
USA	Landers	Desperation	52.2	41.1
USA	Landers	Toolik	23.2	20.3
USA	Landers	Elusive	24.2	19.0
USA	Landers	Schrader	7.0	5.7
USA	Swain	Mountain	48.8	10.0
USA	Swain	Little Rock	40.3	10.0
USA	Swain	Cedar	15.2	4.5
USA	Swain	Dunnigan	17.9	5.0
USA	Swain	Thrush	16.5	5.0
USA	Swain	Kjostad	74.1	2.3
USA	Swain	Meander	25.1	7.55
*Adapted from Landers et al. 1997				

Table 1. Post and pre-industrial mercury flux (change in accumulation) in sediments (adapted from Landers, 1997).

■ ATMOSPHERIC CONTINENTAL PATHWAYS

▸ *The Global Atmospheric Mercury Cycle/Anthropogenic vs Natural Emissions*

Fitzgerald (1986) estimated that the 'close-to-steady-state' annual Hg inputs to the atmosphere were 5000-6000 Mg/yr. Mason, Fitzgerald and Morel (1994) improved on this former estimate and is the most widely accepted mercury budget in the science community to date. Mason et al. (1994) suggested at least 70% of the emissions are from anthropogenic sources and 4000 tonnes were emitted from land based anthropogenic sources of which 2000 tonnes entered the global pool. In their analysis of the global mass balance of mercury, 5000 tonnes of mercury are cycled to and from the global atmosphere each year, compared to an estimated 1600 tonnes before the industrial period (Figures 4 and 5). This is a 3 fold increase.

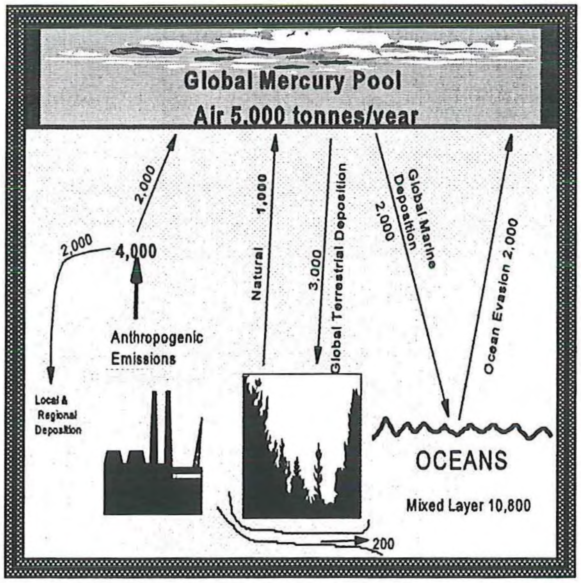

Figure 4. Current mercury budget and cycle (adapted from Mason et al., 1994).

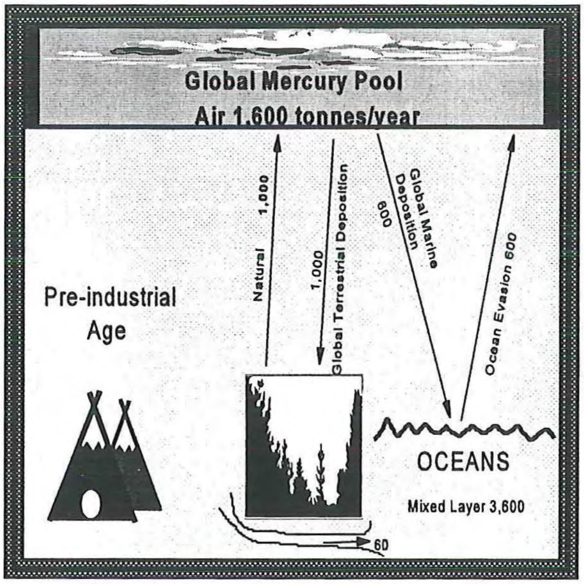

Figure 5. Pre-industrial mercury budget and cycle (adapted from Mason et al., 1994).

- Of the total 5,000 tonnes that enter the global atmospheric pool, 2,000 are from anthropogenic land based atmospheric sources, 2000 from the oceans (much of which is recycled anthropogenic fall-out), and 1000 tonnes from natural terrestrial sources. An additional 2000 tonnes is emitted to the atmosphere from anthropogenic land based sources and deposited locally or regionally.

Pirrone et al., (1996) estimated global anthropogenic mercury emissions of about 2200 Mg/yr, with a one-year atmospheric residence time, this annual input is balanced by an average global deposition rate of about 10 ug/m^2/yr with higher deposition in forested areas of the northern hemisphere. The estimates by Pirrone et al. (1996) are an important up-dating from the earlier estimates by Nriagu and Pacyna (1988), and though further refinement and the use of more up-to-date emissions estimates, as well as a more complete list of sources, suggest that the global estimate of 4,000 Mg/yr for anthropogenic emissions may be an upper estimate. In such a case, US emissions from all sources would range from about 4 to 8 percent of total global anthropogenic emissions. However, we must remember that the relative amounts of global, regional, and local deposition depend greatly on the species of mercury emitted and its subsequent atmospheric chemistry (EPMAP, 1994).

- Global atmospheric mercury budgets are still based on sparse measurements in various regions of the world, at different times, using different apparatus and analytical laboratories. In order to tighten up global estimates on mercury emissions and wet and dry deposition, further consistent measurements are needed.

Despite the research to date, the accuracy of global budget calculations remains dependent upon an improved inventory of emissions. Although reports coming from Canada, the United States and the Scandinavian countries contain increasingly reliable numbers, some of the potentially major players, such as China, India and Russia, remain highly under represented with respect to publications in the scientific journals. Based solely on coal use, both of these countries are likely to produce significant Hg emissions. For instance, in Russia, approximately 20% of the primary energy is supplied by coal (Clarke, 1996), while in China, 75% of the country's total energy requirements are fuelled by coal (Daniel, 1994). Within the scope of trilateral cooperation between Canada, Mexico and the United States, data for Mexico and Canada are still needed to define atmospheric Hg inputs and refine the present deposition and risk models. To narrow the knowledge gaps on Hg cycling, compatible international networks are necessary.

▸ *Atmospheric Mercury Emissions in Mexico, Canada and the United States*

Porcella (1996) uses newer emission estimates for municipal waste combustion (about 20 percent less) and medical waste combustion sources (about 80 percent less). Power generation emissions are about 10 percent less than USEPA 1994 numbers, because recent and quite accurate data on actual emissions were used from Chu and Porcella (1995). These estimates considered other data from Pai and Heisler 1996; Pirrone et al., 1996; USEPA, 1994; Porcella et al., 1996, and NPRI of Environment Canada, 1996. The estimates by the USEPA (1994) were the most accurate estimates up until present. The USEPA in 1994 estimated that approximately 83 percent of the total anthropogenic atmospheric emissions of 240 tonnes in the United States were from combustion sources, primarily utility boilers (21%), municipal waste combustion (23%), medical waste incineration (27%) and commercial/industrial boilers (12%). North American mercury emissions estimates shown in Figure 6 were adapted from Pirrone et al., 1996.

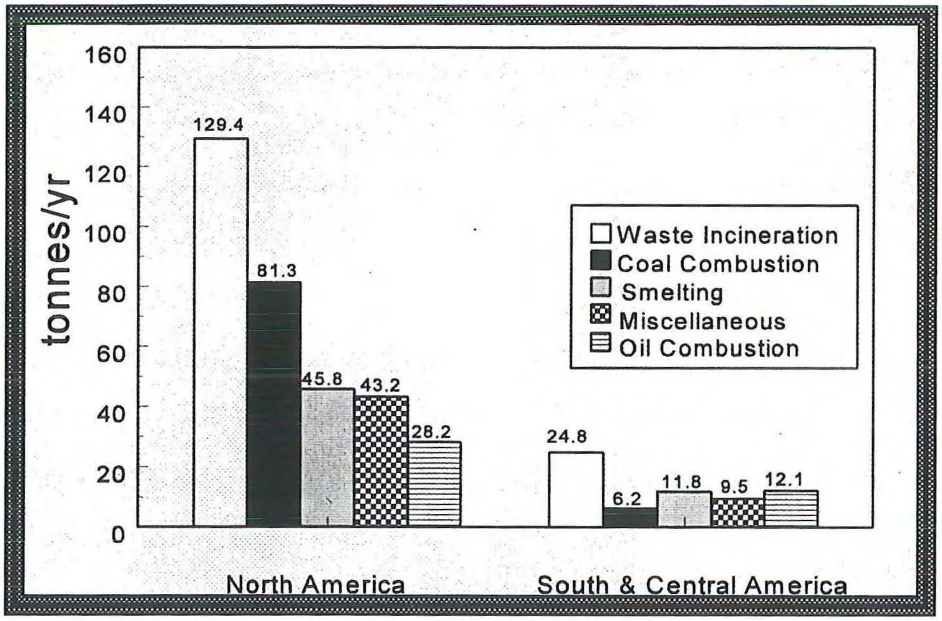

Figure 6. Annual atmospheric mercury emissions (adapted from: Pirrone et al., 1996)

The EPA also estimated in addition to combustion sources, that 13 percent of mercury emissions in the US were from manufacturing point sources, 2 percent from miscellaneous sources and the rest from area sources.

Potential sources of anthropogenic mercury emissions to the atmosphere differ for each NAFTA country. In Canada, it has traditionally been the smelting industry that emit the most mercury. Anthropogenic emissions of mercury to the atmosphere in Canada are reported to have decreased from 38.8 tonnes in 1990 to about 20 tonnes in 1995, and less than 10 tonnes in 1996 (NAMTF, 1997). However, actual data for sectors showing mercury reductions, other than for the smelting industry, are not yet available other than draft. Fourteen chlor-alkali plants still operate in the United States, 3 in Mexico and 1 in Canada. Annual consumption of refined mercury by chlorine and caustic soda manufacturers in the US is 135 tonnes, Mexico 2 tonnes and Canada 0.5 tonnes.

The reporting criteria for mercury emissions in Canada and the US do not account for smaller sources. The minimum reportable quantity of mercury for each facility "manufacturing, processing or otherwise using", is 10 tonnes and 1% of production, however; if the mercury is a by-product such as in combustion of fuel, the 1% criterion does not apply, and only greater than the 10 tonne minimum requires reporting (NAMTF, 1997). Thus, the Canada, US emission estimates are off by an unknown factor and emissions in Mexico are relatively unknown.

▸ *Mercury Deposition*

Since 1977, the US National Atmospheric Deposition Program (NADP) has been providing information about geographical patterns and temporal trends in the deposition of acidic chemicals and nutrients. Over the past 5 years, a number of activities have been undertaken within the NADP to implement a network for mercury deposition (Lindberg and Vermette, 1995). The Mercury Deposition Network has operated as a transition network over the past year and was officially recognized as an NADP sub-network in January 1996 (to be designated as NADP/MDN). Support for the network has been from individual site sponsors. The NADP/MDN is anticipated to operate for a minimum of five years, to be followed by a review and assessment of the program. The objective of the MDN is to develop a regional database on the weekly concentrations and fluxes of total mercury in precipitation to better describe the spatial and temporal trends in wet deposition of mercury. Aside from recording larger scale trends, the MDN and its participating sites will be used to address local vs regional trends, to serve as a point of comparison for smaller networks (i.e. state networks), and to augment more inclusive monitoring and research efforts (i.e. methyl mercury and/or other metals in wet deposition, sampling ambient air or watershed studies). The NADP/MDN is coordinated through NADP's coordination office at Colorado State University. Laboratory analysis and site liaison is contracted to Frontier Geosciences Incorporated in the United States. With the assistance of the coordination office, laboratory and MDN advisory committee, siting criteria, quality assurance (QA) plan, site operations plan and a training video have been developed for the network. The NADP/MDN operates on a weekly collection schedule, Tuesday-to-Tuesday.

Recent MDN mercury data show temporal, seasonal and spatial variability across the United States. The NADP/MDN currently consists of approximately 18 US sites, and 8 proposed additional ones in Canada.

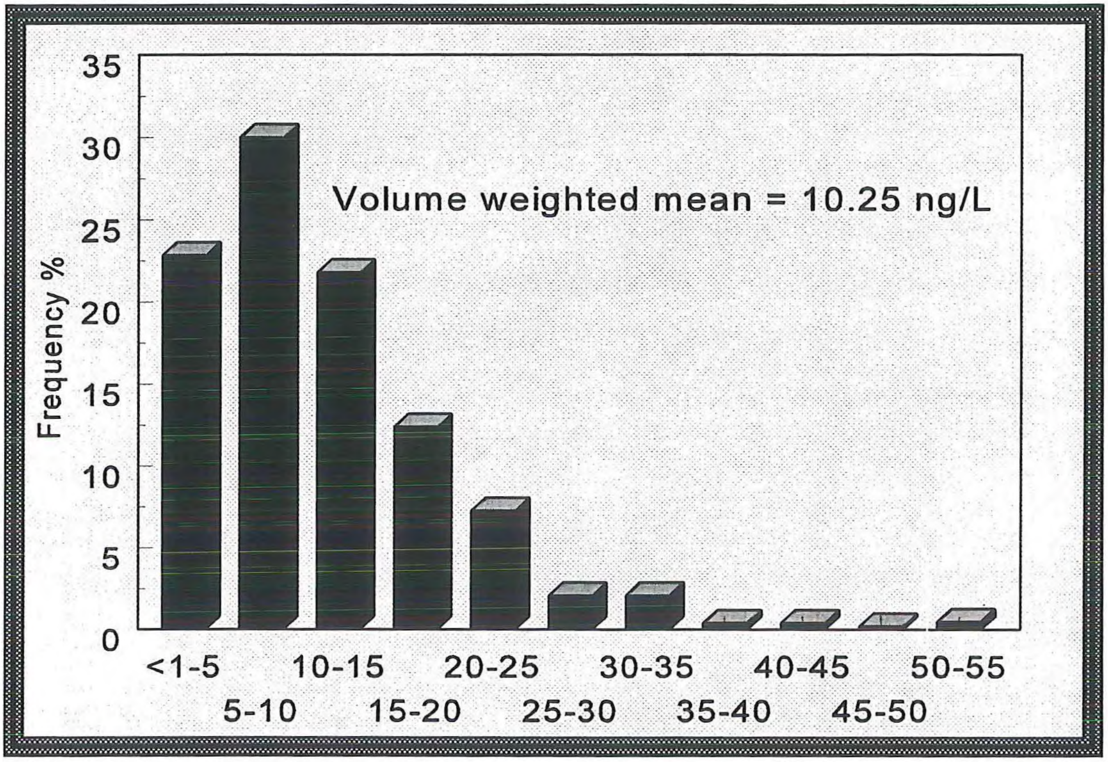

Figure 7. Frequency distribution of mercury concentrations in rainwater (NADP, MDN 1995-1996).

- **Preliminary precipitation results for the NADP/MDN transitional year for the period February 1995 to February 1996, had an average volume weighted mean total mercury concentration of 10.25 ng/L with values between 5-10 ng/L being recorded the most often (Figure 7).**

  The majority of sites in the MDN are located in the eastern region of the United States and only recently have there been sites added to eastern Canada and the northwestern United States.

- **Seasonal values were variable with highest concentrations and deposition occurring in summer (Figure 8). This temporal pattern may possibly be explained by higher ozone levels in the summer that oxidise Hg^0, to HgII. Generally, wet and dry deposition occurs after oxidation of Hg(0) to HgII; for example, Hg(0) is weakly soluble in water, and oxidation is needed to dissolve Hg in precipitation.**

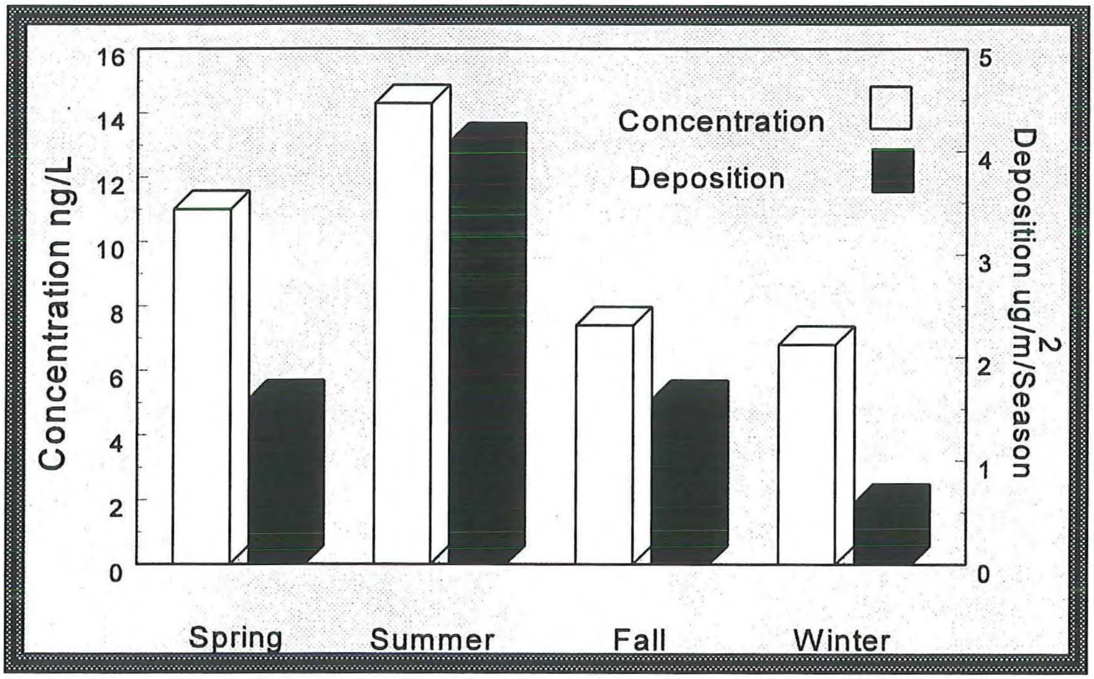

Figure 8. Seasonal variation in wet mercury deposition in the United States during the transition period of February 1995 to February 1996 (NADP, MDN, 1997).

- **Wet deposition of mercury ranged between 3.97 and 12.89 ug/m^2/yr during the NADP/MDN transition period (Figure 9). The FAMS project in Florida was conducted between 1992 and 1995 and showed mercury deposition up to 20 ug/m^2/yr (Guentzel et al, 1995). There are obvious differences in spatial patterns in deposition as well as concentrations which were 3X greater at sites in the northeast region of US and Florida.**

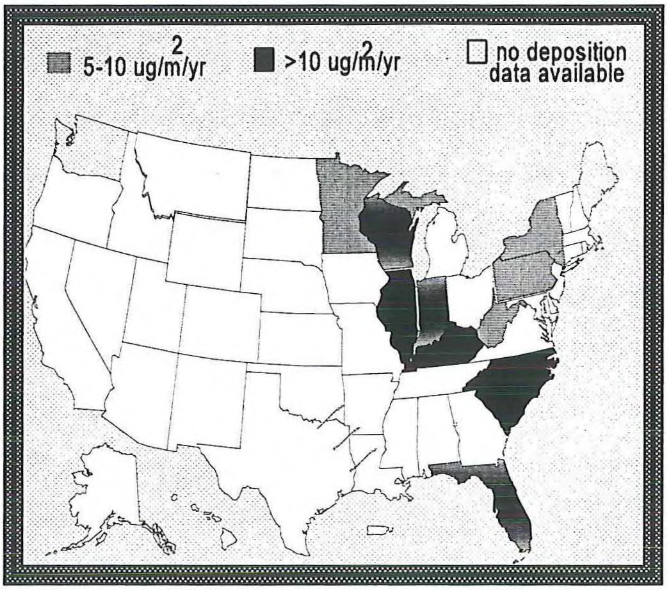

Figure 9. Mercury wet deposition at operational sites in the NADP/MDN during transition period, 1995-1996 (Vermette et al., 1996; and Guentzel et al., 1995).

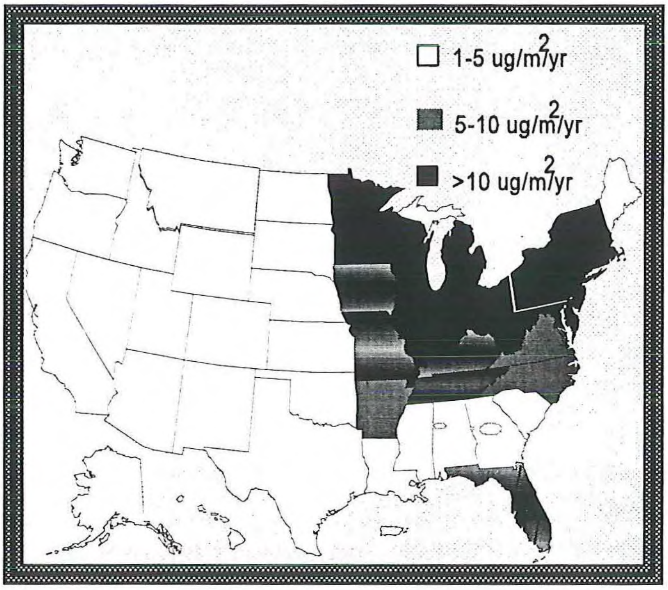

Figure 10. Model prediction for wet mercury deposition in the United States (USEPA, 1994).

▸ *Measuring Continental Air Pollutants:International Mercury Monitoring Networks*

In Mexico and Canada data are needed to define mercury atmospheric inputs and refine the present deposition and risk models. To narrow the knowledge gaps on mercury cycling, international compatible networks are needed. Fitzgerald, (1994) recognised the need for a global mercury network and proposed AMNET, a network designed to monitor elemental gas phase mercury on an international scale. While the idea of a global network for mercury received a lot of support from the international community, the global benifits were not fully appreciated and as yet AMNET has not been started. There are large differences suggested in mercury deposition between the Northern and Southern hemispheres for which the mechanism is not fully understood and only partially illustrated with individual studies (Figure 11).

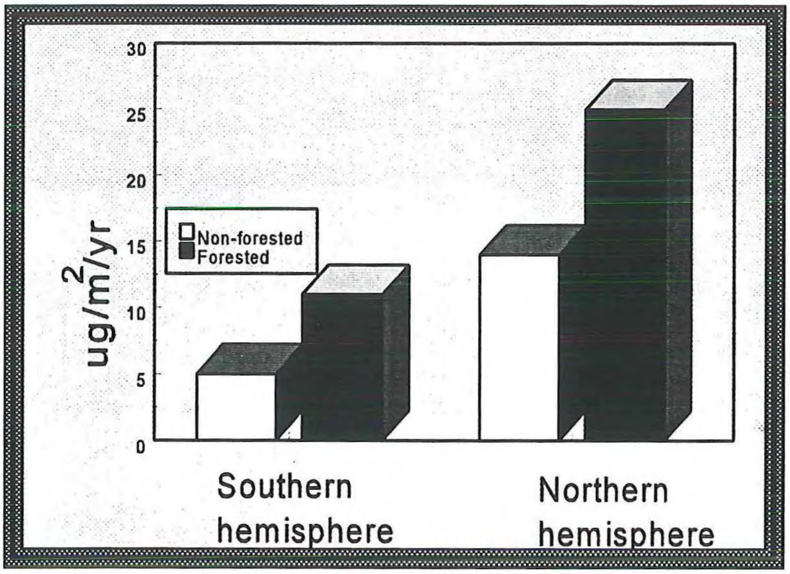

Figure 11. Projected hemispheric trends in mercury deposition (Porcella, 1996).

In 1996, a hemispheric mercury deposition network (headed by Steven Lindberg of ORNL) was proposed for wet mercury deposition within the Americas (AMT, 1996). An independent group from Mexico, Canada, the United States and Brazil worked together with the

CEC and EMAN Canada to plan the project. Although sediment data indicate an increase in past mercury deposition, present transport and deposition are relatively unknown, particularly within the Americas. This knowledge gap is a significant impediment to understanding the global mercury cycle, regional loading and what may be needed for future regional controls. Representatives from 14 agencies in conjunction with EMAN Canada and the US National Atmospheric Deposition Program/Mercury Deposition Network proposed the initial foundation stages of an "Americas Mercury Deposition Network" (AMDN), (Pilgrim et al., 1997). The objectives of the network are:

- **Training,** the AMDN will provide training and technology transfer through the development and sharing of expertise and equipment for high quality Hg wet deposition measurements;
- **Measurements**, the AMDN will provide measurements of the wet deposition of mercury for the purpose of quantifying inputs and describing seasonality in loading for representative ecosystems in the regions of each site;
- **Integration,** the AMDN will integrate wet Hg deposition measurements into ongoing research programs to add value to related biogeochemical and atmospheric research;
- **Collaboration,** the AMDN will establish a foundation for future expansion of collaborative research on the fate, transport, and effects of air toxics throughout the hemisphere, and for predicting the effectiveness of actions to reduce regional antropogenic emissions.
- **Data**, the AMDN will develop a fully quality assured and reliable data base for sites in the Americas which will be widely available to anyone interested. These data will establish a baseline of Hg wet deposition loadings that can be related to current emissions and used to predict and monitor the effectiveness of controls.

The Americas network intended to start first by, gathering preliminary data for a one year period on the spatial and temporal variability in mercury wet deposition from Brazil to Canada (Phase 1). The long-term goal is to develop funding to operate a network for at least 5 years (Phase 2) and to expand the network and number of sites and technology to other countries and to include measurements relevant to dry deposition of mercury (Phase 3). Initially, only mercury will be monitored but a potential expansion to include other hazardous and bioaccumulative pollutants (organics and metals) will be considered in Phases 2 and 3.

- **Since the Americas network was visioned, progress has been made in Canada, with 2 sites supported by the US-Canada Gulf of Maine Council, a further 4 sites are sponsored by EMAN Canada and 2 by the Quebec government, giving a total of 8 Canadian sites expected to be operating in Canada through the NADP in 1997.**
- **This is the start of the first ever hemispheric mercury wet deposition network in this part of the world.**

▸ *Northeast States for Coordinated Air Use Management Mercury Project*

This regional mercury modelling project will:

i. Update EPAÕs inventory of air emissions from major and area sources and mercury emissions in the Northeast

ii. Characterize mercury deposition in the Northeast from both long range transport and local sources

iii. Evaluate the impact on mercury deposition of various state and federal control strategies, including the co-control benefits of reducing ozone and particulate matter levels.

Northeast States Activities To Reduce Mercury Emissions

The Northeast states will establish mercury measurement and control programs, including: fish tissue monitoring, air and deposition monitoring, monitoring and control of mercury emissions from industrial sources, and recycling and resource conservation initiatives. Most states have established fish consumption advisories for pregnant women and children due to mercury levels measured in certain freshwater fish. Identifying mercury as a hazardous air contaminant under state air regulations to achieve the most stringent hazardous emission rate. Conducting an inventory of airborne sources of mercury. Implementing the recent federal mercury emission standards for municipal waste combustors and medical waste incinerators, so that emissions will be cut in half by the 2000. Forming in-state task forces to assess, evaluate, and communicate all mercury-related public health and environmental information

Northeast States Activities To Reduce Mercury Levels in the Waste Stream Include:

Encouraging industries to participate in voluntary recycling programs for fluorescent lamps; Promoting recycling by lessening the regulatory requirements for fluorescent lamps destined for recycling; Requiring the recycling of certain types of batteries; Altering the definition of hazardous waste to facilitate the recycling of batteries and fluorescent lamps; Adopting legislation prohibiting the sale of packaging materials containing mercury; Promoting recycling programs for communities to reduce the incineration of household waste; and Requiring manufacturers to inform consumers of products containing mercury.

■ PRODUCT PATHWAYS FOR MERCURY

▸ *Imports and Exports of Mercury Products*

Mercury has been reduced in commercial products within the last decade. Mercury imports into Mexico in 1994-1995 were from Germany, Spain and the United States. Mercury imports to Canada have fallen from 40-50 tonnes in 1985 to about six tonnes in 1995, whereas exports have increased (Figure 11). Research is now underway to determine if the multinational companies that manufacture products containing mercury, including those that export batteries, are Canadian or US supplied (NAMTF, 1997). In Canada, the average mercury content in fluorescent lamps has fallen from 48.2 mg in 1985 to 27.0 mg in 1995, with an industry target to further reduce the mercury content to 15.0 mg.

The mercury available for use in the U.S. comes from five main sources: primary production (mining), by-product production, secondary production (recovery), sales from excess government

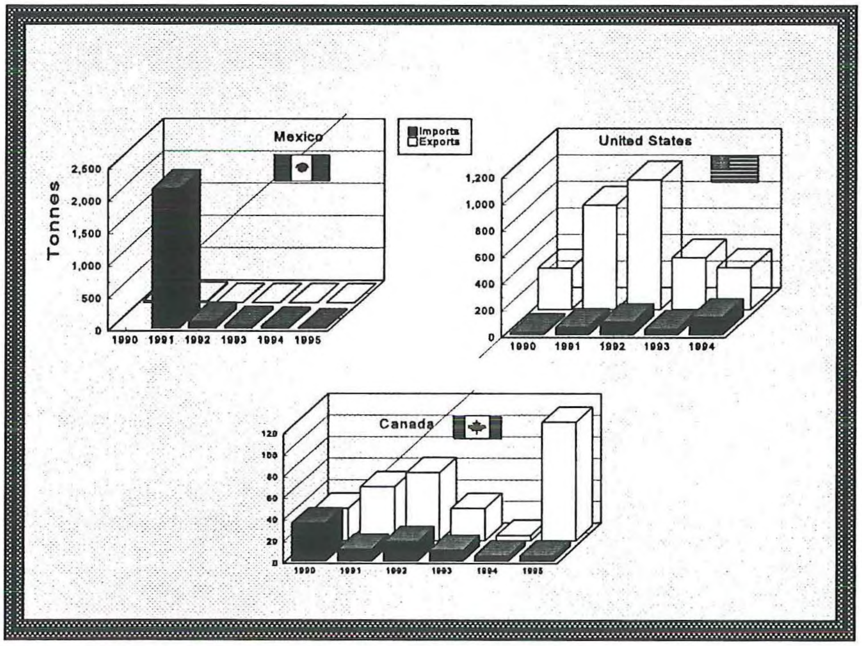

Figure 11. Mercury exports and imports in the 3 NAFTA countries 1990-1995 (NAMTF, 1997).

stocks including the Department of Energy (DOE) and Department of Defense (DOD), and imports. Most recently, there has been a sharp drop in Federal sales. In July 1994, DOD suspended future sales of mercury from their stockpile until the environmental implications of these sales are addressed. In general, the industrial manufacturers that use mercury are shifting away from mercury except for uses for which mercury is considered essential. This shift is seen primarily as a result of Federal bans on mercury additives in paints and pesticides; industry efforts to reduce the amount of mercury in batteries; increasing state regulation of mercury emissions sources and mercury in products; and state-mandated recycling programs.

▸ *Mercury Mining in México*

Mercury mines are located mainly, at the North East and Center West of the Mexican Republic. The richest mercury deposits are located in the central region of Mexico. In the pacific zone is important the production from Guerrero. (Main fields are located in the mineral resources council map). According to Salas classification, mercury can be found in large fields (Nuevo Mercurio, Zacatecas y Hizuco, Guerrero), medium fields (Piloncillos, Chihuahua, El Cuarenta y Cuencamé, Durango, Sain Alto y Mineral de Canoas, Zac. y Huahuaxtla, Gro.) and small fields (Pedernales y Batopilas, Chihuahua, San Bernardo, Durango, Atarjea, Guanajuato, San Joaquín Queretaro y Guadalcázar, San Luis Potosí.), (SECOFI, 1996).

According to National Statistics Institute (INEGI) data, mercury production started in Mexico in 1891. Maximum production of 1014 tonnes was reached in 1942 , 308 tonnes in 1991, 10 tonnes in 1994, and since 1995 no mercury has been produced. A study by the general mining coordination of the Trade ministry (SECOFI) states that mercury production is not reported after 1995, it should not be put aside.

▸ *Products Containing Mercury*

Fluorescent Lighting

Mercury is essential to the operation of a fluorescent light tube. Elemental mercury is introduced into the tube when it is manufactured, and acts as a multi-photon source, producing ultraviolet light when an electrical current is passed through the tube. Over time, the mercury in the tube reacts with phosphorus powder, which coats the inside surface of the tube, and loses its efficacy. Historically, manufacturers added mercury in quantities sufficient to ensure an adequate supply of available mercury in the tube. Recent advances in the development of fluorescent tubes has allowed manufactures to reduce the amount of mercury necessary to account for an adequate lifespan of the tube. Mercury releases from improper fluorescent light tube disposal are projected to have declined to an estimated 14.7 tons per year in 1995, and by the year 2000, releases are expected to be down to 11.6 tons as a result of recycling programs and changes in design technology. This data assumes that the mercury content in fluorescent tubes is reduced to 15 mg per four foot tube by the year 2000. In 1995, a typical fluorescent light tube contained approximately 22.8 mg of mercury.

Thermostats

Thermostats use elemental mercury as part of the actual switching component that prompts a heating, ventilating, and air conditioning system to start or stop. A mercury thermostat is constructed by adding mercury into a glass tube containing metal wire contacts , and then sealing the tube. The mercury, which is liquid, rolls between the contacts which are attached to a metal strip that is part of an electric circuit. The mercury works as a switch by completing or breaking the electric circuit when the ambient temperature varies from a set point, in turn activating or deactivating a system. Since the mercury is contained is a sealed glass bulb inside the thermostat, it is not released during normal use. Mercury-containing thermostats usually last 30 to 50 years, and their disposal usually occurs when buildings are remodelled or demolished. Mercury free thermostats are available; however, they do not last as long or work as well as mercury thermostats.

Thermometers

Mercury containing thermometers contain approximately 0.5 to 2.25 grams of elemental mercury in a small vacuum tube of uniform bore with a mercury containing chamber at one end and a temperature scale on the side. During the manufacture process, elemental mercury is sealed in the chamber, where it expands during heating. Elemental mercury has the unique characteristic of having a liquid physical state at ambient temperature ranges, making it ideal for this application. The EPA has estimated that 32.5 tons of mercury associated with fever thermometers was discarded in 1985, 16.9 tons in 1995, and 16.8 tons in the year 2000. Substitutions for mercury-containing fever thermometers began to replace mercury thermometers in 1984, and the use of mercury thermometers have continued to decline since that time.

Batteries

Electrical energy from batteries is released when an electrical circuit is completed between two electrodes, an anode and a cathode. Energy flows from the anode and returns to the battery cell through the cathode. Mercury has been used in batteries to serve either of two purposes: as a component in the zinc amalgam that forms the electrical anode of alkaline batteries, or as an additive to purify the electrodes. Advances in battery manufacturing technologies has led to recent reductions in the mercury content in batteries. In mercury oxide batteries, mercury accounts for approximately 35-40% of the weight of the battery. The mercury content of these batteries cannot be reduced without reducing the energy content of the battery. In 1996 the mercury oxide battery is manufactured in limited quantities for medical and military applications. Mercury is also used in batteries as an additive to inhibit and or suppress side reactions, and to prevent corrosion. By 1997 only button batteries and special application mercuric oxide batteries will contain mercury.

■ ECOSYSTEM PATHWAYS AND FATE OF MERCURY

▸ *Mercury Methylation in the Aquatic Environment*

Mercury emissions have an important atmospheric phase that because of it's global nature contributes to the total risk to organisms. In most circumstances, fish consumption is the main methyl mercury exposure route to humans and to fish-eating birds and mammals. Therefore, the key to understanding risk from mercury is to determine the linkage of mercury inputs to fish accumulation, a link dominated by methylation. The potential bioavailablity of metals depend on speciation, and that can be influenced by geochemical conditions, thus the accumulation in biota can vary between waterbodies. In the case of mercury, it is the organic form that is most toxic, whereas with other metals it is usually the free ion inorganic form that is most toxic. Among all natural environments, the aquatic ecosystems are characterized by the highest Hg concentrations. It is in these environments that, via microbial activity at various levels, a significant fraction of the inorganic Hg introduced is transformed into methylmercury (CH_3Hg), the most toxic and most bioavailable form of Hg for living organisms (Rudd et al., 1980; Xun et al., 1987; Korthals & Winfrey, 1987; Kerry et al., 1991). Methylmercury is readily accumulated in the organisms as a result of both its strong affinity for proteins and slow elimination rates. These biochemical characteristics peculiar to CH_3Hg result in an efficient bioaccumulation of methylmercury in the aquatic organisms and a marked biomagnification of the methylmercury concentrations from the base to the top of the aquatic food chain (Philips & Gregory, 1979; Allard & Stokes, 1989; Cabana et al., 1994, Tremblay et al., 1996). Consequently, flesh of predatory fish bears the highest methylmercury concentrations in aquatic ecosystems (Wren et al., 1983; MacCrimmon et al., 1983, Meili, 1991; Verdon et al., 1991; Cabana et al., 1994).

Except in cases where Hg point source discharges are substantial and at Hg contaminated sites, organisms are exposed to chronic, low-level Hg concentrations. Because very little Hg(0) dissolves in water, HgII comprises almost all the aqueous Hg and is present in two major forms:

inorganic HgII and monomethylHgII (MMHg). In lakes, MMHg is produced by microorganisms (largely, sulfate-reducing bacteria) in a process called methylation; enzymes in other bacteria can remove the methyl group in a process called demethylation. Recent evidence indicates that ultraviolet radiation can play an important role in demethylation (Rudd, 1995). Usually demethylators also reduce the Hg to elemental Hg(0). These reactions are illustrated by (Hudson et al., 1994, Gilmour et al., 1992, Winfrey and Rudd, 1990). The rate of production depends on several important factors such as sulfate concentrations, dissolved organic carbon (DOC), and other water quality variables (Hudson et al., 1994). Although some methyl mercury deposits from the air (e.g., Bloom and Watras, 1989; Fitzgerald et al., 1991), this fraction is probably largely of marine origin (Prestbo, E., Pers. Comm. with Porcella), net methylation in the aquatic environment is by far the main source of the Hg accumulated in biota.

▸ *Bioaccumulation of Methylmercury in Wildlife*

Fish appear to bind methyl mercury strongly, and its biological half-life in fish is on the order of 2 years (Wiener and Spry, 1996). It is through fish consumption that mercury exposure in humans and in fish-eating birds and mammals takes place. Therefore, the key to understanding risk from mercury is the linkage of mercury inputs to fish accumulation, a link dominated by methylation. This link should lead to related responses between changes in inputs and biotic mercury, mostly methyl mercury (Bloom, 1992; Watras and Bloom, 1992), concentrations. Biotic archives are rare. Newton et al. (1993) analyzed collected bird livers in Britain over a 25 year period showing a strong decrease in Hg concentration over time. This decrease coincides with the decline in industrial use described in several areas [Lindqvist et al., 1991a; Engstrom and Swain (In Press)]. Species with an aquatic-based food web (grey heron) showed a stronger decrease than the terrestrial based species (sparrowhawk and kestrel). The terrestrial food chain was likely contaminated by mercuric fungicides added to seed grain; however, the grey heron would likely obtain MMHg according to the processes outlined earlier.

A good data base exists for aquatic biota in the English-Wabigoon River system in Canada, which received a Hg point source discharge from a chlor-alkali plant (Parks and Hamilton, 1987). Sharp decreases in Hg content in two predaceous fish and a crayfish show that cessation of the point source caused a reduction in biotic Hg. However, Hg levels have not decreased below values reported in sites remote from point sources, indicating a continued supply from sediments or from global background. Other time series data for fish exist, but are of short duration. Swain E. B., (personal communication) suggests that Minnesota fish mercury varies in a manner consistent with the patterns described in Benoit et al. (1994). These results emphasize the paucity of data on biotic Hg time series and reinforce the need to establish careful time series of Hg accumulation in biological populations. Small numbers of museum samples make it difficult to obtain this time series data for biota.

There are a variety of fish eating birds and mammals that bioaccumulate mercury and may carry body burdens that are considered a hazard to their health by laboratory standards. However, some populations may have always had higher mercury concentrations than others. The spatial trends in mercury concentrations that different populations exhibit are important in understanding the mercury issue. For example, it was shown that belugas and ringed seals from the eastern arctic had lower levels of mercury than belugas and ringed seals from the western arctic and was suggested related to the geology (Wagemann, 1995). The 0.5 ppm guideline for mercury in fish is established for human protection. However, due to the amount of fish consumed per body weight, concentrations of mercury lower than 0.5 ppm in the diet of piscivorous birds have been shown to produce reproductive and behavioural affects (Scheuhammer, 1995).

- **Mercury is absorbed by fish mainly through their food. Sweden has an objective to reduce mercury in fish to 0.5 ppm by reducing atmospheric emissions of mercury. The Swedish EPA suggest that the increased mercury inputs to lakes over the past century has caused a rise in the Hg concentration in a one kilo standard size pike from the natural background levels of 0.05-0.3 ppm to current regional levels of 0.5 to 1.0 ppm (Figure 12).**

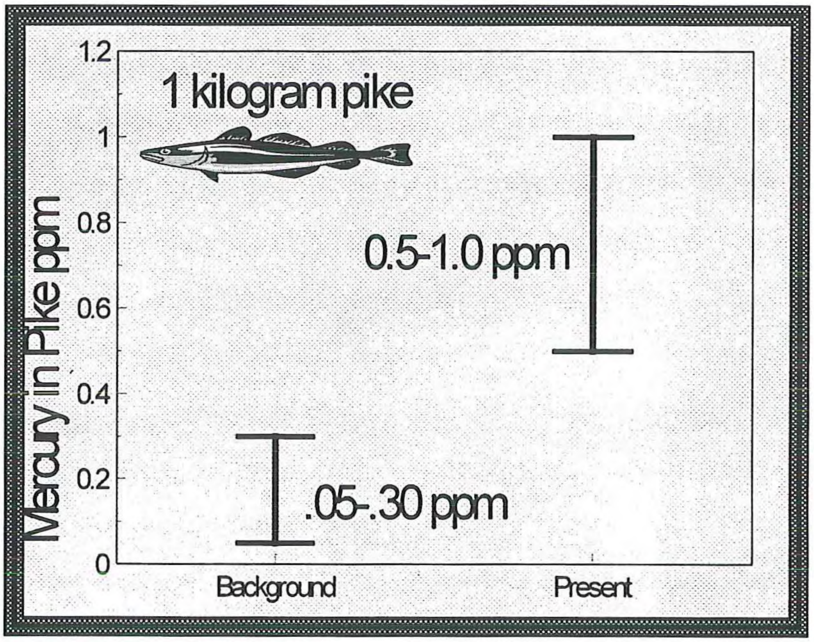

Figure 12. Estimated change in mercury concentrations in a standard 1 kilogram pike in Sweden (Swedish Environmental Protection Agency).

▸ ***Risks to Wildlife Reproduction***

Piscivorous species in regions with higher mercury deposition, geological anomalies and also those in areas that favour methylation, such as partially acidified watersheds or watersheds with large wetlands high in DOC (Rudd, pers. comm.) are more likely most at risk from mercury contamination. Species such as yellow perch and bass occupy a high trophic level and have a greater ability to accumulate mercury. Species that feed on a higher tropic level fish, such as perch, will bioccumulate the most mercury (higher BAF) and will be most at risk. For example, loons feeding on yellow perch will accumulate more mercury than mergansers feeding on salmon parr if they consume the same quantity of fish.

Body burdens of mercury can be good indicators of the health of fish eating mammals and birds. Mercury is excreted in the feathers of birds or hair of mammals thus are good indicators of the concentrations in other body organs. Monteiro and Furness, 1995 suggested seabirds to be good indicators of mercury pollution, concentrations in feathers are typically below 10-15 ppm ww, liver 10-15 ppm dw, and eggs 2 ppm dw. Eisler, 1987 suggested 5 ppm ww as a benchmark in feathers protective of birds.

Evers et. al, 1997 published the findings of the loon mercury research conducted by the Canadian and US Wildlife agencies (Figure 13).

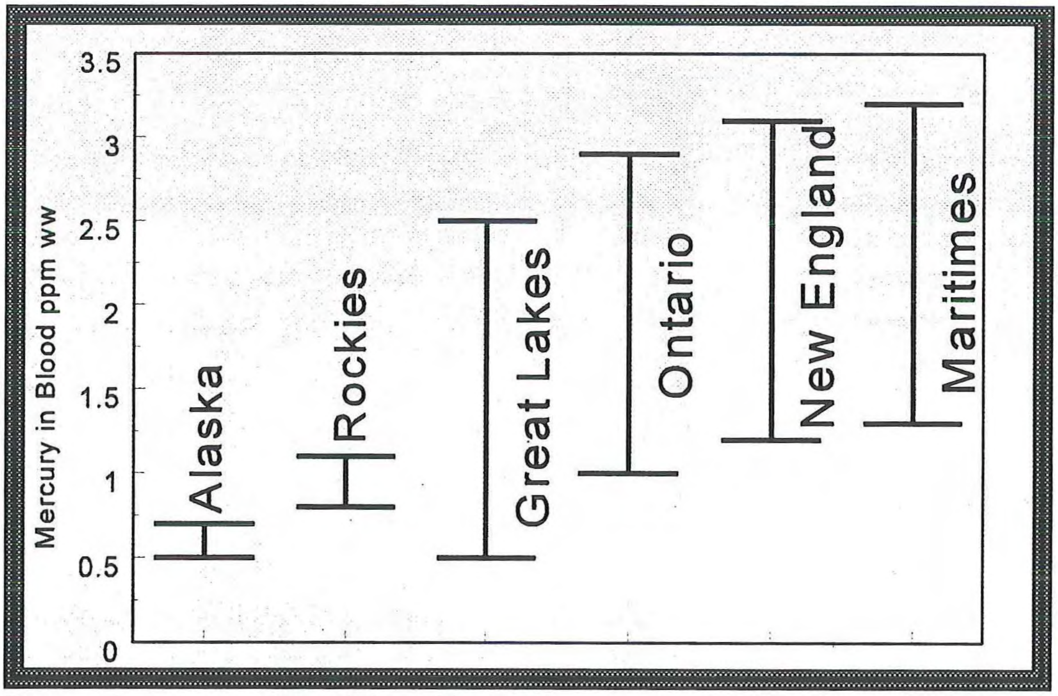

Figure 13. Mercury in adult loon blood (Evers et al., 1997).

A significant increase in mercury in the blood of loons was observed from west to east. This correlates well with higher deposition and higher mercury in the sediments of the northeast United States. In a recent study in Atlantic Canada, mercury concentrations in loons were found to be the highest yet observed in North America (Burgess, 1996). Wider scale studies and effects on population productivity now need to be determined.

Between 1984-1990, 221 Common Loons were collected in Minnesota and examined for mercury contamination (Ensor et al., 1992). Of the 221 examined in that study, 128 were found dead or dying and 93 were live captured. Mercury concentrations in the feathers ranged between 0.08-83 ppm ww with a geometric mean of 3.6 ppm ww. Twenty two percent had mercury concentrations in the liver above 13 ppm ww which is considered a benchmark associated with impaired reproduction. The mean mercury concentration in feathers was 8.67 for 41 adults examined with a high of 29 ppm, and 2.66 ppm for 91 juveniles examined. Meyer et al. (1995) examined the mercury content of blood and feathers from 35 adult loons in Wisconsin. The study showed that loons from lakes with low buffering capacity had higher blood levels of mercury.

HUMAN HEALTH

▸ *Exposure Pathways*

Currently in health research, exposure assessment has became the key element of analysis, and for environmental exposures with a long term effect, retrospective exposure assessment is now developing (Correa et al., 1994). Research has demonstrated the need to consider all the routes of human exposure (Ott, 1985; Smith, 1988). This is most important if we want to understand the different ways long range transport of pollutants can be related to human and biota health effects. The basic conceptual model is simple (Figure 14). Exposure factors and patterns do vary by culture, activity and region. National approaches have been developed (EPA, Exposure factors handbook, 1989), although should be carefully adapted. Environmental, nutritional and genetic factors are related to birth defects (Cordero, 1994), and birth defects are being detected through active and passive surveillance systems in the region (Santos-Burgoa et al., Epidemiology, 1996) in areas with high industrial and water pollution; not yet causally related, these are clues for research.

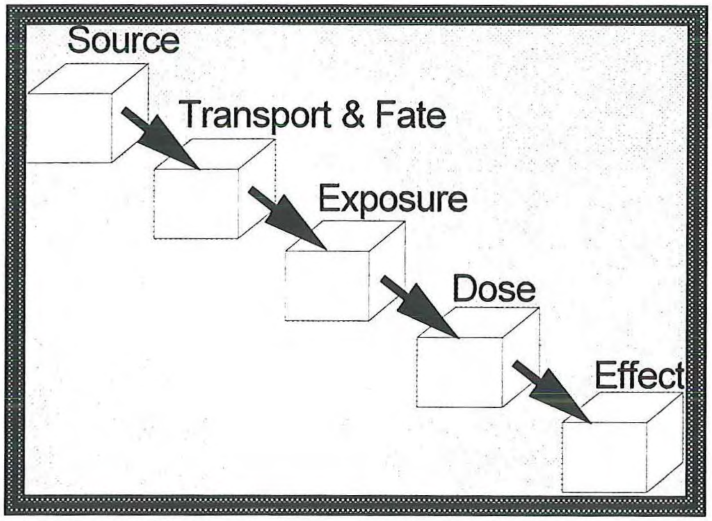

Figure 14. Conceptual exposure-risk model.

However other more basic reproductive effects of mercury should be considered, such as infertility, as has been documented in heavily exposed populations (Rchootin and Olsen, 1983), with increased risk for infertility associated to mercury exposure of 30% in males and 70% in women. More subtle research has pointed out a delayed time to pregnancy from mercury exposure (Baird, et al., 1986).

▸ ***Symptoms of Methylmercury Poisoning*** (Clarkson, 1991; Fitzgerald and Clarkson, 1991)

¥ Signs and symptoms of mercury toxicity in adults range from paraesthesia through ataxia, deafness, and death depending on the degree of exposure;

¥ Selective focal damage occurs in the brain when adults are exposed, but diffuse damage is found in children and the fetus as the formation of the brain is widely damaged;

¥ Signs and symptoms are more severe in children and occur at exposures 10-fold lower than adults. They include motor retardation and severe damage to brain function. Blood levels of 0.04 to 0.08 µg/l in mothers of children produce these effects;

¥ Binding of the thiol groups interferes with cell division and formation of nerves such that partially formed neurons are found in the brain of children. Specifically, depolymerization of the microtubles rich in thiols that are necessary for forming the microstructure in the brain occurs;

¥ The damage to the brain appears to be irreversible; however, administration of compounds that are high in thiols may reduce the amounts of methylmercury that is absorbed.

Three characteristics of the mercury poisoning syndrome make it difficult to link chronic mercury exposure in the general population with neurological effects found in individual patients. These are: A silent period before the appearance of the disease of weeks to several months follows ingestion as the mercury is transformed from the methylated to the ionic form and bioaccumulated in the brain; difficulty of associating neurological damage diagnosed in a single patient and mercury exposures several months before; and scanty information of mercury levels in fish in domestic water involved in sports fishing and general consumption rates of fresh water and marine fish.

▸ *Developmental Effects*

Methylmercury is a potent toxin that causes impairment of the central nervous system (CNS) and developmental toxicity in humans (USEPA, 1996; WHO, 1990; Clarkson, 1991). Methylmercury is also a fetotoxin, which affects embryonic development and causes fetal malformations, as shown in animals. Although the placenta may represent a certain barrier to Hg, transfer of alkyl Hg is possible, being more important than that of aryl or inorganic Hg; certainly it has been associated to teratologic abnormalities in newborn infants. The significance of this for the North American Region is still to be considered. The primary route of exposure for humans and wildlife to methylmercury is consumption of contaminated fish.

Although the toxic effects of mercury have been known for over a century, two poisoning epidemics link in utero exposure to CH_3Hg in food to neurological disease in humans and animals. One of the earliest recorded poisoning was from consumption of fish and shellfish contaminated with CH_3Hg in Minamata Bay and later in Niigata, Japan. In a review of the extensive body of neurotoxicologic data collected after the Minamata outbreak, Watanabe and Satoh (1996) determined that CH_3Hg has acquired the unique status among hazardous chemicals in our environment in that (a) existence of developmental neurotoxicity is apparent in both humans and animals; (b) a relatively large body of neurologic and behavioural data is available in both humans and animals compared to most other chemicals and the ongoing large-scale epidemiologic studies [in the Seychelles and Faroe Islands] are evaluating behavioural functions in more specific ways than routine neurologic and psychological tests. The authors concluded that because of the extensive information available on CH_3Hg it can be used as a model for developing predictive tests of human risk from in utero exposure to other chemicals of unknown neurotoxicity.

Limitations of the Iraqi study (small number of affected mother-child pairs, difficulty in determining exact age of the children, lack of data on the background frequency of abnormal responses in the Iraqi population, and possible overestimation of maternal CH_3Hg dose during critical periods of prenatal development) have instigated additional investigations of the effects of low-level CH_3Hg exposure on prenatal neurodevelopment in the Seychelles and Faroe Islands. The goal of the Seychelles Island study is to establish whether there is a relationship between fetal exposure to CH_3Hg from dietary fish, as measured by maternal hair levels during pregnancy and measures of development in the offspring during early childhood (at 6.5, 29 and 66 months of age) (Marsh et al., 1995). Preliminary results of the Seychelles Island study of infants up to the age of 29 months suggest no traceable effects except in one test indicating a decrease in the activity level only in boys. Since the results of the tests at 66 months have not been analyzed, the investigators caution full interpretation. It is still unclear if an association between fetal mercury exposure at these levels and neurodevelopment exist (Myers et al., 1995, page 714). This is particularly important if the subtle effects of fetal mercury exposure may be undetectable when the children are young and appear later in life. In contrast, findings from the Faroe Island studies suggest there may possibly be inter-generational risks from exposure to low levels of CH_3Hg. The detailed findings of these two are not expected to be available for two or more years.

▸ *Chronic Exposure and Effects in Adults*

In the Brazilian Amazon, a significant proportion of the Hg present in the aquatic environments has been attributed to goldmining operations (Veiga, 1990; Malm et al, 1995) and to weathering of soils after slash and burn practices (Roulet et al., 1997). An estimated five million people who regularly consume fish are exposed to methylmercury contamination in Brazil, representing a major concern since exposure probably started 20 to 40 years ago. Even if exposure levels rarely exceed the WHO recommended limit of 50 ug/g (a limit deemed adequate for the prevention of neurological effects in adults), recent findings indicate deleterious effects on the nervous system of exposed populations (Lebel et al., 1996a, Neurotoxicology 17 (1); 1996b

(Hamburg or New Orleans abstracts; 1997 (WASP in press). Seeking to examine neurofunctional and clinical manifestations of nervous system dysfunction in relation to hair Hg concentrations at levels below 50 ug/g, the team of Lebel et al (1996a, b) conducted a battery of tests sensitive to visual and motor functions (contrast sensitivity, grip strength, fatigue, manual dexterity).

Although the hair concentrations for the population studied were relatively low (median around 12 ug/g), the researchers found association between diminished neurophysiological functions and increasing hair Hg levels. The significance of these findings lies in the fact that other populations, such as the aboriginal people of northern Québec or others relying on fish as an important nutritional source (particularly fishermen), may be affected in the same manner. While previous studies of the Cree, instigated by elevated methylmercury levels in fish following the construction of hydroelectric reservoirs and conducted using standard clinical criteria, yielded inconclusive results (Wheatly, 1979; MacKeown-Eyssen and Reudy, 1983; Spitzer, 1988) the documented hair Hg concentrations of some portions of this population are comparable to those of the Brazilian investigation (MacKeown-Eyssen and Reudy, 1983). At present, hair Hg levels deemed acceptable by the Cree Board of Health and Social Services of James Bay have been set at 0 - 30 ug/g (James Bay Mercury Committee 1993). In view of the above, and in considering that the James Bay region, prior to flooding had very low levels of soil Hg as compared to the Amazon soils which have relatively high background concentrations (Hg burdens of 10 000 to 30 000 ug/m^2 for the upper 20 cm have been reported by Roulet et al. 1997), it is not inconceivable that other populations living in seemingly low risk regions are in fact suffering the effects of low level (yet long term) intoxication.

▸ *Reference Dose*

Risk is not only based on the benchmark mercury concentration in fish (0.5 ppm) but also the amount of fish eaten. In 1989, the US EPA had a daily RfD of 0.3 ug/kgBW/day and dose was tightened to 0.1 ug/kgBW/day in 1995. The RfD is an estimate of the lowest daily intake that will not result in any health effects. Because this revised USEPA daily RfD was thought by some toxicologists to be in conflict with the findings of the Seychelles Island study, it was requested that it be reviewed in light of this. A decision is suppose to be made by 1998. Because of the widespread contamination of fish and potential for adverse health effects from methylmercury exposure, especially in pregnant women and children, several risk management activities to limit fish consumption have been undertaken by state and local agencies, the US Food and Drug Administration (FDA), US EPA, and WHO. Since the Iraqi study represents the best data currently available to quantify developmental neurotoxicity from methylmercury exposure, public health agencies have utilized these data to estimate corresponding acceptable daily exposure levels

In the US, consumption advisories have been issued in 37 states due to methylmercury contamination of freshwater fish. In a national assessment of risks associated with consumption of freshwater fish, the USEPA used a hypothetical local exposure analysis of mercury emitted from a source located near a water body. From this modelling scenario, it was determined that subsistence fisherman and their children receive the highest levels of mercury, and certain wildlife species exceed protective wildlife criterion. Comparison of existing methylmercury consumption guidelines in the U.S to dietary survey data showed that the acceptable daily intake is exceeded by women with above average fish consumption rates. For example, using an average concentration of methylmercury in fish of 0.134 ppm, a 100 gram serving (WHO recommended limit for sensitive populations) would contain 13.4 μg mercury per serving. The quantities of fish and shellfish consumed by women of child-bearing age based on the dietary survey at the 95th, 97th, and 99th percentile are 111, 133 and 175 grams per day, respectively. Estimates of methylmercury exposure in 62 kg women consuming fish at the 95, 97 and 99 percentile are shown in (Table 2).

Adverse effects due to methylmercury exposure from fish consumption in North America is not likely to be detected in the exposed general population due to limitations in study size and absence of clinical assessment of developmental neurotoxic effects. However, the biogeochemical cycling of mercury, the pathways of exposure and the toxicity of methylmercury all justify the public health concerns regarding exposure to methylmercury, especially in sensitive populations eating fish regularly. It is suggested that reduction in long-range transport of mercury and associated atmospheric deposition of mercury could reduce the public health risks associated with increasing levels of methylmercury in fish.

Table 2. Percentile consumption by women of child-bearing age (g/day fish) methylmercury exposure assuming average 0.134 ppm in fish (ug/day) corresponding daily intake (µg/kg/day) for a 62 kg women margin of safety compared to USEPA RfD of 0.1 ug/kg/day.

95th Percentile	97th	99th
111 (g of fish eaten)	133	175
14.8	17.8	23.5
0.24	0.29	0.38
2.4	2.9	3.8

The current EPA RfD is based on a study of Iraqi infants that had been exposed in utero to methylmercury-treated seed grain that was mistakenly used in home-baked bread. Among the signs noted in the infants exposed during fetal development were cerebral palsy, altered muscle tone and deep tendon reflexes as well as delayed developmental milestones, i.e., walking by 18 months and taking by 24 months. A benchmark dose of 11 ppm in hair; equivalent to maternal blood levels of 47 microgram/L and body burdens of 69 ug or daily intake of 1.1 ug/kg-day was established based on the Iraqi data using a ratio of 250:1 was used to correlate hair concentrations to concurrent blood concentrations in the derivation of the critical effect. An uncertainty factor of 10 is applied to account for variability in human population, in particular the wide variation in biological half-life of methylmercury and the variation that occurs in the hair to blood ratio and for lack of a two-generation reproductive study and lack of data for possible chronic manifestations of the adult paraesthesia that was observed during gestation.

State fish consumption advisories are often issued to address local conditions and are generally based on the Food and Drug Administrations (FDA) action level for commercial fishing of 1 ppm. The current FDA action level initially proposed as a guideline for fish and shellfish sold in interstate commerce in response to the Japanese poisoning epidemics was changed to specifically address methylmercury in 1984. The FDA established a TDI based on a weekly tolerance of 0.3 mg of total mercury per person, of which no more than 0.2 mg should be present as methylmercury. The tolerable level for methylmercury would correspond to 230 ug/week for a 70 kg person or 33 ug/kg/day. In recognition of the developmental effects, FDA also warns pregnant women and women of child-bearing age to limit their consumption of fish known to have high levels of mercury.

In 1990, based on the Iraqi outbreak, the IPSC of the World Health Organization determined that a daily intake of 3-7 ug/kg of methylmercury would cause adverse effects of the nervous system, manifested as a 5 percent increases in the incidence of paraesthesia. In addition, they concluded that developmental effects characterized by motor retardation or signs of CNS toxicity could be detected as increases over background incidence at maternal hair level of 10-20 ppm mercury.

The Seychelles and Faroe Island health studies are major initiatives evaluating risks to children whose parents have been exposed to low concentrations of mercury in their diet over their life-times. Findings from the Faroe Island studies (which are continuing) suggest there may be inter-generational risks from exposure to low levels of methyl mercury, whereas the Seychelles Islands suggest there are no traceable effects.

▸ *Risk Groups*

Since current environmental health policies for fish consumption, the health risk during pregnancy requires consideration of (a) an acceptable intake of fish containing methylmercury, (b) the fish consumption patterns in women of child-bearing age, and the levels of methylmercury in the fish consumed by this population. Ongoing investigations evaluating risk to children exposed prenatally to methylmercury are attempting to identify the threshold at which developmental neurotoxicity are indistinguishable from background levels. This information will be useful in reducing public health risks only when combined with fish consumption patterns, residual levels of methylmercury in commonly consumed fish, and use of biomarkers to determine existing methylmercury body burdens. In contrast, although there is evidence that commonly consumed freshwater fish and marine fish bioaccumulate mercury, there are few cases of human poisoning.

Because methylmercury may produce more subtle neurodevelopmental effects such as impairment of sensory or cognitive systems (e.g., delayed development in children, memory loss, loss of IQ points) at low levels of exposure through fish consumption, there are potentially a significant population in North America at increased risk. In particular, risks to women of child-bearing age whose exposure during critical periods of prenatal development (after the seventh-week of gestation) exceeds the effect level or threshold. Since the central nervous system continues to develop up to age 14, infants who are breast fed and children up to the age 14 are also at risk because they have higher exposures to methylmercury on a per kilogram body weight basis compared with adults (USEPA, 1996). Based on measured mercury levels in marine fish combined with demographic data and dietary survey data, the USEPA also conducted a risk assessment based on eating marine fish only. This assessment found that (a) that 30% of the general population consume fish at least once during a three-day period and (b) 50% of women of child-bearing age are being exposed to methylmercury at levels at or above the reference dose (see Table 2) from consuming marine fish; of this population it is predicted that a large fraction of pregnant women are consuming fish with mercury levels at or above the reference dose.

■ POLICY AND SCIENCE INTERFACE

▸ *Uncertainties and the Weight of Scientific Evidence*

While numerous environmental pollutants (e.g., lead, dioxin, PCBs) have been the subject of considerable public concern and subsequent legislation, Hg has not, as yet, gained such publicity. Although, mercury, as outlined in the preceding sections, represents a cause for concern due to its particular nature (i.e., persistence and bioavailability), some uncertainties with respect to its biogeochemical cycling and potential chronic effects on humans and ecosystems have not in the past permitted a smooth interface between science and policy decisions on this issue.

- ♦ The first uncertainty, and perhaps the most important is "data reliability", much of the older mercury data are inaccurate due to analytical difficulties and sample contamination. Unlike lead which exists at levels in the ppm range (making it easily measurable by atomic absorption techniques), Hg concentrations in the atmosphere and water tend to lie in the ppt (parts per trillion) to low ppb range. It has only been in the past 5 - 10 years that technical advances have permitted researchers to produce reliable mercury results for the majority of environmental compartments of concern.

- ♦ The second uncertainty is the "geographic extent of anthropogenic influences and contributions related to natural sources".

- ♦ A third uncertainty is the "transfer of Hg to the aquatic food chain". The processes controlling the bioavailability of Hg are not fully understood, whereby some lakes receiving the same deposition have higher methylmercury in the biota than others.

- ♦ A fourth uncertainty is "the effect of low level exposure to methylmercury", Hg environmental policy is based on the fact that deleterious health effects related to methylmercury tend to be viewed solely in terms of clinical signs of intoxication. The critical uncertainty is identifying the level at which sensitive neurobehavorial consequences of prenatal exposure to methylmercury occur, and the extent to which the public, especially sensitive populations (e.g. pregnant women), are exposed to methylmercury from fish consumption. Although, it has been generally accepted that low environmental concentrations are not a cause for alarm, since they do not yield serious health problems like those experienced in the Minimata and Iraqi exposures. Studies have shown that low level exposure to methylmercury can indeed result in chronic health effects. Because fish consumption typically occurs throughout a lifetime, chronic effects from long term low level exposure to methylmercury need to be carefully evaluated.

- **Anthropogenic emissions of mercury are very significant to elevated levels observed in the environment and should not be masked under the umbrella of natural emissions.**

JPAC Hearings

In July 1996, the Environment Ministers of the NAFTA countries through the JPAC public hearings in Toronto, Canada, were briefed on the need to further develop international partnerships in the study of mercury (JPAC, 1996). There is now an excellent opportunity to collaborate and further develop the recommendations from these public hearing and workshops. Although, progress has been made on paper under the Resolution for the Sound Management of Chemicals and through the CEC Mercury Task Force on the status of mercury in each country, there is yet to develop, a science based mercury program shared between the NAFTA countries. At present there is a lack of interface between mercury science and policy decisions.

- ### *Sound Management of Chemicals*

One area of cooperation is the Sound Management of Chemicals initiative of the Parties, established in the fall of 1995 under Resolution #95-5 of the Council for Environmental Cooperation. In accordance with the directives of the resolution, the three countries formed a Working Group, which identified mercury as a priority persistent and toxic substance for which it would develop a fast-track action plan. The Working Group, in turn, designated a North American Task Force on Mercury to assist it with preparation of the North American Regional Action Plan (NARAP) on Mercury. However, the Expert Panel suggests that the present priority given to mercury as compared to other pollutants under the Sound Management of Chemicals needs to be drastically strengthened from what was presented in Mexico City in 1996 by the North American Task Force. The listed recommendations will help to further develop a strategy for mercury within the NAFTA countries.

- ### *International Cooperation on Mercury*

There is an excellent opportunity for the Commission for Environmental Cooperation to form linkages with other international initiatives on Long Range Transport of Atmospheric Pollutants and Risk need to be strengthened. At present this international cooperation is partially fulfilled by the Chair of the CEC Working Group, an opportunity exists to further develop an international committee the NAFTA countries and other global mercury initiatives lead by the organizations like the UNECE, the OECD, the WHO, EPRI, PAO, and experts that developed the US EPA Report to Congress. The mercury group recognises the recommendations made by the Ecological Monitoring and Assessment Network of Canada to the NAFTA Environment Ministers to build ecosystem based science initiatives.

- **To reduce risk to humans and ecosystem health, Canada, Mexico and the United States need to develop cooperative programs on a variety of environmental issues. However, mercury because of the evidence of dramatic increases in the global environment, and considering it's very volatile and toxic nature, should be given higher priority by the CEC.**

■ RECOMMENDATIONS

⇨ To enhance cooperation between Canada, the United States and Mexico in the study of mercury, it is suggested that:

● ***Atmospheric Measurements***

- Because mercury is a global pollutant larger scaled networks are needed than what presently exist. Therefore, the Commission for Environmental Cooperation should develop a mechanism to facilitate mercury monitoring at an international scale;
- A tri-national compatible deposition and gas phase mercury network is recommended. The development of this network can provide a number of science opportunities, and add value by appropriate training and technology transfer, compatible measurements, research integration, data base development and collaboration;
- Analyze the chemical transformation of mercury in the atmosphere and parameters that influence it's transport and deposition.

Emission Inventories

- Mercury emission estimates for Mexico need to be developed;
- Smaller sources in Canada and the US need be accounted for in emission inventories;
- Mercury atmospheric speciation and radio-isotope tracing are essential to developing future controls on the appropriate sources.

- ***Historic Sediment Profile Records***
  - Evaluate the spatial historic deposition patterns of mercury in Mexico through recontruction of Hg deposition by appropriate lake sediment and peat coreprofiles, and dendrochronology.

- ***Holistic Ecosystem Approach***
  - Utilize the existing national ecological programs in Mexico (NIE), Canada (EMAN) and the United States (EMAP) to implement ecosystem based mercury monitoring and to fulfil the commitments of the 1995 Tri-National Ecological Workshop.

  Biological and Chemical Measurements

  - Wildlife Data, further ecosystem based studies on chronic effects on the reproduction of fish-eating birds and animals is needed for all 3 countries.
  - Fish Data, regional data for mercury in fish are needed for remote freshwater lakes in Mexico.
  - Determine factors affecting methylation in different lakes.

- ***Human Health***

- Human Health, the health advisories are sufficient for Canada and the United States but should be determined if they are needed in Mexico.

- Neurobehavioral effects of mercury should be considered, more so in regions where there is a high concentration of human populations in developing ages. Such is the case of Mexico, where 49% of the population are in the reproductive age (15 to 44 years of age) and 37% are children under 15 years of age. (Secretaría de Salud, Estadísticas Vitales, 1991);

- The definition of effects at very low levels of mercury;

- The identification of population based reproductive effects form mercury concentrations, emphasizing teratogenic, and fertility;

- The identification of developmental effects;

- The ratio of Long Range Transport through Continental Pollutant Pathways (CPP) mercury and the total human exposure. This ratio could or should be expressed as a correlation coefficient, adjustable for regional characteristics;

- The different regional patterns of human and biological exposures; in the case of human population, emphasize the different inhalation and ingestion routes, including but not only, the consumption of fish contaminated with mercury;

- Estimate the bias of effects of individual exposures from CPP while assuming a nonlinear disease - exposure dynamic in populations generally exposed to low levels of concentrations attributable to CPP.

- ***Technology Transfer***

  ▸ In order to implement these recommendations, the mercury group encourages the transfer of standard analytical procedures, methodologies and Hg equipment between the 3 countries.

- ***Transcontinental Atmospheric Pollution Mandates***

  ▸ Those organizations with an international mandate to cooperate on environmental issues need to form a committee to address the global issue of mercury pollution, the CEC should take a lead role in setting-up such a committee.

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Case Study

Persistent Organic Pollutants: General Characteristics and Continental Pathways in North America

Authors:

Terry F. Bidleman, Atmospheric Environment Service, Downsview, Canada
Derek C.G. Muir, Freshwater Institute, Winnipeg, Canada
Frank Wania, WECC Wania Environmental Chemists Corp., Toronto, Canada

Reviewed by:
Don Mackay, Trent University, Peterborough, Canada

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1.0 DEFINITION, EXAMPLES, AND GENERAL CHARACTERISTICS OF POPS

Persistent Organic Pollutants (POPs) are organic compounds of natural or anthropogenic origin which are degraded in the environment very slowly or not at all. They include deliberately produced and emitted chemicals such as organochlorine pesticides (e.g. DDT, chlordane, toxaphene, hexachlorocyclohexanes, and endosulfan), industrial chemicals with a variety of applications (e.g. polychlorinated biphenyls, naphthalenes, and diphenylethers), as well as undesirable side-products of combustion (e.g. PAHs) and bleaching processes (e.g. chlorinated phenols). The latter include the polychlorinated dibenzo-*p*-dioxins and dibenzofurans (PCDD/DFs), which are described in detail in a separate case study. Although POPs comprise a very diverse group of chemicals, they have a number of generic characteristics:

Persistence: POPs resist photolytic, chemical and biological degradation. Persistence is often very media-specific. Whereas degradation in the atmosphere can occur in the range of weeks or even shorter (e.g. Kwok et al. 1995, Anderson and Hites, 1996), degradation of some POPs in soil or sediments may occur on the time scale of decades or be entirely non-existent (Kjeller and Rappe 1995, Wong et al. 1995, McLachlan et al. 1996, Howdeshell and Hites 1996).

High lipid solubility: POPs tend to be very apolar compounds with low water solubility and high affinity to lipids. They thus have a tendency to partition into soils, sediments, and fatty tissues of living organisms. Some POPs biomagnify in biological systems, i.e. their concentrations increase with trophic level within a food chain.

Semi-volatility: POPs have an intermediate volatility which sets them apart from both volatile organic compounds (VOCs, CFCs) and involatile organic chemicals such as polymers and humic substances. They can be present in the atmosphere both in gaseous form and associated with particles. They also can re-evaporate after being deposited to the Earth's surface, and are thus able to cycle repeatedly between atmosphere and surface.

Toxicity: The majority of POPs are toxic substances. A wide range of effects are seen after exposure to POPs. These include, among other things, effects on reproduction, development, cytochrome P450-dependent enzymes, porphyrins, the immune system, the adrenals, the thyroid gland and thyroid hormone levels and vitamin A levels. Many POPs also cause visible changes in the liver, including hypertrophy, lesions and in some cases, tumors.

2.0 MULTIMEDIA PARTITIONING AND PHYSICAL CHEMICAL PROPERTIES

Because of their particular physical-chemical characterisitics, POPs are not primarily atmospheric contaminants, but can occur in virtually all environmental compartments, dynamically exchanging among them (Mackay 1991). Transport and accumulation of POPs in air, water, soil, plants and animals is governed by, and can be described with the help of, a small number of cardinal physical-chemical properties and partition coefficients.

Vapour Pressure and Water Solubility

Vapour pressure (Pa) and water solubility (mol/m^3) are parameters describing the maximum, or saturation, concentrations of a specific compound in air and water. Vapour pressure is frequently used to describe the condensation of POPs onto atmospheric particles in ambient air (Bidleman 1988, Pankow and Bidleman 1992). Vapour pressures and water solubilities for solid compounds (P_S, S_S) and those that are liquids at ambient temperature are experimentally accessible (OECD 1981), whereas for compounds that melt above ambient temperature, properties of the hypothetical subcooled liquid (P_L, S_L) can be estimated (Hinckley et al. 1990). The properties of the liquid-phase compound, not the solid, is the proper descriptor of environmental behaviour.

Henry's Law Constant

The Henry's law constant (H, Pa m^3/mol) or its dimensionless version (H/RT) expresses the equilibrium partitioning of a compound between air and water. Applications include scavenging of vapour-phase POPs by rain and fog droplets (Bidleman 1988), air-lake and air-sea gas exchange (Bidleman and McConnell 1996) and gas exchange with moist soil (Jury et al. 1983, 1994a,b). Henry's law constants are estimated by the ratio of P_S/S_S (or P_L/S_L), or are measured directly (e.g. Alaee et al. 1996, Kucklick et al. 1991, ten Hulscher et al. 1992, Fendinger et al. 1989).

Octanol/Water Partition Coefficient

Hydrophobic compounds sorb primarily to the organic matter in soils, sediments and particles in water, and partition into the lipids of organisms. N-octanol is widely employed as a generic surrogate for a variety of organic and fatty materials in the environment and in organisms, and the octanol/water partition coefficient K_{OW} is used to describe sorption to organic matter and bioconcentration from water into the lipids of organisms. Bioconcentration factors (BCF = concentration in lipid/concentration in water) and partition coefficient between soil or sediment organic carbon and water (K_{OC}) are directly correlated to K_{OW} (Eisenreich 1987, Elzerman and Coates 1987).

Octanol/Air Partition Coefficient

Recently the octanol/air partition coefficient (K_{OA}) has been applied to partitioning of POPs between air and lipid phases, such as vegetation (Muir et al. 1993, Simonich and Hites 1995, Tremolada et al. 1996) and organic films on aerosols (Finizio et al. 1996). On a log-log scale, K_{OA} is negatively correlated with P_L (Harner and Bidleman, 1996). Values of K_{OA} are measured by generator column techniques (Harner and Bidleman 1996, Harner and Mackay 1995) or estimated from the ratio $K_{OW}RT/H$ (Harner and Bidleman 1996).

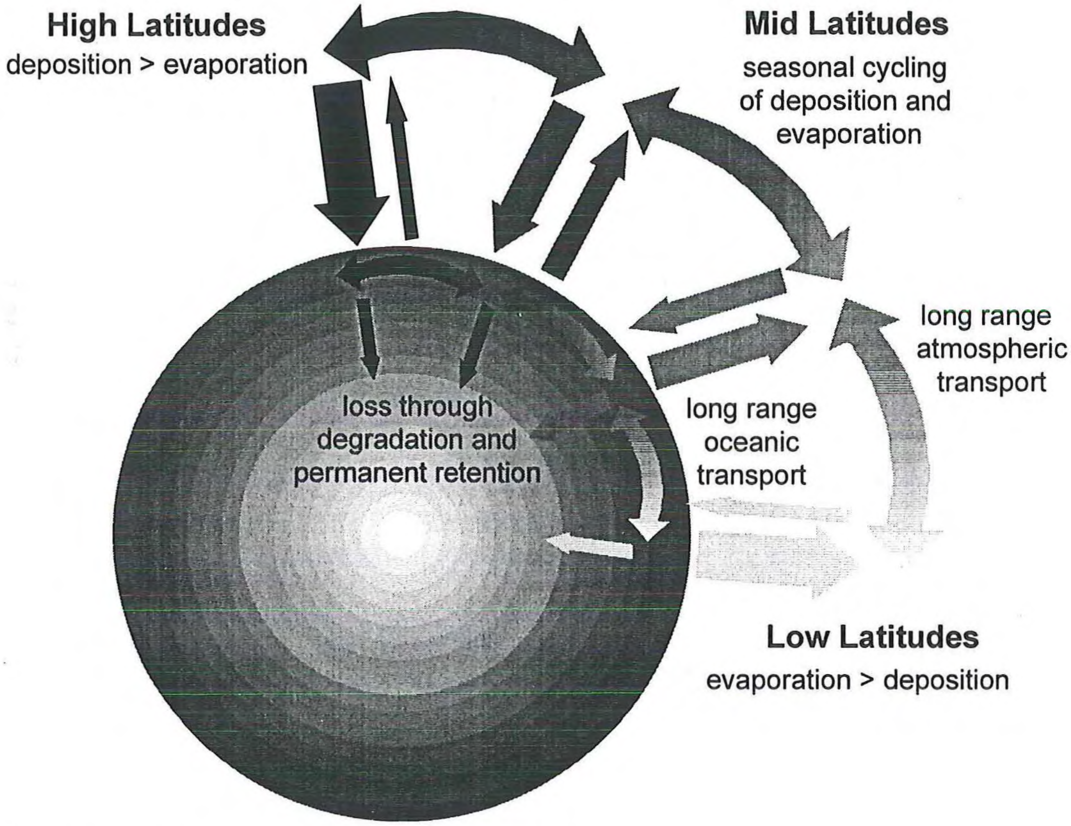
Temperature dependence of physical chemical properties

These physical-chemical properties are dependent on temperature to a varying degree. Temperature effects are greatest for vapour pressure and K_{OA}, intermediate for Henry's law constant, and least for water solubility and K_{OW}. Physicochemical properties data for many POPs are available at 20-25°, but are often lacking at other temperatures. In particular, few measurements have been made of Henry's law constants at different temperatures.

3.0 CONTINENTAL PATHWAYS OF POPS IN NORTH AMERICA

3.1 General Overview

The large scale behaviour and distribution of POPs is controlled by (1) the point of discharge into the environment, (2) the movements of the atmosphere and the oceans, (3) the rate of exchange processes between the atmosphere and the Earth's surface, and (4) the rate of chemical loss from various environmental phases.



Discharge into the Environment

No matter how efficient the atmospheric translocation processes are in dispersing contaminants, concentrations in environmental sample will always be influenced by the history of chemical usage in the surroundings of the sampling site. This has to be accounted for in any attempt to understand and interpret large scale distribution patterns. Not surprisingly, several studies of global distribution patterns of persistent organic contaminants found a strong correlation with the use of a chemical in a country (Calamari et al. 1994, 1995, Tremolada et al. 1995, Simonich and Hites 1995). This is particularly true for less mobile chemical which tend to stay close to their point of release into the global environment.

Movement in the Atmosphere and Oceans

Chemicals are transported in the global environment as a result of the general circulation in the atmosphere and the oceans. The movement of these media is unaffected by the presence and long-term inventory of PCBs in the water column of Lake Superior, Jeremiason et al. (1994) estimated that 26,500 kg have been volatilized from the lake over a twelve-year period, or an average of 2210 kg per year. Net volatilization of PCBs from lakes Superior, Michigan (Hornbuckle et al. 1994, 1995) and Ontario (Hoff et al. 1996) is also implied by measurements of PCBs in surface water and the atmosphere over the lake. By comparison, a report to the Ontario Ministry of Environment and Energy (Johnson et al. 1992) estimated that only 1000 kg of PCBs was emitted annually into the air within all of eastern North America by leaking transformers (440 kg/y), stationary fuel combustion (450 kg/y), landfills (69 kg/y) and incinerators (42 kg/y).

Harrad et al. (1994) estimated that 93% of the contemporary environmental burden of PCBs in the U.K. is held in soil. Their inventory of current PCB sources to the atmosphere were: volatilization from soils 88%, transformer and capacitory leaks 9%, refuse-derived fuel 2% and other industrial sources 1%. A model of PCB loss from soils in the U.K. indicated that concentrations peaked in the late 1960s and have since declined with a half-time of 10-20 years (Harner et al. 1995). It was estimated that 25,000 tonnes of PCBs have been lost from U.K. soils between 1970 and 1990 (Alcock at el. 1993, Jones et al.1995). Clearly re-emission of PCBs from contaminated water bodies and soil is large compared to releases from point sources.

The re-emission of previously deposited material is not restricted to PCBs. Revolatilization of pesticides has been observed in the Great Lakes (Hoff et al. 1996, Ridal et al. 1996, 1997) and even the Arctic Ocean (Bidleman et al. 1995b, Jantunen and Bidleman, 1996). Of course, not just material deposited during times of much higher atmospheric concentrations is slowly evaporating from soils, but also POPs that had been directly applied to soils as a pesticide (e.g. Boul et al. 1994, Spencer et al.. 1996, Jantunen et al. 1997) or with sludge amendments (e.g. McLachlan et al. 1996).

3.3 Long Range Atmospheric Transport of POPs

As an exposure medium for POPs the atmosphere is generally insignificant, but not as a means of transportation. In fact, POPs are transported worldwide through the atmosphere, as indicated by measurements in air and precipitation from the Great Lakes region (Hoff et al. 1996) the Arctic (e.g. Gregor 1991, Bidleman et al. 1995a, Oehme et al. 1996, Fellin et al. 1996, Jantunen and Bidleman 1996) and over the oceans (Iwata et al. 1993, Schreitmüller and Ballschmiter 1995). Levels of PCBs in ambient air are highest in cities (Halsall et al. 1995, Cotham and Bidleman 1995) followed by rural and remote locations. Atmospheric OC pesticide concentrations should be highest in regions of recent use. Levels of toxaphene in air, measured in 1994-95 in South Carolina (Bidleman et al. 1997) and 1995-96 in Alabama (Jantunen et al. 1997) are several times higher than Great Lakes values. A clear south-to-north gradient in airborne toxaphene in the USA was found by Rice et al. (1986). Measurements made in 1981, a year before the ban of toxaphene, showed that concentrations in Mississippi were 25 times higher than those in Michigan. Few atmospheric measurements of pesticides have been conducted in tropical countries. Limited measurements of airborne DDT in equatorial Africa (Congo, Ngabe and Bidleman 1992) and Central America (Belize, Alegria et al. 1996) during the 1990s are similar to median values found in a U.S. survey before DDT was banned (Majewski and Capel 1995). The Belize air samples show levels of DDT, aldrin and dieldrin that are greatly elevated compared to Great Lakes values (Alegria et al. 1996).

The Two Primary Modes of Atmospheric Transport

It is suggested to distinguish two principle modes of atmospheric transport from source to depositional regions - "single-hop" and "multi-hop" transport (see Table). As most POPs are present in the atmosphere both as vapours and attached to particles, they should be transported either way.

	Single "hop"	**multiple "hop"**
modes of transport	passive transport with atmosphere ("classical atmospheric long range transport")	cycling between atmosphere and surface, passive transport with atmosphere during time periods spend in atmosphere ("grass-hopping")
pathways	from source regions to downwind regions, particularly regions favouring deposition (e.g. high precipitation)	from warm to cold regions (predominantly, but not exclusively downwind)
time scale of transport process in atmosphere	• from emission into atmosphere until deposition to the surface • determined by atmospheric residence time of a substance (i.e. its rates of deposition and degradation): days to weeks	• from initial release into the environment until degradation or permanent retention • determined by the overall persistence in the environment: years to decades
chemical prerequisites for long range transport	• persistence in the atmosphere • emissions occur directly into atmosphere	• persistence in the atmosphere • significant volatility
meteorological prerequisites for long range transport	meteorological conditions favourable to long range atmospheric transport, i.e. periods of fast moving air masses without precipitation	conditions favourable to the evaporation from the surface into the atmosphere, e.g. warm temperatures
potential sources of chemical to the atmosphere	• atmospheric emission only	• atmospheric emissions • emissions to water and soils • re-cycling of previously deposited and/or widely dispersed material
deposition mechanisms	• wet and dry particle deposition	• wet and dry particle deposition • gas adsorption to water, soils, vegetation and snow/ice
examples of chemicals subject to this sort of transport	• POPs with very low volatility such as highly chlorinated PCDD/Fs • particulate matter, heavy metals other than mercury, most radionuclides	• POPs which occur in the vapour phase at ambient temperatures • elemental mercury

Some very involatile chemicals are present in the atmosphere associated entirely with aerosol particles. Once deposited, they can only re-enter the atmosphere under special circumstances, such as a dust storm. These chemicals can undergo long range atmopsheric transport, but they have to make the long journey with a single "hop". This "short-cut" transport occurs on the time scale of days to a maximum of a few weeks. Particle-associated POPs, i.e. especially the least volatile ones such as the highly chlorinated PCDD/Fs, are subject to that type of transport. In the case of chemicals which are volatile enough to re-evaporate after being deposited the journey can proceed in countless steps. This so-called “grass hopping” may involve convoluted detours and back-tracking. The journey may last for decades until a molecule is eventually degraded or permanently retained. Some persistent DDT molecules released in the 1950s may still be cycling in the world’s environment today.

3.4 Seasonal Cycles of Air Concentrations

Seasonal cycles have been observed in the concentrations of POPs in the atmosphere (Hermanson and Hites 1989, Manchester-Neesvig and Andren 1989, Hoff et al. 1992a,b, 1993, Halsall et al. 1994, 1995, Oehme et al. 1996, Kaupp et al. 1996) and in the rates and direction of air-water exchange (McConnell et al. 1993, Hornbuckle et al. 1994, Ridal et al. 1996). Recently, even diurnal variation of atmospheric POP concentrations could be observed (Wallace and Hites 1996). Most of these cycles can be explained, at least partially, with the "recycling" of POPs from lakes and soils, the chemicals being volatilized and adsorbed during rises and falls in the local temperature. The logarithm of vapour phase concentrations has been found to be linealy related to reciprocal ambient temperature in several urban and rural locations (Larsson et al. 1989, Panshin and Hites 1994, Monosmith and Hermanson 1996, Poissant and Koprivnjak 1996, Kaupp et al. 1996). In addition to temperature, relative humidity plays a role in exchange with vegetation (Hornbuckle and Eisenreich 1996).

3.5 The Latitudinal Fractionation Process

As early as 1974 it was suggested that certain organic chemicals may migrate through the global atmospheric environment and condense in low temperature regions (Rappe 1974). Over the last 20 years this hypothesis has been restated and revised (Goldberg 1975, Ottar 1981, Rahn and Heidam 1981) and was fully discussed in a review by Wania and Mackay in 1993 who suggested that a process may occur in which POPs may be fractionated during their journey towards the poles because of differing migration velocities. A chemical may have a specific environmental condensation temperature or range of temperatures, and even a latitudinal range of condensation, dictated by the temperature-dependence of its atmosphere-to-surface partitioning. This “global fractionation” process will cause a shift in the relative composition of contaminant mixtures along a temperature or latitudinal gradient with more volatile constituents remaining more in the air phase and migrating faster, with less volatile chemicals partitioning more into the non-gaseous liquid or solid media of water, soils and vegetation. This could be tested by searching for evidence of:

- “inverted” latitudinal concentration gradients of relatively mobile chemicals in non-gaseous media, i.e. concentrations increasing from warm source to cold destination.
- compositional changes of contaminant mixtures to the more volatile constituents at higher latitudes.

Inverted Latitudinal Concentration Gradients of Relatively Mobile Chemicals

Vegetation has been used extensively to investigate global distribution patterns. Calamari et al. (1991) were among the earliest to identify an inverted gradient in concentrations of hexachlorobenzene (HCB) in foliage. Simonich and Hites (1995) analysed tree bark samples from all over the world and found such a gradient for the four most volatile chemicals (HCB, α-hexachlorocyclohexane (α-HCH), γ-HCH, pentachloroanisol) in a set of 22 POPs. Less volatile compounds showed no significant latitudinal dependence. On a smaller scale similar patterns were also observed in lichen from Ontario, where samples from the northwestern part of the province were distinguished from most southern locations by higher proportions of more volatile organochlorine chemicals such as the HCH isomers, pentachloroanisol and chlorobenzenes (Muir et al. 1993)

A striking example of an inverted latitudinal concentration profile is the α-HCH concentration gradient in sea water from the Western Pacific extended towards the North with measurements in the Canadian Arctic (Wania and Mackay 1996). In a study of organochlorine pesticides in surface sediments of eight Canadian lakes along a mid-continental transect from 49°N to 82°N, concentrations of ΣDDT were found to decline with increasing northern latitude, whereas levels of chlorobenzenes were generally higher in the surface sediments of the arctic lakes than in NW Ontario (Muir et al. 1995). Toxaphene, chlordanes, dieldrin, and HCHs did not show consistent latitudinal gradients but had highest concentrations in lakes at 63°N and 75°N.

Compositional Changes of Contaminant Mixtures Along Latitudinal Gradients

The PCBs are ideal for detecting latitudinal, compositional changes of contaminant mixtures, because of their wide range of volatility. Such data for fish liver from lakes and rivers across Canada (Muir et al. 1990) were key for formulating the global fractionation hypothesis (Wania and Mackay 1993). Since then Muir et al. (1995, 1996) presented additional evidence from Canadian lake sediments which cover an even wider latitudinal scale than the burbot data. Statistical analysis revealed that sediments from high arctic lakes could be distinguished by greater proportions of lower chlorinated PCBs (Muir et al. 1996a). Similar shifts in the composition of PCBs indicating a higher proportion of the tri- to pentachlorobiphenyls in the Arctic were reported by König et al. (1995) in zooplankton from Canadian lakes ranging from the Great Lakes region to Ellesmere Island. Finally, changes of PCB congener patterns with time and latitude, which agree with the cold condensation/global fractionation theory were observed in mosses sampled in Norway (Lead et al. 1996).

Changes in congener pattern with latitude were also reported for PCDD/Fs. Surface sediment samples from the Barents Sea had a higher proportion of less chlorinated congeners of PCDDs and especially PCDFs than sediments from the North Sea (Oehme et al. 1993). While the PCDF profile was dominated by OCDF in the North Sea sediments, TCDF was the most abundant congener group in the Arctic samples.

3.6 Time Profiles of Atmospheric Concentrations and Deposition in North America

The initial response of atmosphere to the ban of a pesticide is a rapid drop in concentration followed by a lengthy period of stabilization. DDT was heavily used in the Mississippi Delta on cotton until its registration was cancelled in 1972. Atmospheric levels of DDT in the Delta dropped from 100 ng/m^3 in 1972 to 12 ng/m^3 in 1974 (Arthur et al. 1976). The summertime concentration of α-HCH in arctic air has declined from 800 pg/m^3 in 1982 to 100 pg/m^3 in 1992-94 (Bidleman et al. 1995b, Jantunen and Bidleman 1996). Stepwise drops in atmospheric concentrations that occurred in the early 1980s and again in 1990 coincide with reductions the use of technical HCH by China and Russia (Y.-F. Li, Atmospheric Environment Service, personal communication).

Depositional Profiles in Sediments and Peat Bogs

The historical record of PCB and OC pesticide deposition from the atmosphere is preserved in the peat bogs of the Great Lakes region and eastern Canada (Rapaport and Eisenreich, 1986, 1988, Rapaport et al. 1985). The rise in peat bog residues during the 1950s to peak values in the mid-1960s (DDT) and mid-1970s (toxaphene) closely tracked the U.S. production record of these pesticides. In the case of DDT, concentrations in peat declined after the 1972 ban, but ten years later were still 10-20% of peak values. Moreover, the DDT residues in the peat during the early 1980s were marked by a high proportion of p,p'-DDT and o,p'-DDT relative to their corresponding DDE degradation products. This led Rapaport et al. (1985) to hypothesize that fresh DDT is being transported to the Great Lakes region, possibly from continued usage in countries south of the U.S. The maximum concentration of PCBs in the peat cores occurred around 1969, close to the years of peak production in the U.S. (1967-70). PCBs in the cores declined afterward in response to the 1971 restriction of use to only closed-system applications.

A similar depositional history for PCBs is seen in sediment cores from Lake Ontario (Eisenreich et al. 1989). A record of toxaphene input to Lake Ontario was provided from dated sediment cores (Howdeshell and Hites 1996). Horizon and maximum years of deposition were 1945 and 1973 (± 6 y), in good agreement with the years in which toxaphene was first produced (1947) and maximum use (1972-74). According to models of Wania and Mackay (1993, 1995, 1996), northward migration of PCBs and other semivolatile compounds to polar regions is retarded by continuing cycles of deposition and revolatilization. This is exemplified in the PCB profiles in lake sediments of the Canadian Arctic, where PCB deposition is first seen about 20 years after production began and highest levels are found in the more recent sediment layers from 1982-90 (Muir et al. 1996a). Similar delays were obseved for OC pesticides in the same lake sediments (Muir et al. 1995).

3.7 Modelling Long Range Transport of POPs

Until recently models of POPs in the environment focussed on simulating chemical fate on a low spatial resolution, aimed more at understanding and synthesizing processes (Mackay 1991) than at identifying and quantifying continental pathways. Continental pathway modelling of POPs is complicated by a number of factors:

1. **emission estimates**: The nature of the sources makes it extremely difficult if not impossible to estimate atmospheric emission with reasonable accuracy. Though it is tricky to translate usage data (Li et al. 1996) into release rates into the atmosphere, the task may be feasible for pesticides. It may however be virtually impossibe to produce reliable emission estimates with high spatial resolution for PCBs and PCDD/Fs. A global mass balance study for PCDD/Fs (Brzuzy and Hites 1996) revealed for example that emission estimates and deposition measurements are mismatched by as much as a factor of four, potentially due to unknown sources.
2. **reversible atmosphere-surface exchange**: Models have to account for the potential of POPs to cycle repeatedly between the atmosphere and the surface. The rate of revolatilisation is

dependent, among other factors, on the amount of chemical on the surface, which in turn is dependent on how much has been deposited and not been lost by processes other than evaporation. The amount on the surface either has to be provided as an input parameter or it is calculated as a part of the model, both of which is a challenging task. Presently no surveys of POP levels in foliage and soils on a continental scale are available that would allow the construction of spatially highly resolved maps of surface concentrations. The latter approach requires modelling long time spans to account for the slow response and build-up time of surface concentrations which may be as long as several decades.

3. **complex pollutant mixtures**: Many of the POPs, such as PCBs, PCDD/Fs, and toxaphene, constitute complex mixtures composed of sometimes more than a hundred individual components with highly variable physical-chemical properties and degradation half-lifes. Models can either try to describe the fate of individual components of a mixture, e.g. a particular PCB congener, or employ the construct of a hypothetical compound with the average properties of the mixture.

Nevertheless, there have been attempts at describing quantitatively the transport of POPs through the atmosphere, both on a continental (North America: Voldner and Schroeder 1989, 1990, Europe: Persson and Ullerstig 1996, Van Jaarsveld et al. 1997) and global scale (Pudykiewicz and Dastoor 1996, Strand and Hov 1995, Wania and Mackay 1995, Wania et al. 1996). Most of these are based on models which were initially developed for acidifying pollutants and later adapted for POPs by including descriptions of reversible air-water (Mackat et al. 1986) and air-soil exchange (Jury et al. 1983, 1994a,b). Many of them have modelled the behaviour of OC pesticides, especially HCHs.

Voldner and Schroeder (1989, 1990) modelled the atmospheric transport of toxaphene from source regions to the Great Lakes. Over 80% of the toxaphene applied in the U.S. during 1976 and 1980 was used in eleven southern states. The model shows that emissions of toxaphene from Texas have a high probability of being carried to the Great Lakes, while releases in Georgia are transported both to the Great Lakes and out over the North Atlantic.

Two investigations modelled the dispersion of lindane in Europe (Persson and Ullerstig 1996, Van Jaarsveld et al. 1997). Although both studies confirmed the potential for long range transport of this OC pesticide, predicted air concentrationa and wet deposition rates were either too low by a factor of three (Van Jaarsveld et al. 1997) or too high by a factor of three to ten (Persson and Ullerstig 1996). In both cases the same emission estimate (Baart et al. 1995) was held responsible for the deviation.

Strand and Hov (1996) and Wania et al. (1996) have modelled global HCH behaviour using zonally averaged two dimensional models, accounting for meridonal transport in atmopshere and oceans. Both investigations identify the world oceans as a major reserviour and the degradation in the atmosphere as a major sink of HCHs in the global environment. Wania et al. (1996)'s model calculation reproduce the inverted α-HCH concentration profile with latitude observed in the seas of the Northern hemisphere.

Use of back trajectories of atmospheric long range transport of POPs in North America

Hoff et al. (1992a,b) measured the annual cycle of OC pesticides in the atmosphere in southern Ontario for 1988-89. Back trajectories for especially high events indicated movement of air masses up to the Great Lakes region from the southern U.S. and the Gulf of Mexico. The study suggests that temperature controls the local air-surface exchange of pesticides, but that episodic transport from source regions is also responsible for delivering pesticides to the Great Lakes region. For HCHs, no such link between elevated air concentrations in Québec and back trajectories with origin in southern areas was observed (Poissant and Koprivnjak 1996).

A problem with locating sources by back air trajectories is that it is not clear where along the route the pesticide entered the atmosphere. For example, suppose a high value for DDT is observed at a Great Lakes site and the air trajectory tracks backward to northern Mexico. From this information alone, there is no way of knowing if the DDT actually came from Mexico or was picked up by volatilization from soils in the southern and central U.S. The challenge is to differentiate new and old pesticide sources. Some use can be made of parent compound/metabolite ratios. For example, a high ratio of DDT/DDE has been invoked as evidence for new DDT transport (Rapaport et al. 1985). However some arid soils contain a high proportion of parent DDT decades after its use was stopped (Hitch and Day 1992). A combination of investigative approaches is needed. These include atmospheric monitoring of pesticides in source and receptor regions, coupled with air trajectory information, and the use of "marker" compounds as indicators of pesticide degradation. The latter include parent/metabolite ratios such as DDT/DDE and enantiomers of chiral pesticides which are metabolized at different rates in soils and aquatic systems (Falconer et al. 1997, Jantunen and Bidleman 1996, Ridal et al. 1997).

4.0 POPs IN BIOLOGICAL SYSTEMS

4.1 Bioaccumulation of POPs

POPs enter organism by varying routes and accumulate in body lipids. Bioaccumulation is the term used to define the net accumulation of POPs from all exposure routes (Thomann 1989) and is usually expressed as the concentration of a POP in an organism on a lipid weight basis divided by the concentration found in water (truly dissolved) or air (gas phase). Biomagnification is the term used to define the increased accumulation of POPs with each trophic level in a food web and is expressed as the concentrations in the organism divided by the concentrations in its food, both on a lipid weight or organic carbon (sediments, soils) basis. In aquatic ecosystems, uptake through ingestion of contaminated food is the main route for bioaccumulation of POPs with $\log K_{OW} > 5$ with only a small fraction being accumulated from gill respiration (Thomann 1989, Thomann et al. 1992).

Bioavailability refers to the extent to which pollutants associated with soils, plants, sediments or suspended or dissolved organic carbon in the water column are available for uptake by biota (Dickson et al. 1994). Many factors such as organic carbon content of soils and sediments, pH and kinetic limitations influence the amount of a contaminant that can be released from food particles in the gut or dissolved into sediment pore-waters, and therefore reduce the environmental bioavailability. Despite being tightly bound to particles, membranes and fat globules most persistent organochlorine contaminants have been shown in laboratory studies with invertebrates, fish, mammals and birds, to be readily assimilated from the diet, and when present in the dissolved phase in water, to be rapidly accumulated from water.

The patterns of POPs in abiotic samples will resemble those from emission sources, although they will have been altered to some extent during long range transport. In biotic samples the POP patterns may change for each trophic level in a food web as more easily metabolized POPs are eliminated and the more persistent POPs are biomagnified (e.g. Muir et al. 1988, Bidleman et al. 1993). Thus, predatory birds and mammals at high trophic levels will mainly be exposed to the most persistent POPs. This means that for ecotoxicological risk assessment purposes, it is also important to analyze the levels of POPs at lower trophic levels, in order to estimate the intake and therefore, exposure of organisms at higher trophic levels.

In the terrestrial ecosystem airborne contaminants bound to particles are deposited onto plants, snow and soil while POPs in the gas phase diffuse into the waxy layers of plants (Welsch-Paul et al. 1995). Soil microorganisms and invertebrates accumulate POPs that are bound to organic matter via direct contact with the soil or from soil ingestion. Accumulation in plants is from dry and wet deposition of particles onto plant surfaces and diffusion from air into waxy layers. Plant uptake of POPs via the roots is very limited (Paterson et al. 1990, 1994). Bioaccumulation is primarily from food ingestion (McLachlan 1996). Herbivorous birds and mammals are exposed to POPs via ingestion of plants and soil while omnivores may ingest terrestrial invertebrates as well as smaller birds and mammals. Animals that eat perennial vegetation, for example lichens and twigs, will be exposed to more POPs than those eating annual herbs because of the accumulation of particles containing POPs on their surfaces. At the top of the food web are the predatory birds and mammals which feed on herbivores and omnivores.

In the freshwater and marine ecosystems POPs that are present in air (as the result of long-range transport or local emissions) are eventually deposited by dryfall, gas absorption and direct precipitation (Mackay et al. 1986). There is also inputs from the terrestrial environment of particle and dissolved phase contaminants in runoff water and (in northern latitudes) in snow melt via rivers and estuaries. Particle bound POPs such as PCBs, PCDD/Fs, and high molecular weight PAHs are associated with suspended particles in the water column (depending upon their hydrophobicity) and eventually sediment out in lake bottoms (Baker et al. 1991). POPs bound to the suspended particles equilibrate between the particle's organic carbon fraction and the water, especially the dissolved carbon fraction. In sediments, POPs equilibrate between the organic carbon of the sediment, the pore water between sediment particles and the overlying water layer.

Aquatic microorganisms, plankton, algae and plants are exposed to POPs via contact with the dissolved fraction in water and possible direct contact with suspended particles (Broman et al. 1996, Stange and Swackhamer 1994, Gobas et al. 1991). Uptake occurs by diffusion into the lipid-rich outer membranes and waxy layers and as well as from food ingestion for microorganisms and zooplankton. Invertebrates bioaccumulate POPs dissolved in the water phase when these come in contact with the gills as well as from ingestion of microorganisms, plankton, suspended particles and sediment. For fish, bioaccumulation will mainly be from food ingestion, whether this is sediment, plankton or other fish but some accumulation will come from gill respiration, depending on how lipophilic the POPs are (Clark et al. 1990). The more lipophilic the POP, the less important gill uptake becomes. Accumulation in mammals and birds will be entirely from food ingestion. A unique characteristic of arctic marine food webs in North America is the presence of a fourth level carnivore, the polar bear, which feeds mainly in seals. Scavengers such as arctic fox and some seabirds (glaucous gull, herring gull) may also feed at the same trophic level as polar bears depending on availability of food.

4.2 Toxicokinetics of POPs

The majority of POPs are, by definition persistent, bioaccumulative and toxic substances. They are taken up by aquatic living organisms via diffusion over the gills and from food via diffusion across the gastrointestinal wall. POPs cross the gill/gut membrane and enter the blood where they are quickly distributed to high lipid tissues such as the liver and adipose tissue. Metabolism and elimination are often slow, leading to a net increase of these substances in the organism over time.

There are species differences in the tissue distribution of POPs, partly due to differences in lipid distribution. For example, fishes such as cod (*Gadus morhue*) and burbot (*Lota lota*) store high concentrations of PCBs in liver (livers are up to 50% lipid) while lake trout (*Salvelinus namaycush*) accumulate PCBs mainly in the fat and have highest burdens in muscle. Lipid dynamics can also affect the distribution of POPs. Marine mammals store PCBs and other persistent OCs in blubber layers while in birds, terrestrial mammals and polar bears storage is mainly in depot fat.

Metabolism of xenobiotics occurs mainly in the liver via a two-phase process. In phase I, xenobiotics are converted by oxidation reactions to metabolites that can undergo phase II reactions. In phase II, the product is conjugated with glucuronic acid or glutathione, for example, to produce water soluble compounds that can be excreted in urine or bile. These processes are catalyzed by liver enzymes such as the cytochrome P450 containing monooxygenases (Nebert and Gonzalez 1987) or mixed function oxidase (MFO) enzymes. Substances that are resistant to metabolism will be selectively accumulated in living organisms. In addition to detoxification, the enzymatic processes can also create reactive intermediates that may be mutagenic and/or carcinogenic, or metabolites that are lipophilic and have retained toxicity, or that have the ability to bind selectively to proteins and accumulate in the organism.

Many POPs form metabolites that are biologically active. DDT is metabolized in living organisms to DDD and further to DDE, both of which are lipophilic, toxic and bioaccumulative (WHO 1989a). In some cases, a methyl sulfone ($MeSO_2$) group is added during metabolism and a number of $MeSO_2$-DDE and $MeSO_2$-PCB congeners have been identified in animals (Letcher et al. 1995). Some congeners of PCB may also form hydroxylated metabolites (Jansson et al. 1975). This type of metabolite has been found to selectively bind to transthyretin, one of the major transport proteins for retinol and thyroid hormones in the blood (Brouwer et al. 1988, 1990, Bergman et al. 1994). Aldrin is metabolized in living organisms to dieldrin by the cytochrome P450-dependent monooxygenase, aldrin epoxidase (WHO 1989b). Chlordane is metabolized to some extent to oxychlordane (WHO, 1984a). Hexachlorobenzene is metabolized to some extent mainly by the liver and may form, among other metabolites, pentachlorophenol, tetrachlorohydroquinone, pentachlorothiophenol and lower chlorinated benzenes (Debets and Strik 1979, Renner 1988).

The major excretion route of POPs and their metabolites in mammals is via the feces. Some of this is direct diffusion over the gut membrane and some from bile excretion of metabolites. In invertebrates and fish, excretion also occurs by diffusion over the gill membranes. Female fish and birds excrete lipophilic POPs via their eggs and female mammals via placental transfer to the fetus and in breast milk. Most marine mammals have very high fat contents in milk in order to facilitate fast growth in the young. Therefore, excretion of POPs via milk is more important than via placental transfer for adult females in marine mammal species. This in turn enhances POP exposure of nursing animals (Espeland et al. 1996, Bernhoft et al. 1996, Polischuk et al. 1995). This is of concern as young animals are more sensitive to the effects of POPs than adults.

The net result of uptake, distribution, metabolism and excretion will determine the POP levels found in an organism. This is in turn affected by other factors. Studies carried out to determine the uptake, distribution, metabolism and excretion of POPs usually use one substance at a time. Wildlife and humans, however, are exposed to complex mixtures of POPs. Very little is known about how different POPs affect each other's toxicokinetics. POPs that induce the hepatic cytochrome P450 MFO enzyme system will affect the metabolism of other xenobiotics, for example. This may lead to an increase in xenobiotic metabolism, thus increasing excretion. For example, studies on Baltic seals show that high body burdens of DDT and PCB are associated with lower relative amounts of the mono-*ortho* CB, 2,3',4,4',5-PeCB (CB 118). At total PCB concentrations (sum of seven) of 50 µg/g lipid weight or higher, CB 118 could not be found (Haraguchi et al. 1992, Olsson et al. 1992). ΣPCB levels in Arctic ringed seal are much lower, xenobiotic metabolism does not seem to be induced and concomitantly, CB 118 is present in higher relative amounts (Norstrom and Muir, 1994). Greater activity of CYP MFO enzyme systems may also explain differences in PCB congener patterns in beluga whales from the St Lawrence river estuary compared to those in the Arctic, with 10-12 times lower levels of PCBs (Muir et al. 1996b).

Thus, the relative amounts and the composition of various POPs in biota, especially mammals and birds, may partly be the result of selective effects on the organism's uptake, metabolism and excretion of POPs and not solely a result of the specific pollution burden of the single contaminant in the food web.

4.3 Specific biological effects of POPs

Reproductive and developmental effects: POPs have a number of effects on the ability of organisms to reproduce and develop normally (for reviews see Brouwer et al. 1995, Barron et al. 1995, Bosveld and Van de Berg 1994, Peterson et al. 1993). Exposure to some POPs may cause embryo- and fetotoxicity, decreased offspring survival, abnormalities in the estrus cycle and sex hormone levels, reduced sperm production, reduced litter sizes and even total reproductive failure in mammals. In birds, some POPs cause decreased egg production, retarded egg production, increased embryo mortality, eggshell thinning, embryonic deformities, growth retardation and reduced egg hatchability as well as detrimental effects on parental behavior. In fish, some POPs cause decreased egg and larval survival, reduced sexual maturation and reduced gonad size. Early life stages of fish are more sensitive than adults to the effects of PCB and PCDD/PCDF. In fish, TCDD caused decreased egg hatchability, increased sac fry mortality and edema in lake, brook (*Salvelinus fontinalis*) and rainbow trout (Walker et al. 1991, Spitsbergen et al. 1991, Walker and Peterson 1991, 1992, 1994) and reduced number of eggs and lethal malformations in young of the zebrafish (*Brachydanio rerio*) (Wannemacher et al. 1992). The same effects are seen in lake trout eggs which received their TCDD exposure via maternal transfer and these effects were dose-dependent (Walker et al. 1994).

DDT has long been known to affect bird reproduction (Peakall 1986, Peakall et al. 1990). p,p'-DDE has been shown to cause significant egg-shell thinning in treated birds and predatory species have been found to be the most sensitive.

Hormone disruption: Some POPs act as hormones or interfere with endocrine systems and are therefore called endocrine or hormone disruptors. The reproductive effects of embryonic or fetal exposure to these may only become obvious at later developmental stages or at sexual maturity. The estrogenic and antiestrogenic effects of POPs are the best studied of these effects. Endocrine disruption is also implicated in thyroid and immune system effects. Environmental estrogens can mimic estrogen by binding to the estrogen receptor and turning it on. Antiestrogens bind to the receptor and block the normal binding of estradiol, thus inhibiting estrogen activity. POPs may also function as androgens or antiandrogens, for e.g. p,p'-DDE is antiandrogenic, binding to the androgen receptor and blocking the binding of endogenous androgens (Kelce et al. 1995). Toxaphene has been shown to be estrogenic in the *in vitro* E-screen test (Soto et al. 1994).

Some effects on the thyroid seem to be related to the ability of some POPs, particularly hydroxy-PCB metabolites, to attach to the binding sites on the transthyretin-retinol-binding protein complex (TTR-RBP) in plasma, thereby disrupting the normal transport of the thyroid hormones triiodothyronine (T3) and thyroxine (T4) as well as vitamin A (retinol) to their target tissues. Some POPs (particularly PCDDs and PCBs) affect vitamin A metabolism and transport in several ways. Hepatic stores may be depleted (Håkansson et al. 1990), possibly due to enhanced retinol metabolism (Adams et al. 1990). Laboratory and field studies have shown that PCB exposure leads to drastic reductions in plasma concentrations of vitamin A (retinol) (Innami et al, 1974) and thyroid hormones (Brouwer et al. 1989, Durham and Brouwer 1989).

Mixed function oxidase enzyme induction: Exposure to organochlorines and some PAHs induces liver cytochrome P450-MFO enzymes, which metabolize xenobiotics and endogenous substances (Nebert and Gonzalez 1987). Cytochrome P450 1A forms are induced by planar aromatic hydrocarbons such as 3-methylcholanthrene, PCDD/PCDF and non-ortho PCBs. This induction is often measured as increases in activity of several enzymes including ethoxyresorufin-O-deethylase (EROD)and aryl hydrocarbon hydroxylase (AHH). Induction of cytochrome P450 1A is mediated by the aromatic hydrocarbon (Ah) receptor which has been found in the cell membranes of all vertebrates studied so far. Cytochrome P450 2B forms are induced by another class of substances, typified by phenobarbital and measured as aminopyrine N-demethylase activity, aldrin epoxidase and pentoxyresorufin O-dealkylase. POPs that induce cytochrome P450 2B are DDT, chlordane and di- to tetrachloro -ortho-substituted PCBs.

Seals and cetaceans, like other mammals, seem to have functional cytochromes P450 1A1 and 1A2 and thus a higher capacity to metabolize planar compounds. They differ from terrestrial mammals in having weak cytochrome P450 2B activity and thus a reduced ability to metabolize xenobiotics that are substrates for these particular MFOs (Boon et al. 1992, Goksryr et al. 1992, Goksryr 1995). Seals seem to have more cytochrome P450 2B than whales (Goksryr 1995).

Organisms lacking functional cytochromes P450 1A or 2B will not be able to eliminate the POPs metabolically, leading to their bioaccumulation. This is particularly the case for fish and invertebrates, making them carriers of many persistent organochlorines in food webs.

Immunosuppression: Immunosuppressive effects may be one of the most sensitive and environmentally relevant effects of POPs (Vos and Luster 1989). Many POPs are known to disrupt humoral- and cell-mediated immunity as well as non-specific responses, leading to immunosuppression. Humoral-mediated immunity involves the body's ability to recognize foreign substances (helper T-cells) and mount a response by stimulating the production of antibodies (B-cells). Cell-mediated immunity is involved in delayed hypersensitivity reactions (for example skin reactions to allergens) and the production of cytotoxic T-cells against tumors and viruses. Most POPs cause multiple effects on the immune system. Effects on immune function occur at exposure levels which do not produce signs of toxicity. Changes occur in both cell-mediated and humoral immunity in both laboratory rodents, chickens and in non-human primates exposed to TCDD. The fetus and lactating newborn animals are particularly sensitive to the immunosuppressive effects of TCDD.

Immunosuppression has been measured in harbor seals fed Baltic fish under semifield experiments and was found to correlate with levels of PCDD/PCDF and coplanar PCBs expressed as TEQs (De Swart et al. 1995, Ross et al. 1995, Ross et al. 1996).

Cancer and mutagenicity: Most POPs are regarded as cancer promoters. TCDD is a powerful tumor promotor in both cell culture and animal studies as well as a potent carcinogen in laboratory animals (Kociba et al. 1978, WHO 1989c, Ahlborg 1993, Lucier et al. 1993 and Huff et al. 1994). Several other PCDD/PCDF congeners as well as coplanar PCBs have been shown to be potent tumor promotors (NIH 1980, Silberhorn et al. 1990, Sargent et al. 1991, Hemming et al. 1993). PCB mixtures and congeners are cancer promoters, however, they are not mutagenic when tested using the Ames test and no DNA adducts are formed (Safe 1994). Toxaphene is mutagenic in the Ames and a potent carcinogen in rats and mice (reviewed in Saleh 1991).

Neurotoxicity: Other effects of POPs on organisms may include neurotoxic effects and neurological and behavioral changes in offspring. Behavioral changes also occur in adult animals, including changes in mating behavior. Numerous animal and human studies demonstrate that PCB exposure leads to neurobehavioral effects, particularly in offspring, and that these effects are not dependent on binding to the Ah receptor (for reviews see Seegal and Shain 1992, Tilson et al. 1990).

Most of the cyclic aliphatic and aromatic, chlorinated pesticides such as chlordane, toxaphene and DDT are neurotoxic. They alter sodium and potassium ion flux across nerve cell membranes and interfere with nerve impulse transmission. In general, they are more toxic to insects than to other animals. Toxaphene is acutely toxic to fish with lethal water concentrations in the range of 5-100 μg/liter and saltwater fish are more sensitive than freshwater fish (Saleh 1991). Laboratory studies have shown that exposure to toxaphene affects behavior and growth in fish (Pollock and Kilgore 1978, Mayer et al. 1975, 1977). Fish injected with environmentally relevant doses of technical toxaphene and released into the wild had reduced long-term survival (Delorme 1995).

5.0 CONCLUSIONS/RECOMMENDATIONS

It is now established beyond all doubt that POPs are subject to global scale transport, and thus obviously to continental transport, but to extents that differ from compound to compound. If a certain amount of a POP is discharged to the environment in Central America, some fraction of it will reach the US and Canada and cause an increase in concentrations in various components of the ecosystem. The reverse process is also likely. Although such transport is known to occur, it is not yet possible to quantify (i) what fraction of the discharged amount will reach a given region and (ii) how long this will take. The effect is incremental in that there is an existing background from numerous global sources at various times. It should be possible to quantify these processes and assign sources for a number of candidate substances, but this will require a long term program of investigations. The key steps are:

1. Continue to determine concentrations in air, soil, water, sediment, precipitation, and biota over all North America.
2. Compile inventories of uses and storage, including the history of use of candidate substances.
3. Determine the relevant properties of the candidate substances over a range of temperatures including environmental degradation half-lifes.
4. Document the conditions for environmental transport in North America, e.g. atmospheric circulation patterns, temperatures, precipitation, deposition, and ocean currents.
5. Combine information in 2 to 4 in an attempt to model continental POP transport using a variety of approaches. Express error and sensitivity of the model calculations, validate their results against data from 1, and refine the model assumptions.
6. Set target concentrations, possibly as a fraction of current levels based on toxicological information.
7. Construct scenarios of future discharge reductions to achieve these targets, i.e. set milestones of concentrations and deadlines for achievement, which have been shown feasible using models.

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Case Study

Dioxin

Authors:

Mark Cohen-CBNS- Queen College, Flushing, USA
Barry Commoner- Queen College, Flushing, USA

Reviewers:
Carlos Santos Burgo Instituto de Salud, Ambiente y Trabajo, Mexico City, Mexico
Alfonso Espitia Cabrera- Universidad Autonoma Metropolitana, Mexico City, Mexico
Tammy Jones- US EPA, Research Triangle Park, USA
Derek Muir- Freshwater Institute, Winnipeg, Canada
David Stone- Dept. of Indian Affairs and Northern Development, Hull, Canada

1. PREFACE

The initial draft of this document was prepared for consideration and discussion by Mark Cohen and Barry Commoner.

Several members of the CEC Continental Pollutant Pathways Expert Advisory Group and others reviewed the initial draft and provided useful information, including Carlos Santos Burgoa, Alfonso Espitia Cabrera, Tammy Jones (through expert group member Gary Foley), Derek Muir, and David Stone. Several others have also provided useful information.

An attempt has been made to incorporate as much of the above input into this interim draft report as possible, although there has not yet been time to fully address each of the contributions.

Due to time constraints, the additional contributors to this document have not yet had the opportunity to review this revised interim draft, and so, it would be unfair to them to list them as co-authors. It is expected that some or all of the above-mentioned contributors to this case study (and possibly others as well) will ultimately be acknowledged as co-authors of this report.

In the coming period, it is intended that this interim draft be reviewed by the above contributors and others, and that the final version and listed coauthors of this case study will evolve as a result of these further reviews and comments.

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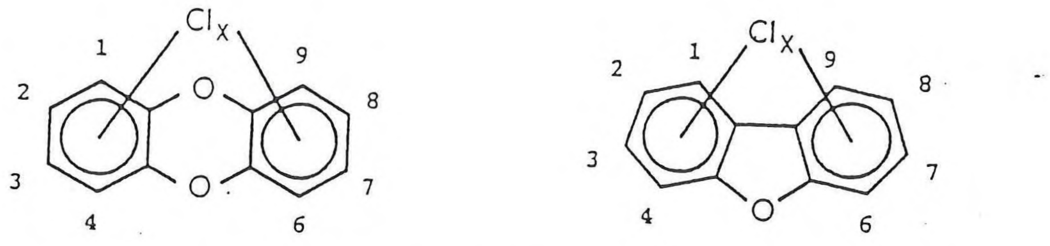
3. INTRODUCTION

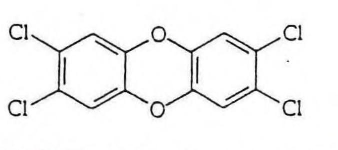
"Dioxin" is the term commonly given to a class of compounds known chemically as polychlorinated dibenzo-p-dioxins and polychlorinated dibenzofurans (PCDD/F's). As discussed below, there are other compounds beside PCDD/F's that exhibit dioxin-like toxicity. Evidence suggests that human exposure to PCDD/F's and dioxin-like compounds can lead to various forms of cancer and various developmental and endocrine-disruption effects.

There are 210 different chlorinated dioxin and furan molecules or "congeners." Each different congener has its chlorines in different positions in the structure. The general structure of PCDD/F molecules is shown in the diagrams below; also, as examples, the specific congeners 2,3,7,8-tetrachlorodibenzo-*p*-dioxin (2,3,7,8-TCDD) and 2,3,7,8-tetrachlorodibenzofuran (2,3,7,8-TCDF) are shown.

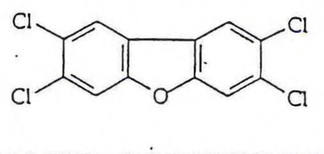
It has been found that only the 17 PCDD/F congeners with chlorines in at least the 2,3,7,8 positions show evidence of dioxin-like toxicity. The toxicity of each congener can be expressed in terms of its estimated, approximate potency relative to that of the most toxic congener (2,3,7,8-tetrachlorodibenzo-*p*-dioxin) by multiplying by its estimated "Toxic Equivalency Factor" or TEF. Values of the TEF's commonly used are given in Table 1 below. The overall PCDD/F toxic potency of a mixture of dioxin and furan congeners is often summarized by multiplying the amount of each congener by its TEF and then summing over all the congeners to get the total Toxic Equivalents (TEQ) in the mixture. The meaning of a result expressed as "TEQ" is that the total overall toxicity of the mixture is presumed to be equivalent to the stated amount of "pure" 2,3,7,8-TCDD.



Structures of (left) polychlorinated-para-dioxins and (right) dibenzofurans.



2,3,7,8-TETRACHLORODIBENZO-P-DIOXIN



2,3,7,8-TETRACHLORODIBENZOFURAN

Table 1. Toxic Equivalency Factors (TEF's) for PCDD/F Congeners
(U.S.EPA 1994a,b)

Dibenzo-*p*-dioxins		**Dibenzofurans**	
Congener(s)	TEF	Congener(s)	TEF
dibenzo-*p*-dioxin molecules with three or less chorine atoms (27 total)	0	dibenzofuran molecules with three or less chlorine atoms (59 total)	0
2,3,7,8-TCDD	1	2,3,7,8-TCDF	0.1
all other TCDD's (21 total)	0	all other TCDF's (37 total)	0
1,2,3,7,8-PeCDD	0.5	2,3,4,7,8-PeCDF	0.5
		1,2,3,7,8-PeCDF	0.05
all other PeCDD's (13 total)	0	all other PeCDF's (26 total)	0
1,2,3,4,7,8-HxCDD	0.1	1,2,3,4,7,8-HxCDF	0.01
1,2,3,6,7,8-HxCDD	0.1	1,2,3,6,7,8-HxCDF	0.01
1,2,3,7,8,9-HxCDD	0.1	1,2,3,7,8,9-HxCDF	0.01
		2,3,4,6,7,8-HxCDF	0.01
all other HxCDD's (7 total)	0	all other HxCDF's (12 total)	0
1,2,3,4,6,7,8-HpCDD	0.01	1,2,3,4,6,7,8-HpCDF	0.01
		1,2,3,4,7,8,9-HpCDF	0.01
all other HpCDD's (1 total)	0	all other HpCDF's (2 total)	0
OCDD	0.001	OCDF	0.001
Abbreviations:		**Abbreviations:**	
TCDD=Tetrachlorodibenzo-*p*-dioxin		TCDF =Tetrachlorodibenzofuran	
PeCDD =Pentachlorodibenzo-*p*-dioxin		PeCDF =Pentachlorodibenzofuran	
HxCDD =Hexachlorodibenzo-*p*-dioxin		HxCDF =Hexachlorodibenzofuran	
HpCDD =Heptachlorodibenzo-*p*-dioxin		HpCDF =Heptachlorodibenzofuran	
OCDD =Octachlorodibenzo-*p*-dioxin		OCDF =Octachlorodibenzofuran	

As mentioned above, there are other persistent organic pollutants with a similar chemical structure to 2,3,7,8-substituted PCDD/F congeners which exhibit dioxin-like toxic effects. For example, there are at least 13 PCB congeners that are considered to exhibit dioxin-like toxicity (3 non-*ortho* PCB's, 8 mono-*ortho* PCB's, and 2 di-*ortho* PCB's[1]). Other compounds, such as chlorinated naphthalenes, chlorinated diphenyl ethers, and a range of brominated analogs, may also exhibit dioxin-like toxicity.

TEF values for the dioxin-like PCB congeners have been estimated (e.g., Ahlborg *et al.*, 1994), although, as with the PCDD/F TEF's, there is some uncertainty in these estimates (e.g., Wenning and Iannuzzi, 1995). With estimated TEF values, the contribution of dioxin-like compounds to the overall dioxin-like potency of a mixture can be assessed relative to the CDD/F congeners present in the mixture.

There have been relatively few side-by-side measurements of PCB's and PCDD/F's in environmental media or tissue samples. The relative importance of PCDD/F's and PCB's to overall dioxin-like potency varies from study to study.

In studies reporting measurements of cow milk, PCDD/F's and PCB's appear to contribute roughly comparable dioxin-like potencies, e.g.:

the Netherlands: Slob *et al.* (1995) and van der Velde *et al.* (1994);

England: Krokos *et al.* (1996); Sewart and Jones (1996);

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[1]. Polychlorinated biphenyls are molecules which have two benzene rings bonded together by a single chemical bond. On each of the benzene rings, there can be chlorine atoms attached in any position. Thus, there are 209 possible PCB "congeners", ranging from biphenyl (with no chlorines) to decachlorobiphenyl (with 10 chlorines). The positions on the benzene rings adjacent to the chemical bond joining the two rings are called the "*ortho*" positions; there are two such *ortho* positions on each benzene ring, each ring has one *ortho* position on each side of the joining bond. If there are no chlorines occupying an ortho position, the two benzene rings exist in essentially the same plane, i.e., they are coplanar. This coplanar, polychlorinated configuration (with at least two chlorines on each ring) is similar to the structure of 2,3,7,8-TCDD. Due to steric considerations, if there *are* chlorines in one or more *ortho* positions, the benzene rings are unable to exist in the same plane (the chlorine atom(s) get in the way); thus, these are somewhat less like 2,3,7,8-TCDD, although mono-*ortho* PCB's and a few di-*ortho* PCB's have been found to exhibit dioxin-like toxicity.

The same rough equivalence has been found in many studies of human milk, e.g.:

- **Japan**: Hirakawa *et al.* (1995);
- **the Netherlands**: Liem *et al.* (1993); Tuinistra et al (1994,1995);
- **England**: Duarte-Davidson *et al.* (1992).

In a comprehensive study of food in the Netherlands, PCDD/F's and PCB's were estimated to contribute approximately the same total dioxin-like potency in the average diet (Theelen *et al.*, 1993).

In measurements in aquatic ecosystems, it has been frequently been found that the dioxin-like PCB's are responsible for the majority of the dioxin-like potency measured, e.g.:

- **Great Lakes region** (fish and fish eating birds): Smith *et al.* (1990);
- **St. Lawrence River** (fish): Brochu *et al.* (1995);
- **the Netherlands** (porpoise): Van Scheppingen *et al.* (1996)

A similar enhanced significance of PCB's (relative to PCDD/F's) appears to hold for many of the measurements made in arctic ecosystems, e.g.:

- Dewaily et al., 1994 (milk samples from Inuit women in arctic Quebec);
- Ayotte et al., 1995 (blood plasma from Inuit in arctic Quebec);
- Hebert et al., 1996 (Canadian arctic caribou fat and other tissues);
- Muir et al., 1996 (a range of Canadian arctic species)],

Pollutant levels in the arctic are discussed in much greater detail in the case study on <u>Persistent Organic Contaminants in the Canadian Arctic</u> (presented elsewhere in this Volume).

In addition, the case study on <u>Persistent Organic Pollutants</u> (POP's) (presented elsewhere in this Volume) provides detailed information about a range dioxin-like POP's and other POP's.

Much remains to be learned about PCDD/F's and other dioxin-like compounds. This case study will focus primarily on PCDD/F's, as an example.[2]

4. SOURCES

PCDD/F's have never been produced intentionally (except in extremely small quantities for use in research). Unfortunately, however, it is formed as an unwanted byproduct in many different types of processes.

The two broad classes of formation situations which are the generally regarded as being the most important are:

(1) combustion processes (e.g., waste incineration, fuel combustion, metallurgical processes);

(2) solution-phase processes (e.g., pulp bleaching with chlorine or chlorine containing compounds, chlorinated organic chemical manufacturing, chlorine production)

Formation of PCDD/F's in any given source results from a complex set of chemical reactions which are influenced by many factors, including, but not limited to, the individual concentrations of many potentially relevant reactants, temperature, and inter- and intra-phase mass transfer processes. PCDD/F formation can be enhanced when certain precursors are present (e.g., chlorophenols; *un*chlorinated dibenzo-p-dioxin), but, significant quantities can also be formed in the absence of the introduction of any known precursors. The relative proportions of different PCDD/F congeners created in different formation situations can be highly variable. While the formation of PCDD/F's in combustion and solution phase processes is exceedingly complex and only poorly understood at the present time, it is obvious that chlorine and/or another chlorine-containing compound is essential to their formation.

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[2]. The narrowed focus of this particular case study on PCDD/F's is not meant to imply that these compounds are necessarily the most important of the dioxin-like POP's.

4.1 Emissions to Air

As mentioned above, PCDD/F's can be formed in many types of combustion systems (e.g., Commoner *et al.*, 1987; Addink and Olie, 1995; Karasek, 1995). In these systems, PCDD/F formation appears to be most favored in the temperature range from about 300 - 450 °C. This is, of course, cooler than most combustion chambers; PCDD/F's are believed to form as the combustion exhaust gases cool *after* they leave the combustion chamber. PCDD/F's can form in the gas phase or on the surface of aerosol particles in the cooling stack gases. Certain compounds (e.g., copper) appear to act as a catalyst in surface-formation reactions.

There have been many attempts to determine the influence that chlorine -- in its various forms -- has on the formation of PCDD/F's in combustion systems. In a broad overview of a range of combustion-process emissions of PCDD/F's, Thomas and Spiro (1995) found a general increase in emissions as the level of chlorine in the material being combusted increased.

In certain situations, a positive correlation appears to be conclusively established. One such situation is the combustion of gasoline in the internal combustion engines of automobiles. When leaded gasoline is used, chlorinated and brominated substances (e.g., ethylene dichloride and ethylene dibromide) are added to the fuel. This is done so that the lead in the gasoline does not build up in the engine and thereby cause damage; during combustion, the chlorinated additives convert most of the lead in the fuel to relatively volatile lead chlorides which are able to escape from the engine. (The fact that the lead escapes from the engine may be good for the engine, but obviously, is not so good for the environment.) Experimental measurements have shown that PCDD/F emissions from automobiles burning leaded gasoline are significantly higher than those from unleaded gasoline (e.g.: Hagenmaier *et al.* 1990; Bingham *et al.* 1989; U.S. EPA 1994a; Marklund *et al.* 1987,1990). These measurements provide strong evidence that chlorinated substances are linked to PCDD/F formation.

In incineration processes, the situation is considerably less straightforward. A recent summary of experimental investigations involving particular combustion situations was provided by Wikstrom and coworkers (1996). These researchers also reported a new set of experimental data concerning this issue. They burned a simulated municipal waste in a laboratory-scale combustor and found that increasing levels of chlorine in the waste led, generally, to increasing PCDD/F emissions in the exhaust gases. In this experiment, inorganic chlorine (calcium chloride) and organic chlorine (polyvinyl chloride) was equally effective in leading to the formation of PCDD/F's.

In general, some studies have found a positive correlation between PCDD/F emissions from incineration and chlorine input, and some studies have not. In one recent review of available data, it was concluded that the available experimental evidence does *not* support the idea that PCDD/F formation generally increases with the amount of chlorine or chlorine-containing substances involved in the process (Rigo *et al.*, 1995). In any given combustion setting, there may be a threshold effect in which PCDD/F formation begins to increase with increasing chlorine input *above a certain level of chlorine input*. This has been observed in several experiments (as summarized by Wikstrom *et al.* 1996). Given the complexity of the physical and chemical processes occurring, it is not surprising that the apparent threshold level appears to vary among different types of incinerators, combustion conditions, and fuels. Due to the complexity of the situation and a lack of data, the relative contribution of various chlorinated substances in the formation of PCDD/F's in many types of combustion systems remains controversial and poorly understood.

Pollution control equipment can capture some of the PCDD/F that has been formed and prevent its immediate escape from the system. The PCDD/F captured in pollution control systems for combustion devices is associated with the captured flyash. A portion of the captured PCDD/F may be re-released to the environment during handling, transport or from its ultimate disposal site (e.g., an ashfill) to the extent that any of the following processes occur: (a) ash particles can become airborne through the action of wind or mechanical action; (b) PCDD/F's can volatilize from the ash; or (c) PCDD/F's or PCDD/F-containing ash particles can be washed into surface or ground water by precipitation.

Certain pollution control systems can actually increase the amount of PCDD/F's formed. For example, some electrostatic precipitators (ESP's) operate at a relatively high temperature — in the range in which PCDD/F formation is favored — and it has been found that more PCDD/F leaves these devices than enters them. Emissions of PCDD/F from municipal waste incinerators equipped with these so-called "hot-sided" ESP's are believed to be extremely large (e.g.: U.S. EPA 1994a; Webster and Connett, 1997).

Quantitative estimates of the amounts of PCDD/F's emitted by various sources are fraught with difficulty for several reasons.

First, for any given type of source (e.g., medical waste incinerators), very few individual facilities have actually been tested for emissions of PCDD/F's.

Second, the tests that have been done show that PCDD/F emissions can vary greatly from test to test on the same facility and from facility to facility of the same type (i.e., similar process and pollution control characteristics).

Third, overall emissions inventories and emissions estimates for given facilities are often made by using the few available tests to estimate <u>emissions factors</u> for a given type of source, with a given type of pollution control equipment. The use of emissions factors to estimate emissions is obviously an approximation. In order to even use such factors, however, one must be able to assemble a complete list of the sources of a given type, with details of each facility's emissions-relevant process characteristics, pollution control devices, and throughput (e.g., tons/day burned in an incinerator). Unfortunately, this does not appear to be possible for many types of sources, even those that are relatively significant.

For example, there are many uncertainties in currently available inventories of medical waste incinerators in the United States. Most State agencies are currently unable or unwilling to assemble an accurate list with relevant details (e.g., location, throughput, pollution control equipment). Because of this basic lack of knowledge, confusion and controversy surrounds the estimate of emissions from medical waste incinerators in the United States. A similar lack of information appears to exist in Canada for medical waste incinerators. The state of knowledge about medical waste incinerators appears to be more advanced in Mexico, with estimates available for the throughput and locations for each of the 10 incinerators known to exist (Cabrera, 1997).

Several general PCDD/F emissions summaries and/or emissions inventories have been presented; examples, in alphabetical order, include the following: (i) Cohen M., B. Commoner, *et al.* (1995); (ii) Fiedler, H., *et al.* (1990,1992); (iii) Harrad, S., *et al.* (1992); (v) Thomas and Spiro (1995, 1996); (vi) Thornton (1994) and Thornton and Weinberg (1995); (iv) USEPA (1994a, 1995, 1996a); (vii) Zook and Rappe (1994)].

Currently, there is not enough information in Canada, Mexico or the United States to develop an accurate emissions inventory for PCDD/F's. For most source classes in each country, the situation is as follows: (a) very few (if any) emissions tests have been made, and so even the development of rough emissions factors is difficult; (b) even if emissions factors were available, adequate facility-level details about location, throughput, process, and pollution control equipment either have not been assembled or are being kept confidential by government or industry groups. Efforts are underway in each country by government agencies and/or non-governmental organizations to improve the accuracy of PCDD/F emissions inventories, although it would appear that more resources need to be allocated to these efforts.

In light of the above, one must be cautious in presenting estimates for PCDD/F emissions. As a rough guide, a *qualitative* summary of approximate, estimated PCDD/F air emissions from various sources in the United States is given in Figure 1. For many of the sources in this Figure, the uncertainties in emissions are very large.

In Figure 1, no estimate was made — due to an extremely high level of uncertainty — for emissions from in-use pentachlorophenol (PCP)-treated wood (e.g., PCP-treated telephone poles). PCP is contaminated with PCDD/F, as an unintentional byproduct of its chemical synthesis. As PCP is very extensively used, emissions may be significant even if only a small fraction of the PCP's PCDD/F burden was emitted to the air.

In the U.S., there are several ongoing PCDD/F inventory efforts, including (but not necessarily limited to) those by the U.S. EPA Office of Air Quality Planning and Standards (U.S. EPA 1995, 1996a), the U.S. EPA's National Center for Environmental Assessment (Cleverly, 1996), and an inventory currently being developed at the Center for the Biology of Natural Systems at Queens College.

Many of the sources shown qualitatively in Figure 1 for the U.S. also exist in Canada and Mexico. However, there may be significant differences in the relative importance of different sources in Canada and Mexico, compared to the situation in the U.S.

In Mexico, initial data on an emissions inventory is being assembled (Cabrera, 1997; Santos-Burgoa, 1997). Initial consideration is being given to the following potential PCDD/F sources currently operating in Mexico: (a) medical waste incinerators; (b) hazardous waste incinerators[3]; (c) the petrochemical industry; (d) cement kilns; (e) combustion of leaded gasoline in automobiles[4]; (f) metallurgical processes (Mexico is a major copper producer); and (g) brick production facilities (where uncontrolled waste combustion is typically used as a heat source).

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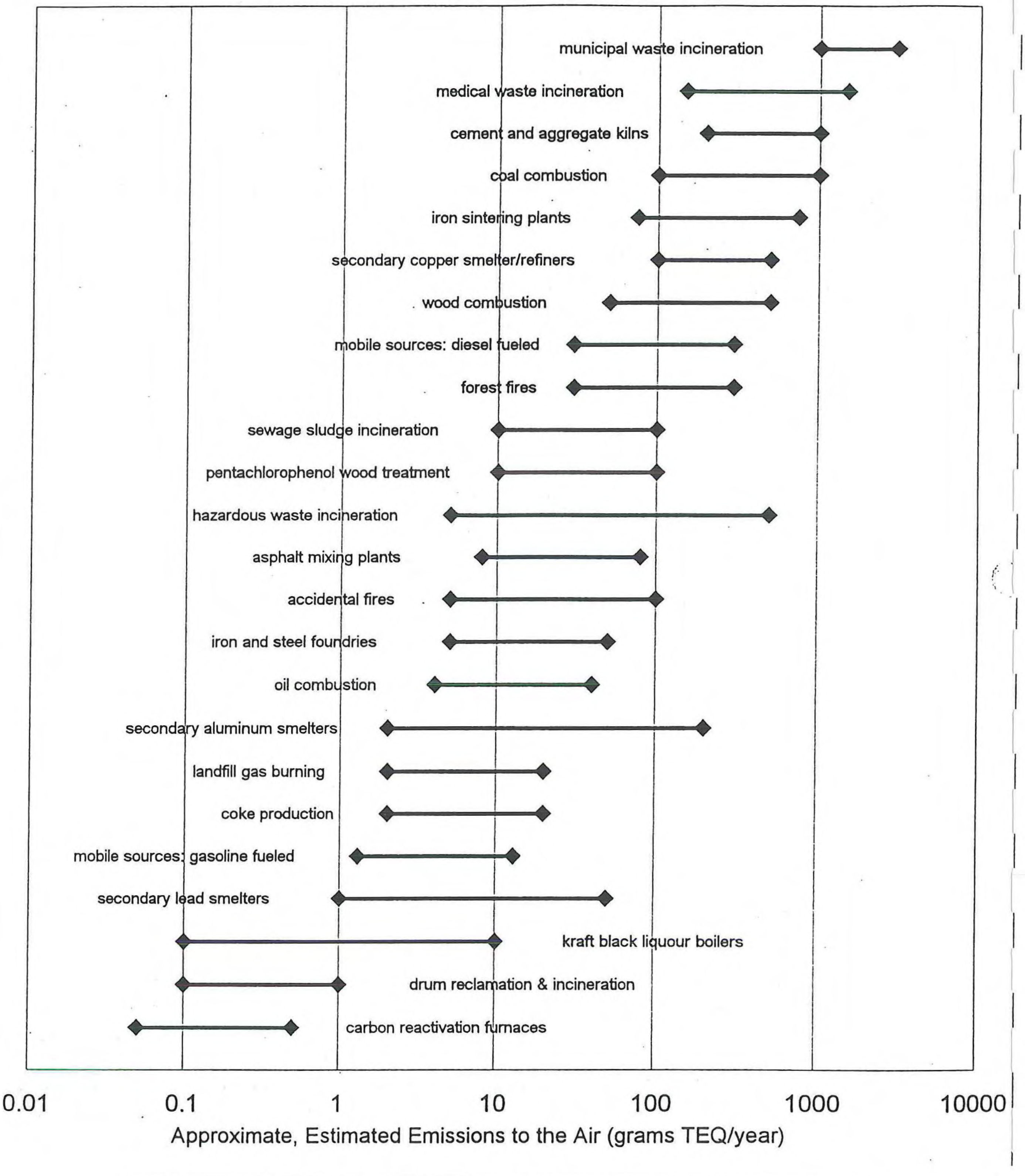
[3]. A new initiative in Mexico has begun in which a system of Integral Centers for the Management and Utilization of Industrial Residues ("Centros Integrales para el Manejo y Aprovechamiento de Residuos Industriales - CIMARI'S") is to be developed. These Centers are expected to be involved in the following aspects of industrial waste management: (a) recycling of oils and solvents; (b) energy recovery from certain combustibles; (c) sludge treatment; (d) treatment of residues with heavy metals; (e) treatment of residues with organic substances; (f) thermal oxidation; and (g) solidified and stabilized waste confinement.

[4]. Although the use of leaded gasoline is being phased out, it is still used somewhat. Thus, emissions from automobiles may play a relatively greater current role as a PCDD/F source in Mexico than it does, for example, in the U.S. and Canada. It should be noted, however, that leaded gasoline is still used as an aviation fuel in the U.S.

Figure 1. Emissions of Dioxin to the Air in the United States: Qualitative Summary



"TEQ" = 2,3,7,8-TCDD Toxic Equivalents
The Indicated "Ranges" for Each Source Show the Approximate Uncertainty in the Emissions Estimate

Future work will attempt to estimate the relative significance of the above sources, and the role of other PCDD/F sources in Mexico.

Many of the sources in the U.S. also exist in Canada, although their relative importance is expected to be somewhat different. A preliminary emissions inventory for Canada was developed by the Center for the Biology of Natural Systems (Cohen *et al.*, 1995). There is a current effort led by the government-sponsored Federal/Provincial Task Force on PCDDs/PCDFs to create a nationwide emissions inventory (Morcos, 1997). A stakeholder workshop about the inventory development is to be held on March 26, 1997 in Ottawa.

Based on their chemical similarity, essentially all of the sources that emit PCDD/F's to the air will also emit other dioxin-like compounds (e.g., dioxin-like PCB's). Emissions data for these other dioxin-like compounds is even more scarce than that for PCDD/F's, however. Congener-specific emissions measurements from sources are practically non-existent.[5] PCB's were extensively used in a range of applications until the 1970's, and residues from these earlier uses are ubiquitous (e.g., PCB-contaminated soil). PCB emissions from these residues of past uses are still occurring. In addition, PCB's are still allowed to be used in certain "closed" applications (e.g., electrical transformers), and there are unintentional, accidental releases from these uses as well.

Thus, the construction of an accurate emissions inventory for PCB's (including dioxin-like PCB's) is perhaps even more complicated than that for PCDD/F's. As a consequence, we are perhaps further away from an accurate emissions inventory for PCB's than we are for PCDD/F's. As an example, in a recent draft PCB emissions inventory for the United States (U.S. EPA 1996a), no congener-specific emissions estimates could be made, emissions from only a few of the source categories considered in Figure 1 could be included, and, no estimates could be made for emissions from PCB-contaminated sites, accidental releases from transformer leaks and fires, or environmental residues from past contamination.

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[5]. Like the PCDD/F's, different PCB congeners exhibit markedly different toxicity characteristics. There are 209 total PCB congeners, of which only 13 are generally considered to be dioxin-like. Based on the commonly used set of estimates by Ahlborg *et al.* (1994), 3,3',4,4',5-PCB (PCB-126) and 3,3',4,4',5,5'-PCB (PCB-169) are considered to have approximate TEF's (their dioxin-like potency relative to 2,3,7,8-TCDD) of 0.1 and 0.01, respectively; all other dioxin-like PCB's are believed to have TEF's of 0.0005 or less, although some of these less toxic congeners are found in such high concentrations that they can contribute significantly to the overall dioxin-like toxicity in particular situations.

4.2 Emissions to Water

[...discussion of at least the following sources to be included...]

pulp and paper mills
organic chemical manufacturing
chlor-alkali production
urban runoff
sewage treatment plants
landfill leachate

4.3 Emissions to Land

In some instances, PCDD/F-containing material is disposed of on land. Current examples of this include disposal of incinerator ash, sewage sludge, and sludge from pulp and paper manufacturing involving chlorine bleaching. In the past, the practice of land disposal of hazardous waste from the chemical manufacturing industry was significant, but, is now restricted. A dramatic example is the Hyde Park landfill in Niagara, New York, about 2000 feet east of the Niagara River, in which an estimated 0.7 - 1.6 tons of 2,3,7,8-TCDD was dumped between 1954 and 1975 (U.S. EPA, 1990). It has been speculated that this site has the most PCDD/F in any single location anywhere on earth[6]. PCDD/F's from this and other sites can be emitted to the air through volatilization and dust erosion processes (wind and mechanical), and to surface water or groundwater. Field and laboratory measurements suggest that PCDD/F's are relatively stable in the soil environment; half-lives appear to be at least on the order of 10 years or longer (Brzuzy and Hites, 1995; Freeman and Schroy, 1989; Hagenmaier *et al.*, 1992; Kapila *et al.*, 1989; Orazio *et al.*, 1992; Puri *et al.*, 1989). The reasons for this stability are the following: PCDD/F's (a) bind relatively tightly to soil particles, (b) are not very water soluble, and (c) are relatively resistant to degradation by microorganisms or by abiotic processes (e.g., photolysis) in the soil environment. Thus, the only way that PCDD/F's can efficiently "leave" the soil environment is when soil particles become airborne (i.e., windblown dust) or waterborne (e.g., soil erosion).

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[6]. To put this amount in perspective, the total amount of PCDD/F emitted to the air in the U.S. is on the order of 5,000 - 10,000 grams TEQ/year — about 10 - 20 pounds TEQ per year. Thus, the amount of dioxin estimated to be in the Hyde Park landfill is the equivalent to approximately 70 - 320 years of current U.S air emissions of PCDD/F.

As mentioned above, leachate and erosion from contaminated waste sites may be an important source of PCDD/F to surface and ground water. While there is unfortunately very little data to make quantitative estimates, waste sites along the Niagara River may be very significant current contributors to the PCDD/F loading of Lake Ontario.

5. EXPOSURE PATHWAYS

PCDD/F's are a ubiquitous contaminant of food, water, air, soil, vegetation and commercial products. Thus, we are exposed to PCDD/F's through the food we eat, the air we breathe, the water we drink, and through dermal (skin) exposure to any contaminated material. Human exposure is defined as the contact of an environmental agent with the human body. Exposure is not equal to dose, but is a realistic approximation to it (Johnson, 1992). Currently in health research, exposure assessment has become a key element of analysis, and for environmental exposures with a long term effect, retrospective exposure assessment is now being developed (Correa et al, 1994). Research has demonstrated the need to consider all the routes of human exposure (e.g., Ott, 1985; Smith, 1988). The most important route of human exposure to PCDD/F is believed to be through food consumption (USEPA 1994a; Zook and Rappe, 1994; Startin, 1994; Schecter, 1994bc).

5.1 Food - Aquatic and Terrestrial

As mentioned above, the most important PCDD/F exposure pathway of most people is believed to be through food, although there have been very few measurements of the PCDD/F levels in various foods. Based on the limited available data, the most important foods contributing to exposure appear to be milk, dairy products, beef[7] and fish (USEPA 1994a; Zook and Rappe, 1994; Startin, 1994; Schecter, 1994bc). Startin (1994) has recently reviewed PCDD/F levels in different foods.

A very approximate summary of the relative PCDD/F contributions of different foods to the diet in the U.S. (from Schecter *et al.*, 1994c) and Canada (from Birmingham *et al.*, 1989) — both based on extremely limited data — is given in Figure 2. Similar results have been found in food surveys done in Germany (Beck *et al.*, 1989; Furst *et al.*, 1990), Britain (Startin 1994), and the Netherlands (Theelan *et al.*, 1993).

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[7]. Other animal products, e.g., eggs, chicken, pork, are also somewhat significant contributors as well, although in the U.S., these are considered to be of lesser importance. A Canadian study estimated that eggs contributed about as much PCDD/F to the diet as beef (Birmingham *et al.*, 1989).

For adults in the U.S., two recent estimates of the total estimated background exposure due to food consumption are 116 pg/day (U.S. EPA 1994a) and 18 - 192 pg/day (Schecter *et al.*, 1994c). Assuming an average adult weight of 65 kilograms, these estimates are equivalent to an exposure of about 2 and 0.3 - 3 pg/kg of body weight, respectively.

For infants, exposure through breast feeding may result in a higher intake of PCDD/F than adults. On a per-kilogram of body weight basis, a breast feeding infant's exposure appears to be *significantly* higher than adults. For infants exposed through breast milk, the exposure has been estimated to be on the order of 35 - 53 pg/kg of body weight (Schecter *et al.*, 1994c).

5.1.1 PCDD/F in Terrestrial Food Chains

Based on their analysis of available data, the EPA estimated that nearly all of the general population's exposure to PCDD/F is through food, of which milk and dairy products account for about 36% and beef for about 32% (USEPA 1994a). These estimates are consistent with those of Schecter *et al.* (1994c), who estimated that dairy products and beef contributed approximately 43% and 48%, respectively, of PCDD/F intake from food for adults[8].

Measurements of PCDD/F's in terrestrial food chains include measurements in beef cattle in the U.S. (Lorber *et al.*, 1996), Caribou in the Canadian arctic (Hebert *et al.*, 1996), and a range of milk and milk products as discussed below.

Given their importance to human exposure, the levels and origin of PCDD/F in dairy products will be discussed here in some detail.

The following measurements have been reported for PCDD/F's in cow's milk in the United States:

(a) a study in the vicinity of the municipal solid waste incinerator in Rutland, Vermont (USEPA 1991), before and after the incinerator began operating, in which a total of 11 milk samples from 4 different sites were analyzed for PCDD/F content;

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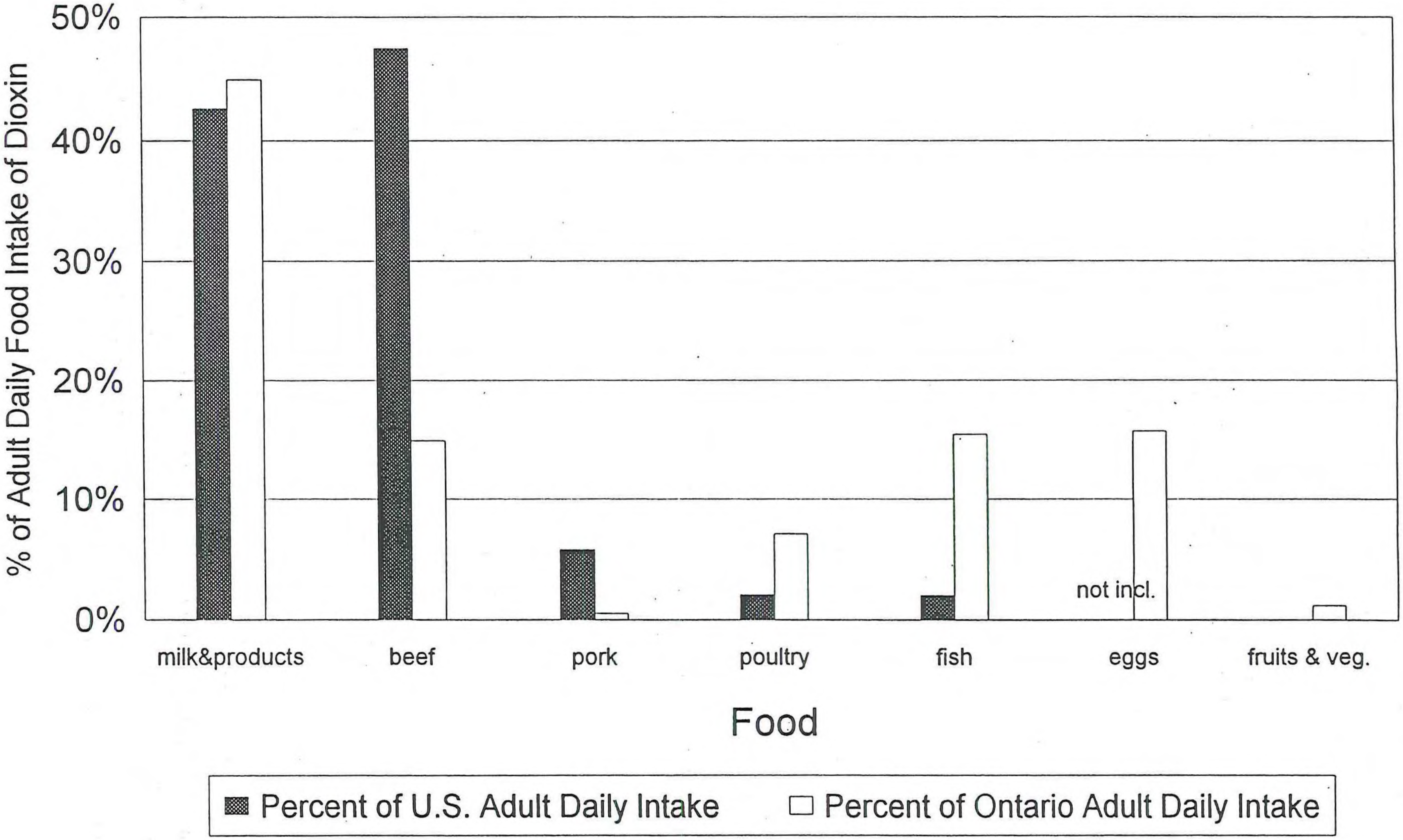
[8]. Schecter *et al.* (1994c) report "low" and "high" estimated exposure rates from food consumption, based on a range of contamination levels in foods. Because the low and high estimates are more than an order of magnitude apart, a geometric (rather than arithmetic) average was used in summarizing this analysis.

Figure 2. Relative Importance of Dioxin Exposure from Different Foods

(Estimates are based on extremely limited data and should be regarded as preliminary approximations only)



U.S. Data: Schecter et al. (1994), Envr Health Persp 102(11): 962-966 (geometric mean of low & high estm's used)
Ontario Data: Birmingham et al. (1989), Chemosphere 19(1-6):507-512.

(b) a study conducted by ENSR at a farm near the Elk River Generating Station in Elk River, Minnesota (ENSR, 1989), in which duplicate samples of cow's milk were collected from a single farm, once in the summer and once in the winter.

(c) a study conducted for the California Air Resources Board (Stanley and Bauer, 1989) in which 8 packaged milk samples were analyzed;

(d) a study by LaFleur *et al.* (1990) in which one sample of milk was analyzed for 2,3,7,8-TCDD, 2,3,7,8-TCDF and 1,2,7,8-TCDF before exposure to bleached paperboard milk cartons and after varying periods of exposure;

(e) a study conducted by the Food and Drug Administration (Glidden *et al.*, 1990) in which a total of approximately 20 milk samples were taken from 15 dairies in 6 states; samples of milk were taken both before and after the milk was placed in cartons;

(f) a study conducted in New York State (Schecter, *et al.*, 1989) in which 3 samples (whole milk, 2% fat milk, and heavy cream) were obtained from a grocery store in Binghamton, New York;

(g) a study conducted at five farms in Connecticut (Eitzer, 1995) in the vicinity of a new municipal solid waste incinerator, before and after the incinerator began operating, in which a total of 29 samples were analyzed.

In Canada, retail milk products purchased between 1985 and 1988 in six different Canadian cities were analyzed for PCDD/F (Ryan *et al.*, 1991). It was found that PCDD/F's from the chlorine-bleached plastic-coated paperboard containers appeared to be leaching into the milk products, adding significantly to the levels of PCDD/F's in the milk.[9]

There are no data available on the PCDD/F content of milk in Mexico.

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[9] Ryan *et al.* (1991) state that recent (ca 1990) efforts in Canada have been successful in producing paperboard containers with substantially lower levels of PCDD/F's. (Among other changes, the substitution of chlorine dioxide for elemental chlorine as a bleaching agent at many pulp/paper manufacturing facilities has led to significant reductions in the levels of PCDD/F's generated in their processes.)

There have been several studies conducted in Europe of PCDD/F in cow's milk, including: Fanelli *et al.* (1980), Rappe *et al.* (1987), Riss *et al.* (1990), Rappe *et al.* (1990a), Beck *et al.* (1990a,b), Ryan *et al.* (1990 & 1991), Schmid and Schlatter (1992), Furst *et al.* (1992 & 1993), DeJong *et al.* (1993), Lassek *et al.* (1993), Slob *et al.* (1995), and Harrison *et al.* (1996).

The U.S. EPA (1994a) estimated the U.S. background PCDD/F concentration in milk using data from LaFleur *et al.* (1990) and the Rutland study (USEPA 1991). These estimates, coupled with estimates of PCDD/F content of other foodstuffs, air, and water, were used to estimate the total exposure of the general population.

The U.S. EPA is currently conducting a nationwide survey of PCDD/F levels in cow's milk, using a nationwide sampling network originally designed to monitor milk contamination by radioactive fallout. Samples are being collected from dairy facilities serving at least one large metropolitan area in each state. The analysis for PCDD/F may be done on regionally-pooled composite samples. In addition, the U.S. Food and Drug Administration is conducting a market-basket survey of PCDD/F in milk, dairy products and fish. These studies will provide much-needed additional data on PCDD/F levels in food in the U.S.

How do PCDD/F's get into dairy and animal products? The major route of exposure of dairy cows, beef cattle and other animals to PCDD/F is thought to be their consumption of PCDD/F-contaminated feed crops (McLachlan *et al.*, 1990; Furst *et al.*, 1993; Stevens and Gerbec, 1988). PCDD/F's enter crops largely from the air; they are not generally taken up by plants from soil or groundwater (McCrady *et al.*, 1990). Dry deposition of vapor-phase material may be the dominant mechanism by which they enter vegetation (Welsch-Pausch *et al.*, 1995). As discussed below, the more toxic tetra- and penta-chloro PCDD/F congeners (i.e., TCDD/F and PeCDD/F) are relatively volatile and exist to a greater extent in the vapor phase in the atmosphere than do the less toxic higher-chlorinated congeners. Thus, it is "unfortunate" that vapor-phase PCDD/F's appear to be more efficiently transferred to vegetation.

Ingestion of contaminated soil is normally of minor importance but can be significant in some situations (Fries 1995ab). Soil contamination by PCDD/F can arise from atmospheric deposition and from the application of PCDD/F-contaminated fertilizers and soil amendment products (e.g., sewage sludge, animal manure, etc.). The use of PCDD/F-contaminated pesticides *may* be another contributor to the levels of PCDD/F's in animal feed crops, although it is probably of lesser importance.

PCDD/F's ingested by most animals are not broken down efficiently by metabolic pathways; for example, approximately 20% of the PCDD/F ingested by dairy cows is excreted in their milk (McLachlan *et al.*, 1990). Recent reviews of PCDD/F's in agricultural systems include those by Fries (1995ab), Hattemer-Frey and Travis (1991), Lorber *et al.* (1994), and McLachlan (1996).

A collaborative project involving the Center for the Biology of Natural Systems at Queens College, the New England Environmental Policy Center (director: Jean Richardson), and the University of Wisconsin's Department of Dairy Science is currently being conducted in which PCDD/F is being measured and modeled in the air, crops, and cow's milk of eight dairy farms in Wisconsin and Vermont. It is hoped that this study will lead to an increased understanding of the modes of entry, relative levels, and fate of PCDD/F in dairy ecosystems.

5.1.2 PCDD/F in Aquatic Food Chains

The primary pathway of human exposure as a result of PCDD/F contamination of aquatic ecosystems is through the consumption of contaminated fish. PCDD/F's can enter aquatic ecosystems as a result of:

> sources which discharge PCDD/F's directly to the water (e.g, a pulp and paper mill using chlorine or a chlorine-containing compound for pulp bleaching);
>
> sources which discharge PCDD/F's to the air, a portion of which may enter a given water body as a result of atmospheric deposition.

PCDD/F's bioconcentrate in the aquatic food chain and can reach significant levels, even though they are rarely (if ever) found above detection levels in the water column. Recent reviews of PCDD/F's in aquatic ecosystems include those by Cook *et al.* (1991), Fletcher and McKay (1993), and Walker and Peterson (1994). Clarke *et al.* (1996) have recently reviewed the levels of PCDD/F's found in fish in the United States and international waterways. Their review indicated that PCDD/F contamination in fish is ubiquitous, with quantifiable levels found worldwide.

> *(Note: probably should add a "synthesis" paragraph here, summarizing key aspects of PCDD/F in aquatic ecosystems)*

A few examples of the range of studies that have been done on PCDD/F's in aquatic ecosystems will be given.

A Canadian survey of fish and sediments near pulp mills was completed in the early 1990's [some of these data were reported by Muir *et al.* (1992); most is contained in reports (e.g., Trudel 1991)]. MacDonald *et al.* (1992) and Yunker and Cretney (1996) have discussed levels and sources in biota near pulp mills in British Columbia.

(...brief discussion of these and possibly other data to be added]

PCDD/Fs have also been reported in marine mammals in Canadian waters (St. Lawrence River estuary, west coast and Arctic) and, of course, in fishes

(... references and brief descriptions of findings to be added...).

A long-term monitoring program of the eggs of herring gulls (a fish eating bird species) has provided useful information on the spatial and temporal variation of PCDD/F's in the Great Lakes regional aquatic ecosystem (e.g., Hebert *et al.*, 1994). Measurements showed a general decline in PCDD/F levels between 1981 and 1984, but no discernable trend between 1984 and 1991. Birds eating fish in Saginaw Bay (Lake Huron) and Lake Ontario showed significantly higher levels of PCDD/F in their eggs than those in other colonies in the Great Lakes region.

There have been several studies of PCDD/Fs in aquatic sediments in Canadian waters (Macdonald *et al.* 1992; Evans *et al.*, 1996; Bourbinierre *et al.*, 1996) and in the Great Lakes (Czcuzwa and Hites 1984; 1986). The sediment studies are very useful for identifying trends in sources and historical inputs. Results from Great Slave Lake (Evans *et al.*, 1996) and Lake Athabasca (Bourbonniere *et al.*, 1996) suggest a small preindustrial historical input of OCDD, presumably due to forest fires.

The fate and distribution of PCDD/F's in aquatic ecosystems has been examined in a series of studies in an experimental mesocosm (Corbet et al., 1983; Servos et al., 1992ab; Friesen et al., 1995). These studies have shown that... *(add brief synthesis and/or description of results)*

5.2 Other Exposure Pathways: Drinking Water, Inhalation, Skin Adsorption

Under typical "background" conditions, i.e., removed from any strong local source of PCDD/F contamination, exposure pathways other than food consumption are believed to be of minor significance.

Using an estimated background drinking water PCDD/F contamination level of 0.0056 pg TEQ/liter (based on 214 measurements) and a typical adult water consumption rate of 1.4 liters/day, the U.S. EPA (1994a) estimated that exposure from water consumption was on the order of 0.008 pg TEQ/day, about 0.01% of the typical exposure due to food consumption.

Using an estimated background air concentration of 0.095 pg TEQ/m^3 (based on 84 measurements) and a typical adult inhalation rate of 23 m^3/day, the U.S. EPA (1994a) estimated that exposure from air inhalation was on the order of 2 pg TEQ/day, perhaps accounting for a few percent of the typical background exposure due to food consumption. A similarly small amount (~ 1 pg/day) was estimated for soil ingestion (U.S. EPA 1994a), under background conditions.[10]

Dermal exposure is believed to be important in some circumstances for certain pollutants (e.g., pesticide exposure of agricultural workers involved in applying pesticides (Granjean, 1990). Investigations into dermal exposure to PCDD/F's have shown that the skin is not an effective barrier against PCDD/F (e.g., see discussion in Montague, 1989). Under background conditions, however, dermal exposure to PCDD/F is not expected to constitute a significant exposure pathway.

Under certain "non-background" situations, however, these other PCDD/F exposure pathways may be important.

In some occupational settings, for example, exposure to PCDD/F (presumably through air and dust inhalation) may be significant. Any occupational environment associated with a significant PCDD/F source (see Figure 1) could involve significant occupational exposure. An example is municipal waste incinerator operation, where it has been found, for some plants, that workers show evidence of a higher level of exposure than the general population (presumably as a result of exposure to PCDD/F- contaminated air and lust) (Schecter 1994b).

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[10]. Soil ingestion (called "pica") is a more significant exposure route for children than it is for adults (Hayes and Rodenbeck, 1992).

Even for some non-occupational situations, non-food exposure routes *may* be important. Ground level concentrations downwind of facilities which emit very large quantities of PCDD/F's can reach significant levels, and people who live or work near such a facility might be significantly exposed through inhalation or dust ingestion.

As an illustration of this, consider the data presented in Figure 3, below. Here, the

inhalation exposures predicted for an adult living and working downwind of PCDD/F sources of varying magnitudes are compared against the range of background food exposures as presented by Schecter (1994c). The inhalation exposure estimates in this figure are based on air dispersion calculations presented by the U.S. EPA (1994a) for the annual average ground-level air concentrations (in the direction of maximum impact) resulting from emissions from a hypothetical facility[11]. It can be seen from this example that persons in close proximity to sources or source complexes emitting on the order of 100 g TEQ per year or larger may be exposed to significant amounts of PCDD/F (i.e., rivaling or surpassing the exposure from food consumption).

Are there any facilities with such large PCDD/F emissions? Certain municipal waste incinerators with "hot-sided" electrostatic precipitators (as discussed above in the section on sources) may have emissions of this magnitude. For example, during a 1992 stack test of a municipal waste incinerator in Columbus, Ohio, the emissions rate was estimated to have been on the order of 1000 g TEQ per year. When the facility was tested more recently, the level fell to approximately 200 grams TEQ per year, possibly the result of "special" waste preparation: the waste was allegedly pre-selected, dried, and plastics were removed (Connett, 1994)[12]. Other similar incinerators have been found to emit PCDD/F at rates of 100 g TEQ/yr or more (Webster and Connett, 1997). Certain large iron sintering plants may emit PCDD/F at a similarly high rate (based on emissions factors extrapolated from European facilities), but, data on their emissions in North America are almost non-existent.

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[11]. The hypothetical facility was modeled after an actual waste incinerator burning 200 metric tons per day. In this example, the facility is surrounded by flat terrain and has a 30.5 meter tall stack. Other important parameters for the simulation include the following: (a) the stack diameter is assumed to be 1.52 meters; (b) the exhaust gas temperature is assumed to be 127 °C; and (c) the exhaust gas exit velocity is assumed to be 8.9 meters/sec. The distance from the source at which the maximum annual-average ground-level concentration was predicted to be approximately 900 meters. A distance of 1 kilometer (with virtually the same annual average concentration as at 900 meters) was chosen for the calculations shown in Figure 3.

[12]. This facility was recently closed down.

cancer risk of 330 per million, well above the "acceptable" level (Commoner, Webster & Shapiro, 1986). If exposure to dioxin-like PCBs is included as well, the risk is 500-1000 per million, according to the U.S. EPA 1994 dioxin assessment (U.S. EPA, 1994b).

6.2.2 Effects on Reproduction and Development

Numerous experiments show that laboratory animals exposed to low (i.e., environmental) levels of PCDD/F's exhibit a number of reproductive and developmental abnormalities, generally due to disruptive effects on the relevant hormone systems. These abnormalities include reduced sperm count, reduced fertility, decreased birth weight, reduced levels of sex hormones, and birth defects[15] (e.g., cleft palate in mice). Similar effects have been observed in wildlife exposed to environmental levels of PCDD/F, for example defective egg development in fish-eating birds in the Great Lakes (U.S. EPA, 1994b).

Some of the effects observed in laboratory animals occur in humans as well. Thus, testosterone levels are reduced in rats exposed experimentally to PCDD/F, and in workers exposed in industrial situations as well (U.S. EPA, 1994b; DeVito *et al.*, 1995). Similarly, maternal exposure to PCDD/F is associated with the decreased birth weight of offspring in both mice and humans (DeVito *et al.*, 1995). Such effects generally occur at considerably lower levels of exposure than the levels that induce increased cancer incidence. Thus, while increased incidence of cancer among laboratory animals has been observed at exposure levels of 944-137,000 pg/kg body weight, non-cancer developmental effects occur at body burdens of 10-12,500 pg/kg -- more than an order of magnitude lower (U.S. EPA, 1994b; DeVito *et al.*, 1995).

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[15]. Environmental, nutritional and genetic factors are related to birth defects (Cordero, 1994), and birth defects are being monitored through active and passive regional surveillance systems (Santos-Burgoa *et al.*, 1996) in areas with high levels of pollution. Causal relationships have not yet been developed, but, this research may lead result in an increased understanding of the toxic effects of PCDD/F and other pollutants.

6.2.3 Other Non-Cancer Effects

The immune system appears to be particularly sensitive to the disruptive effects of PCDD/F's, leading, for example, to increased susceptibility to viral infections in mice. Nervous system defects may also occur; for example, low level exposure of pregnant monkeys to PCDD/F's results in defective learning behavior. Endometriosis, the proliferation of cells that line the uterus outside that organ occurs at an enhanced rate in monkeys exposed to low doses of PCDD/F's (DeVito *et al.*, 1995).

In spite of all that has been learned about the range of non-cancer health consequences of exposure to PCDD/F and other dioxin-like substances, there are still many uncertainties.

7. CONTINENTAL POLLUTANT PATHWAYS

There are many pathways through which human exposure to PCDD/F's can result from transport of material over continental distances.

7.1 Long Range Atmospheric Transport of PCDD/F's

The ability of different PCDD/F congeners to be transported long distances in the atmosphere depends on their atmospheric "lifetimes." Many different phenomena influence the fate of PCDD/F congeners in the atmosphere. The primary mechanisms which limit the atmospheric lifetime (and hence transport) of PCDD/F's are believed to be:

- chemical transformation, including photolytic destruction by ultraviolet light and chemical reaction (e.g., with hydroxyl radical); and

- fallout (also called "deposition") as a result of precipitation ("wet deposition") and as a result of the impaction of particles and the adsorption/absorption of gases at the earth's surface ("dry deposition").

7.1.1 Vapor/Particle Partitioning

The fate of "semivolatile" pollutants (such as PCDD/F's) at any given location in the atmosphere depends intimately on whether the pollutant is in the vapor or particle phase (Junge, 1978; Bidleman, 1988). The extent of vapor/particle partitioning depends intimately on the physical-chemical properties of the pollutant, the ambient temperature, and the detailed nature of the local atmospheric aerosol.

Because of particularly low volatility, PCDD/F congeners with six, seven, and eight chlorines tend to exist primarily in the particle phase in the atmosphere. Those with four and five chlorines — including some of the most toxic congeners — are believed to exist in significant proportions in both the vapor and particle phase in the atmosphere, based on both theoretical considerations and experimental evidence.

These tetra-chloro (TCDD/F) and penta-chloro (PeCDD/F) congeners may move relatively freely between the two phases, depending on the local atmospheric conditions (i.e., they may be "exchangeable.") There is some experimental evidence which suggests that atmospheric PCDD/F is exchangeable. Eitzer and Hites (1989a and 1989c) measured the amounts of vapor-phase and particulate-phase PCDD/F in the atmosphere of Bloomington, Indiana and found that while there was no temperature-related effect on the total concentration of PCDD/F in the atmosphere, the proportions in the two phases were dependent on the ambient temperature at the time of the measurement. Furthermore, in agreement with theory, it was found that the vapor/particle partitioning of each of the congeners was, in general, critically dependent on each congener's subcooled liquid vapor pressure (Eitzer and Hites, 1989a). Chemical Transformation in the Atmosphere

When a PCDD/F molecule is in the particle phase, it is believed to be much less vulnerable to photolytic destruction by ultraviolet light (Koester and Hites, 1992a) and chemical reactions in the atmosphere than when it is in the vapor phase. In the vapor phase, the rates of destruction by ultraviolet light and chemical reaction depend, or course, on the detailed characteristics of the atmospheric micro-environment at any given location, and, will be different for different congeners at the same location. It is not possible to predict the rate of photolytic destruction or chemical reaction of a vapor-phase (or particle-phase) PCDD/F congener at any given location in the atmosphere with great accuracy at the present time, *even* if details about the concentrations of reactants and electromagnetic spectrum were known at that location. Rough approximations can be made, however. Atkinson and coworkers (Atkinson, 1991; Kwok *et al.*, 1994; Kwok *et al.*, 1995) have considered available theoretical and experimental evidence and have concluded the following:

- Reactions of gas-phase PCDD/F's with ozone (O_3), nitrate radical (NO_3), or the HO_2 radical are not expected to be important atmospheric loss mechanisms;

- Reaction of gas-phase PCDD/F's with hydroxyl radical (OH) is an important atmospheric loss mechanism, with reaction rates varying among different congeners;

- Photolysis

    - *may* be somewhat important for vapor-phase tetrachloro-dibenzo-p-dioxins (TCDD's), although less important than reaction with hydroxyl radical;

    - is probably not important for pentachloro-dibenzo-p-dioxins (PeCDD's);

    - because OH radical reactions are predicted to be slower with PCDF's than with PCDD's, photolysis may be relatively more important for PCDF's.

Building upon the work of Atkinson and coworkers (e.g., Kwok and Atkinson, 1995 and references cited therein), Meylan and Howard (1993; 1996ab) have developed an improved estimation methodology for the prediction of hydroxyl radical reaction rates with gas-phase PCDD/F's and other compounds. Using reaction rates estimated with their methodology, together with an estimated average hydroxyl radical concentration of 1.5×10^6 molecules per cm^3, atmospheric vapor-phase half-lives (relative only to hydroxyl radical attack) for 2,3,7,8-TCDD, 2,3,7,8-TCDF, 1,2,3,7,8-PeCDD, 1,2,3,7,8-PeCDF, and 2,3,4,7,8-PeCDF are estimated to be 5.3, 65.0, 6.2, 143.5, and 143.5 days, respectively.

Given a lack of data, it is difficult to make quantitative estimates of the half-lives of vapor-phase PCDD/F molecules with respect to photolysis (i.e., destruction by ultraviolet light). Based on experimental measurements, Dulin *et al.* (1986) estimated the half-life of 2,3,7,8-TCDD *in water* to be on the order of 4-5 days in the summer at a 40° latitude. Extrapolation of solution-phase photolysis rates to the vapor phase is not straightforward, as the processes occurring in solution can be more complex than the "simple" photolytic phenomena in the vapor phase. Nevertheless, as assumed by Atkinson (1991) the vapor-phase half-life of 2,3,7,8-TCDD relative to photolysis *may* be similar to that in the aqueous phase; if so, the rate of photolytic destruction of vapor-phase 2,3,7,8-TCDD would be of the same order as the destruction due to reaction with hydroxyl radical.

Photolysis rates for different PCDD/F congeners appear to decrease, generally, with increasing numbers of chlorines (e.g., Choudry and Webster, 1989; Sivils *et al.*, 1994), and so, photolysis of PeCDD/F congeners (those with five chlorines) would be expected to be slower than TCDD/F congeners (with four chlorines). In solutions, it has been found that OCDF is more vulnerable to photolytic destruction than OCDD (Wagenaar *et al*, 1995). If this relative vulnerability is true for other dibenzofurans, the vapor-phase photolysis of 2,3,7,8-TCDF may be more rapid than that of 2,3,7,8-TCDD, and the rate for PeCDF's may be more rapid than that for PeCDD's.

A large area of uncertainty concerns the products of the atmospheric degradation of PCDD/F's. If the atmospheric degradation of a polychlorinated PCDD/F molecule results in the same compound with one less chlorine, then the toxicity can be increased (as can be seen in Table 1 at the beginning of this case study, toxicities of PCDD/F compounds generally increase as the number of chlorines decrease). Degradative dechlorination has been observed in the environment (e.g., Miller *et al.*, 1989).

It has been observed that while the emissions of PCDD/F's are relatively equally distributed among the different homologue groups, the atmospheric concentrations and deposition tend to be heavily skewed towards the higher chlorinated congeners. This suggests that the dechlorination of higher-chlorinated congeners leading to the more toxic tetra- and penta-chlorinated congeners is not, ultimately, of major importance.

Perhaps the reason for this is that the rate of destruction increases as the number of chlorines falls, so, once the degradation starts, there is a cascade of dechlorination events that proceeds relatively quickly. In other words, once the conditions in the local atmosphere are "severe" enough to degrade a higher-chlorinated congener to a lower-chlorinated congener, they are more than sufficient to degrade the resulting lower-chlorinated congener. Thus, in these circumstances, the potentially more toxic intermediates may have very short lifetimes.

7.1.2 Atmospheric Deposition of PCDD/F's

Dry deposition at the earth's surface is a relatively important loss process for vapor-phase PCDD/F's, but, this process does not deplete the total atmospheric burden of PCDD/F very rapidly. Due to very limited water solubility, wet deposition of vapor phase PCDD/F's is not expected to be an important fate pathway.

Thus, based on the chemical transformation mechanisms discussed above, even if they existed completely in the vapor phase (which they don't) the tetra- and penta-chloro dibenzo-p-dioxin and dibenzofuran congeners are expected to have atmospheric lifetimes of 1-2 days or longer.

Portions of the tetra- and penta- congeners and essentially all of the hexa-, hepta- and octa-chloro congeners are associated with particles in the atmosphere, and fate processes affecting particulate pollutants will govern their atmospheric lifetime.

Particles in the atmosphere are generally classified into three separate size ranges: the "nucleation range", comprising particles with diameters less than approximately 0.05 µm (1 µm = 1×10^{-6} meters); the "accumulation range", comprising particles with diameters roughly between 0.05 µm and about 2 µm; and the "coarse particle range", consisting of particles with diameters greater than about 2 µm in diameter (e.g., Lodge, 1981; Prospero *et al.*, 1983; Seinfeld, 1986; Pacyna, 1995). Particles in the two smallest ranges are sometimes grouped together in a "fine particle" category.

Particles in the nucleation range are subject to coagulation and incorporation into larger particles; thus, the material contained in them is generally transferred to larger particles, generally in the accumulation range. The time scale for such processes is on the order of hours (Pacyna, 1995). Particles greater than about 10 µm -- generated primarily by mechanical actions and wind erosion -- have relatively fast sedimentation rates, and can be removed relatively quickly by dry deposition from the atmosphere. Typical residence times for such large particles are on the order of a few hours. Particles in the accumulation range, on the order of 1 µm in size, do not efficiently coagulate into larger particles, and do not have large sedimentation velocities. They can be removed by wet and dry deposition processes, but, have atmospheric lifetimes on the order of 7 - 10 days (Pacyna, 1995; Prospero, 1983). Since the fate of nucleation range particles is often to end up quickly incorporated into the accumulation range, the atmospheric lifetime of the *pollutants* associated with these small particles is also on the order of 7-10 days. Manchester-Neesvig and Andren (1989) used an average residence time for atmospheric particles in the Northern Hemisphere of 6 days in their estimation of the particle-mediated atmospheric lifetime of particle-bound PCB's.

Windblown soil particles are often relatively large, and fall into the coarse particle range. However, a fraction of such aerosols are fine particles (e.g., Pacyna, 1995).

Particles emitted from combustion and incineration processes will generally fall into the fine-particle category. Vapor/particle partitioning is often considered a surface phenomenon, and partitioning to particles is often assumed to scale with particle surface area. That is, when vapor-phase material condenses onto particles, it is considered to do so relatively equally onto each available surface, on a per-area basis. The most significant portion of the surface area available for adsorption is with the fine particles. Thus, for exchangeable, semivolatile compounds that partition between the particle and vapor phases, a significant fraction of their particle-associated mass will lie in the fine-particle range.

Measurements of the size of particles to which semivolatile compounds in the atmosphere are absorbed are difficult, and many factors can bias the results (e.g., Poster *et al.*, 1995). For PCDD/F's, such a measurement was attempted by Kaupp *et al.* (1994). In this study, three samples were collected in a rural area (at the University of Bayreuth, Germany), during periods with mean ambient temperatures from 17.4 - 21.9 °C. PCDD/F's were found in both the vapor- and particle phase. Approximately 90% of particle-phase PCDD/F's were associated with particles less than

1.35 µm in diameter; approximately 50 - 60% of the particle-phase PCDD/F's were associated with particles less than 0.45 µm in diameter. It was found that smaller particles tended to have higher concentrations of PCDD/F's than bigger particles, consistent with a surface-area-related partitioning phenomena.

Thus, to the extent that they are associated with particles, PCDD/F's are expected — on the basis of theoretical and experimental considerations — to exist predominantly in the fine particle range, with sizes less than approximately 2 µm.

The atmospheric lifetime of pollution associated with such particles will be highly variable depending on meteorological conditions, source characteristics, and particle characteristics, but, average atmospheric lifetimes of such particle-associated pollution will be on the order of 1 week (or more). Depending on the meteorological conditions, such particles could travel hundreds to thousands of kilometers in the atmosphere before they are removed by wet or dry deposition processes.

7.1.3 Summary: Atmospheric Lifetime of PCDD/F's

Based on the foregoing considerations, higher chlorinated PCDD/F congeners (i.e., those with 6, 7 and 8 chlorines) are expected to exist largely associated with small particles in the atmosphere, and will have atmospheric lifetimes of roughly one week. Significant fractions of the more toxic tetra- and penta-chloro dioxin congeners are expected to exist in both the vapor and particle phases in the atmosphere. Their atmospheric lifetimes are expected to be on the order of a few days to a week. Over such periods, PCDD/F's can be transported in the atmosphere over regional and continental scales.

7.1.4 Modeling the Long-Range Atmospheric Transport of PCDD/F's

In an air pollution model, an attempt is made to mathematically express and combine all the relevant factors to quantitatively estimate the fate and transport of atmospheric pollutants. One useful type of information that can be obtained from modeling is information about source-receptor relationships; i.e., estimates of the relative contribution of specific sources or source regions to the concentrations or deposition at a given receptor. If an eventual policy goal is to lower the impact at the receptor, then the sources contributing to that impact must first be characterized.

There have been few attempts to model the regional and long-range transport of PCDD/F's in the atmosphere. These include the following:

- Rappe *et al.* (1989) and Tysklind *et al.* (1993) compared measured atmospheric concentrations of PCDD/F at locations in Sweden to modeled back-trajectories of the air masses sampled, and found evidence that long-range atmospheric transport of these compounds was occurring.

- Van Jaarsveld and Schutter (1993) modeled regional and long-range transport and deposition of PCDD/F's in portions of Europe by adapting a Lagrangian model developed originally for analysis of acidic deposition. Specific source-receptor relationships were not reported.

- In a current study, the short-, medium-, and long-range transport of PCDD/F's from specific sources and source regions to dairy farms in Wisconsin and Vermont is being modeled. Measurements of PCDD/F's in the air above the farms, in the crops, and in the milk are also being made and will be used to test the validity of the model.[16]

- Currently, the U.S. EPA is attempting to simulate the long-range transport of PCDD/F's using the RELMAP air pollution model (Cleverly, 1996).

- In a study of the sources of PCDD/F's to the Great Lakes (Cohen *et al.* 1995), the regional and long-range atmospheric transport of PCDD/F's from air emissions sources in the United States and Canada was analyzed using a model developed by the U.S. National Oceanic and Atmospheric Administration (Draxler 1991, 1992, 1994). A few details of the results of this study will be described below.

In the Great Lakes study, algorithms were added to the NOAA model (HYSPLIT) to simulate the atmospheric vapor/particle partitioning behavior of PCDD/F's. Relatively good agreement was found between predicted and measured concentrations of atmospheric PCDD/F at Dorset, Ontario.

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[16]. This ongoing work is a collaborative project with the principal involvement of the following people: *Mark Cohen*, *Barry Commoner*, and others at the Center for the Biology of Natural Systems at Queens College; *Jean Richardson* of the New England Environmental Policy Center and the University of Vermont; *Sarah Flack* of Farm Management Consulting in Vermont; *Randy Shaver*, *Janet Reisterer* and others at the University of Wisconsin-Madison Department of Dairy Science; *Bruce Maisel* of ENSR Consulting; *Mark Horrigan*, *John Stanley* and others at the Midwest Research Institute.

An attempt to characterize source-receptor relationships was made in this analysis. Approximately half of the predicted atmospheric deposition of PCDD/F's (on a TEQ basis) was linked to sources in states and provinces immediately adjoining the Great Lakes; the other half was associated with sources in the rest of the U.S. and Canada. Sources in Mexico and the rest of the world were not considered; if they had been included, it is expected that they would have made a lesser but perhaps not insignificant contribution to the total predicted loadings.

In Figure 4, an example of the cumulative relationship between the sources' distance from a particular Lake (in this case Lake Michigan) and the relative PCDD/F contribution (on a total TEQ basis) is shown. For this lake (similar to other Lakes), it was found that about half of the cumulative PCDD/F deposition comes from sources about 300 miles (480 kilometers) or less from the center of the Lake — that is, located in the U.S. states and the province of Ontario that border the Lake. The remaining half of the total deposition comes from sources as far as 1,500 miles (2,400 kilometers) away.

The extent to which the different sources contribute to the PCDD/F deposited in the Great Lakes was found to depend not only on the amounts they emit and their distances from the lakes, but also on their geographic location. A higher percentage of the PCDD/F emitted from sources to the south and west of the lakes is deposited in them than from the sources to the north and east. Figure 5 illustrates this effect in the case of Lake Michigan, again, on a total TEQ basis.

Another illustration of the effect of the weather pattern in influencing the efficiency with which a given source contributes PCDD/F to a given receptor is given in Figure 6, again with Lake Michigan as the example. For this purpose the entire U.S. and Southern Canada area was divided into 20,000 squares (each 270 square miles in area). Using the air pollution model, the percent of individual PCDD/F congeners emitted from each square's center point that would be transported through the atmosphere and deposited in Lake Michigan was estimated (i.e., the "air transfer coefficient" was estimated for a series of potential source locations to a given receptor, Lake Michigan). In Figure 6, the results for a particular congener (2,3,4,7,8-pentachloro-dibenzofuran) are shown. The figure maps the geographic distribution of six successive ranges of air transfer coefficient. It shows that 2,3,4,7,8-PeCDF transport and eventual deposition to Lake Michigan is most efficient for sources to the west and southwest of the Lake, and least efficient for sources to the northeast and southeast of the lake. This was found for the other PCDD/F congeners as well, and is reflective of the general southwest-to-northeast and west-to-east prevailing weather patterns in the region.

Figure 4. The Effect of Distance of Sources on Cumulative Deposition of Dioxin in Lake Michigan

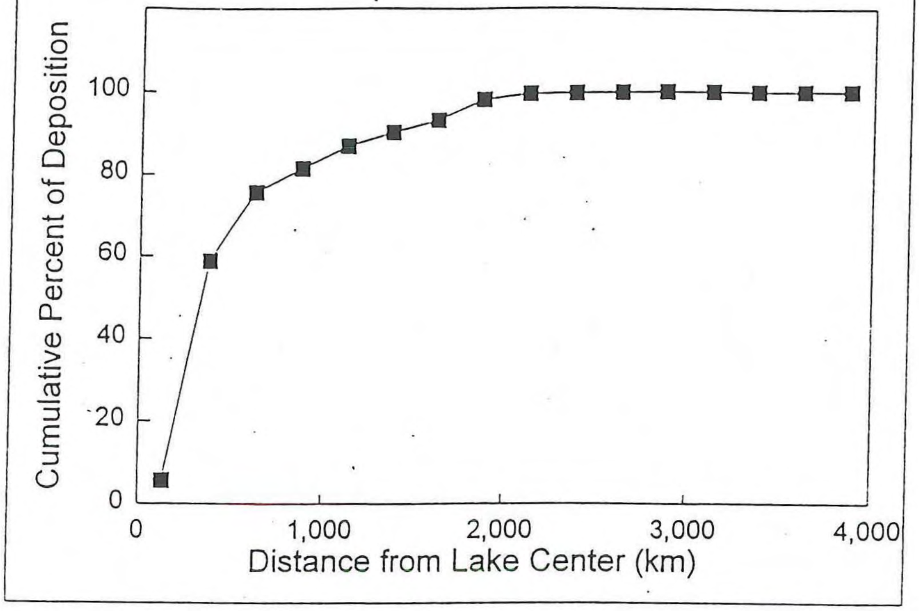


Figure 5. The Relation Between the Directional Orientation of Dioxin Sources and their Emissions and Deposition in Lake Michigan

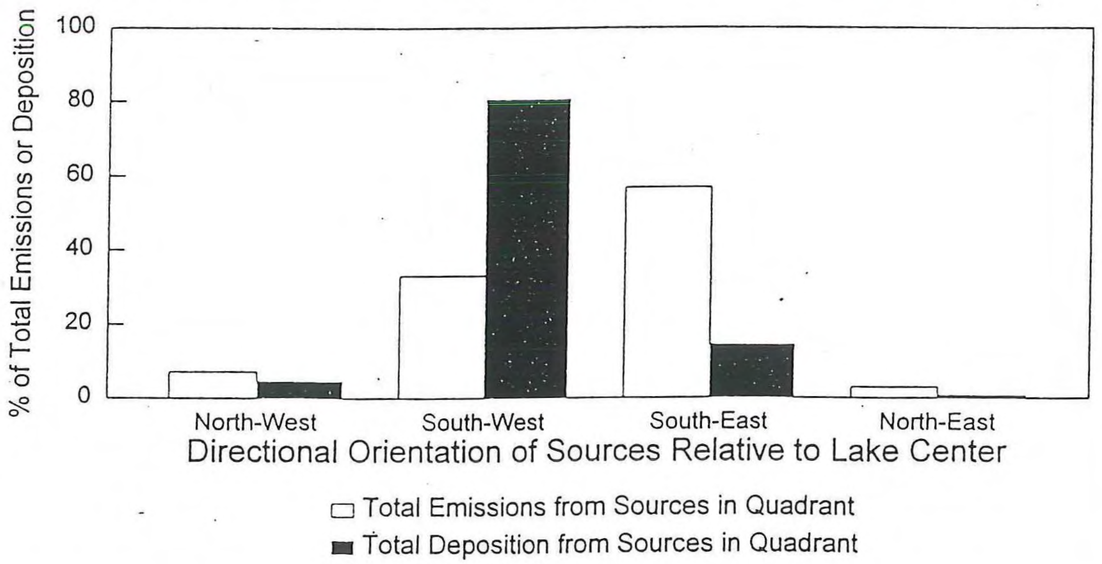
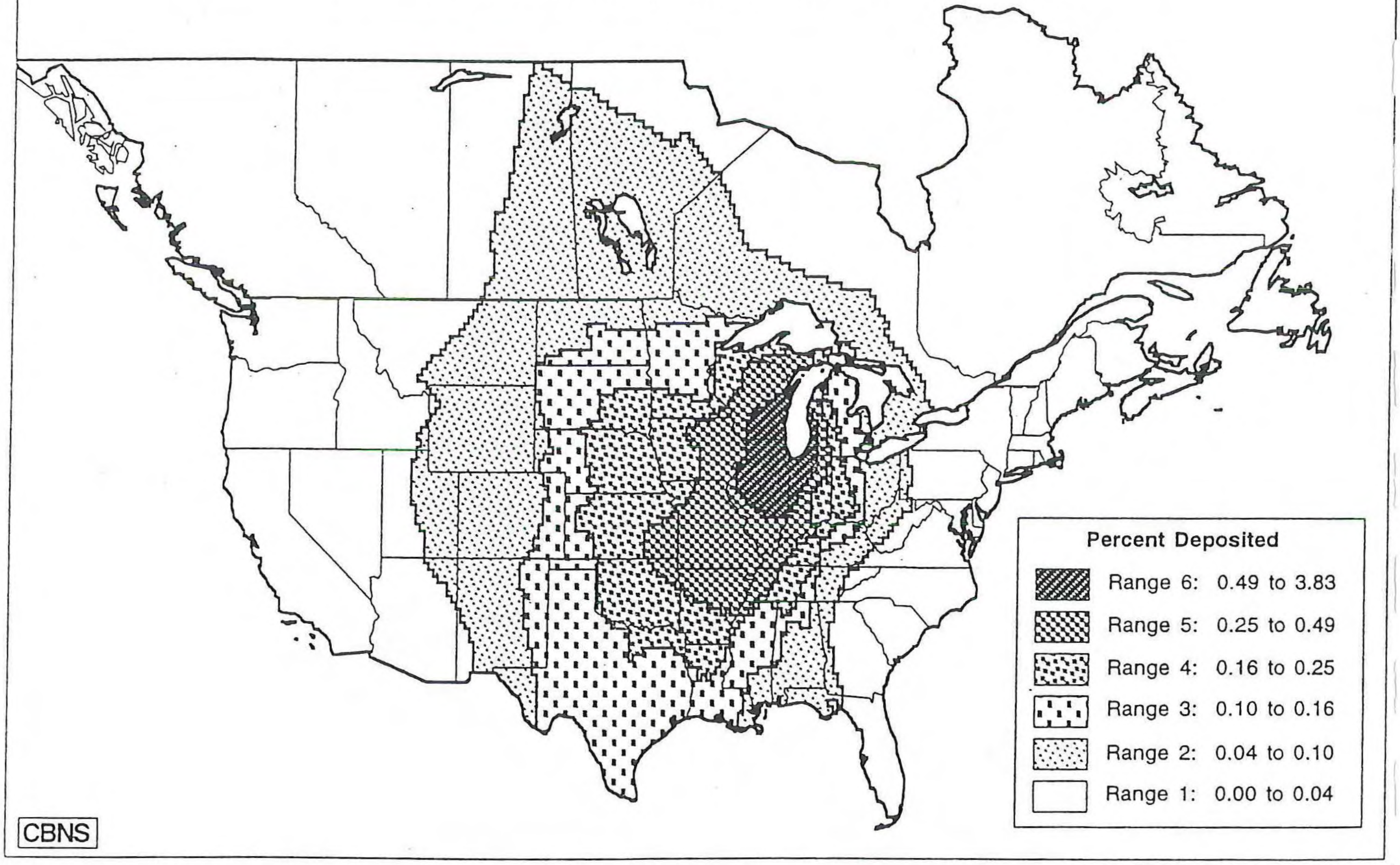


Fig. 6: GEOGRAPHIC DISTRIBUTION OF AIR TRANSFER COEFFICIENTS FOR 2,3,4,7,8-PeCDF.
Percent of Emissions Deposited in Lake Michigan in 1993.



7.2 Ocean Transport Pathways for PCDD/F's

There have been relatively few measurements of PCDD/F levels in ocean water, sediments, or biota, and most of the measurements are for near-coastal waters and estuaries (e.g., Hagen *et al.*, 1995; Rappe *et al.*, 1990b; Hashimoto *et al.*, 1995; Jarman *et al.*, 1996). Most measurements of persistent toxic compounds in marine ecosystems have involved PCB's and pesticides (e.g., Iwata *et al.*, 1993; Tatsukawa, 1992).

The recent measurements of Matsumura and coworkers (1995) in the western North Pacific may have been the first reported measurements of PCDD/F's in open ocean seawater. In the one site for which data was given, TCDD, PeCDD, HxCDD, HpCDD and OCDD were detected at levels of 0.04, 0.06, 0.21, 0.35, and 0.39 pg/liter, respectively.

PCDD/F's have been found in ocean-fish-eating animals, including polar bears and ringed seals in the arctic (Muir *et al.*, 1992; Norstrom and Muir, 1994) and albatross at Midway Atoll, stated to have an extensive feeding range covering much of the subtropical and northern Pacific ocean (Jones *et al.*, 1996).

The ocean may play a role in transporting PCDD/F's over continental distances. First, there are direct discharges of PCDD/F's from industrial sources to rivers leading to an ocean (tributary rivers) or directly to an ocean. PCDD/F contamination in the ocean can be dispersed and transported by currents over large distances. In a similar manner, PCDD/F's discharged by sewage treatment plants to tributary rivers or oceans — as a result of discharges to the facility or possibly even as a result of the formation of PCDD/F's in the wastewater chlorination process — may be transported over long distances in the oceans. Finally, there are many pathways by which air emissions may find their way into tributary rivers or oceans. A portion of the PCDD/F emitted to the air is deposited directly onto tributary rivers or oceans; a portion of the PCDD/F emitted to the air is deposited by precipitation which finds its way to tributary rivers or oceans; and finally, a portion of the PCDD/F emitted to the air is deposited to the earth's surface even without precipitation ("dry deposition") but can be delivered to tributary rivers or oceans by runoff or wind.

Unfortunately, little is known about the relative importance of different PCDD/F sources to ocean contamination, although it is likely that both atmospheric deposition and coastal water effluent discharges are important. Moreover, little is known about the fate and transport of PCDD/F's in ocean ecosystems. Thus, it is difficult to assess the overall significance of ocean transport of PCDD/F's *over continental distances.*

PCDD/F's in marine water are expected to partition significantly to suspended sediment, similar to its expected behavior in fresh water, although there are few data on this phenomenon available.

Based on measurements and theoretical considerations, Broman and coworkers (1991) estimated that approximately 25% of the PCDD/F in the surface layers of the Baltic Sea was truly dissolved, and 75% was associated with suspended particles. Thus, sedimentation may be a dominant fate process for PCDD/F's emitted from coastal liquid effluent discharges. Thus, for PCDD/F in coastal liquid effluent discharges (i.e., from sewage treatment plants), sedimentation may be a dominant fate process and may limit the long-range transport in the ocean.

Fish feeding in a coastal area contaminated with PCDD/F can become contaminated with PCDD/F. If some of these fish then migrate to areas 100's or 1000's of kilometers away, then this would constitute a potential pathway for PCDD/F transport over continental distances. Salmon, shad, sturgeon, orange roughy, mackerel, tuna are examples of fish widely consumed by humans that migrate long distances in the ocean *[Does anyone know which of these ocean fish (or others) that would be most likely feed in potentially contaminated near-shore waters or eat other aquatic organisms that feed in potentially contaminated near-shore waters?]*

The principal human health consequence of contamination of the oceans by PCDD/F and the possible widespread dispersal of such contamination would probably be increased dietary exposure to PCDD/F's through consumption of fish (and possibly sea vegetables).

7.3 The Potential Impact of Trade and Commerce on Exposure to PCDD/F's

Discussion of the continental transport of PCDD/F reflects concerns that an action taken in one place -- for example, operation of a trash-burning incinerator -- may result in the increased exposure of the population of another region or country. This issue can arise through processes other than the actual transport of airborne or waterborne PCDD/F from one place to another. It may arise as well through trade in industrial facilities that emit PCDD/F's; through commerce in PCDD/F-contaminated food products; and even as a result of commerce in commodities, such as organochlorine hazardous waste, chlorinated plastics, or leaded gasoline, which, when burned generate PCDD/F emissions.

The following example illustrates the potential effect of international trade in industrial facilities on PCDD/F exposure. As a result of citizen pressure, the concerns of the medical community and the initiatives of regulatory agencies, PCDD/F emissions from medical waste incinerators have been under increasing scrutiny in the United States. In the last few years increasing numbers of existing incinerators have been shut down and investment in new ones has decreased. One result may be increased export of incinerators from the United States to Mexico. If 100 such newly imported incinerators were to operate there, they would emit roughly 10-100 g TEQ of PCDD/F per year[17], contributing significantly to the exposure of the Mexican population[18]. On the other hand, if the transfer of medical waste disposal technology between the United States and Mexico were based on autoclaves rather than incinerators, the trade could have a beneficial effect on the Mexican environment.

As indicated above, ingestion of dairy products (such as cheese) and beef represent important paths of exposure to PCDD/F. Such products (in contrast with milk) are readily transported as items of national[19] and international trade, efficiently exposing the population of one region to PCDD/F generated in another region. Thus, in North America, the exposure of the respective populations of Mexico, the United States and Canada to some degree will reflect the balance of trade in such foods among the three countries[20], as well as their relative levels of PCDD/F emissions. The typical

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[17]. A typical medical waste incinerator's PCDD/F emission rate is on the order of 0.1 to 1 gram TEQ/year.

[18]. How large is this transferred emission amount relative to the long-range transport of PCDD/F through the environment (e.g., the atmosphere)? In order to answer this question, a detailed modeling study, for example, of the air transport of PCDD/F between sources in the U.S. and receptors in Mexico would need to be carried out. Such a study has not yet been done. A very rough, qualitative comparison can be made, however, in the following way. It has been estimated that the total air emissions of PCDD/F in the United States is on the order of 5,000 - 10,000 g TEQ/year (with very large uncertainties). Of these emissions, it was estimated that on the order of 40 g TEQ/year (~ 1% of the emissions) were transported to and deposited in the Great Lakes (Cohen *et al.* 1995). Given the general nature of prevailing weather patterns and the geographical distribution of PCDD/F sources, it is *not* likely that the total transport and deposition of PCDD/F through the air *from the U.S. to Mexico* would be much higher than this, and, may in fact be much lower than this. This amount is on the same order of magnitude as the amount emitted by the exported sources in this illustration.

[19]. It has been estimated that in the United States, the average food product travels on the order of 1000 - 1500 km between the site of original production and eventual human consumption [Reference].

[20]. U.S. Exports to Mexico during fiscal year 1996 were reported to include:

(a) 4,675 metric tons of cheese ($13.4 million); $120 million, total, of dairy products;

(b) 81,000 live cattle & calves ($42 million); 47,000 tonnes of beef & veal ($130 million)

Exports from Mexico to the U.S. for fiscal year 1996 were reported to include:

(a) $18 million, total, of dairy products;

person's exposure to PCDD/F's may depend more on the level of contamination at a dairy farm 100's or 1000's of kilometers away (producing milk for cheese) than on any local source of PCDD/F's near where they live or work.

Finally, trade in materials that can serve as precursors to the generation of PCDD/F's may also carry with it an effect on the exposure of the population of the importing country. For example, in the United States the disposal of hazardous waste, which if burned emits significant amount of PCDD/F to the air, has become an important item of commerce. One result is that hazardous waste produced in the U.S. has become a major fuel in the cement industry, and, if exported to the Mexican or Canadian cement industry, would to some degree become a component in these countries' environmental PCDD/F levels. A similar issue arises when disposable polyvinyl chloride products -- major items of commerce -- produced in one country and exported another country are eventually discarded and burned in local incinerators, contributing to environmental levels of PCDD/F.

However complex and subject to debate, such processes should be considered in efforts to remedy the impact of PCDD/F's on health and environmental quality.

8. POLICY IMPLICATIONS

8.1 The Need for Remedial Action

In U.S. EPA regulatory procedures, action to remedy the effects of an environmental pollutant is generally based on the establishment of standards that govern the emissions of the relevant classes of sources. (In the case of PCDD/F's, incinerators for example.) At present, PCDD/F standards are based on an assessment of the risk of cancer. A lifetime (70 year) risk of one in a million has been generally regarded as "acceptable" in siting individual facilities, although risk levels 10 times greater have sometimes been allowed.

As noted above, data on the PCDD/F content of food intake and measurements of the PCDD/F body burden lead to the conclusion that the general U.S. population is exposed to a cancer risk of several hundred per million, or -- if the effect of dioxin-like PCBs is included -- 500-1000 per

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(b) 600,000 live cattle and calves ($180 million).

[Source of data: Foreign Agric. Trade of the U.S., Economic Research Service, USDA] million (U.S. EPA 1994b). It seems reasonable to conclude, therefore, that action to sharply reduce the exposure to PCDD/F's and other dioxin-like substances is mandated by the observed levels. As noted earlier, non-cancer effects due to exposure to PCDD/F's are likely to occur at levels at least an order of magnitude below those that give rise to the increased risk of cancer, intensifying the need for remedial action.

The Canadian government's guideline for human exposure is 10 pg TEQ per kg of body weight per day. This guideline is several times greater than the general level of exposure, ~1-2 pg TEQ/kg/day, which, as noted above, applies as well to the U.S. population. Accordingly, current Canadian regulations do not call for remedial action at this general level of exposure to protect human health. However, since air transport of PCDD/F from U.S. sources may be responsible for a large part of the exposure of the Canadian population (based on the analysis of Cohen *et al.*, 1995), remedial action in the U.S., taken in response to U.S. standards, would probably reduce exposure in Canada as well as the United States.

The foregoing considerations have important implications for environmental policy. When, for example — as it is now — the decision to operate a waste-burning incinerator is based on the direct health-hazard impact experienced only in the nearby area, the public policy issues come down to a straightforward question: Should the community build the incinerator that would expose the people of the community, themselves, to this hazard? Since, in this case, the risk would be self-imposed, the community at risk, through its elected officials or a referendum, could decide whether or not to accept it.

The situation regarding the general population's exposure to PCDD/F is not consistent with the above decision-making methodology. Because of the dominance of air transport in exposure to PCDD/F's and their ability to be transported long distances in the atmosphere, exposure as a result of pollutant transport over regional and continental distances is strongly suspected to be a significant problem. That is, the PCDD/F emitted by any one incinerator (or other source) is combined with that from many other sources, and their collective impact is visited upon people everywhere — whether or not they chose to build an incinerator — through food produced on distant farms. Given the prevailing weather patterns, for example, it is likely that PCDD/F emitted to the air by U.S. sources makes a significant contribution to the PCDD/F content of milk produced on Canadian dairy farms. The same may be true of the impact of Mexican sources on dairy farms in the southwest region of the United States.[21]

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[21] . As an aid to national and international policy formulation, these and other potential source-receptor-exposure relationships need to be accurately characterized. A model-based analysis of the atmospheric transport and deposition of PCDD/F's emitted by sources in North America, with particular attention to the contamination of agricultural food chains, would be of great use in elucidating the relative importance of PCDD/F transport over local, regional and continental scales in leading to human exposure. Such an analysis would be greatly assisted if a sufficient number of environmental measurements of PCDD/F's were made to allow the model's validity to be tested. As noted above, information on emissions sources is relatively limited; the analysis could be

Thus, the policy debate must be greatly broadened. While the people in any given community would of course benefit by persuading officials to shut down the local incinerator in favor of alternatives such as recycling, the same remedial action would have to be taken on a much wider scale to significantly reduce the given communities exposure to PCDD/F. Since the impact of any one incinerator is part of the collective impact of many of them, action to reduce the hazard must be collective as well. In sum, policy decisions about PCDD/F sources should not be considered to be strictly local; regional, national, and international inputs into the decisions are necessary.

8.2 Opportunities for Remedial Action

At present, the regulatory measures that are applicable to sources that generate PCDD/F emissions — and which therefore contribute to long-range continental air transport — are based on the strategy of pollution *control*; to meet regulatory standards, a device capable of sufficiently recapturing or destroying the PCDD/F before it is emitted is attached to the source. Thus, in its latest documentation of PCDD/F emission regulations (for hazardous waste incinerators: U.S. EPA, 1996b), U.S. EPA requires the installation of a control device that represents the "Maximum Achievable Control Technology." This strategy has been practiced, at least in the major source of airborne PCDD/F — municipal waste incinerators — and the industry has sharply improved the efficiency of their control systems in recent years. The most stringent PCDD/F emission standard for municipal waste incinerators, 0.1 ng/m^3, is that recently enacted in Germany. Very extensive and costly controls are required to meet such a limit.

An alternative approach to remedial action is based on the principle of pollution *prevention* rather than control. It has been promulgated most notably by the International Joint Commission, which has called for "zero emission" or "virtual elimination" of PCDD/F's and many other persistent toxic compounds in the Great Lakes. Unlike the control strategy, the goal of the prevention approach is to change the technology of the production process in which the pollutant originates, so that the source no longer produces it. This strategy is designed to break the link, inherent in the technology of the facility, between the production of a "good" and the production of PCDD/F. Thus, a municipal waste incinerator produces a good in the form of trash disposal (and in some cases, electric power) and, at the same time, generates PCDD/F's. The aim of pollution prevention is to find a way to produce the good that does <u>not</u> produce PCDD/F's.

For fundamental, thermodynamic reasons, it is extremely difficult to achieve zero emission — or even to approach it — by means of a control device. The effort needed to accomplish each

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greatly improved if accurate, geographically and temporally resolved, congener-specific PCDD/F emissions inventories in North America could be assembled.

incremental level of efficiency (i.e., the percent of the pollutant recaptured or destroyed) rises dramatically as the device's required efficiency is increased. In practice, this effect is expressed economically; the cost of a control system rises disproportionately relative to increases in its efficiency. For example, it has been found that the cost of a power plant's sulfur dioxide control system rises from $50 per kW of capacity at 70% efficiency to $2,000 per kW at 90% efficiency. To reach 99.9% efficiency, the system would cost on the order of $4,000 per kW— about 10 times the cost of the power plant itself. Similarly, it has been reported that to achieve the 0.1 ng/m^3 PCDD/F emission rate now required of German municipal waste incinerators, the cost of the control system amounts to about two-thirds of the total cost of the facility. Thus, to approach the goal of zero emission by means of improved PCDD/F control systems, the cost of building and operating the sources would need to rise dramatically.

The task of achieving zero emission through the strategy of pollution prevention is not encumbered by the serious economic constraint that is built into the control strategy. Here, too, there is an economic issue: How will the cost of achieving the economic good produced by a PCDD/F-generating facility change if a PCDD/F-free technology is used instead to produce the same good? This approach has been used to evaluate the technological and economic feasibility of substituting technologies with dramatically reduced PCDD/F emissions (in some cases, essentially zero emissions) for the production technologies responsible for the major loadings of PCDD/F to the Great Lakes (Commoner *et al.* 1996a). The analysis of replacing municipal waste incinerators in the Great Lakes states and the province of Ontario with an essentially PCDD/F-free technology that produces the same good — i.e., the disposal of an equal amount of waste — may be used as an example.

In 1993 the communities in the Great Lakes region burned 11.8 million tons of residential waste in 54 incinerators, at an average cost to the communities of $57.58 per ton (the tipping fee). A technologically feasible alternative is to use intensive recycling programs to dispose of this same amount of waste instead of continuing to operate the incinerators. The cost of this substitution is the cost of operating the recycling system (additional costs for collection, recyclables processing, and public education are necessary), plus the cost of retiring the bonded debt carried by the incinerators. But this substitution involves two economic gains: the incinerator tip fees are eliminated, and (based on their current market value) the materials recovered by the recycling system would yield an income. When these factors are taken into account, the Great Lakes communities would <u>save</u> about $500 million per year in trash disposal costs if the switch were made to the recycling alternative. There would be a net gain of nearly 3600 waste processing jobs in the region, and 21,000 additional jobs could be created if enterprises were set up to make use of the recovered materials. Finally, since the power produced by the incinerator is now sold to utilities well above the utilities' own cost of power production, closing the incinerators would reduce these costs and hence consumers' electricity rates.

The cost of replacing the Great Lakes region's 609 hospital waste incinerators (as of 1995) — another major source of airborne PCDD/F — with autoclaves (and subsequent landfilling of the

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Draft Interim Report to the Secrrtariat of the Commission for Environmental Cooperation

Case Study

Ozone and Particulate Matter in the Atmosphere

Authors:

Gerardo M. Mejia- Instituto Tecnologico de Monterrey, Mexico
Jim McTaggart-Cowan- Royal Roads University, Victoria, Canada
Bruce Hicks- National Oceanic Atmospheric Administration, Silver Springs, USA
Carlos Santos-Burgoa- Instituto de Salud, Ambiente y Trabajo, Mexico City, Mexico

TABLE OF CONTENTS

1. INTRODUCTION

The recent approval of the North American Free Trade Agreement (NAFTA) among Canada, the United States and Mexico focused the interest in developing studies to evaluate the environmental impact of NAFTA. Great interest to study the transport and dynamics of air pollutants in North America. These studies will be valuable to evaluate the impact of increasing traffic and industrial activities; to identify, characterize, and quantify sources of air pollutants; to locate properly industrial and urban developments in the continent; and to direct policy decisions to prevent air pollution.

A wide range of pollutants are released to the atmosphere through North America. In the last decades great effort has been focused to characterize emissions and quantify the amounts of pollutants emitted by natural and anthropogenic sources. These emissions change continuously in time and, in particular, anthropogenic emissions also change in their chemical composition and amounts released, mainly caused by the continuous change of population, industrial activities, vehicle fleet, and new technologies in use. The pollutants emitted to the troposphere, or that vaporize from solid or liquid wastes, are transported by wind, reacting in the atmosphere to create other pollutants. Some of these pollutants have short average residence time in the atmosphere, like SO2, and may react fast, but other pollutants may exist long periods of time in the troposphere, like CFC's and POP's, being transported in some cases to the stratosphere. These pollutants may react in the stratosphere, causing severe changes to its chemical composition, like stratospheric ozone depletion whose effects will last for several years. Major pollutants in the air include particles emitted directly, particles below 2.5 microns in diameter generated through chemical reactions, carbon monoxide (CO), carbon dioxide (CO2), nitrogen oxides (NOx), sulfur oxides (SOx) and reactive organic gases (ROG). These pollutants react in many different ways to produce photochemical pollutants, acids, and particles. Other pollutants are emitted to the atmosphere and some of them may represent a serious health hazard. In this document we will refer only to those that contribute to ozone and particulate matter formation in the atmosphere.

Several studies have been conducted in urban and regional areas of Canada, the United States and Mexico to study the dynamics of air pollutants. However, very few air quality studies exist with the collaboration of institutions of two or three of the countries. This lack of joint studies has caused great controversy in assigning responsibilities in the impact of air pollutant emissions that may affect other country. This has become an issue of great interest in the Mexico-United States border area, where it is expected a dramatic increase of industrial, traffic and commercial activities in the next year. Local and federal governments of North American cities and states have interest in evaluating the current conditions of air quality in the area, since few air quality and meteorology monitoring stations exist. It is desirable to develop and evaluate cost-effective air pollution control strategies in areas where it is known that air quality problem exist, as well as finding the best areas where industries may be allocated with minimum damage to the environment. Of great interest is to determine the best location for highways interconnecting the border area. Pollutants emitted by mobile sources may cause a significant damage to areas or animals that may be sensitive to specific unknown the impact of long range transport of air pollutants. For example, the impact in Mexico of SO2 emissions from the northeast coast of the United States. To solve air pollution problems, institutions of the three countries must work together to develop joint studies to evaluate the impact of long range transport of air pollutants. However, this requires to design and share standardized data bases of emissions, meteorology, air quality, land use, topography, and other data necessary to apply air quality models at regional and continental scale. It is important to note that an appropriate continental air quality model may not exist, but can be developed bases upon previous models designed for urban and regional studies of air pollutants dynamics.

The problem of ozone formation in the troposphere has been detected in large cities of North America where monitoring stations exist. The solution of the problem represents a major challenge for scientists and policy makers in actual days. Moreover, it is unknown the impact in rural areas when ozone or precursors of ozone are transported. To achieve a better understanding of ozone formation in the continent a tropospheric ozone research plan has been set for North America, where institutions of Canada, the US, and Mexico created the North America Research Strategy for Tropospheric Ozone (NARSTO), a partnership to achieve a better understanding of ozone dynamics in the country, coordinating and enhancing policy-scientific research to determine effective strategies for local and regional ozone management. This partnership among the three countries is an effort to develop planned and coordinated activities to understand the problem, to create standard data bases and to develop models to study ozone dynamics in the continent.

Several regions of North America have shown high levels of particles in the atmosphere. Despite the health hazard that particles represent, in particular those below 2.5 microns in diameter, very few studies have been performed to detect the main sources of emission and to study their dynamics in the border region. Recently, the increasing interest of local authorities of cities have focused the attention of different sectors to develop control strategies to improve air quality in their regions and their nearby rural area. For example, great interest exist to study the effect of emissions of border areas in visibility degradation at the Grand Canyon, in the US, and at the Big Bend/Maderas del Carmen National Parks, in the US/Mexico border. The problems detected in the Mexico/US border area are of great concern for the border cities economies, since alternate fuels, inspection/maintenance programs, better fuels availability are air pollution control strategies that may lead to policy decisions. The decisions taken may have a big economic and politic impact in the border area development. Although we know that the main sources of air pollutants are transportation, transportation fuel, and power production, at this time, little is known about the amounts of particles emitted to the atmosphere by different sources and about the physical, biogenic, and chemical composition of particles. These parameters, as well as a better understanding of the meteorology of the region, are important data to achieve a better understanding of the transport and fate of particles in the atmosphere, as well as the associated transport of organic compounds sorbed into the particles.

2. DYNAMICS OF POLLUTANTS IN THE ATMOSPHERE

Pollutants in the atmosphere react in many different ways, depending if they are present in the gas or aqueous phase or if they are sorbed in particulate matter. Pollutants may be directly emitted from the source, e.g., SO2, CO, CO2, NO, ROGte s, and respirable particles produced in fires, dust, mining, etc.; or may be produced by reactions in the atmosphere of other pollutants, e.g., ozone, NO2, sulfate, nitrate, etc., producing even respirable particles. The chemical kinetics of pollutants in the air is extremely non-linear, which makes necessary the use of advanced chemical kinetic mathematical models to study the dynamics of pollutants in the air and their transport.

In the case of ozone, the basic cycle starts when a molecule of NO2 photodecomposes and produces a molecule of NO and a free oxygen, which rapidly reacts with O2 to produce ozone. Interestingly, the ozone oxidize the NO to regenerate NO2 and the molecule of O2, completing the cycle. The photodecomposition of NO2 is driven by the radiation received in the lower troposphere, that changes in a given region with the time of the day and seasonal station of the year. The regeneration of NO2 is slower than its dissociation during the day and this explains why ozone concentrations increase during the day, reaching a peak after noon, and decay at night. It is important to note that NO destroys ozone. If ozone is transported by wind during the day to rural areas or to the upper troposphere, it is not destroyed and may accumulate to the next day or may remain in rural areas until it is destroyed by other mechanisms of reaction. This may explain high ozone levels that have been measured in some rural areas that receive the pollutants transported from urban areas. Furthermore, NOx may be transported to agricultural areas where natural organic serve as catalyist to produce ozone.

Hydrocarbons comprehend a great variety of compounds, being the most important the groups of volatile organic compounds (VOC's), and the subgroup of reactive organic gases (ROG's), and persistent organic pollutants (POP's). The group of ROG's is the most active and, consequently, the most important in studies of atmospheric chemistry. The VOC's emitted react at different speeds in the atmosphere, depending on their chemical composition. Some of them react in a few hours, being the ROG's specially active in oxidizing NO to NO2 through different chain reactions, increasing ozone formation indirectly. VOC's may be sorbed in particles an transported to rural areas, where they are desorbed when temperature rises during the day, enhancing ozone formation in rural areas. At this time it is not well known the health hazard that represent particles that sorb VOC's and semivolatile organic compounds (SVOC's) when they are inhaled.

Particles are the most complex of the pollutants in the atmosphere, being present in solid or liquid phase or a combination. They may be composed of inorganic or organic compounds, or may have biogenic origin. For this, their physical and chemical properties vary greatly in the same urban or rural area. Primary particles are emitted directly to the atmosphere, react with gases or coagulate with other particles and finally, subject to gravity, they fall out, if they are big enough, by dry or wet deposition. The particles that remain suspended are small in their aerodynamic diameter and may be inhaled by humans or animals. Suspended particles are usually less than 10 microns in size, being those particles less than 2.5 microns the most dangerous for humans as they are trapped in the lungs. Particles accelerate reactions in the atmosphere, in particular in liquid phase where SO2 and NOx may be deposed in water drops from the gaseous phase. In the water drop, they are oxidized to nitrate and sulfate giving an acid property to the droplet, acidifying lands and surface waters, like rivers and lakes, when the drop falls as rain. The oxidation may be catalyzed by different HC present in the water drop or directly by ozone or other oxidant agents that may be absorbed in the water drop too. If the drop does not fall as rain, it evaporates and very fine particles result with heterogeneous chemical composition. These particles also add directly to visibility concerns.

From the discussion above is observed that the chemistry of ozone, HC and particles in the atmosphere is related in different ways. First, to study the dynamics of these pollutants in the air, emission sources must be identified and quantified. Also, in a urban or rural area study emission sources are common. Second, HC produce ozone indirectly and may contribute to ozone formation in rural areas or regions far away from where they are emitted. Third, NOx produce ozone, but may be absorbed in water drops and is deposed as nitrate. This process is important in studying the dynamics of ozone and aerosols and the fate of NOx and SO2. Fourth, water absorbs different gases present in the atmosphere, accelerating reactions that form particles and acids. Finally, pollutants are transported in the troposphere by winds and follow the same pathways. These factors must be considered when air quality studies are made in urban and regional areas. While there are obvious major sources of air pollutants such as transportation, in the case of transboundary or continental studies a great amount of data is necessary to find the best cost-effective control strategies related to point sources to improve air quality.

3. HEALTH EFFECTS

The effects of air pollution on human health is one of the most controversial and important problems that human beings face today. En some occasions, concentrations of air pollutants have reached high levels in periods of hours or days. The effects on people depend on the concentration of the pollutant, the period of exposure and the population exposed. The most affected people with these periods of high pollution are children, elderly and those with health problems, usually people with respiratory problems.

Determining the relationship between health effects and the long-term exposure to an air pollutant is a difficult task. Specifically, it is desirable to determine the levels of morbidity and mortality of population exposed to a pollutant. This task is difficult because factors such as personal habits, hereditary, diet, living conditions, smoking, physical condition, etc., must be separated of the effect of air pollution. Relationships dose-response are the common mean of describing the effects of air pollution in a given population exposed for a given period of time. Obtaining these expressions is not easy. Large amounts of data obtained usually from hospitals must be analyzed to find dose-response values. In these analysis is assumed, in many cases incorrectly, that the factors vary randomly or they are approximately the same for the group.

Air pollutants are usually related to diseases in the respiratory system. Pollutants may have an acute reaction, such as irritative bronchitis, or chronic reaction, such as chronic bronchitis or pulmonary emphysema. Acute bronchitis is a short-lasting inflammation of the bronchial system while chronic bronchitis is a sustained inflammation usually accompanied by cough and shortness of breath. Pulmonary emphysema is a condition where alveolar walls are destroyed. A cure for emphysema is not known. These diseases may be caused by different pollutants. Among the most important are ozone and particulate matter in the air.

The effect of ozone and other photochemical oxidants, such as peroxyacilnitrates or peroxialkilnitrates, in the pulmonary system is not well understood yet. At concentrations of 1 ppm produces a narrowing of the airways of the lung. The exposure to concentrations typical of urban air, 0.1 to 0.2 ppm (the US standard is 0.120 ppm and the Mexican standard is 0.110 ppm), apparently accelerates aging of lung tissue. Another effect of photochemical oxidants is eye irritation.

The health effects of particulate matter (PM) have been well understood from the occupational studies of dust exposure. It was thought that critical air pollution episodes where the only dangerous general population concern in relation to PM (Schwartz, 1990). However more recently the analysis of currently accepted air concentration of fine PM has demonstrated the effects on morbidity and mortality, even at low levels. The consistency in morbidity is important (Pope, 1996; Santos-Burgoa, 1995); however there is a consistent increase of total population mortality in 1% for each 10 micrograms/cubic meter of PM10 (fine fraction). This has been shown consistently throughout several studies in Canada, United States, Europe (Pope et al, 1996) and Mexico (Loomis et al, 1996). This has been shown in many different locations regardless of the diversity of contents of PM. This has had a regulatory effect that has been translated into a new effort to regulate lower levels of exposure (EPA, 1997), and at lower fractions of PM (PM of 2.5 microns.).

Particles are trapped in different regions of the respiratory system. They may be trapped by the nasal cavity, the trachea, the bronchial tubes and the lungs. Within the lungs particles can be trapped in the bronchioles and the alveoli. In the respiratory system particles are deposited by means of four different mechanisms: interception, when the particle hits a surface because its trajectory is close to the surface; impaction, the momentum of the particle keeps it in a predetermined trajectory hitting a surface; sedimentation, when the particle settles by gravity forces; and diffusion, when the particle hits a surface caused by the Brownian motion induced by the impact of gas molecules.

Air quality standards of PM10 or of total suspended particles (TSP) are expressed in terms of micrograms/cubic. However, deposition and injury in the respiratory system may not depend just on the mass concentrations to which population is exposed. The physical properties and chemical composition of the particles may have a role, although at present the correlations are primarly with concentration, particularly of PM2.5, and on a first order independent of chemical and physical properties other than size. Most of the particles with diameters larger than 10 microns are removed in the head region, and only particles with diameters less than 2.5 microns penetrate into the lungs and deposit in the alveoli. For this reason, particle size distribution must be considered in air quality standards. Recent studies show that about 20 to 30% of particles inhaled in the range of diameter 0.1 to 1.0 microns deposit in the respiratory system, about 50% particles with diameter around 2.0 microns deposit, and 90 to 100% of particles with diameter 10 microns are trapped. The most important health hazard is caused by particle less than 2.5 microns, since they can deposit in the alveoli. From these fractions trapped in the respiratory system, more than 60% of particles of 1 micron reach the lowest levels of the lungs, while most of the particles in the size of 10 microns wind up getting deposited in the first four splits of the air passage.

Particles in the air may be emitted, react, grow, sediment and be transported from different areas. Hence, the receptor may capture particles coming from different sources, some of them coming form short range distance, within few kilometers, and others from long range distance, several hundreds to thousands of kilometers, even at continental scale. For this, researchers and policy makers face a great challenge deciding the impact of the contribution of different sources in air quality in a given region. While traditional practice would suggest that deciding the contribution of different sources is critical to evaluate effectiveness of different strategies to improve air quality and, consequently, to reduce human exposure to different pollutants, in this case the major sources, on a first order, are known fossil fuel combustion processes, thus action needs to be taken right away.

Human exposure is defined as the contact of an environmental agent with the human body. Exposure is not equal to dose, but is a realistic approximation to it (Johnson, 1992). Currently in health research, exposure assessment has became the key element of analysis, and for environmental exposures with a long term effect, retrospective exposure assessment is now developing (Correa et al, 1994). Research has demonstrated the need to consider all the routes of human exposure (Ott, 1985, Smith 1988) This is important if we want to understand the different ways long range transport of pollutants can be related to human and biota health effects. Specifically in the case of particulates, its daily and hourly variations are significant for health effects, as they have been demonstrated to be acute as well as chronic (Pope, 1996). The route for exposure is inhalation, and ingestion of water and food or soil ingestion through pica of children (Hayes L and Rodenbeck S, J Env Health, 1992) is the main medium for other chemicals. Human exposure to particulate matter is complex, as it is dependent not only on the general environment concentration, but on its concentration in different human settings (micro environments). In fact in Mexico it has been shown that the correlation of air monitoring stations concentration to human exposure is statistically significant, although not the only factor that explains human exposure to particulates since it only explains 30% of the total variance (Santos-Burgoa, 1997).

4. EMISSION SOURCES

4.1 Mobile sources.

The most important combined source of NOx, HC and CO pollutants are vehicles. Despite being the most common and simple source of pollution, emissions of a vehicle fleet is hard to estimate, since every single vehicle have different composition of emissions, depending on mechanical conditions, fuel used, engine design, engine repair, and, even, driving patterns. For this, mathematical models are used to estimate emissions of a vehicle fleet. These models consider the different vehicle types and year in the fleet, their mechanical conditions, the specific emission factors of each model, the driving patterns, the average speed, the average mileage per year, vehicle aging, and other factors. In the case of emissions factors, the values considered were obtained from extensive studies in US vehicles for conditions specific of the US. From different models available, EPA recommends the use of MOBILE 5 as the best current model to estimate emissions of a vehicle fleet in the US. Based upon this model, specific MOBILE 5 models have been developed for some cities in Mexico. However, to use these models is necessary to obtain the necessary data for the model, which most of the times does not exist at this time for the Mexican cities and vehicle fleet. To use the models developed for Mexico, data of yearly mileage, mechanical conditions, average speed, etc., of the fleet have been estimated. This estimation of data may produce unrealistic estimates of emissions of vehicles, with the corresponding consequences in uncertainties evaluating air pollution control strategies. Furthermore, HC are have different kinetic reactivities to produce ozone that makes the problem of studying the dynamics of ozone more complex, since HC need to be speciated and their composition depend on fuel used and mechanical conditions of the vehicle fleet.

Another problem arising from vehicle emissions is that NOx are oxidized in gaseous or aqueous phase, they are transported in the air, ant their final fate is their inhalation or deposition as nitrate, PM2.5 or as an acid, with the corresponding damage to soil and surface waters. Other gaseous pollutants in the air may form other particles that may directly be inhaled or may sorb VOC's, transport them long distances, desorbing them, causing damage to rural areas where anthropogenic sources of air pollutants do not exist and may be inhaled then as well. Evaluating the impact of vehicle emissions in other states or countries have become an issue of great interest for policy makers. However, to decide about the best policies that must be enforced to prevent and control air pollution maintaining the economic grow requires of a complex air quality model. Such a model must consider anthropogenic as well as natural emissions, the meteorology of the region, the land use, the incoming solar and UV radiation, topography of the region, etc., in order to produce reliable predictions of the dynamics of air pollutants.

4.2 Surface and stationary sources.

Surface sources are those who emit pollutants at ground level in a given extension of land, for example, crops, the ocean, fires, use of hydrocarbons or wood for space heating, and the forest.

Among the most important pollutants emitted by these sources are biogenic components, inorganic particles, CH4, N2O, HC, NOx and SO2. Some of these pollutants are precursors of ozone and secondary particles and it is important to consider the influence of this emissions in regional or continental studies of air pollutants transport. Surface sources change along the regions in the countries and their emissions are difficult to measure. Like mobile sources, emissions of surface sources are estimated using emissions factors that, as in the case of mobile sources, need to be validated for their use in other states or countries different from where the emission factors were obtained.

Contrary to mobile sources, emissions of stationary sources are concentrated and control strategies may be easily implemented. However, the cost associated to implement a given strategy needs to be evaluated in regards of its effectiveness to improve air quality in a given region. New plants are designed with state-of-the-science technologies to control air pollution, in such a way that emission standards are accomplished; but existing old plants have manufacture technologies that did not considered environmental aspects in their design. In the latter case the most desirable approach is pollution prevention, where usually a change in technology will improve economics of these plants. In some cases the cost of retrofit plants with new technologies may be very costly. This could lead the plant to be out of business for the high investment cost necessary to adapt new air pollution control technologies to the existing process. This issue is very delicate for policy makers who decide to enforce some emission regulation that may lead to the closing of a plant. Nevertheless, it is necessary to find ways to encourage change of old technologies.

Fossil-fueled power plants are among the most important stationary sources of air pollutants. These plants are located in different parts of North America and the emissions of different pollutants change depending on the operation conditions of the facility and the fuel used. Traditionally monitored pollutants emitted by power plant are CO2, NOx, SO2, CO and HC, and for coal-fired plants also heavy metals and radioactive materials. These pollutants are transported by winds and react to produce ozone, respirable particles and acid rain. For this, decisions about fuel usage, location of plants and permitted amount of pollutants released to the atmosphere is a matter of great interest for policy makers. In the US, emissions of these plants have been calculated extensively using methodologies based upon emission factors that are characteristic of each plant and fuel used (Schrock and Baechler, 1995). In Mexico emission factors obtained for US plants have been used and this may introduce serious errors in the estimated emissions unless the emission factors are validated or calculated for Mexican plants.

Uncertainty in the emissions of power plants have caused great controversy in areas where other stationary sources of SO2 exist. For example in some areas of the Mexico-Texas border is necessary to evaluate the best cost-effective strategies to control SO2 air pollution, but a comprehensive study that considers emissions, meteorology, reactions, deposition and other factors has not been made yet, mostly caused by lack of information of the area. This issue is of great concern, since power plants in North America may have a great effect in air quality in the continent.

5. OZONE AND PARTICULATE MATTER IN THE TROPOSPHERE

5.1 Ozone.

The issue of ozone pollution in surface air is one of the most perplexing to face today's air pollution regulators. First, the issue involves consideration of many chemicals, both natural and man-made, that react to form ozone as an end product. Second, the issue is politically charged, since many of these contributing chemicals derive from activities that are fundamentally identified with maintaining our standard of living (the power industry, automobiles, etc.). Third, the ozone question is confusing to the public, since in the upper atmosphere man-made chemicals are destroying the ozone layer, which is bad, and in the lower atmosphere different man-made chemicals are generating ozone, which is also bad. Fourth, the issue is intimately intertwined with that of nitrogen oxides and their influence on ecosystems through excessive deposition of nutrients, contributing to eutrophication. The issue is fraught with danger of imposing inadequate or cost ineffective controls, because the effects are so complex and because so many chemicals are involved.

The dominant surface-layer ozone concern relates to human health, although ecological impacts are also well known. Ozone affects the lungs and respiration of animals; it also affects foliage. Studies of these effects have been convincing. There is no longer any argument about whether ozone causes damage to living organisms; the question now is how to reduce the levels of exposure. The problem is that regulating any one of the causative chemicals alone can have sometimes surprising results, and that not all of the chemicals involved are amenable to local control. It is not that ozone itself is a persistent chemical in the air that is transported for long distances (although indeed ozone can be transported from one region to another), but that some of its precursors do indeed fit into that category and that effective ozone reduction strategies must take this reality into account.

Some ozone in surface air originates from higher levels in the atmosphere (stratosphere/troposphere exchange), but most is generated locally from chemicals emitted by societal activities. The key ozone precursors are various oxides of nitrogen (generically referred to as NOx) and a wide variety of volatile organic compounds (VOCs). Ozone is generated as a by-product of atmospheric chemical reactions involving these chemicals. All of these chemicals, and ozone itself, are susceptible to atmospheric transport, but some of the NOx and VOC ozone precursors can be carried for especially long times and great distances. In general, we are no longer worried about each area of concern as a separate issue, but need to understand how local upwind emissions combine with the pollution brought in by long range transport to aggravate local ozone exposures.

The spate of control measures of the 1960s and 1970s focused attention on the need to reduce local VOC emissions, primarily from private automobiles, to start reducing ozone exposures in areas where population is centered. These strategies worked in those parts of the continent where they were targeted -- specifically cities and urban areas -- but did little to relieve ozone pollution in rural areas. Now, we are faced with steadily growing NOx levels, on a continental scale, that will modify the improved ozone environments that have already been attained in some areas and will inject an unavoidable reason for more careful attention to the physical, chemical and meteorological processes that are in play. Experience has taught well that the concept of uniform, continental-wide application of standardized control strategies is a reflection of political fairness and legal simplification that fails to recognize the differences in the environments that contribute to surface ozone pollution. Clearly, solar radiation intensity varies with latitude, and all chemical reactions that are promoted by ultraviolet radiation will be affected accordingly. Among these reactions are many of those that lead to ozone generation from its precursors. Moreover, the composition of the natural "chemical soup" that contributes to ozone production varies greatly from place to place, with striking differences with longitude. Thus, ozone control strategies are likely to need recognition that what works well in one location will not necessarily work as well in others. The range of situations encountered across this continent is so wide that control strategies should be based on only the very best understanding, or else the ozone reduction targets will not be met and the consequences of any regulatory errors will be propagated downwind to affect other areas, in another jurisdiction if not in another country.

There is a newly emerging complexity that might well modify the way in which regulators are thinking about ozone in surface air. The same nitrogen oxides that enter into the ozone generation process are themselves deposited to ecosystems, where they frequently serve as unwelcome nutrients. Coastal ecosystems downwind of densely populated areas are especially vulnerable -- such as the mid-Atlantic seaboard of the USA. This coastal region of this continent is already showing the unmistakable effects of excessive nitrogen nutrients, some of which arrive via the atmospheric pathway from air pollution sources far upwind. Regulating for ozone must take into account the fact that the decisions made will have consequences on the coastal eutrophication issue, as well. Likewise, regulators considering strategies to improve coastal aquatic habitats must be cognizant of the benefits to be accrued as NOx emissions into the atmosphere are reduced. This is a prime example of how different regulatory issues can overlap, and of how the most appropriate decisions must be based on an integrated understanding of the processes, costs, and benefits involved.

High ozone concentrations are found in several areas of North America. Recognizing the complexity of the problem, the National Research Council concludes that there is necessary a coordinated program to reduce tropospheric ozone in the continent by identifying the key sources of precursos, and by elucidating the chemical, physical, and meteorological processes that control ozone formation. As direct response to this conclusion was created the North American Research Strategy for Tropospheric Ozone (NARSTO), a partnership of universities, government institutions, industries, contractors, and environmental associations interested in ozone research and control. NARSTO is organized in groups and teams that share information and promote and coordinate research activities to cover gaps in knowledge to achieve a full understanding of ozone dynamics and transport in the continent. The teams are organized to cover monitoring and data collection, emission estimates, modeling chemistry and transport, analysis and assesment, quality systems and data management. These teams report to a science and resource and planning group that depends from the executive comittee and the executive assembly.

Research goals include:

- Review of available state of the science tools for measurementes.
- Provide ozone distribution and trends for North America.
- Evaluate data of monitoring sites.
- Improve meteorological modeling and knowledge of chemical mechanisms.
- Improve estimates of natural and antrophogenic emissions of ozone precursos.
- Develop and compare air quality models.

Policy and science assesment questions address by NARSTO are the following:

- What changes have ocurred in tropospheric ozone scince over the last decade that might alter or confirm the course of current ozone air qulity management strategies?
- How manageable is the ozone problem for a given area?
- Are existing emission control measures helping to bring the ozone problem under control?
- What are the relationships between the on-going efforts to manage tropospheric ozone pollution and the scientific understanding of the issues?
- What are the alternate approaches for reducing current and future high ozone concentrations on urban and regional scales?
- How can we best track and asses the progress and efectiveness of our ozone management efforts?
- Will our efforts to manage ozone help or hinder efforts to mitigate other environmental problems, such as acid rain, fine particles, and global climate change, and vice versa?
- What are the most significant research developments in tropospheric ozone science over the last decade?
- How does ozone accumulation on urban and regional scales depend on the source dimension and location? How does it depend on the relative contribution from local and regional sources?

- What are the most recent assesment of the relative contributions of VOCs, NOx, and CO to ozone accumulation on urban and regional scales in North America?

- What are the strengths and limitations of the current scientific methods and tools in assessing tropospheric ozone issues and developing management strategies?

- What approaches are required to determine historic concentrations trends of ozone and its precursors on urban and regional scales? What is required to demonstrate the effectiveness of emission control strategies over time?

- What are the relationships between the control strategies designed to manage tropospheric ozone and those designes to manage other pollutant regimes of interest?

These questions addressed by NARSTO give a clear idea of the necessary effort that must be dedicated in the near future to achieve a better understanding of ozone formation and tranport in the atmosphere. Considering the spread of conditions in North America, and the long-range aspects of air pollution, NAFTA/CEC focuses on the coupled ozone/eutrophication issue as a prime example of how the member states must work together to achieve the continental environmental standards that are desired, while maintaining economic growth and prosperity.

5.2 Particulate matter.

An aerosol is a suspension of particles in the air, each particle consisting of a given number of components. The study of the chemical and physical properties of particles are critical to understand their dynamics and fate in the troposphere. Important physical properties are their size distribution and density. These properties are critical to calculate the rate of removal from the atmosphere. The chemical properties are important to understand the reaction mechanisms that particles follow, how they transform and change size over time, and their capability to serve as nuclei for water.

Settling velocities and transport of particles depend greatly in the size and density of the particle. The particle in the atmosphere is subject to the gravity force, the buoyancy force and the drag force. The gravity and buoyancy forces are constant for a particle of a given size and density suspended in air. The drag force depends on the settling velocity, and this velocity depends on the size and density of the particle. For this reason, the size distribution is critical to determine the distance a particle travels before it is removed from the atmosphere. Particles in the range below 10 microns remain suspended in the air easily due to the Brownian motion. In this case the particles are bombarded by molecules of the surrounding fluid causing a random motion of the particles in the air. The particles subject to Brownian motion travel long distances before settling and they are the most important size to be considered in long range transport of air pollutants.

Visibility degradation is an important phenomenon caused by aerosols. The problem is becoming important in recreative areas where visibility is affected by emissions of pollutants. The visibility degradation depends on the size of the particle. Gaseous pollutants like SO2 and NOx react in the atmosphere, they oxidize and become sulfate and nitrate, respectively. These pollutants may be absorbed easily in water, oxidized, and left after the water evaporates. The result is a particle that may remain suspended in the air for several days or more before settling. Currently, great controversy exist about the levels of contribution of Mexican and US industry in visibility degradation in National Parks close to the border.

Particles may be composed of many different chemicals. Important components in particles are heavy metals, light metals, biogenic components, VOCs, and water. The chemical composition is used to identify the possible sources that emitted a given particle using the technique of mass balance component analysis; however this is limited by the increase resulting from "background levels" and not considering meteorology variables of the region. Notwithstanding this inherent limitations, this technique is widely used to identify the origin of a particle and determine contribution of different sources to the degradation of air quality in a given region. However, this technique does not take into account other factors, like wind patterns and settling, that may affect the results of the model. More complex models that consider aerosol formation and its dynamics in the atmosphere are necessary to achieve more reliable results. This is of great importance in studies of continental transport of air pollutants where the impact of emissions of different countries is evaluated. For example, acid rain deposition of SO2 and NOx has become critical in rural areas. In this case emissions from urban areas are transported by wind during several days, traveling several hundreds of kilometers before they depose in earth or surface waters. During these days pollutants may cross different states, regions and countries. For the above, controversy exist about the possible sources that may be affecting the region and policy decisions are difficult to enforce between the countries affected. An important characteristic of particles is that VOCs may be sorbed on them. For this effect, VOCs are transported and released in other areas where they react promoting, for example, ozone formation or desorb chemicals that may represent a health hazard. It is important to note that the very significant morbidity and mortality evidence associated with the fine respirable particles is, at this time, dependent only on size (the smaller the worse) not on their chemical composition.

6. MODELING OZONE AND PARTICULATE DYNAMICS

People is exposed to pollutants through different pathways. A key question to address on long range transport of particles and ozone precursors is in what magnitude influence human exposure. In the case of air pollutants, they may come from regions far away from their final fate. This is of great importance when is necessary to improve air quality and the emissions affecting the region are in another state or country. In this case, pollutants may be transported long distances before they are deposed from the atmosphere. However, to address this problem at a continental scale is necessary a great amount of meteorology and emission data that, at this time, exist only for some areas of North America, and sometimes with certain levels of uncertainty. This uncertainty is caused mainly for the high cost of monitoring a large region, being necessary to rely in stochastic or deterministic models to estimate data for areas where monitoring data do not exist. This uncertainty may be reduced using better models, usually of the deterministic type. These models have been widely used to study the dynamics of ozone and particles in urban areas.

Two important aspects must be overcame to apply air quality models to regional areas. One aspect is that the necessary information to run the model must be collected or, if it does not exist, estimated. The other aspect is to develop appropriate air quality models that can be used in large extensions in the continent. The collection of information also needs the design of standard management procedures of data that must be collected from different states and countries, with the goal to be used in air quality studies. The air quality models must consider the shape of the earth, the chemistry of pollutants in the atmosphere, the emissions of the different sources in the domain, and the meteorology of the region. Also, air quality models must be flexible to be applied to different regions and reliable in the information they produce.

A powerful tool to study the dynamics of ozone, acids and fine respirable particulates are photochemical air quality models. These models solve the complex set of partial differential equations which describe the chemical reactions of air pollutants and their transport in the atmosphere. In the case of ozone formation, the differential equations are highly non-linear, which makes the mathematical solution more complex. However, recent advances in computer hardware and software have allowed the solution of these equations in reasonable time. An extensively tested model may predict with good accuracy the dynamics of air pollutants in the three spatial dimensions, its response to perturbations and may be used to assess air pollution control strategies.

To apply a photochemical air quality model to a given region it is necessary to determine a series of parameters and inputs characteristic of the region. These parameters and data include meteorology, topography, solar radiation, land use, emission inventory of mobile and stationary sources, etc. The parameters and data may be obtained usually form government agencies or from other studies. An emission inventory is critical to identify, characterize and quantify emissions, as well as a necessary input to the model to study the dynamics of pollutant emissions. However, estimating an emission inventory is not a simple task. In the case of North America, the US and Canada have worked in the last decades to develop methodologies to estimate and manage emission inventory data of mobile and stationary sources. In the case of an air quality study the geographic location of the source is important to assign the emission to a specific cell in the domain. These emission data and the geographic location of sources is readily available in the US and Canada but, in the case of Mexico, the emission inventory data is being estimated adapting methodologies developed in other countries. In some specific cases, the data need to be gathered from different institutions or estimated from fuel consumption and emission factors.

Several studies of urban air pollution have been performed in North America. In Mexico only one extensive study of ozone dynamics was made in Mexico City, where several Mexican and US institutions participated. Although great interest exist in some areas of the Mexico US border to study impact of air pollutants emitted in both sides of the border, only one preliminary regional application of a photochemical model has been made in the Mexican-US border area. The scope, lessons and opportunities of the latter project are described in the following paragraphs.

6.1 Case Study: Ozone Dynamics in the Mexico-Texas Border Region.

In this project a photochemical air quality model was applied to the Mexico-Texas border region to study the dynamics and fate of air pollutants. This region has several important urban and industrial areas that experience high levels of air pollution. The border region is defined as the region within 100 miles at each side of the political boundary; however, cities close to the border, and which may have an impact in air quality, were considered in the model. This is important to develop integrated cost effective air pollution control strategies for the region. For the above, the problem of understanding the dynamics of air pollutants becomes more complex and, furthermore, evaluating air pollution control strategies becomes a big challenge. The region is relatively sparse, but presents concentrated sources of emissions (e.g., cities, power plants and smelters with tall stacks). Also, the composition of emissions can vary greatly from one region to another, due to the distribution of local sources and to the different fuels and types of air pollution controls used. For example, the fraction of cars with catalytic converter differs between the two countries as well as fuel composition. Furthermore, the relative abundance of ROG and NOx may change from one region to another and can be critical in determining the most effective control strategies.

Several air pollution studies have been conducted in the area of Tijuana/San Diego and Juarez/El Paso. Results of these studies show high levels of ozone (well above the air quality standard) as well as high levels of TSP and PM10 have been detected in some areas Despite the importance of PM2.5, little is know about its concentration levels. Recent studies have focused the attention in evaluating air pollution impact in regional areas due to emissions in urban areas, for example, visibility degradation at the Big Bend National Park. In this project the CIT Photochemical Model was applied to the Texas-Mexico border area. To apply this model is necessary to gather information of both countries to analyze air pollution problems in an integrated form. Air quality, emission and meteorology data are readily available in the US. In Mexico, part of this information exist and other is being generated. To applied the model, the information needed was collected or estimated form different agencies. In particular, emission inventories of mobile and stationary sources were estimated using fuel consumption data and emission factors calculated for the Monterrey Metropolitan Area.

In 1994 the vehicular fleet in Mexican border cities with Texas accounted for more than 468,000 vehicles, which includes private and public service vehicles in different categories: cars, trucks, buses, and motorcycles. It was estimated that Ciudad Juarez had approximately 58.3% of the vehicular fleet, followed by the area of Reynosa-Matamoros with 36.6%. The vehicular fleet estimated did not include vehicles from other states which may had been in transit in the region.

An important characteristic of the vehicular fleet of this region is the high number of US vehicles in Mexican cities: in average, there are seven US cars per each Mexican car. We may expect that US cars circulating in Mexican border cities will have a better emission control technology than

Mexican cars; however, maintenance of the US cars running in Mexico is poor and the emission control equipment may not be operating appropriately. Furthermore, poorly tuned US cars can emit more pollutants than a Mexican car. We found also a direct relationship between population and the number of vehicles. Another important characteristic of the vehicular fleet is that about 60% of the Mexican vehicles are models 1983 or older. Only about 8% are models 1990 and above. The distribution of models is similar in the different cities along the border.

Distribution of fuel consumption data are important to estimate emission of air pollutants in different areas of the border region. In the north of Mexico natural gas is commonly used as fuel in the industry. The average total consumption of natural gas of Cd. Juarez, Piedras Negras, Nuevo Laredo, Reynosa and Matamoros for 1992, 1993 and 1994 was 1,800,000 cubic meters/day. Cd. Juarez consumed about 76% followed by Nuevo Laredo and Reynosa. Gasoline shows an increase in consumption of Magna-SIN (unleaded) and a decrease in NOVA, (a gasoline with lead). In July 1994 the consumption of Nova represented only 6% of the total consumption. The consumption of diesel in border cities has been stable for 1992, 1993 and 1994.

Studying the dynamics of air pollution and evaluating air pollution control strategies for the border region is not a simple task. The region comprises urban and rural areas of different characteristics. While most part of the region is arid and dry, some areas close to the Gulf of Mexico are humid and used for agriculture. In the region, few anthropogenic sources of air pollutants exist in rural areas, being the most concentrated in urban areas. Although air pollution problems have only been detected in some areas where monitoring stations exist (Tijuana-San Diego, Juarez-El Paso and the Monterrey metropolitan area), it is desirable to study the dispersion of air pollutants and study the impact in rural areas.

6.1.1 Objectives

The primary objectives of this study were to apply a photochemical air quality model to better understand the dynamics of photochemical pollutants (ozone, acids) in the Mexico-Texas border region and to make a preliminary assessment of air pollution control strategies for the region. Several secondary objectives were accomplished during this project: modeling tools and their application were transferred between the two institutions, graduate students were trained in air quality modeling, emission, meteorology and air quality data were gathered or estimated for the region, and a Geographic Information System data base was created to process data for the model and to visualize the results.

6.1.2 Photochemical Model Application

The CIT Photochemical Airshed Model was used to study the dynamics of ozone and acids along the Mexico-Texas border area. The model considered 80x136 cells, that covered an extension of 1000 by 1700 kms, i.e., the grid size was 12.5x12.5 kms, in the horizontal directions. In the vertical direction six layers of different width were considered. The width of the ground layer was 30 m, the second 70m, the third 200m, the fourth 400m, the fifth 800m, and the ninth 1600m, given a total of 3,100m height. The selection of widths was based on previous air quality studies. The domain's edges were 100 kms east of Houston, 550 kms west of Juarez/El Paso to include important SO2 and other pollutant emissions of Arizona that may have some effect in air quality in Juarez/El Paso. The edge in the south was 150 kms south of the Monterrey Metropolitan Area and 100 kms north of the Dallas/Fort Worth area. To test the model, the week from July 18 to July 24 1993 that had peak levels of ozone in the Monterrey Metropolitan Area was chosen. This period showed also low cloud coverage that reduces corrections when solar radiation is calculated. Also, very low rain was present in the domain. The CIT model solves the advection-diffusion equations using a regular grid.

To apply the model, an emission inventory of NOx, SO2, CO and VOC of stationary and mobile sources of the Mexican states of Tamaulipas, Nuevo Leon, Coahuila and Chihuahua was estimated. This emission inventory was estimated using fuel consumption data and emission factors for different mobile and stationary sources. The emission factors were obtained in a previous study of the Monterrey Metropolitan Area. This inventory was integrated with the emission inventories of Texas, New Mexico, and Arizona. Emissions inventories of these US states were obtained from Texas Natural Resource Conservation Commission and similar agencies in the other states.

As part of the project, land use data were integrated from the US and Mexico to be used in the model. Digital maps of land use 1:250,000 of the US Geological Survey (USGS) and maps 1:1,000,000 from Instituto Nacional de Estadistica, Geografia e Informatica were used for this purpose. The data were integrated according to the classification of land use of the model, which is consistent with the USGS classification.

Meteorology data of the National Climatic Data Center, in the US, and of the Sistema Meteorologico Nacional and Servicio a la Navegacion en el Espacio Aereo Mexicano, in Mexico, were used to obtain wind field, humidity and temperature data in the domain using a diagnostic model. Air quality data in the domain were estimated using information from US and Mexican monitoring stations. The data were using an interpolant computer code specifically designed to generate data for the CIT photochemical model.

A data base was created in a Geographic Information System (GIS) using ARC/INFO where emissions, land use and meteorology data were loaded. The political division of the states and counties of the states, the computational grid, and population density were included in the GIS data base. Air quality and meteorology monitoring stations were located geographically using the GIS. For example, Figure 1 shows the geographical distribution of elevated stationary SO2 emission sources along the border area. Similar information exist for CO, HC, and NOx, for stationary and mobile sources. Then, with ARC/INFO, an automatic data logging procedure was developed to create the data files for the CIT model assigning stationary and mobile source emissions into the mathematical grid, used by the model, from information located geographically and by counties in the region under study. This procedure minimizes errors when the data are transported from their location in counties to the grid used by the model. For example, Figure 2 shows CO emissions of mobile sources of each county in the domain and Figure 3 shows the same emissions loaded into the computational domain. It is important to note in Figures 2 and 3 that the scales of intensity are different because the data are received in tons/year/km2 and the model uses ppmv-m/min. A similar procedure was followed to locate air quality and meteorological stations in the grid to generate the data files necessary to run the model.

The model was tested to predict ozone concentrations and to evaluate air pollution control strategies. As expected, the model predicted that the Juarez/El Paso and Monterrey metropolitan areas showed the higher levels of ozone concentrations in the region, as can be seen in Figure 4. The national US standard of ozone concentration is 120 ppb. Measurements of ozone concentrations in El Paso are around 200 ppb during summer time and 100 to 130 ppb in the Monterrey metropolitan area. Figure 3 shows that similar values are predicted by the model.

Once the photochemical model is tested and validated comparing predictions with actual measurements of air pollutants concentrations, the model is a powerful tool to evaluate air pollution control strategies for ozone. To control this pollutant, actions can be taken reducing emissions of HC, NOx or both. Different strategies can be proposed to reduce ozone to given level, for example the standard of 120 ppb, and different strategies may be enforced if we want to reduce peak or average ozone levels in a given region. However, it is important to evaluate not only the effectiveness of the strategy to control ozone, but its cost too, i.e., the model can be used to assess cost-effective air pollution control strategies.

In the project a preliminary evaluation of air pollution control strategies was made. Results show that NOx control seems to be the most reasonable approach to reduce ozone concentration in areas that present peak ozone levels, like Juarez/El Paso and the Monterrey metropolitan Area. The case of no-control is useful to compare the predictions of the model with actual measurements of monitoring stations, as well as possible areas of high contamination which may be included in monitoring projects to validate results of the model. Also, the case of no-control shows predictions of air quality for the region, something that would be extremely expensive to obtain from directly monitoring air quality data. The cases of controlling HC, NOx or both show the impact in air quality of each strategy. The reduction of these pollutants depend on the strategy proposed. The reductions proposed here are used only to show the response of the model. From these examples, we can say that NOx control seems to be the most effective in reducing levels of ozone, but the cost of each strategy needs to be evaluated to support decisions about which strategy can be enforced and what results are expected.

Although the results obtained with the model seem reasonable, the application shows that is necessary to improve the emission inventory and meteorology information to obtain more reliable results.

6.1.3 Lessons, Opportunities, and Limitations

The project enhanced the research capacity of the working team. Students were trained in air quality modeling using state-of-the science photochemical models. Also, students were trained in

GIS software usage to handle large amounts of data to create the necessary data files for the model as well as in scientific visualization for better interpretation of results obtained from the photochemical model.

The project has been the foundation for other projects for the border area. A project will start in September to study ozone dynamics in the area of Juarez/El Paso and other project is underway in the area of Reynosa-McAllen/Matamoros-Brownsville to study the dynamics of ozone and PM10.

Several limitations were found in the development of the project. The emission inventories are less proven, and are subject to significant evaluation. Moreover, in the case of Mexico, the emission inventory was estimated from fuel consumption data using calculated emission factors obtained from a previous study of the Monterrey metropolitan area. These factors were obtained analyzing data of the vehicular verification program and of environmental emission audits of several companies. These emission factors must be calculated more extensively for the border region in order to improve our results, in particular in the case of Mexican vehicles. Also, emissions of ROG need to be speciated according with the process where fuel is consumed and their diurnal variation must be considered. In this project a typical speciation for ROG in the region was used and diurnal variations in emissions were neglected.

Meteorology data were available for several monitoring stations along the border region, but more data are necessary to improve the results obtained from a diagnostic meteorology model. Due to the extension of cells (12.5x12.5 kms), effects of topography can not be taken into account with great detail to predict wind fields in areas where hills are found; for example, close to El Paso/Juarez and Monterrey metropolitan area. It is possible to consider effects of topography if data are available in digitized format to minimize error reading from maps. Nevertheless, the results show the general trend in ozone formation and transport along the border area.

Air quality data was very limited since few monitoring stations exist; in particular for the case of Mexico only two cities are monitored in the domain (Monterrey and Juarez). The effect of wind in air quality may be significant if the grid size is reduced, in particular where hills are found. This may affect the predicted areas of air pollution impact along the border area.

Solar incoming radiation data may be improved too. In the project information of meteorology stations to interpolate cloud coverage in the region was used, then the model estimates correction factors for the incoming radiation. Scaling factors for UV radiation were taken as 1, i.e., UV radiation data were not corrected. To obtain more reliable results, incoming UV scaling factors must be obtained from experimental studies along the region.

For Mexico, land use data were obtained from maps, assigning to each county the land use in accordance with the model classification. This may be improved if the data were digitized and errors minimized when the data are loaded into the GIS. In the case of the US, digitized data were obtained and were loaded directly into the GIS:

The CIT Photochemical Model applied to the Mexico-Texas border region uses a regular grid, therefore, low resolution is obtained in urban areas since squares of approximately 150 squared kms are assumed to be uniform in emissions and air pollutant concentrations. For better predictions of the dynamics of air pollutants, it is desirable to use a more refined grid in urban areas, and a coarse grid in rural areas to reduce the number of operations. This is possible with a multiscale model. Further development of appropriate models for regional applications must be developed.

Despite the limitations found, the application of the photochemical model to study the dynamics of ozone and acids in the region attracted the attention of several US and Mexican government agencies and institutions. Results of this project have been presented in seminars, congresses, workshops, and symposiums, and published in proceedings, extended memories or briefing reports. This project has been a foundation for future air quality studies in the border region.

7. AIR POLLUTION CONTROL STRATEGIES

Several air pollution control strategies may be enforced to reach a given standard of air quality for a given urban area or region. The key issue is to find the best cost-effective strategy that will lead to the desired air quality. For this purpose air quality strategies are evaluated in terms of their technical, economical, and social feasibility. Of course, the evaluation is not simple and usually air quality models are used as tools to evaluate predicted changes in air quality when a strategy or a set of strategies are proposed for a given area under study. As a consequence, the effectiveness of strategies change with the region under study since the meteorology, amount and type of pollutant, solar radiation, land use, etc. are different in each region under study. This issue is very important when large range transport of air pollutants is studies. Strategies that may be effective in a region under study to improve its air quality may not apply in other region and, the most important, air quality in one region may be affected from the transport of pollutants emitted in another region. The role that the study of continental pollutant pathways take in this point becomes a matter of directing policies to protect areas controlling emissions in other regions. The problem is delicate when emissions of one country may be affecting other country. The role of the Commission for Environmental Cooperation becomes very important to study and solve problems already existing, that will be solved only with the existing good will and cooperation of Canada, the United States and Mexico. Despite this commitment of cooperation of the three countries, little is know about long range transport of air pollutants in North America.

Mobile sources are major contributors of ozone, acid rain, and respirable particulates precursors and strategies involve use of better vehicle technologies, retirement of old cars, retrofit of vehicles, move to electric vehicles, use of alternate fuels, commitment to supply renewable fuels, inspection and maintenance programs, mechanical training, use of public transportation and control of evaporative emissions in cars, during fueling, and in gas stations. Controlling stationary sources of emissions may be easier since emissions are concentrated in major sources like power plants, mining and melting industries. General strategies require the use of devices to control directly emitted particle emissions, use of low NOx production burners, fuel changes to reduce sulfur and hydrocarbon emissions, use of catalytic converters, use of electric motors, and other. Controlling natural sources is difficult. For example, it is necessary to preserve natural areas that may contribute with HC that are ozone precursors, but with reforestation programs we prevent erosion and suspension of particles by wind. An important issue are new sources of air pollution. New industries are being located in the countries, traffic activities are increasing and new highways are under construction. It is important to determine the best location of new sources to minimize the impact in air quality in the continent.

Evaluating and enforcing air pollution control strategies in border areas is not simple. In the case of the Mexico-US border the problem is more complex than in the US-Canada border. Some of the major differences are technologies and age of the vehicular fleet, fuel formulations, type of industries, highway communications, average income of population, land use, commercial activities, and social behavior. All these differences increase the difficulty to evaluate air pollution control strategies in the Mexico-US border area. The same border areas share common airshades, natural resources, and are influenced by long range transport of pollutants emitted in either country. Of course, the same effect may happen in areas within a country. As a consequence, only through integrated studies with the participation of institutions of the two countries feasible solutions to improve air quality in border areas or within a country will be accomplished.

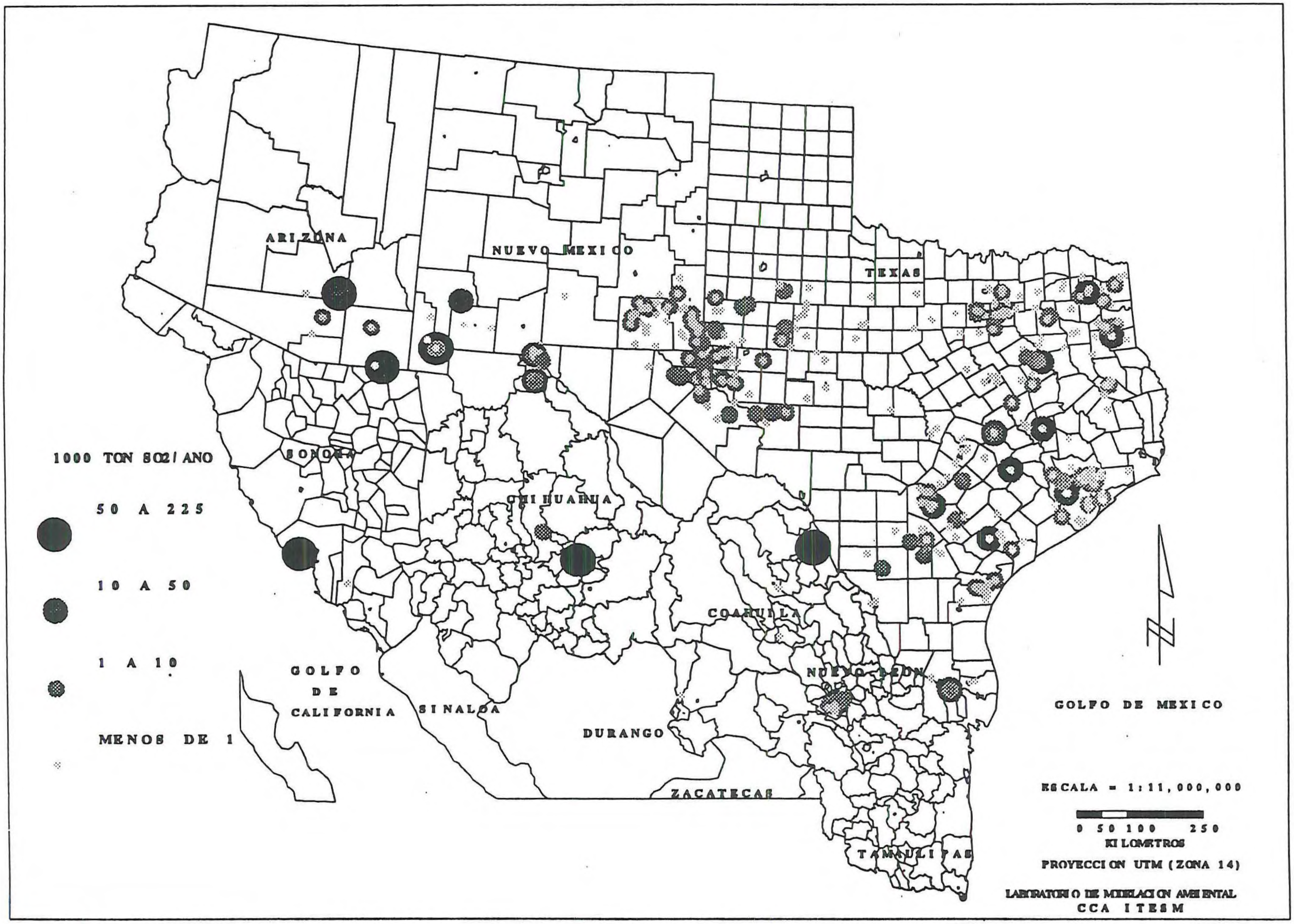

Figura 1. Emisiones puntuales de SO_2 en el dominio de modelación.

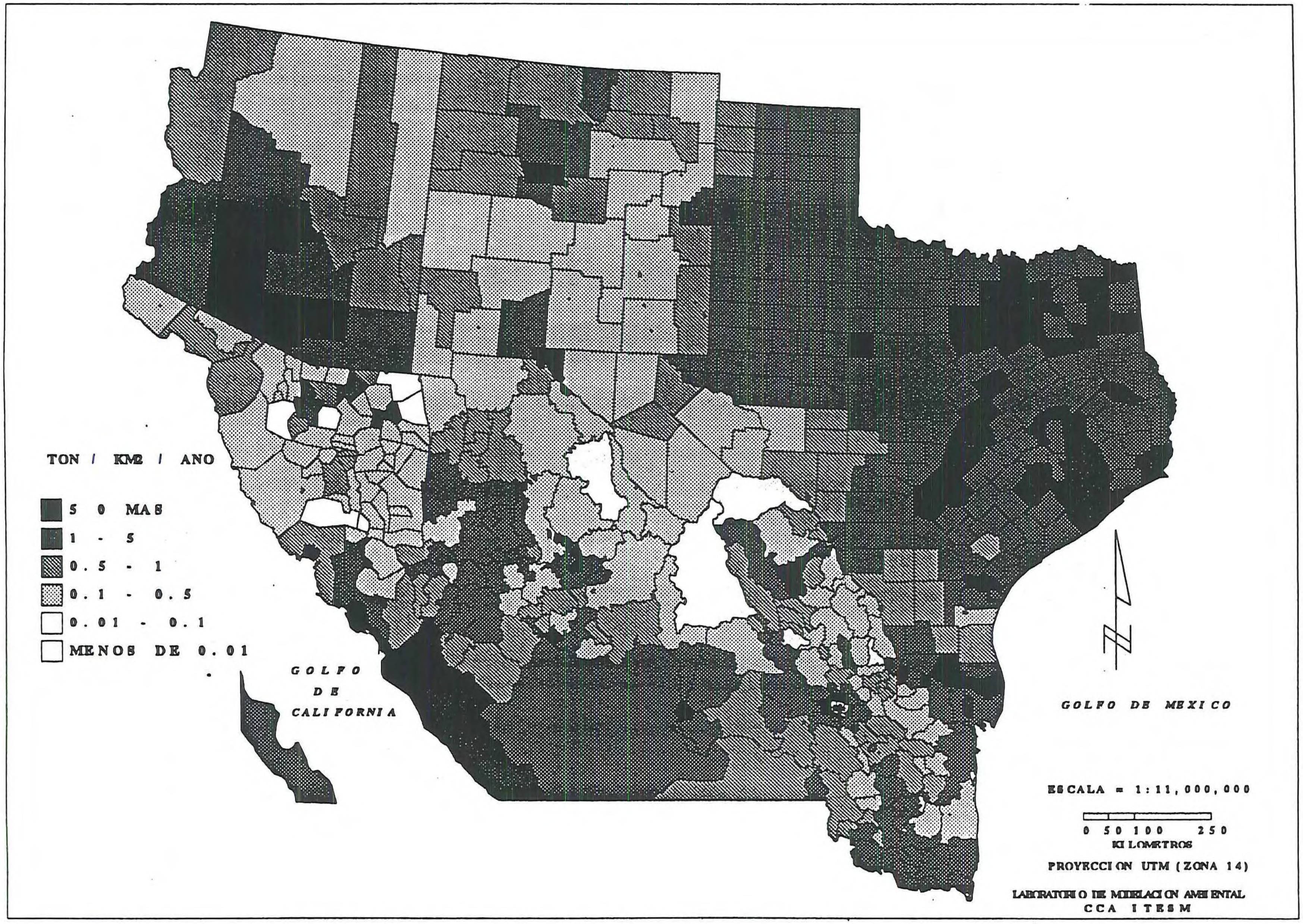

Figura 2. Emisiones de CO debidas a fuentes móviles.

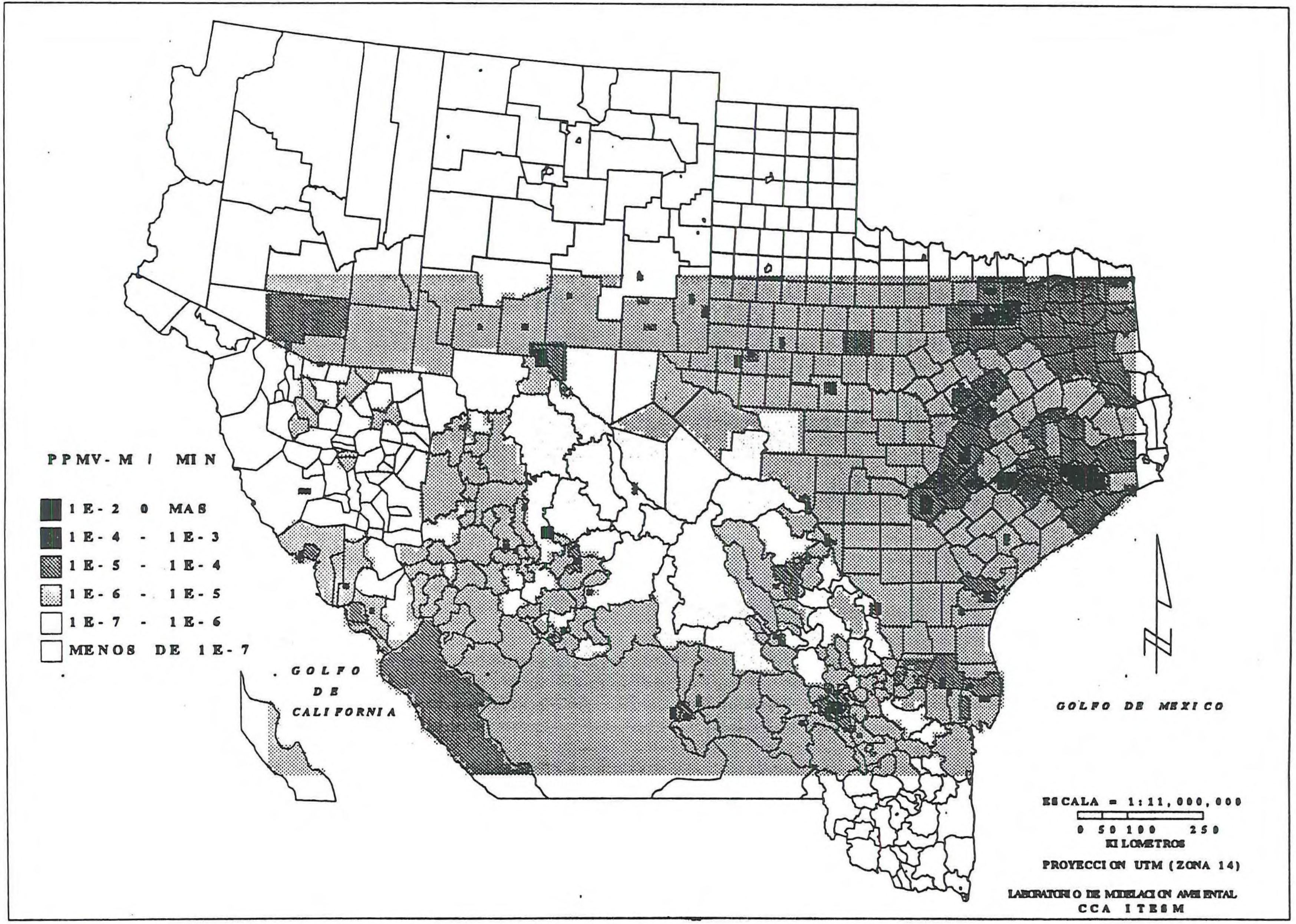

Figura 3. Emisiones de CO debidas a fuentes móviles trasladadas a la malla computacional.

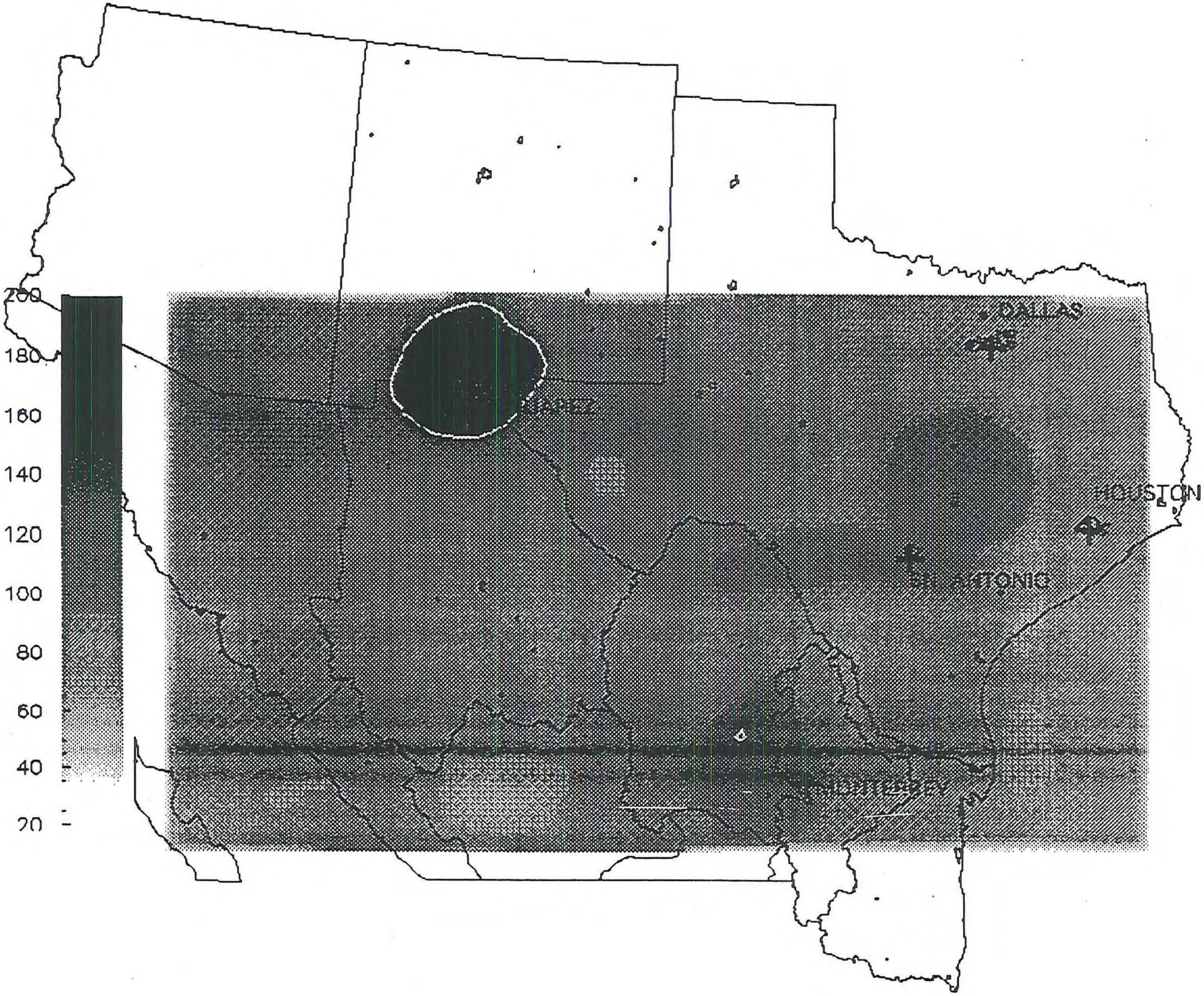

Figura 4. Concentración de ozono en ppb calculada a las 15 hrs. (18/07/96).

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Case Study

Acid Rain

Authors:

Tom Brydges- Environment Canada, Burlington, Canada
Humberto Bravo- Centro de Ciencias de la Atmosferica, Mexico City, Mexico
Rudolfo Sosa Echeverria- Centro de Ciencias de la Atmosferica, Mexico City, Mexico

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1.0 INTRODUCTION

In the late 1960's, Svante Oden, using data collected in Sweden, described the phenomenon of acid rain caused by the long range transport of pollutants. Acid rain is caused by pollutants, such as sulfur dioxide (SO_2) and nitrogen oxides (NOx), which in the atmosphere are converted chemically to sulfuric acid and nitric acid. Diluted forms of these acids fall to earth as rain, hail, drizzle, freezing rain or snow (wet deposition), or are deposited as acid gas or dust (dry deposition). Normal rain is slightly acidic, due to the presence of carbon dioxide, but acid rain can be as much as 100 times more acidic due to the presence of sulfuric and nitric acids. Although natural sources of SO_2 and NOx emissions existing in North America, most of the emissions come from human activity.

Acid rain is an in an interesting case study because the three countries are at different stages of dealing with the problem. Acid rain became a Canadian environmental issue in the 1970's. Scientists had noted losses of fish populations in some highly acidified lakes of northern Ontario and acid rain was documented in the Muskoka/Haliburton area of south central Ontario.

Acid rain became a Canada/USA bilateral issue when the United States sent a diplomatic note to Canada in November 1978 requesting negotiations aimed at developing appropriate emission control programs. The Canada/US Air Quality Accord was formally signed in 1991.

In more recent times, information has been gathered in Mexico documenting low pH rain due to mineral acids. Acid rain was first detected in the Mexico City metropolitan area in 1980. It is also prevalent in the Vera Cruz area on Mexico's east coast. The existence of acid rain in Vera Cruz demonstrates the problem of long range transport of acid rain precursors, as there are no significant contributing sources in this area.

2.0 EFFECTS OF ACID RAIN

Extensive damage to ecosystems, particularly sensitive surface waters, was the first major concern in Canada. In eastern Canada, more than 700,000 lakes sensitive to acidification are exposed to elevated levels of atmospheric sulfate deposition.

Damage to other components of the environment, such as forests and building materials, became of concern in both Canada and the United States. Several detailed assessments have been published in both countries. (RMCC, 1990; NAPAP, 1990)

An example of the effects of acid rain is provided by Mexico's Mayan Monuments, designated as "Monuments to Mankind" by UNESCO. The Monuments are constructed of limestone, which is highly susceptible to acidic solutions. The Monuments now show the effects of acidic precipitation, and permanent damage will result if acid rain persists in the area. Upper wind back trajectors ending at the Yucatan Peninsula is presented in the figure 1 (Bravo et al.,. 1991.)

In 1986, the National University of Mexico, the Governments of Mexico and the United States launched a cooperative initiative to investigate the effects of acid rain in the Gulf of Mexico. Specifically, the study was designed to examine impacts in air quality in the area of interaction between land and sea. The study results demonstrated that the Gulf of Mexico air was highly polluted, with significant concentrations of anthropogenic aerosols, as well as dust, SO_4 and NO_3. In addition, it was illustrated that the dry deposition of aerosols is a continuous and important process affecting air-to-Gulf fluxes of air pollutants. Rain samples, especially those collected near large port cities in Mexico, were found to be extremely acidic, with large amounts of SO_4 and NO_3 aerosols present, in addition to sea salt. Further study is needed to assess the ecological and environmental impacts of acid rain on the Gulf of Mexico (Parungo et al.,1990;.Paringo and Miller 1988).

3.0 ACID RAIN CONTROLS

Emission control programs for SO_2 and NOx have been extensive and are quite complex in both Canada and the United States.

3.1 Canada

Although there were numerous industry specific control actions beginning in the 1960's, the seven eastern Provinces agreed in 1984 to reduce their total SO_2 emissions to 2.3 million metric tons by 1994. Eastern Canadian emissions of SO_2 have fallen by 56% from 3.8 million metric tons in 1980 to 1.7 million tons in 1994. The target of reducing these emissions to 2.3 million metric tons by the year 1994 was surpassed. Each of the seven easternmost provinces met its individual target.

The emission reductions are a result of industrial process changes, installation of scrubbers, and fuel-switching in the last five years. The largest emission source in Canada is the smelting of metal ores, which accounted for 50% of total eastern Canada SO_2 emissions in 1994. Power generation and other sources contributed 20% and 30% respectively.

Canada is committed to a permanent national limit on SO_2 emissions of 3.2 million metric tons by the year 2000. Canada met this goal in 1993. By 1994, its SO_2 emissions were down to approximately 2.7 million metric tons, a 41% reduction from the 1980 level of 4.6 million metric tons.

Total Canadian NOx emissions have remained relatively constant from 1980 to 1990. Mobile sources (i.e., on-road transportation such as cars, truck) accounted for 35% of total Canadian NOx emissions in 1990. Electric utilities and industrial sources contributed 12% and 23%, respectively, while other sources contributed the remaining 30%. Under the Canada-United States Air Quality

Agreement, Canada is committed to a 10% reduction in projected NOx emissions from stationary sources by the year 2000.

3.2 United States

The 1990 Clean Air Act includes an innovative program to reduce acid air pollutants (acid snow, acid fog or mist, acid gas and acid dust, all referred to here as "acid rain"). The Act takes a new nationwide approach to the acid rain problem. The law sets up a market based system designed to lower sulfur dioxide pollution levels. Beginning in the year 2000, annual releases of sulfur dioxide will be about 40% lower than the 1980 levels. Reducing sulfur dioxide releases should cause a major reduction in acid rain.

Phase I of the acid rain reduction program went into effect in 1995. The U.S. is committed to reducing its annual SO_2 emissions by the year 2000 to 14.4 million metric tons (15.8 million tons), 9.1 million metric tons (10 million tons) below 1980 levels. Big coal-burning boilers in 110 power plants in 21 Midwest, Appalachian, southeastern and northeastern States will have to reduce releases of sulfur dioxide. In 2000, Phase II of the acid rain program goes into effect, further reducing the sulfur dioxide releases from the big coal-burning power plants and covering other smaller polluters. Total sulfur dioxide releases for the country's power plants will be permanently limited to the level set by the Clean Air Act for the year 2000.

Reductions in sulfur dioxide releases will be obtained through a program of emission (release) allowances. The Environmental Protection Agency (EPA) will issue allowances to power plants covered by the acid rain program; each allowance is worth one ton of sulfur dioxide released from the smokestack. To obtain reductions in sulfur dioxide pollution, allowances are set below the current level of sulfur dioxide releases. Plants may only release as much sulfur dioxide as they have allowances. If a plant expects to release more sulfur dioxide than it has allowances, it has to get more allowances, perhaps by buying them from another power plant that has reduced its sulfur dioxide releases below its number of allowances and therefore has allowances to sell or trade. Allowances can also be bought and sold by "middlemen", such as brokers, or by anyone who wants to take part in the allowances market. Allowances can be traded and sold nationwide. There are stiff penalties for plants which release more pollutants than their allowances cover.

The acid rain program provides bonus allowances to power plants for (among other things) installing clean coal technology that reduces sulfur dioxide releases, using renewable energy sources (solar, wind, etc.) or encouraging energy conservation by customers so that less power needs to be produced. All power plants under the acid rain program will have to install continuous emission monitoring systems (CEMS), machines that keep track of how much sulfur dioxide and nitrogen oxides the plant is releasing. A power plant's program for meeting its sulfur dioxide and nitrogen oxide limit will appear on the plant's permit, which will be filed with the State and EPA.

To cut down on nitrogen oxide pollution, EPA will require power plants to reduce their nitrogen oxide releases, and will require reductions in nitrogen oxide releases from new cars. Reducing nitrogen oxide releases will reduce both acid rain and smog formation. The flexible market-based acid rain reduction program is expected to be a model for pollution control efforts in the United States and other countries.

3.3 Mexico

Mexico has not yet initiated any emission controls specifically to control long range transport of SO_2 or NOx.

4.0 BENEFITS OF CONTROLS

4.1 Canada

Canada has an objective of no more than 20 kg/ha/hr of wet sulfate deposition falling in any sensitive area. The area of eastern Canada receiving 20 kg/ha or more of wet sulfate per year has declined by nearly 59%, from 0.71 million km^2 in 1980 to about 0.29 million km^2 in 1993.

Lake sulfate levels are indicators of the primary acidifying agent (sulfuric acid) in acid rain. Sulfate levels are declining in most of the lakes in Ontario and Quebec, whereas the sulfate in most lakes in the Atlantic Region is stable. Lake sulfate levels respond to a reduction in SO2 emissions, However, there is a time lag of possibly many years before this translates into widespread regional improvement in lake pH or alkalinity. After a further time lag, such improvements are likely to result in changes to fish populations and other biota.

Lake acidity levels are indicated by pH (a direct measure of acidity) and alkalinity (the capability of water to buffer or neutralize acidity). Between 1981 and 1994, increases in pH or alkalinity indicated improvements in lake water acidity. A total of 202 lakes were monitored for acid rain effects in Ontario, Quebec and the Atlantic Region between 1981 and 1994. Of these, 33% showed some improvements in acidity. Acidity levels were stable in 56% of the lakes and 11% were becoming worse.

Only in the Sudbury region of Ontario did the majority of monitored lakes show an improvement in acidity. This is attributed to the substantial control of SO2 emissions from Sudbury's nickel and smelters. Despite significant SO2 controls in eastern Canada, the acidity of most lakes showed little change (stable) in the remainder of Ontario, Quebec and the Atlantic Region due to the continuing transboundary flow of acidifying emissions from the Untied States.

5.0 RECOMMENDATIONS

1. Continue to monitor effects and benefits of control programs in Canada and the USA

2. Enhance terrestrial sampling of acid rain in Mexico.

3. Development of inventory of acid rain precursors is of great need in Mexico

4. Effects of acid rain on Mayan Monuments and other monuments of historical heritage in North America needs to be defined.

5. Conduct a second cruise on Gulf of Mexico basin in a joint research project by Mexico/USA and Canada, participating with the University of Mexico (Seccion de Contaminacion Ambiental), NOAA from the USA and an Ad Hoc research group from Canada.

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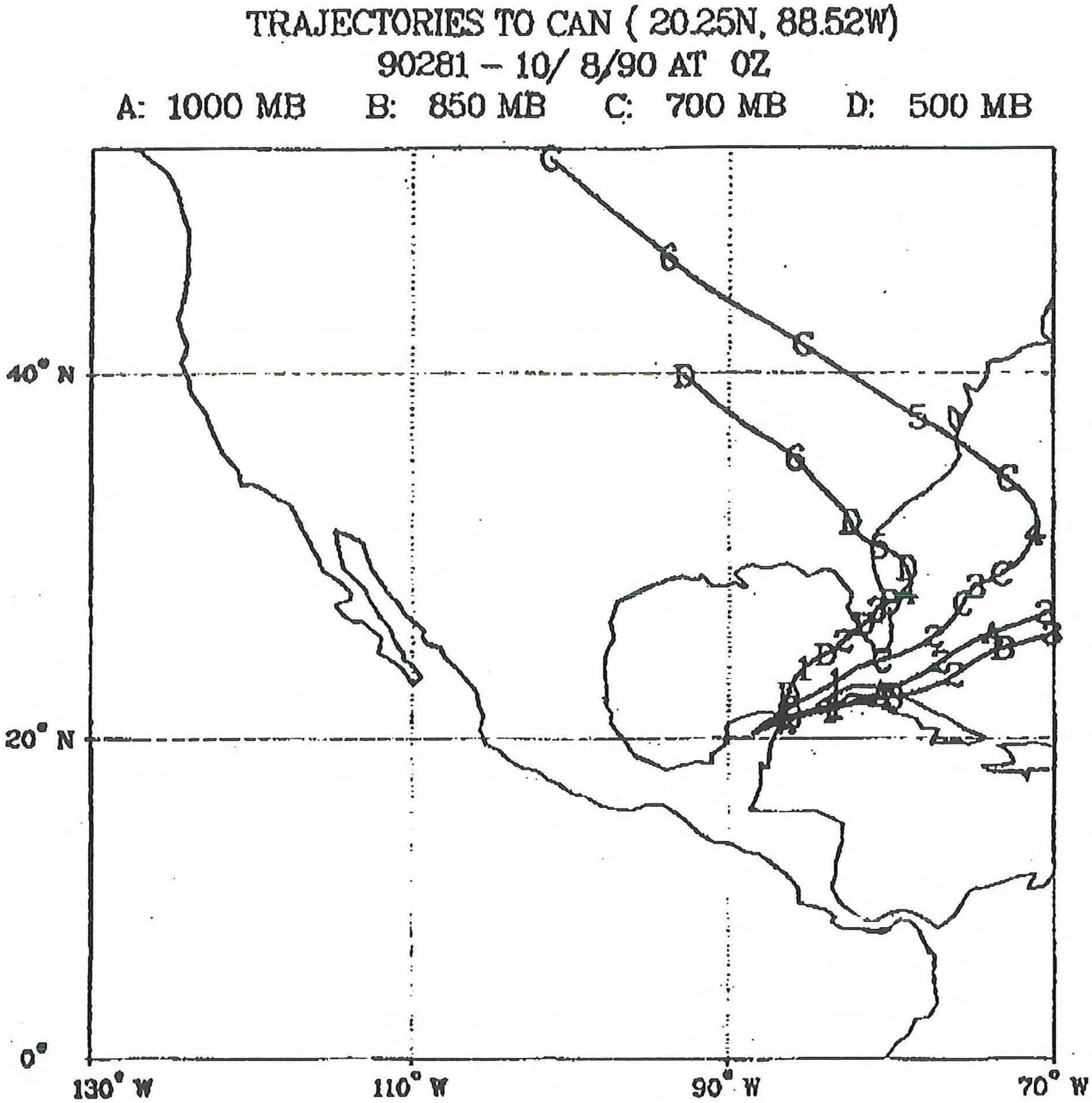
Figure 1. Upper wind back trajectory ending at the Peninsula de Yacatan in 10/8/90.



H. CASE STUDIES ON SOURCE-RECEPTOR RELATIONSHIPS

Draft Interim Report to the Secretariat fo the Commission for Environmental Cooperation

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Case Study

Source Receptor Relationships

Authors:
James W.S. Young- SENES Consultants Limited, Richmond Hill, Canada
Zivorad R. Radonjic- SENES Consultants Limited, Richmond Hill, Canada
Diane V. Michelangeli- SENES Consultants Limited, Richmond Hill, Canada
Francisco Guzman- Instututo Mexicano del Petroleo, Mexico City, Mexico
Carlos Santos-Burgoa-Instituto de Salud, Ambiente y Trabajo, Mexico City, Mexico

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1.0 INTRODUCTION

1.1 Background

In support of the Commission on Environmental Cooperation's initiative on "Continental Pollutant Pathways", an elucidation of the primary source - receptor links among Canada, the United States and Mexico is needed to allow the development of recommendations regarding transboundary and border environmental issues. This case study presents a scoping unit transfer matrix for Canada, the United States and Mexico which was produced using a simplified air dispersion/deposition model. This matrix consists of 100 receptor points and 99 emission sources covering the three countries..

An estimate is to be given of the magnitude of the error involved from the use of this simplified model by comparing the model output to the observed wet deposition and measured air concentration data.

1.2 Approach

After reviewing a number of simple approaches to this Long Range Transport (LRT) modelling problem, SENES decided that the best approach would be to use a fully configured LRT model. The model selected was CALPUFF (Version 4.0) which was developed by the U.S. EPA in the 1995 and finalized in the 1996 to provide a modelling system that was capable of (1) treating time varying point and area sources, (2) modelling domains from tens to hundreds of kilometres from a source, (3) predicting for averaging times ranging from one hour to one year, (4) being applied to inert pollutants and those subject to linear removal and chemical conversion mechanisms, and (5) being applied in rough or complex terrain situations.

This model can also be used in regulatory studies to assess the impact of sulphur oxides emitted from major point and area sources over mesoscale transport distances.

The formulation used was very simple - pollutants were assumed to be emitted from either (1) the emissions centroid of each state/province where enough information existed to calculate the location or (2) major cities. The receptors were also selected arbitrarily as the geographical centre of each state/province. This means that the source-receptor links derived should give a broad conceptual picture of acidic deposition but may be quite inaccurate in location and size of maxima. This accuracy is tested as part of this case study.

2.0 THE CALPUFF/CALMET MODELLING PACKAGE

The CALPUFF/CALMET modelling system includes (1) meteorological wind field generators, (2) a dispersion model with chemical removal, wet and dry deposition, complex terrain algorithms, building downwash, plume fumigation and other effects, and (3) postprocessing programs for the output fields of meteorological data, concentrations and deposition fluxes.

The first part of the modelling system contains programs (METSCAN, READ62, SMERGE, PXTRACT, PMERGE) for preprocessing the meteorological data. CALMET is a meteorological model which includes a diagnostic wind field generator containing an objective analysis and parameterized treatments of slope flows, kinematic terrain effects, terrain blocking effects, a divergence minimization procedure, and a micrometeorological model for overland and overwater boundary layers. CALMET can handle the curvature of the earth correction as well as the wind rotation due to the fact that the position of grid North changes.

The second part of the modelling package contains the actual dispersion/deposition processor. CALPUFF is a non-steady-state Lagrangian Gaussian puff model containing modules for complex terrain effects, overwater transport, coastal interaction effects, building downwash, wet and dry removal, and simple chemical transformation.

The third part of the modelling package contains two post-processing programs: PRTMET - which can be used to select portions of the meteorological information and CALPOST - which can be used for extracting different averaging periods for predicted concentrations and wet/dry deposition fluxes. In this particular application, a SENES postprocessor (SUM) was also used to produce output in the form of a Transfer Matrix.

2.1 Modelling Domain

The meteorological grid used was 100 by 100 km within a domain size of 5600 by 5300 km. The co-ordinate origin was at Latitude 19° North and Longitude 130° West. The computational grid can be the same size or smaller. For this study, the predictions were made at discrete receptors and no computational grid had to be defined. Figure 2.1 presents the modelling domain and the meteorological grid.

2.2 Meteorological Preprocessors

METSCAN is a meteorological preprocessor which performs quality assurance checks on the hourly surface meteorological data in the NCDC CD-144 format which is used as input to the SMERGE.

READ62 is a preprocessor program which reads and processes the twice-daily upper air wind and temperature sounding data available from the National Climatic Data Center (NCDC). A new version of READ62 extracts the data from the NCDC CD ROM format. All Canadian, U.S.A. and Mexico upper air data were available on the same CD ROM. The output of READ62 is required as an input into the CALMET meteorological preprocessor. READ62 scans the upper air data for completeness, with warning messages to flag missing or incomplete soundings.

PXTRACT is a preprocessor which extracts precipitation data from stations and time periods from the fixed length, formatted precipitation data file in the U.S. NCDC TD3240 format. SENES Consultants has designed a series of programs to transfer the Canadian AES format directly into the CALMET input format (directly compatible with PMERGE output). The Mexico data were handled in the same way as the Canadian data.

PMERGE reads the processed precipitation data from PXTRACT and reformats the data into the required format for the CALMET meteorological processor with the precipitation data sorted by hour rather than by station. PMERGE resolves Aaccumulation periods@ and flags suspicious or missing data. The output goes directly to CALMET as the precipitation input file (PRECIP.DAT).

SMERGE reads all surface data, combines them together and orders all stations by hour. The output goes directly to CALMET as the surface input file (SURF.DAT).

CALMET is the meteorological preprocessor which computes the time and space interpolated fields of the meteorological variables (wind field, mixing heights, etc.) required by CALPUFF to describe mesoscale transport and dispersion processes. CALMET uses individual upper air data files created by READ62, the SURF.DAT merged hourly surface meteorological data files and the merged precipitation data file, PRECIP.DAT. A single output file containing the gridded meteorological field is produced which serves as the input file to CALPUFF (for this project, the size of the CALPUFF meteorological input file in binary format is approximately 2 GB).

Other meteorological preprocessors that have been written by SENES in order to use the CALPUFF/CALMET modelling system include:

ORDER re-orders the sequence of hourly surface elements by row from the original Canadian AES data so that the order matches that required by the CD144 format.

AESCD changes the re-ordered Canadian AES hourly surface data into the CD144 format.

PRW changes the U.S. NCDC present hourly weather format into the CD144 format.

CANPRW changes the Canadian AES present hourly weather format into the CD144 format.

PRECIP converts the Canadian hourly horizontal precipitation format into a vertical hourly precipitation format.

DAILY distributes daily amounts of precipitation to the correct hours based on the hourly present weather records.

FINAL converts the hourly precipitation data into the PMERGE output format.

LAMCON calculates the Lambert Conformal co-ordinates required by CALMET from latitude and longitude for sources, receptors, surface stations and upper air stations.

LAND averages the National Land Use and Land Cover Inventory categories supplied by Environment Canada (originally from NCAR) over a one degree grid square.

LANDF assigns ones land use category to each grid point.

LANDPICK matches the modelling grid to the land use grid to determine the final land use categories for modelling.

2.3 Dispersion Processor

CALPUFF is a multi-layer, multi-species non-steady-state puff dispersion model which can simulate the effects of time- and space-varying meteorological conditions on pollutant transport, transformation and removal. It is a Lagrangian, Gaussian, variable trajectory, puff superposition model designed to account for spatial and temporal variations in transport, diffusion, chemical transformation and removal mechanisms encountered on the regional scale. With the puff superposition approach, a continuous plume is modelled as a series of discrete puffs. Each puff is transported independently of other puffs. A puff is subject to growth by diffusion, chemical transformations, wet removal by precipitation, and dry deposition at the surface. Up to five pollutants [sulphur dioxide (SO_2); sulphate (SO_4); nitrogen oxide ($NO_x = NO, NO_2$); nitric acid ($2HNO_3$); and nitrate (NOG_3)] may be modelled simultaneously.

2.4 Chemical Transformations

The chemical processes modelled in CALPUFF are the conversion of sulphur dioxide to sulphate and the conversion of nitrogen oxides to nitrate aerosol. Some of the technical algorithms are briefly described in U.S. EPA (1995):

Dry Deposition: *A full resistance model is provided in CALPUFF for the computation of dry deposition rates of gases and particulate matter as a function of geophysical parameters, meteorological conditions and pollutant species. Options are provided to allow user-specified, diurnally varying deposition velocities to be used for one or more pollutants instead of the resistance model (e.g. for sensitivity testing) or to by-pass the dry deposition model completely.*

Wet Deposition: *An empirical scavenging coefficient approach is used in CALPUFF to compute the depletion and wet deposition fluxes due to precipitation scavenging. The scavenging coefficients are specified as a function of the pollutant and precipitation type (i.e. frozen vs. liquid precipitation).*

Chemical Transformation: *CALPUFF includes options for parameterizing chemical transformation effects using the five species scheme (SO_2, SO_4, NO_x, HNO_3 and NO_3) employed in the MESOPUFF II model or a set of user-specified, diurnally-varying transformation rates.*

2.5 Post-Processor

SUM converts the output from CALPUFF, in the form of CALPUFF.LST files, into the Transfer Matrix format.

2.6 File Sizes

The size of each surface data file as received was 0.707 MB or a total of ~76 MB for the 107 meteorological stations used for this study. After preprocessing this was reduced to a CALMET input file (SURF.DAT) of about ~75 MB. The size of the raw upper air data file was ~175 MB (~2.5 MB per station for 70 stations). After preprocessing this was reduced to an input file of ~30 MB. The raw precipitation data files totalled 20 to 30 MB. After preprocessing the CALMET input file (PRECIP.DAT) size was increased to ~10 MB.

2.7 Caveats

The major caveat with this work is the representation of the sources of emissions by single point sources for each state or province. The data presented should not be used for control strategy development but rather to give national source-receptor links among the various states and provinces in North America. Errors up to a factor of 5 can be expected.

3.0 EMISSION SOURCES

For this study 100 sources are modelled and they are presented in Figure 3.1. Details of each source used is detailed in Tables 3.1, 3.2 and 3.3 which give the source name, source codes, and the sulphur dioxide emissions rates. We have also assumed that all emission sources are 50 metres in height with a 1 m/s exit velocity and 293°K exit temperature.

3.1 Canada

The Canadian emission sources were selected from a database on a 50 km grid, supplied via Yohannes Mariam by the emission inventory staff of Environment Canada in Ottawa. For the purposes of this scoping exercise each province or sub-province was given one emission source located at its emissions weighted centroid. Table 3.1 details the source locations and emission rates used for Canada. The total emissions for each province were supplied by Peggy Hallward of Environment Canada.

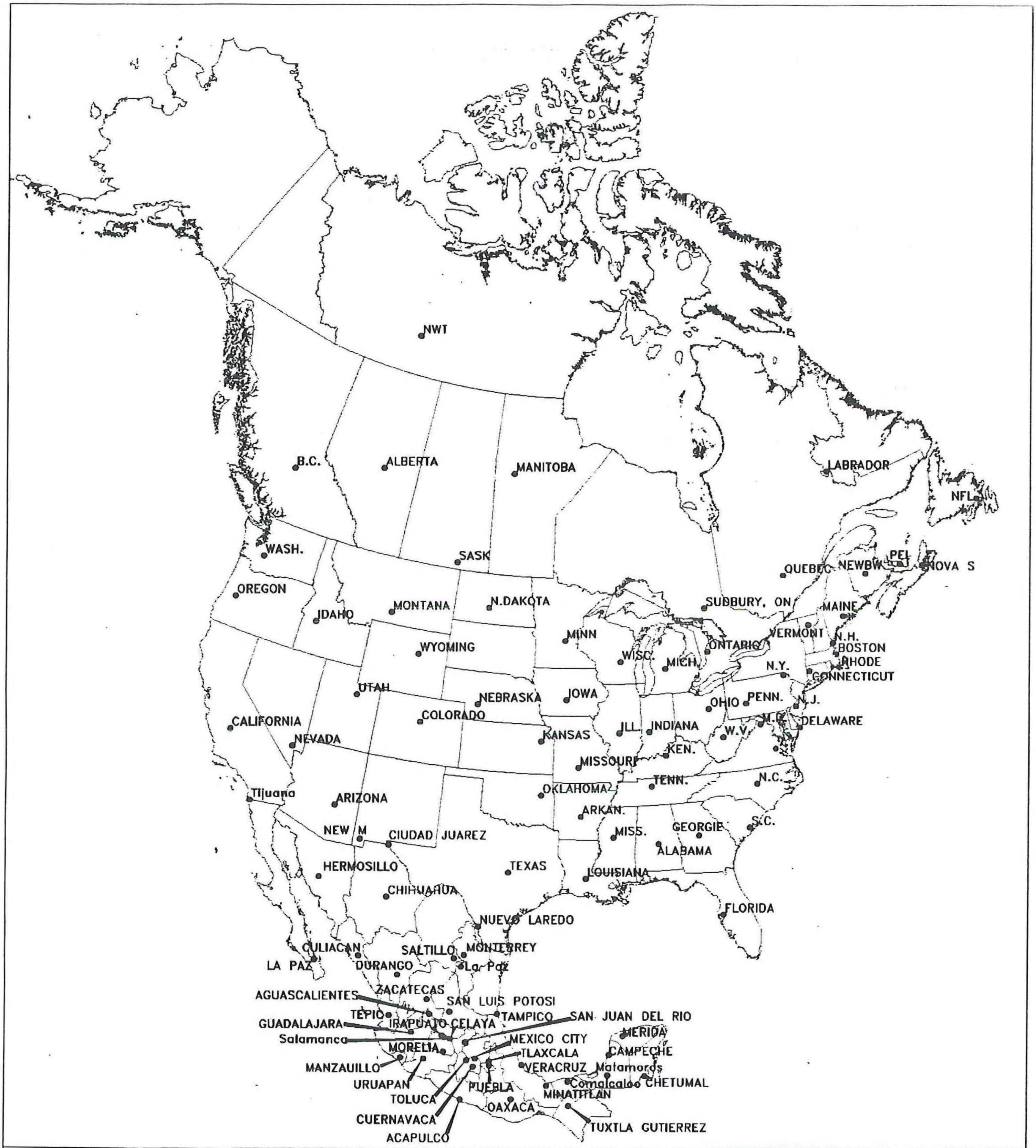
3.2 United States

The U.S. source information was also obtained from Yohannes Mariam of Environment Canada in Ottawa. Initially there were about 61,000 sources: those east of 92°W longitude were aggregated into 2° x 2° grid squares south of 36°N latitude, and 0.5° x 0.5° grid squares north of it. Those between 92°W and 102°W longitude were aggregated into 1° x 1° grid squares.

The first step was to select those U.S. grid squares with SO_2 emissions greater than 15,000 tonnes per year. These were used to define the emissions centroid for each state. The total emissions for each state were then assigned to that emissions centroid. Table 3.2 details the source emission rates used for the United States. The totals by state were calculated by prorating the 1994 emissions (EPA-454/R-95-011) by the 1990 total as shown in Table 3.2.

3.3 Mexico

The SO_2 emissions data for Mexico were supplied by members of the working group and were assigned to the closest major industrial area. Table 3.3 details the source names, codes and emission rates used for Mexico. The detailed assessment of the emissions is presented in Appendix B.



2131-2/06MAR87/XRF_SRC.DWG/M.T.N.

FIGURE 3.1

SOURCE LOCATIONS

Table 3.1

Canadian Total SO_2 Emissions by Province

CANADA (1990) - Environment Canada, August 1996 prepared by Peggy Hallward

Source Name	Source Code	Emission Rate (kilotonnes/year)	Emission Rate (grams/second)
Alberta	ALB	567	17980
British Columbia	BC	104	3298
Manitoba	MAN	516	16362
New Brunswick	NEWB	181	5740
Newfoundland	NFL	57	1807
Nova Scotia	NOVS	178	5644
Ontario (excluding Sudbury)	ONT	574	18202
Ontario (Sudbury)	ONTS	618	19597
Prince Edward Island	PEI	3	95
Quebec	QUE	391	12399
Saskatchewan	SASK	88	2790
Yukon & NWT	NWT	17	539
TOTAL		**3294**	**104453**

Table 3.2

American Total SO_2 Emissions by State

USA (1990 estimated) - National Air Pollutant Emissions Trends, EPA-454/R-95-011

Source Name	Source Code	1994 Emission Rate (short tons/year)	1990 Emission Rate [1994*22433/21119]	Emission Rate (grams/second)
Alabama	AL	740000	786042	21287
Alaska	AK	2000	2124	58
Arizona	AZ	336000	356906	9666
Arkansas	AR	108000	114720	3107
California	CA	204000	216693	5868
Colorado	CO	106000	112595	3049
Connecticut	CT	55000	58422	1582
Delaware	DE	95000	100911	2733
District of Columbia	-	7000	7436	201
Florida	FL	831000	882704	23905
Georgia	GA	793000	842340	22812
Hawaii	-	20000	21244	575
Idaho	ID	37000	39302	1064
Illinois	IL	1188000	1261916	34175
Indiana	IN	1754000	1863132	50457
Iowa	IA	255000	270866	7336
Kansas	KS	117000	124280	3366
Kentucky	KY	1063000	1129139	30579
Louisiana	LA	434000	461003	12485
Maine	ME	80000	84978	2301
Maryland	MD	363000	385585	10442
Massachusetts	MA	205000	217755	5897
Michigan	MI	523000	555540	15045
Minnesota	MN	143000	151897	4114
Mississippi	MS	234000	248559	6731
Missouri	MO	717000	761611	20626
Montana	MT	82000	87102	2359
Nebraska	NE	70000	74355	2014
Nevada	NV	63000	66920	1812
New Hampshire	NH	64000	67982	1841
New Jersey	NJ	163000	173142	4689
New Mexico	NM	264000	280426	7594
New York	NY	500000	531109	14383
North Carolina	NC	536000	569349	15419
North Dakota	ND	214000	227315	6156
Ohio	OH	2408000	2557823	69271
Oklahoma	OK	146000	155084	4200
Oregon	OR	55000	58422	1582
Pennsylvania	PA	1328000	1410627	38202
Rhode Island	-	6000	6373	173
South Carolina	SC	277000	294235	7968
South Dakota	SD	41000	43551	1179
Tennessee	TN	925000	982552	26609
Texas	TX	1300000	1380885	37397
Utah	UT	78000	82853	2244
Vermont	VT	5000	5311	144
Virginia	VA	355000	377088	10212
Washington	WA	172000	182702	4948
West Virginia	WV	1204000	1278912	34635
Wisconsin	WI	321000	340972	9234
Wyoming	WY	132000	140213	3797
TOTAL		**21119000**	**22433000**	**607527**
			(actual 1990 emissions total)	

Table 3.3

Mexican Total SO_2 Emissions by State

MEXICO (1994) - Estimated by SENES Consultants Limited based on data supplied by Francisco Guzman and Carlos Santos-Burgoa

Source Name	Source Code	Emission Rate (tonnes/year)	Emission Rate (grams/second)
Aquascalientes	AGU	779	25
Baja California	TIJ	108149	3429
Baja California Sur	PAZ	25263	801
Campeche	CAM	108665	3446
Chiapas	TUX	779	25
Chihuahua	JUA	6484	206
Coahuila	SAL	733	23
Colima	MAZ	204332	6479
Durango	DUR	574	18
Estad de Mexico	MEX*	75055	2380
Guanajuato	CEL	20900	663
Guerrero	ACA	733	23
Hidalgo	HID	777385	24651
Iraputo	IRA	2127	67
Jalisco	GUA	11324	359
Michoacan	MOR	943	30
Morelos	CUE	733	23
Nayarit	TEP	733	23
Nuevo Leon	MON	325482	10321
Oaxaca	OAX	8811	279
Puebla	PUE	4133	131
Queretaro	SAN	6815	216
Quintana Roo	CHE	733	23
Salamanca	SAC	304103	9643
San Luis Potosi	SLU	3897	124
Sinaloa	CUL	138101	4379
Sonora	HER	196973	6246
Tabasco	COC	3676	117
Tamaulipas	TAM	233768	7413
Tlaxcala	TAL	3753	119
Toluca	TOL	18367	582
Urapuato	URU	95667	3034
Veracruz	VER	396786	12582
Yucatan	MER	29630	940
Zacatecas	ZAC	533	17
TOTAL		**3116919**	**98838**

Note: * - Hidalgo+Estad de Mexico the same location.

4.0 METEOROLOGICAL DATA AND DATA REPLACEMENTS

For this project, three types of meteorological data are used: surface observations, hourly precipitation data and upper air measurements. Because of the scope of the project and the fact that there are always missing data, parameter gaps were replaced in a variety of ways described specifically for each kind of observation.

4.1 Hourly Surface Observations

One hundred and nineteen (119) meteorological surface stations were used to create the wind fields in the study area. This included 49 Canadian, 51 U.S.A. and 7 Mexican stations. The locations of the stations used are presented in Figure 4.1. Table 4.1 details the information about the surface stations used in the meteorological preprocessor. This table presents the station number used in Figure 4.1, the State/Province Code, the WMO Station Number, the latitude and longitude (in decimal degrees), the time zone and the elevations (in metres).

The SMERGE processor does not accept missing data so that every station had to be scanned for missing hours which then have to be (1) interpolated from the surrounding hours if the gap is small or (2) replaced with data from the closest station if the gap is too large. The present weather codes for type of precipitation then have to be merged into the file for the stations.

4.2 Precipitation Observations

The same 119 stations listed in Table 4.1 were also used for precipitation. The location of these stations is given in Figure 4.1.

Processing U.S. data was quite easy because the format corresponds to TD3280, the format required by the PMERGE program. The Canadian precipitation data, on the other hand, followed the sequence outlined below:

- 22 of the 49 Canadian stations had measured 24 hourly precipitation rates which could be converted simply to the required format using a SENES designed preprocessor

- for the remaining 27 stations, which only measured daily precipitation, the following procedure was established. Based on the daily precipitation amounts, those hours for which precipitation was observed were determined from the surface weather observations. The total daily amount was divided equally among the hours observed to have precipitation. This approach is very reasonable for calculating annual and 24 hour concentrations.

FIGURE 4.1

HOURLY SURFACE AND PRECIPITATION OBSERVATION STATIONS USED

700 0 700 1400 2100 kilometres

SCALE 1:35 000 000

32131-2/03MAR97/XRF_SURF.DWG/M.T.N

Table 4.1
Surface and Precipitation Meteorological Stations

No	Station Name	State Code	Station Number	Latitude (decimal)	Longitude (decimal)	Time Zone	Elevation (m)
1	BAIE COMEAU A	CAN	7040440	49.133	68.200	5	21.0
2	BAKER LAKE A	CAN	2300500	64.300	96.083	6	18.0
3	BIG TROUT LAKE	CAN	6010738	53.833	89.867	5	220.0
4	CALGARY INT'L A	CAN	3031093	51.117	114.017	7	1084.0
5	CARTWRIGHT	CAN	8501100	53.700	57.033	4	14.0
6	CHIBOUGAMAU-CHAPAIS A	CAN	7091404	49.767	74.533	5	388.0
7	CHURCHILL A	CAN	5060600	58.750	94.067	6	35.0
8	CHURCHILL FALLS A	CAN	8501132	53.550	64.100	4	440.0
9	COPPERMINE A	CAN	2200902	67.817	115.133	7	22.0
10	CORAL HARBOUR A	CAN	2301000	64.200	83.367	5	64.0
11	DEER LAKE A	CAN	8401501	49.217	57.400	4	22.0
12	EDMONTON NAMAO A	CAN	3012210	53.667	113.467	7	688.0
13	FORT MCMURRAY A	CAN	3062693	56.650	111.217	7	369.0
14	FORT NELSON A	CAN	1192940	58.833	122.583	8	382.0
15	FORT RELIANCE	CAN	2201900	62.717	109.167	6	164.0
16	FORT SMITH A	CAN	2202200	60.017	111.950	7	203.0
17	FREDERICTON A	CAN	8101500	45.867	66.533	4	17.0
18	GASPE A	CAN	7052605	48.767	64.483	5	33.0
19	GERALDTON A	CAN	6042716	49.783	86.933	5	349.0
20	GOOSE A	CAN	8501900	53.317	60.417	4	46.0
21	GRANDE PRAIRIE A	CAN	3072920	55.183	118.883	7	669.0
22	HAY RIVER A	CAN	2202400	60.833	115.783	7	166.0
23	INUKJUAK	CAN	7103282	58.450	78.117	5	3.0
24	KENORA A	CAN	6034075	49.783	94.367	5	407.0
25	KUUJJUAQ A	CAN	7113534	58.100	68.417	5	34.0
26	LA GRANDE RIVIERE A	CAN	7093715	53.633	77.700	5	191.0
27	LA RONGE A	CAN	4064150	55.150	105.267	6	375.0
28	MAYO A	CAN	2100700	63.617	135.867	8	504.0
29	MONTREAL MIRABEL INT'L A	CAN	7035290	45.683	74.033	5	82.0
30	NAIN A	CAN	8502800	56.550	61.683	4	6.0
31	NORMAN WELLS A	CAN	2202800	65.283	126.800	8	67.0
32	PRINCE ALBERT A	CAN	4056240	53.217	105.683	6	428.0
33	PRINCE GEORGE A	CAN	1096450	53.883	122.683	8	676.0
34	PRINCE RUPERT A	CAN	1066481	54.300	130.433	8	34.0
35	RANKIN INLET A	CAN	2303401	62.817	92.117	5	32.0
36	ROUYN A	CAN	7086720	48.217	78.833	5	301.0
37	SABLE ISLAND	CAN	8204700	43.933	60.017	4	4.0
38	SAINT JOHN A	CAN	8104900	45.317	65.883	4	109.0
39	SCHEFFERVILLE A	CAN	7117825	54.800	66.817	5	522.0
40	SHEARWATER A	CAN	8205090	44.633	63.500	4	51.0
41	ST JOHN'S A	CAN	8403506	47.617	52.733	4	132.0
42	THE PAS A	CAN	5052880	53.967	101.100	6	271.0
43	THUNDER BAY A	CAN	6048261	48.367	89.317	5	199.0
44	TORONTO LESTER B. PEARSO	CAN	6158733	43.667	79.633	5	173.0
45	VAL D'OR A	CAN	7098600	48.067	77.783	5	337.0
46	VANCOUVER INT'L A	CAN	1108447	49.183	123.167	8	3.0
47	WABUSH LAKE A	CAN	8504175	52.933	66.867	4	551.0
48	WATSON LAKE A	CAN	2101200	60.117	128.817	8	690.0
49	WHITEHORSE A	CAN	2101300	60.717	135.067	8	703.0
50	YARMOUTH A	CAN	8206500	43.833	66.083	4	43.0
51	YELLOWKNIFE A	CAN	2204100	62.467	114.450	7	205.0
52	ABILENE	TX	13968	32.417	99.683	6	543.8
53	ALLENTOWN BETHLEHEM	PA	14737	40.650	75.433	5	118.9
54	ALPENA PHELPS COL AP	MI	94849	45.067	83.567	5	210.0
55	AMARILLO INT'L AP	TX	23047	35.233	101.700	6	1098.5
56	ASTORIA/CLATSOP COUNTY ARPT	OR	94224	46.150	123.883	8	7.0
57	ATLANTA HARTSFIELD	GA	13874	33.650	84.433	6	307.8
58	ATLANTIC CITY INTL	NJ	93730	39.450	74.567	5	19.5
59	BILLINGS/LOGAN INT'L ARPT	MT	24033	45.800	108.533	7	1088.0
60	BISMARCK WSFO AP	ND	24011	46.767	100.750	6	502.0
61	BOISE/AIR TERMINAL	ID	24131	43.567	116.217	7	874.0
62	BROWNSVILLE	TX	12919	25.9	97.433	6	5.8
63	CEDAR CITY/FAA AIRPORT	UT	93129	37.700	113.100	7	1714.0
64	CHARLESTON WSO AP	SC	13880	32.900	80.033	6	12.2
65	CHARLOTTE DOUGLAS AP	NC	13881	35.217	80.933	5	219.5
66	CHERRY POINT MCAS	NC	13754	34.900	76.883	5	11.0
67	COLORADO SPRINGS/MUNICIPAL AR	CO	93037	38.817	104.717	7	1881.0
68	COLUMBIA REGIONL AP	MO	3945	38.817	92.217	6	271.3
69	COLUMBUS WSO AP	OH	14821	40.000	82.883	5	247.7
70	DALLAS/FT WORTH AP	TX	3927	32.900	97.033	6	167.6
71	EL PASO	TX	23044	31.8	106.4	7	1194.2
72	ELKO/MUNICIPAL ARPT	NV	24121	40.833	115.783	8	1565.0
73	FRESNO/AIR TERMINAL	CA	93193	36.767	119.717	8	100.0
74	FT SMITH MUNICIPL AP	AR	13964	35.333	94.367	6	136.9
75	GALLUP/FAA AIRPORT	NM	23081	35.517	108.783	7	1972.0
76	GRAND JUNCTION/WALKER FIELD	CO	23066	39.117	108.533	7	1475.0
77	HUSTON	TX	12960	29.087	95.32	6	29.3
78	INDIANAPOLIS INTL AP	IN	93819	39.733	86.267	5	241.3
79	JACKSON THOMPSON FLD	MS	3940	32.317	90.083	6	94.5
80	JACKSONVILLE/INT'L ARPT	FL	13889	30.500	81.700	5	9.0
81	KEY WEST/INT'L ARPT	FL	12836	24.550	81.750	5	6.0
82	LAS VEGAS/MCCARRAN INT'L ARPT	NV	23169	36.083	115.167	8	664.0
83	LEXINGTON BLUEGRASS	KY	93820	38.033	84.600	5	294.4
84	MEDFORD/JACKSON COUNTY ARPT	OR	24225	42.383	122.883	8	405.0
85	MIDLAND	TX	23023	31.95	102.183	6	870.2
86	MISSOULA/JOHNSON-BELL FLD	MT	24153	46.917	114.083	7	972.0
87	MONTGOMERY DANNELLY	AL	13895	32.300	86.400	6	67.4
88	NASHVILLE METRO AP	TN	13897	36.117	86.683	6	179.8
89	NEW ORLEANS	LA	12916	29.983	90.25	6	20
90	NORTH PLATTE BRD FLD	NE	24023	41.133	100.683	6	845.8
91	OCEANA NAS	VA	13769	36.817	76.033	5	8.0
92	OKLA CITY-WSFO AP	OK	13967	35.400	97.600	6	390.1
93	PHOENIX/SKY HARBOR INT'L ARPT	AZ	23183	33.433	112.017	7	337.0
94	PITTSBURGH GR P'BURG	PA	94823	40.500	80.217	5	346.6
95	POCATELL/MUNICIPAL ARPT	ID	24156	45.683	118.850	8	1365.0
96	PORTLAND INT'L JTPRT	ME	14764	43.650	70.300	5	13.7
97	PROVIDENCE GREEN ST	RI	14765	41.733	71.433	5	15.5
98	RAPID CITY REGINL AP	SD	24090	44.050	103.067	7	963.8
99	RENO/CANNON INT'L ARPT	NV	23185	39.500	119.783	8	1341.0
100	ROANOKE WOODRUM AP	VA	13741	37.317	79.967	5	350.2
101	ROCK SPRINGS/FAA AIRPORT	WY	24027	41.600	109.067	7	2100.0
102	RUSSELL MUNI ARPT	KS	93997	38.867	98.817	6	568.1
103	SAN ANTONIO	TX	12921	29.533	98.437	6	240.2
104	SAN DIEGO/LINDBERGH FIELD	CA	23188	32.733	117.167	8	9.0
105	SEATTLE/SEATTLE-TACOMA INT'L	WT	24233	47.450	122.300	8	137.0
106	SHREVERPORT WSO AP	LA	13957	32.467	93.817	6	77.4
107	SPOKANE/INT'L ARPT	WT	24157	47.633	117.533	8	595.0
108	SPRINGFIELD CAPTL AP	IL	93822	39.850	89.683	6	181.1
109	SYRACUSE HANCOCK AP	NY	14771	43.117	76.117	5	125.0
110	TALLAHASSEE/MUNICIPAL ARPT	FL	93805	30.383	84.367	5	21.0
111	TAMPA	FL	12842	27.967	82.533	5	3
112	TUSCON	AZ	23234	37.767	119.717	8	779
113	ACAPULCO/G. ALVAREZ	MX	768056	16.767	99.750	6	5.0
114	LA PAZ INTL AIRPORT	MX	764055	24.067	110.367	7	21.0
115	MERIDA INTL ARPT	MX	766440	20.983	89.650	6	10.0
116	MEXICO CITY/JUAREZ	MX	766790	19.433	99.083	6	2234.0
117	MONTERREY	MX	763944	25.767	100.100	6	387.0
118	PUERTO VALLARTA	MX	766014	20.667	105.267	6	6.0
119	VARACRUZ	MX	766910	19.200	96.133	6	-

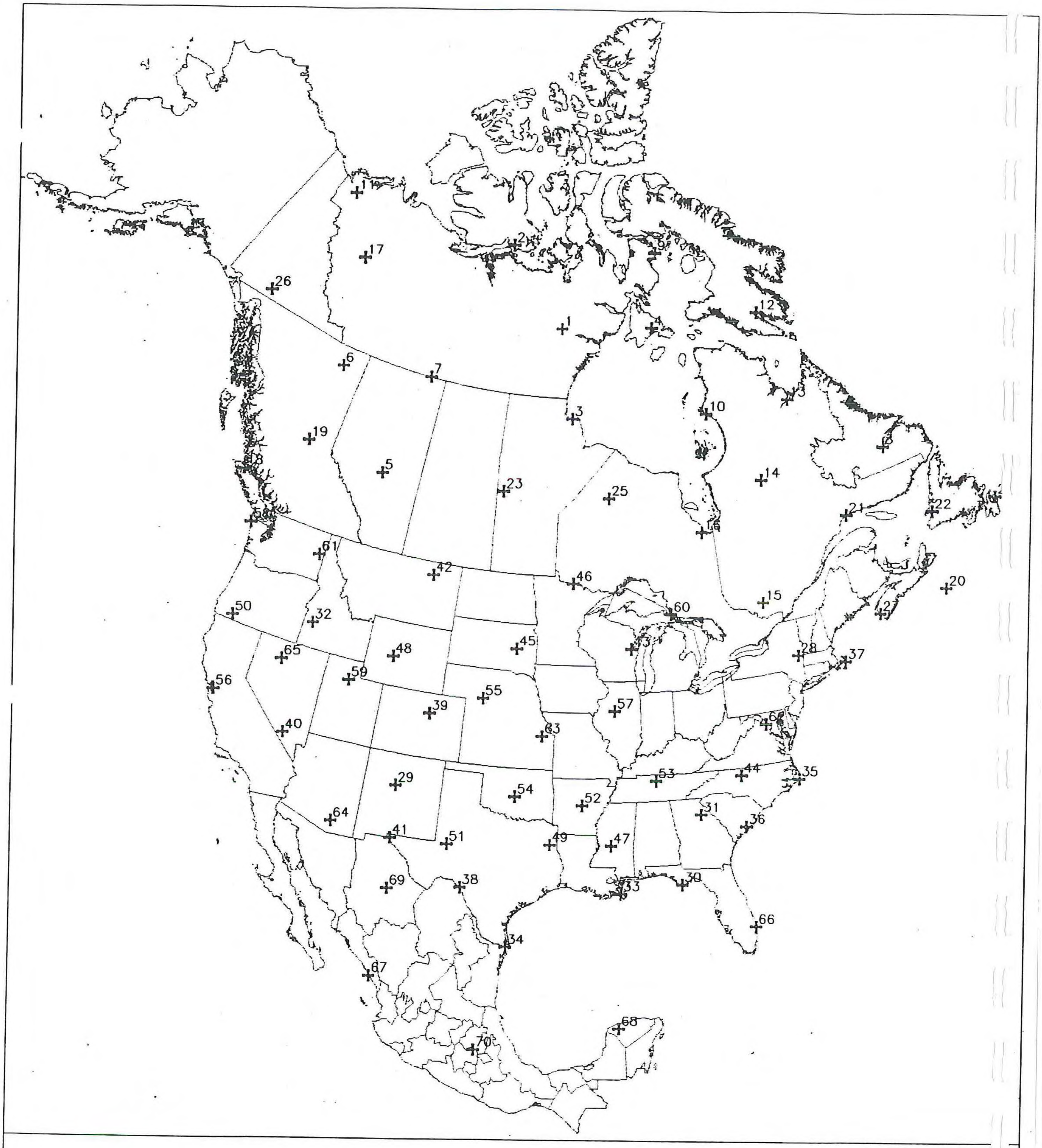


FIGURE 4.2

UPPER AIR OBSERVATION STATIONS USED

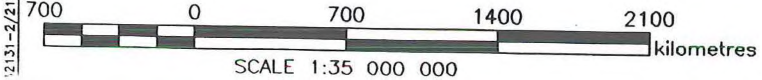


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Table 4.2

Upper Air Stations

No.	Canadian Station Name	State Code	Station Number	Latitude (decimal)	Longitude (decimal)	Time Zone
1	BAKER LAKE (UA)	NWT	16910	64.300	96.00	5
2	CAMBRIDGE BAY	NWT	26005	69.100	105.12	6
3	CHURCHILL	MB	15901	58.750	94.07	6
4	CORAL HARBOUR	NWT	16801	64.200	83.37	5
5	EDMONTON/STONY PLAIN	AB	25145	53.550	114.10	7
6	FORT NELSON UA	BC	25262	58.830	122.60	8
7	FT SMITH (UA)	NWT	26118	60.030	111.95	7
8	GOOSE/GOOSE BAY	NF	15601	53.300	60.37	4
9	HALL BEACH/HALL LK	NWT	16895	68.780	81.25	5
10	INUKJUAK	PQ	15704	58.450	78.12	5
11	INUVIK (UA)	NWT	22258	68.320	133.53	8
12	IQALUIT (UA)	NWT	16607	63.750	68.55	5
13	KUUJJUAQ (UA)	PQ	15641	58.100	68.42	5
14	LA GRANDE IV	PQ	15708	53.750	73.67	5
15	MANIWAKI	PQ	4734	46.380	75.97	5
16	MOOSONEE	PQ	15803	51.270	80.65	5
17	NORMAN WELLS (UA)	NWT	26214	65.280	126.75	8
18	PORT HARDY	BC	25223	50.680	127.37	8
19	PRINCE GEORGE	BC	25206	53.880	122.68	8
20	SABLE ISLAND	NS	14642	43.930	60.02	4
21	SEPT ILES (UA)	PQ	15636	50.220	66.27	5
22	STEPHENVILLE/HARMON AFB	NF	14503	48.530	58.55	4
23	THE PAS	MB	25004	53.970	101.10	6
24	TORBAY/ST JOHNS	NF	14531	47.670	52.75	4
25	TROUT LAKE	ON	15806	53.830	89.87	5
26	WHITEHORSE	YK	26316	60.720	135.07	8
27	YARMOUTH	NS	94620	43.870	66.05	4
No.	American Station Name	State Code	Station Number	Latitude (decimal)	Longitude (decimal)	Time Zone
28	ALBANY	NY	14735	42.750	73.80	5
29	ALBUQUERQUE	NM	23050	35.050	106.62	7
30	APALACHICOLA	FL	12832	29.730	85.03	5
31	ATHENS	GA	13873	33.950	83.32	5
32	BOISE	ID	24131	43.570	116.22	7
33	BOOTHVILLE	LA	12884	29.330	89.40	6
34	BROWNSVILLE	TX	12919	25.900	97.43	6
35	CAPE HATTERAS	NC	93729	35.270	75.55	5
36	CHARLESTON	SC	13880	32.900	80.03	5
37	CHATHAM	MA	14684	41.670	69.97	5
38	DEL RIO	TX	22010	29.370	100.92	6
39	DENVER/STAPLETON ARPT	CO	23062	39.770	104.88	7
40	DESERT ROCK/MERCURY	NV	3160	36.620	116.02	8
41	EL PASO	TX	23044	31.800	106.40	7
42	GLASGOW	MT	94008	48.220	106.62	7
43	GREEN BAY	WI	14898	44.480	88.13	6
44	GREENSBORO	NC	13723	36.080	79.95	5
45	HURON	SD	14936	44.380	98.22	6
46	INTERNATIONAL FALLS	MN	14918	48.570	93.38	6
47	JACKSON/THOMPSON FLD	MS	3940	32.320	90.08	6
48	LANDER	WY	24021	42.820	108.73	7
49	LONGVIEW	TX	3951	32.350	94.65	6
50	MEDFORD	OR	24225	42.370	122.87	8
51	MIDLAND	TX	23023	31.930	102.20	6
52	N LITTLE ROCK	AR	3952	34.830	92.27	6
53	NASHVILLE	TN	13897	36.250	86.57	6
54	NORMAN	OK	3948	35.230	97.47	6
55	NORTH PLATTE	NE	24023	41.130	100.68	6
56	OAKLAND INT AP	CA	23230	37.750	122.22	8
57	PEORIA	IL	14842	40.670	89.68	6
58	QUILLAYUTE	WA	94240	47.950	124.55	8
59	SALT LAKE CITY	UT	24127	40.770	111.97	7
60	SAULT STE MARIE	MI	14847	46.470	84.37	5
61	SPOKANE	WA	24157	47.630	117.53	8
62	STERLING(WASH DULLES)	VA	93734	38.980	77.47	5
63	TOPEKA	KS	13996	39.070	95.62	6
64	TUSCON	AZ	23160	32.120	110.93	7
65	WINNEMUCCA	NV	24128	40.900	117.80	8
66	W. PALM BEACH	FL	12844	26.680	80.12	5
No.	Mexican Station Name	State Code	Station Number	Latitude (decimal)	Longitude (decimal)	Time Zone
67	MAZATLAN SINALOA	MZT	22009	23.180	106.42	7
68	MERIDA IAP	MID	12878	20.950	89.65	6
69	CHIHUAHUA	MCV	22007	28.700	106.07	6
70	MEXICO CITY/INT APT	MEX	11903	19.430	99.07	6

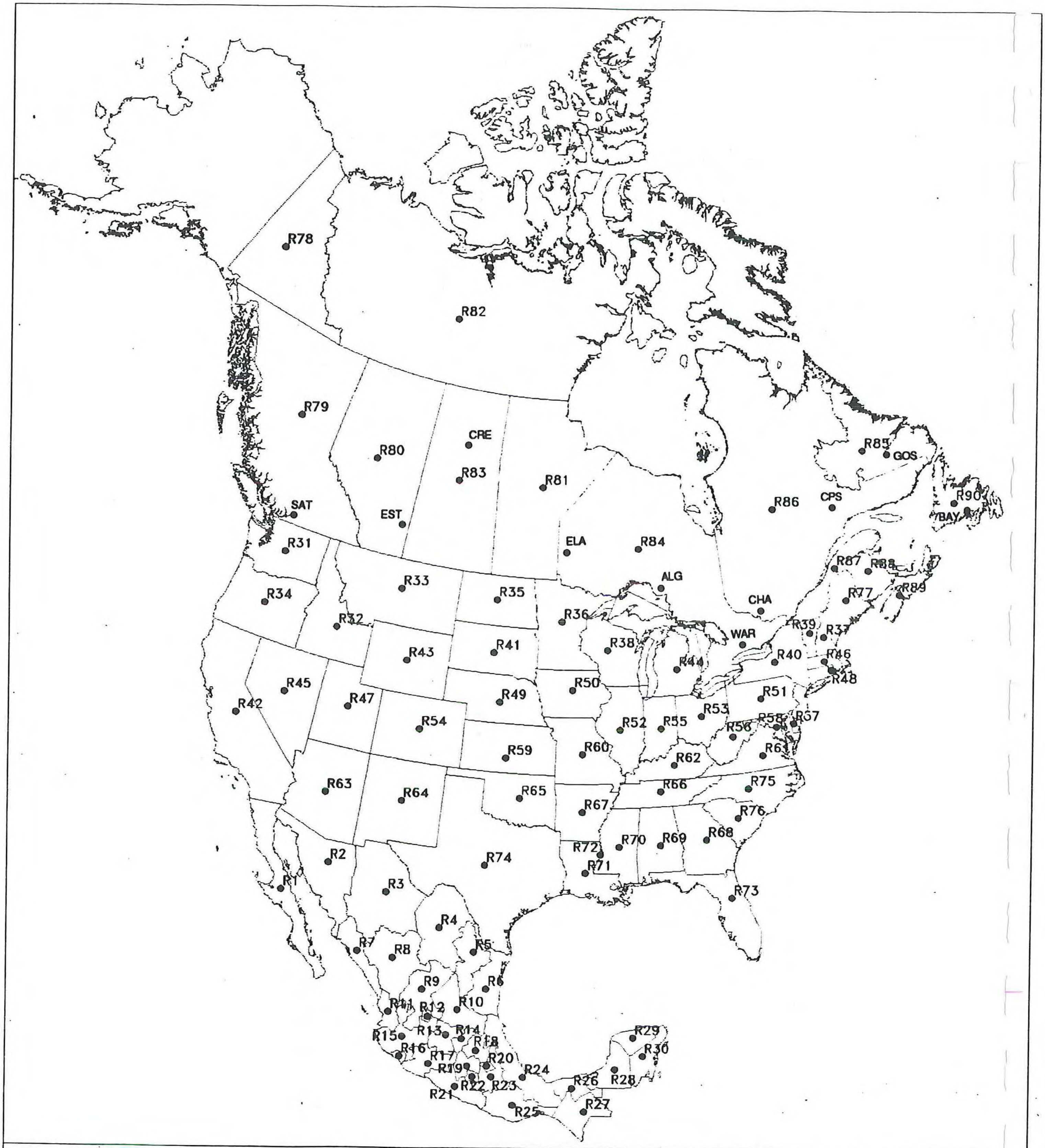

FIGURE 4.3

RECEPTOR LOCATIONS USED

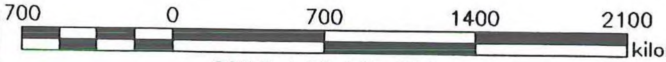

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Table 4.3

Province/State - Receptor Connections used for Modelling

Province/State	Receptor No.
CANADA	
Alberta	R80
British Columbia	R79
Labrador	R85
Manitoba	R81
New Brunswick	R88
Newfoundland	R90
Nova Scotia	R89
Ontario	R84
Prince Edward Island	R87
Quebec	R86
Saskatchewan	R83
Yukon & NWT	R82
Receptors Used for Validation	
Algoma	ALG
Bay D'Espoir	BAY
Chalk River	CHA
Chapais	CPS
Cree Lake	CRE
E.L.A	ELA
Goose Bay	GOS
Esther	EST
Saturna	SAT
Warsaw Caves	WAR
MEXICO	
Aquascalientes	R12
Baja California	R1
Baja California Sur	R1
Campeche	R28
Chiapas	R27
Chihuahua	R3
Coahuila	R4
Colima	R16
Durango	R8
Estad de Mexico	R19
Guanajuato	R13
Guerrero	R21
Hidalgo	R18
Jalisco	R15
Michoacan	R17
Morelos	R22
Nayarit	R11
Nuevo Leon	R5
Oaxaca	R25
Puebla	R23
Queretaro	R14
Quintana Roo	R30
San Luis Potosi	R10
Sinaloa	R7
Sonora	R2
Tabasco	R26
Tamaulipas	R6
Tlaxcala	R20
Veracruz	R24
Yucatan	R29
Zacatecas	R9

State	Receptor No.
U.S.A	
Alabama	R69
Alaska	R78
Arizona	R63
Arkansas	R67
California	R42
Colorado	R54
Connecticut	R48
Delaware	R57
District of Columbia	NM
Florida	R73
Georgia	R68
Hawaii	NM
Idaho	R32
Illinois	R52
Indiana	R55
Iowa	R50
Kansas	R59
Kentucky	R62
Louisiana	R71
Maine	R77
Maryland	R58
Massachusetts	R46
Michigan	R44
Minnesota	R36
Mississippi	R70
Missouri	R60
Montana	R33
Nebraska	R49
Nevada	R45
New Hampshire	R37
New Jersey	R57
New Mexico	R64
New York	R40
North Carolina	R75
North Dakota	R35
Ohio	R53
Oklahoma	R65
Oregon	R34
Pennsylvania	R51
Rhode Island	R46
South Carolina	R76
South Dakota	R41
Tennessee	R66
Texas	R74
Utah	R47
Vermont	R39
Virginia	R61
Washington	R31
West Virginia	R56
Wisconsin	R38
Wyoming	R43

Note: NM - Not Modelled

5.0 CALIBRATION AND SENSITIVITY ANALYSIS

5.1 Calibration

The model was run for the year 1990, using the 1990 emissions inventory to the extent possible. In Mexico the data has not yet been stratified for 1990 so that the emissions used represent emissions from 1994 - 1996. The model predictions were compared to the 1990 measurements at the available measurement points.

Wet Deposition

The comparison of predicted against observed wet SO_4 deposition at 10 locations is presented in Table 5.1 and the model performance for this parameter in Table 5.2. Table 5.1 shows that many of the ten locations are well predicted while others are significantly under or over predicted. As will be shown later, this is due primarily to the distribution of emission sources and their characteristics used in the scoping model.

Based on the results presented in Table 5.1, the EPA protocol (U.S. EPA 1992) and software for calculating various model performance statistics (Harrison 1992), the CALPUFF model scores are presented in Table 5.2.

Table 5.2 reveals that the CALPUFF model over predicts the observed mean wet SO_4 deposition by 35%. The table also shows that CALPUFF over predicts variances by 139%.

The fractional bias has been selected as the basic measure of performance in this evaluation, because it has two desirable features. First, the fractional bias is symmetrical and bounded values for the fractional bias range between -2.0 (extreme underprediction) and +2.0 (extreme overprediction). Second, the fractional bias is a dimensionless number which is convenient for comparison of the results for studies involving different concentration levels or even different pollutants.

Figure 5.1 is a graphical illustration of model performance in which the fractional bias of the standard deviation (y-axis) is plotted against the fractional bias of the average (x-axis). The CALPUFF model is close to the centre of the graph and is relatively free of bias even for the gross simplifications used for emission sources for this work.

Values of the fractional bias that are equal to -0.67 are equivalent to an underprediction by a factor of two, while values that are equal to +0.67 are equivalent to an overprediction by a factor of two.

Table 5.1

MEASUREMENTS vs. PREDICTIONS - SO_4 WET DEPOSITION (kg/ha)

St. (short)	Station	Measurements			Background Deposition	Rec. No.[1]	CALPUFF Wet SO_4 Deposition[2]
		Year	Precipitation (cm)	Wet SO_4Deposition			
ALG	Algoma	1990	123.9	25.3	5.0	ALG	32.7
BAY	Bay Despoir	1990	172.7	9.7	6.9	BAY	7.7
CHA	Chalk River	1990	86.3	18.1	3.5	CHA	52.8
CPS	Chapais	1990	118.0	17.2	4.7	CPS	9.8
CRE	Cree Lake	1990	44.9	1.9	1.8	CRE	1.8
ELA	E.L.A	1990	54.5	4.7	2.2	ELA	4.1
GOS	Goose Bay	1990	85.2	5.0	3.4	GOS	3.4
EST	Esther	1990	40.8	6.8	1.6	EST	1.6
SAT	Saturna	1990	98.3	6.8	3.9	SAT	3.9
WAR	Warsaw Caves	1990	96.4	23.1	3.9	WAR	42.6

Note:

[1] - Closest modelled receptor used.

[2] - Background deposition added to CALPUFF predictions.

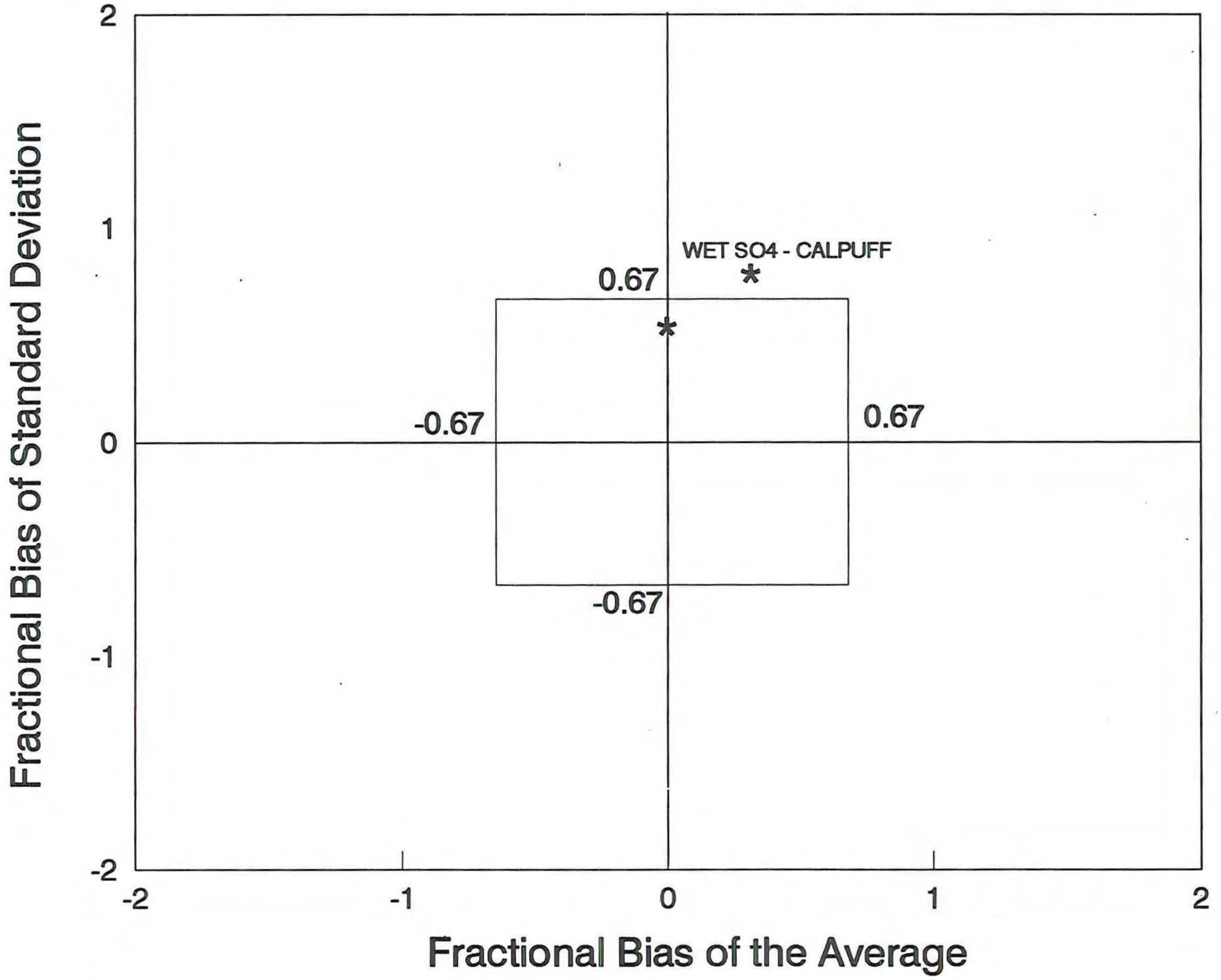
Table 5.2

MODEL VALIDATION RESULTS FOR SO_4 WET DEPOSITIONS

Model Statistics	CALPUFF
Mean of predictions (P)	16.1
Mean of observations (O)	11.9
Fractional bias of the average	0.3
S.D. of predictions	18.9
S.D. of observations	7.9
Fractional bias of standard deviations	0.79
FOR PAIRED DATA:	
Within factor of two (%)	80.0
Slope d(p)/d(O)	1.7
Correlation coefficient (r)	0.82
Unweighted MRE (%)	60.9
Weighted MRE (%)	70.2
Robust MRE (%)	55.3

Figure 5.1

Wet SO4 Deposition Fractional Bias Plot for CALPUFF



For paired observation and prediction data CALPUFF has 80% within a factor of two. The slope of the scattergram is 1.7 for CALPUFF. A correlation coefficient of 0.82 for CALPUFF was determined.

The unintegrated mean relative error (MRE) between observations and predictions was 60.9% for the CALPUFF model. An MRE score weighted proportionately to $(P + O)^2$ (to emphasize the highest wet SO_4 deposition) was 70.2% for the CALPUFF model.

A robustly weighted MRE (to de-emphasize any outliers) was 55.3% for the CALPUFF model. The ideal model would have a Robust MRE equal to 0%.

In general, the model scored well for the scoping emissions input and receptors for SO_4 deposition.

Figure 5.2A presents the observed SO_4 deposition for a portion of eastern North America and Figure 5.2B presents the pattern as predicted by the scoping model. Figure 5.2B represents the major features of the wet deposition pattern with some obvious locational shifts of maxima resulting from the simplistic distribution of sources used. For example, the predicted major peak over Missouri should really be over the Texas/Arkansas border.

5.2 SENSITIVITY RUNS

Point vs. Area Source Representation

Previous work indicates that the wet deposition predictions improve using a point source rather than an area source representation (except for SO_2); the dry deposition and concentration predictions get worse. For this preliminary characterization a point source representation was used.

Dry Deposition Switch

Dry deposition switch was set to "on" for CALPUFF modelling.

Meteorology Input

The impact of leaving one major station out near a critical receptor is minimal for annual averages but may be significant for shorter predictions (hourly and daily). Since this preliminary modelling focusses on annual averages, missing meteorology is not considered to be a problem.

Constant Atmospheric Pressure

A constant value for atmospheric pressure was not used for this work. The actual hourly pressure data was part of the input stream.

Moderate vs. Light Precipitation

Since the scavenging rate is a direct function of rainfall rate, the actual hourly rainfall was used.

Type of Precipitation

The actual hourly precipitation type was input for stations in eastern U.S.A., Manitoba, Ontario, Quebec and the Atlantic provinces. For the remaining stations, including Mexico, the type of precipitation (liquid or solid) was determined based on the hourly temperature measurements, since incorporating weather data into surface stations is a very time consuming step. The long-term deposition results are not affected by this approach.

Relative Humidity

The actual hourly relative humidity was an input for this work rather than fixing an average value for the year.

Sea-Salt Correction

For comparison of modelled with measured data, the sea-salt corrected data were used.

5.3 Location of Emission Points

In order to demonstrate the importance of the location of emission points, a series of sensitivity runs were carried out for a U.S.A. state - Pennsylvania. In the scoping run, a single point located at "PENN" emitted 38,202 g/s of sulphur dioxide. The "home state" receptor (R51) should have a deposition of 20 to 50 kg/SO_4/ha/year according to Figure 5.2A, with a best guess of 30 kg/ha/year. Figure 5.3A presents the location of the source and receptor. Table 5.3 shows the model predicted wet deposition at receptor R51 resulting from all sources (127.6) and from the "PENN" source along (115). The predicted value is ~4.3 times the observed value.

Figure 5.3B shows the location of the four largest sources in Pennsylvania. The first sensitivity run used the four source locations and the actual emissions as outlined in Table 5.4 which represented 71% of the total state emissions.

FIGURE 5.2a

1990 OBSERVED WET EXCESS SO_4 DEPOSITION (KG/HA/YR)

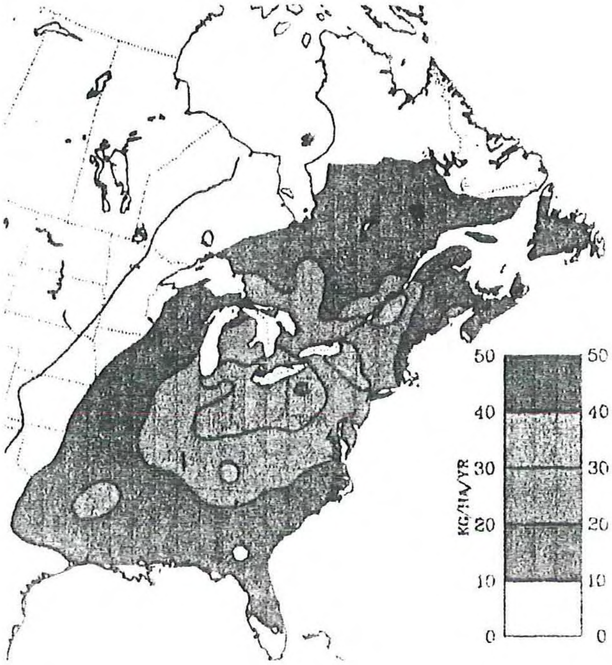
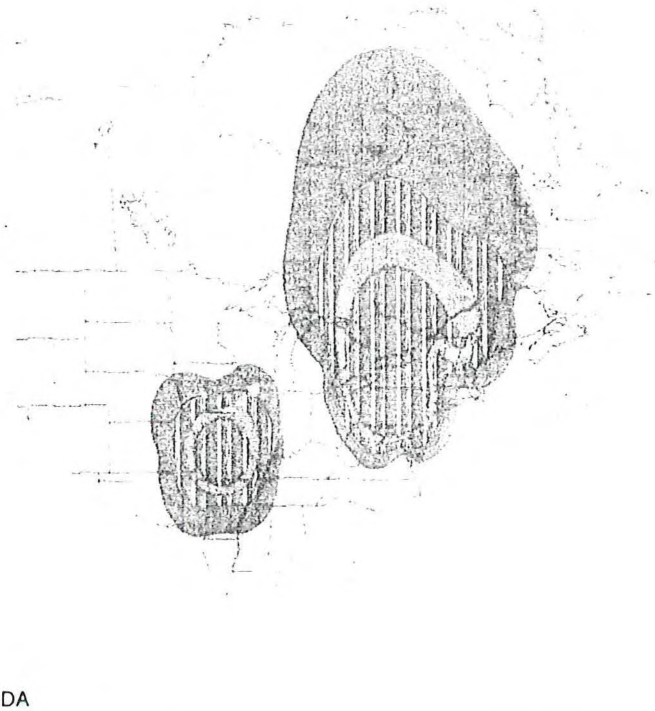


FIGURE 5.2b

1990 CALPUFF PREDICTED WET EXCESS SO_4 DEPOSITION (KG/HA/YR)



SOURCE: ENVIRONMENT CANADA

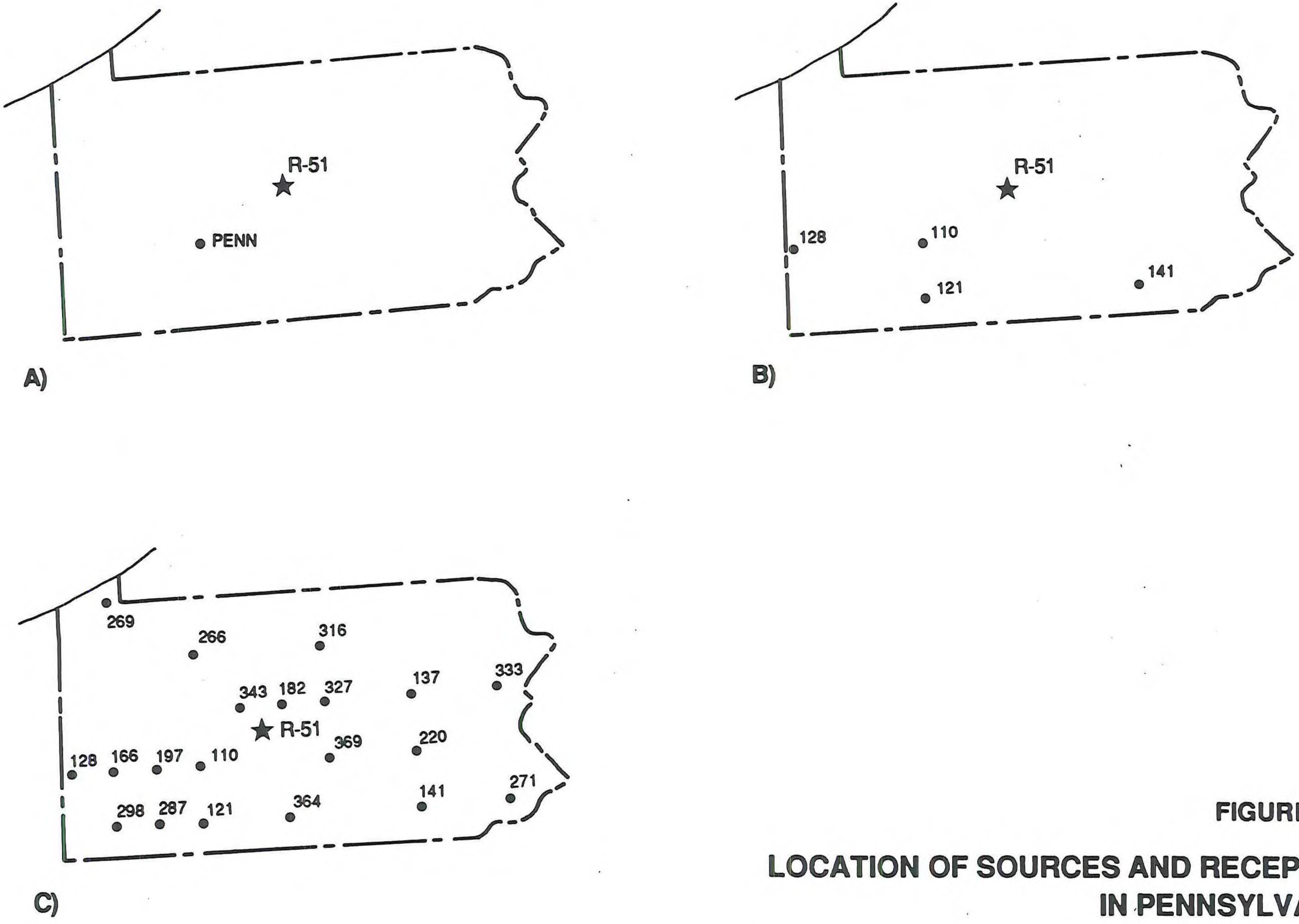


FIGURE 5.3

LOCATION OF SOURCES AND RECEPTOR IN PENNSYLVANIA

Table 5.3

SINGLE SOURCE IN PENNSYLVANIA

Receptor	Incremental SO_4 Wet Deposition (kg/h/year)		Contribution from Non-Pennsylvania States
	Total Contribution from All Sources	Pennsylvania State Only	
51	127.6	115.0	12.6

Table 5.4

ACTUAL EMISSIONS FOR FOUR SOURCES IN PENNSYLVANIA

SRC Number	SO_2 Emissions (g/s)
110	10347
128	5295
141	4613
121	7026
Total	27281

This represents 71% of the state emission (38202 g/s).

Table 5.5

FOUR SOURCES IN PENNSYLVANIA (ACTUAL EMISSIONS ONLY)

Receptor	**Incremental SO_4 Wet Deposition (kg/h/year)**		**Total**
	All Sources Except Pennsylvania	**Pennsylvania Only from Four Sources**	
51	12.6	63.7	76.3

Table 5.6
ACTUAL SOURCE DISTRIBUTION IN PENNSYLVANIA

No.	Source Name	Emissions (g/s)
1	110	10347
2	128	5295
3	121	7026
4	169	615
5	166	2313
6	197	1617
7	287	564
8	298	203
9	364	180
10	343	251
11	266	729
12	182	2081
13	327	72
14	316	106
15	369	155
16	141	4613
17	220	1196
18	271	669
19	333	299
Total		38276

See Figure 3 for locations.

Table 5.7
NINETEEN SOURCES IN PENNSYLVANIA

Receptor	Incremental SO_4 Wet Deposition (kg/h/year)		Total
	All Sources Except Pennsylvania	Pennsylvania Only	
51	12.6	71.9	84.5

Table 5.5 shows that this distribution predicts a sulphate deposition of 76.3 at receptor R51 (2.5 times the approximate measured value of 30).

The second sensitivity run used the actual distribution of the 19 major sources in Pennsylvania as presented in Table 5.6. The total actually 74 g/s larger (error of less than 0.2%) than the state total used in the modelling (due to two difference sources of emissions information).

Table 5.7 presents the predicted results at R51 as 84.5 kg SO_4/ha/year, a factor of 2.8 above the observed value.

The third and fourth sensitivity runs, used a modified stack height of 70 m (closer to the actual) with an exit velocity of 25 m/s and an exit temperature of 303°K.

The four source scenario was used and for the third sensitivity run, only Source 110 was modified. This predicted a maximum wet SO_4 deposition at Receptor 51 of 45.2 kg/ha/year.

The fourth sensitivity run used modified source characteristics for Sources 110, 121, 128 and 141 and predicted a wet deposition of 43.1 kg/ha/year.

Comparing these values against the observed value shows excellent agreement (both within a factor of 1.5).

The major conclusion from these runs is that source locations and characteristics are very important parameters in simulating acid deposition impacts. At the very least, these parameters must be available for modelling simulations in North America.

6.0 TRANSFER MATRICES

6.1 What is a Transfer Matrix?

For the purposes of estimating changes in concentration and depositions due to emission changes, the output of long range transport models can be conveniently expressed by an atmospheric 'transfer matrix'. The matrix formulation assumes that the deposition at a receptor location is the sum of partial contributions, each of which is proportional to the emissions from a source or a group of sources. The coefficient of proportionality connects the source region with the receptor. The array of coefficients connecting deposition at a receptor with a unit of emission in a source region constitutes a unit transfer matrix and indicates the strength of the atmospheric linkage between the source and the receptor. A full description of the mathematics involved can be found in Young and Shaw (1986).

In addition, a background deposition from natural sources and from anthropogenic sources

Table 6.1

Example Wet SO_4 Deposition - (kg/ha/year)

Source	Emis(g/s)	Src. Code	Receptor ----> 1	2	3	4	5	6	7	8	9	10	11	12
1	17980	ALB	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
2	3298	BC	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
3	0	LAB	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
4	16362	MAN	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
5	5740	NEWB	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
6	1807	NFL	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
7	5644	NOVS	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
8	18202	ONT	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
9	19597	ONTS	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
10	95	PEI	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
11	12399	QUE	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
12	2790	SASK	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
13	539	NWT	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
14	21287	AL	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
15	58	AK	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
16	3107	AR	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
17	9666	AZ	8.26E-07	1.12E-06	1.28E-06	1.22E-06	1.09E-06	8.86E-07	9.36E-07	9.96E-07	8.78E-07	7.87E-07	7.17E-07	7.41E-07
18	5868	CA	2.78E-04	9.51E-04	1.31E-03	9.39E-04	5.55E-04	2.17E-04	3.32E-04	4.03E-04	2.18E-04	1.30E-04	9.51E-05	1.03E-04
19	3049	CO	1.22E-07	6.28E-07	8.32E-07	4.42E-07	1.92E-07	4.79E-08	1.21E-07	1.46E-07	5.46E-08	0.00E+00	0.00E+00	0.00E+00
20	1582	CT	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
21	2733	DE	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
22	23905	FL	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
23	22812	GA	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
24	7336	IA	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
25	1064	ID	0.00E+00	1.01E-05	1.81E-05	1.06E-05	4.48E-06	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
26	34175	IL	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
27	50457	IN	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
28	3366	KS	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
29	30579	KY	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
30	12485	LA	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
31	5897	MA	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
32	10442	MD	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
33	2301	ME	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
34	15045	MI	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
35	4114	MN	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
36	2359	MO	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
37	6731	MS	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
38	2359	MT	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
39	15419	NC	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
40	6156	ND	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
41	2014	NE	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
42	1841	NH	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
43	4689	NJ	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
44	7594	NM	3.05E-05	7.43E-05	9.64E-05	7.43E-05	5.06E-05	2.71E-05	3.72E-05	4.26E-05	2.82E-05	2.00E-05	1.57E-05	1.66E-05
45	1812	NV	1.06E-05	2.21E-05	3.17E-05	3.08E-05	2.55E-05	1.67E-05	1.54E-05	1.87E-05	1.49E-05	1.23E-05	9.19E-06	1.04E-05
46	14383	NY	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
47	69271	OH	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
48	4200	OK	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
49	1582	OR	3.21E-06	2.09E-05	2.64E-05	1.46E-05	7.13E-06	2.90E-06	3.86E-06	4.79E-06	2.69E-06	1.69E-06	1.16E-06	1.29E-06
50	38202	PA	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
51	173	RI	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
52	7968	SC	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
53	26609	TN	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
54	37397	TX	2.87E-05	3.74E-05	4.23E-05	4.04E-05	3.69E-05	3.06E-05	3.20E-05	3.40E-05	3.04E-05	2.76E-05	2.54E-05	2.61E-05
55	2244	UT	3.34E-07	1.64E-06	2.58E-06	1.83E-06	1.03E-06	3.28E-07	4.35E-07	5.88E-07	2.81E-07	0.00E+00	0.00E+00	0.00E+00
56	10212	VA	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
57	144	VT	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
58	4948	WA	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
59	9234	WI	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
60	3797	WY	0.00E+00	7.37E-07	1.66E-06	9.83E-07	3.76E-07	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
61	23	ACA	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
62	25	AGU	1.19E-10	1.45E-10	1.58E-10	1.55E-10	1.45E-10	1.27E-10	1.30E-10	1.36E-10	1.26E-10	1.17E-10	1.10E-10	1.13E-10
63	3446	CAM	2.06E-11	2.37E-11	2.49E-11	2.42E-11	2.27E-11	2.05E-11	2.16E-11	2.21E-11	2.06E-11	1.94E-11	1.88E-11	1.90E-11
64	662.7	CEL	6.22E-15	1.19E-14	1.39E-14	1.11E-14	7.89E-15	4.59E-15	6.69E-15	7.29E-15	4.99E-15	3.57E-15	3.23E-15	3.24E-15
65	23	CHE	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
66	0	CHI	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
67	117	COC	0.00E+00	1.36E-10	6.13E-10	2.70E-09	3.23E-09	7.32E-10	9.99E-11	1.71E-10	1.67E-10	1.90E-10	6.89E-11	1.16E-10
68	23	CUE	8.95E-16	1.10E-15	1.20E-15	1.17E-15	1.09E-15	9.43E-16	9.75E-16	1.02E-15	9.36E-16	8.72E-16	8.14E-16	8.33E-16
69	4379	CUL	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
70	18	DUR	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
71	359	GUA	6.68E-10	8.19E-10	5.67E-09	1.00E-08	1.11E-08	5.49E-09	7.18E-10	7.50E-10	6.86E-10	6.35E-10	6.00E-10	6.10E-10
72	6246	HER	2.73E-03	1.12E-02	2.29E-02	2.17E-02	1.47E-02	6.13E-03	5.93E-03	8.49E-03	5.30E-03	3.52E-03	2.08E-03	2.59E-03
73	67.4	IRA	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
74	206	JUA	7.35E-08	1.01E-07	1.15E-07	6.32E-05	9.39E-05	8.08E-08	8.43E-08	9.00E-08	7.95E-08	7.15E-08	6.47E-08	6.67E-08
75	0	LAP	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
76	0	MAT	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
77	6479	MAZ	4.70E-05	1.76E-04	2.46E-04	1.60E-04	8.55E-05	2.93E-05	5.64E-05	6.74E-05	3.27E-05	1.74E-05	1.34E-05	2.05E-04
78	940	MER	8.54E-14	9.68E-14	1.02E-13	9.78E-14	9.36E-14	8.53E-14	8.88E-14	9.07E-14	8.58E-14	8.09E-14	7.91E-14	7.94E-14
79	26449	MEX	4.95E-06	6.35E-06	7.01E-06	6.74E-06	9.07E-03	9.73E-02	5.42E-06	5.71E-06	2.48E-02	5.08E-01	4.34E-06	6.88E-02
80	0	MIN	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
81	10321	MON	6.42E-08	1.05E-07	1.21E-07	1.04E-07	8.27E-08	5.58E-08	7.08E-08	7.61E-08	5.84E-08	4.61E-08	4.21E-08	4.26E-08
82	29.9	MOR	3.29E-10	3.92E-10	4.16E-10	3.98E-10	3.71E-10	3.23E-10	3.47E-10	3.56E-10	3.26E-10	3.02E-10	2.92E-10	2.93E-10
83	0	NUE	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
84	279	OAX	0.00E+00	0.00E+00	0.00E+00	0.00E+00	6.08E-05	1.54E-02	0.00E+00	0.00E+00	0.00E+00	1.43E-02	0.00E+00	0.00E+00
85	801	PAZ	0.00E+00	2.15E-13	1.31E-04	8.81E-04	8.17E-04	4.93E-13	1.41E-13	2.44E-13	2.26E-13	2.45E-13	8.73E-14	1.47E-13
86	131	PUE	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	6.37E-04	0.00E+00	0.00E+00	0.00E+00	1.52E-03	0.00E+00	2.45E-05
87	9643.1	SAC	6.62E-09	1.04E-08	1.23E-08	1.13E-08	9.47E-09	6.83E-09	7.68E-09	8.37E-09	6.82E-09	5.69E-09	5.04E-09	5.23E-09
88	23	SAL	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
89	216	SAN	2.81E-12	3.72E-12	4.02E-12	3.72E-12	3.24E-12	2.59E-12	2.96E-12	3.09E-12	2.66E-12	1.85E-05	2.20E-12	1.70E-04
90	124	SLU	4.03E-09	5.06E-09	5.37E-09	7.58E-09	9.03E-09	3.74E-09	4.19E-09	4.33E-09	3.82E-09	3.43E-09	3.29E-09	3.30E-09
91	119	TAL	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	5.56E-05	0.00E+00	0.00E+00	0.00E+00	1.80E-04	0.00E+00	3.93E-06
92	7413	TAM	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
93	23	TEP	3.31E-14	5.20E-14	5.77E-14	4.90E-14	3.86E-14	2.65E-14	3.47E-14	3.66E-14	2.81E-14	2.21E-14	2.07E-14	2.07E-14
94	3429	TIJ	8.71E-06	2.92E-05	1.17E-04	2.61E-04	2.62E-04	9.02E-05	1.04E-05	3.87E-05	3.00E-05	2.70E-05	2.99E-06	3.25E-06
95	581.9	TOL	1.36E-13	1.68E-13	1.83E-13	1.78E-13	2.52E-05	2.61E-04	1.48E-13	1.55E-13	1.96E-04	2.26E-03	1.24E-13	5.16E-04
96	25	TUX	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
97	3033.6	URU	2.40E-05	9.34E-05	1.37E-04	8.95E-05	4.67E-05	1.56E-05	3.09E-05	3.76E-05	1.64E-04	6.98E-04	5.25E-05	3.09E-03
98	12582	VER	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
99	17	ZAC	1.92E-10	2.38E-10	2.62E-10	2.58E-10	2.41E-10	2.11E-10	2.14E-10	2.24E-10	2.09E-10	1.94E-10	1.80E-10	1.87E-10
Total Wet SO_4 Loading-No Backg.			3.17E-03	1.27E-02	2.50E-02	2.42E-02	2.58E-02	1.20E-01	6.46E-03	9.15E-03	3.09E-02	5.31E-01	2.30E-03	7.55E-02
Total Wet SO_4 Loading-With Backg.			4.80	4.81	4.83	4.82	4.83	4.92	4.81	4.81	4.83	5.33	4.80	4.88
			1	2	3	4	5	6	7	8	9	10	11	12

outside of the 84 source areas must be added to the values predicted by using the matrix.

The background is relatively small in high deposition regions and dominates in remote regions. It is usually the product of a constant background concentration (in precipitation) times the precipitation at the receptor. An average background of 4.8 kg/ha/year was used for this scoping model run. This background was developed based on 17 observation stations used for model validation in the report "*A Detailed Source-Receptor Matrix for Ontario, Quebec and the Atlantic Provinces*".

6.2 Matrices Produced for this Project

The unit transfer matrices were produced for each emission source and 100 discrete receptors.

Table 6.1 represents part of the unit transfer loading matrix for wet sulphate deposition (using the 1990 SO_2 emissions). The full loading matrix for the 99 sources and the 106 receptors for modelled wet SO_4 deposition (without and with background) is presented in Appendix A.1.

7.0 RESULTS FOR NORTH AMERICA

7.1 Canada

The transfer matrix presented in Appendix A produces wet sulphate depositions at each of the receptors. Table 7.1 summarizes the five highest predicted wet SO_4 loadings in each country of North America. The largest loading in Canada is predicted to be PEI at approximately 22 kg SO_4/ha/year. Table 7.1 identifies the 10 major contributors to the loading in PEI in 1990 as the superstack in Sudbury. The largest contributor to wet SO_4 loadings in Quebec are sources in Ontario. Table 7.1A gives some sense of the importance of receptor location showing that a maximum of 54 kg/ha/year was predicted for the Chapais monitoring station with the largest contributor being Ontario and a value of 33 kg/ha/year for the Algoma monitoring station in Ontario with the largest contributor being Quebec. Looking at the top ten contributors in Canada, one clearly sees evidence of continental transport from the U.S.A.

7.2 U.S.A.

Examining the U.S.A. part of Table 7.1, one identifies that the largest predicted loading of sulphate to a receptor in the U.S.A. is 128 kg/ha/year at the Pennsylvania receptor. While this station was examined in detail in the sensitivity studies, Table 7.1 still shows that Pennsylvania has the largest impact on itself. This is generally true for most receptor areas. Table 7.1 also shows that Canada (ONT) is the largest contributor to sulphate loadings in New Hampshire and Vermont and that Mexico (Estad de Mexico, Sonora and Sineloa) are the contributors to loadings

Table 7.1

HIGHEST WET SO_4 LOADINGS (kg/ha/year) WITHIN EACH COUNTRY AND RANKED SOURCE CONTRIBUTION

	Country	U.S.A					Canada					Mexico				
Receptor-->	Number	37	39	51	60	67	84	86	87	88	89	13	14	17	19	20
	State/Prov.	New Hampshire	Vermont	Pennsylvania	Missouri	Arkansas	Ontario	Quebec	PEI	New Brunswick	Nova Scotia	Guanajuato	Queretaro	Michoacan	Estad de Mexico	Tlaxcala
Wet SO_4 Loading (kg/ha/y)-->		19.03	47.80	127.61	76.17	16.91	7.63	15.15	21.88	6.04	5.04	5.45	5.55	8.11	5.47	5.58
Ranking of Contribution																
1		ONT	ONT	PA	TX	AR	MAN	ONT	Sudbury	ALB	MI	MEX	MEX	MEX	MEX	PUE
2		MI	MI	MD	AR	TX	ALB	MI	ALB	Sudbury	IN	OAX	OAX	MAZ	OAX	OAX
3		MAN	MAN	MI	AZ	MS	NM	IN	MN	NM	ALB	URU	TOL	TOL	PUE	MEX
4		WI	WI	ONT	HER	MEX	AZ	WI	WI	MI	IL	TOL	PUE	URU	TAL	TAL
5		ALB	VT	OH	MEX	LA	OK	IL	SASK	IN	KY	HER	URU	SAN	TOL	TOL
6		ND	AL	NY	NM	AZ	HER	MN	ND	ND	FL	MAZ	HER	TAL	MAZ	TX
7		IL	MN	IN	MO	NM	BC	IA	QUE	IL	GA	PUE	TAL	PUE	URU	NM
8		NY	IL	ALB	OK	HER	WY	CO	MI	KY	AL	TAL	MAZ	TX	TX	NV
9		IN	IN	WI	CA	OK	SASK	BC	ONT	FL	MAN	SAN	CA	CA	CA	HER
10		WY	ND	ND	WY	CUL	KS	AZ	NWT	BC	BC	CA	TX	NM	NM	AZ

Table 7.1a

	Country	Canada	
Receptor->	Number	91(ALG)	93(CHA)
	State/Prov.	Ontario	Quebec
Wet SO_4 Loading (kg/ha/y)-->		32.51	54.11
Ranking of Contribution			
1		QUE	ONT
2		MAN	MAN
3		NEWB	MI
4		BC	WI
5		PEI	Sudbury
6		WA	IN
7		NWT	IL
8		OR	ALB
9		CA	KY
10		ID	FL

in Arkansas.

7.3 Mexico

Due to reduced precipitation in Mexico, Table 7.1 shows relatively low sulphate loadings in that country. The maximum loadings of 8 kg/ha/year in Michoacan is due to the Estad de Mexico. It is also clear from Table 7.1 that California, Texas, New Mexico, Nevada and Arizona contribute to wet sulphate deposition in Mexico.

8.0 EXAMPLA APPLICATION - NORTH AMERICAN VOLCANOES

8.1 Introduction

Since the unit transfer matrix provides the meteorological link between source regions and receptor points, it can be used to look at natural or man made emissions. For this study, volcanoes in North America have been selected as an example case.

8.2 Source Term

This application looked at three volcanoes. The first was Mount St. Augustine (about 285 km southwest of Anchorage) which when active in 1976 is estimated to have emitted 8,760 kT/year - 277,778 g/s (Lepel et al. 1978) and in 1986 was passively degassing approximately 443 kT/year - 14,047 g/s (Kodosky et al. 1991).

The second was Mount St. Helens in the State of Washington which emitted in 1980 as a quiescent plume 1.0 kT/day or 365 kT/year - 11,547 g/s.

The third was Popo (about 100 km southwest of Mexico City) which emitted from 5 to 120 kT/day during a conference in Mexico City in January 1997. Using the average over three days as 46 kT/day, gives an approximate emission rate of 16,790 T/year - 532,411 g/s.

8.3 Transfer Matrix Model (TMM) Estimated Loading

Using the TMM calculated for this project (Section 6.0) and looking at the ten largest source-receptor links, the following wet sulphate loadings are estimated for each receptor:

Mt. St. Augustine (Source Location - Alaska, U.S.A.)

Receptor	Country	Estimated Loading Wet SO_4 kg/ha/yr	
		Active	Passive
1	Ontario	8.28	0.419
2	New York	3.44	0.174
3	Michigan	2.86	0.145
4	Pennsylvania	2.81	0.142
5	Quebec	2.21	0.112
6	Vermont	1.93	0.0975
7	PEI	1.89	0.0958
8	Ohio	1.73	0.0877
9	Illinois	1.60	0.0809
10	Maryland	1.52	0.0767

Mt. St. Helens (Source Location - Washington, U.S.A.)

Receptor	Country	Estimated Loading Wet SO_4 kg/ha/yr
		Passive
1	Illinois	0.189
2	Quebec	0.0730
3	Ontario	0.0376
4	PEI	0.0302
5	Vermont	0.0214
6	Maine	0.0207
7	New Brunswick	0.0175
8	New York	0.0174
9	New Hampshire	0.0176
10	Michigan	0.0166

Popocatepetl (Source Location - Mexico City, Mexico)

Receptor	Country	Estimated Loading Wet SO_4 kg/ha/yr
		Passive
1	Michoacam	65.5
2	Missouri	18.0
3	Arkansas	17.6
4	Querataru	14.1
5	Estad de Mexico	12.0
6	Guanajuatu	10.7
7	San Luis Potosi	10.2
8	Guerreru	8.41
9	Hidalgu	5.80
10	Morrelsos	3.50

8.4 Results

This application of the transfer matrix to estimating the impact of natural soruces is very instructive. It clearly demonstrates the existence of Contintental Pollutant Pathways from Alaska, U.S.A., to Prince Edward Island, Canada from Washington, U.S.a. to Quebec, Canada; and from the Estad de Mexico, Mexico to Missouri, U.S.A. It also shows that the active volcanoes can contribute significant amounts of sulphate to the enviornment of North America. Popocateptl in its passive state emits more sulphure dioxide than all of the man-made emissions in North America.

9.0 SUMMARY AND RECOMMENDATIONS

9.1 Summary

Based on the CALPUFF model validation the predicted annual wet depositions for sulphur compounds are in reasonable agreement with the measurements. The other predicted parameters are also thought to be reasonable for policy setting purposes.

The comparison with the RADM model (which includes a finely gridded emissions inventory and state-of-the-art chemical and physical processes) is very encouraging. The CALPUFF results compare very well with the RADM results for wet deposition.

9.2 Recommendations

1. In order to support intercountry negotiations, further receptors in each country should be simulated.

2. The simulation should be undertaken using about five major point sources in each state each with the correct source characteristics (height, exit velocity, exit temperature).

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APPENDIX A.1

WET SULPHATE DEPOSITION MATRIX

Table A.1

Wet SO_4 Deposition - (kg/ha/year)

Receptor --->

Source	Emis(g/s)	Src. Code	1	2	3	4	5	6	7	8	9	10	11	12	13	14
1	17980	ALB	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
2	3298	BC	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
3	0	LAB	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
4	16362	MAN	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
5	5740	NEWB	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
6	1807	NFL	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
7	5644	NOVS	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
8	18202	ONT	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
9	19597	ONTS	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
10	95	PEI	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
11	12399	QUE	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
12	2790	SASK	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
13	539	NWT	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
14	21287	AL	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
15	58	AK	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
16	3107	AR	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
17	9666	AZ	8.26E-07	1.12E-06	1.28E-06	1.22E-06	1.09E-06	8.86E-07	9.36E-07	9.96E-07	8.78E-07	7.87E-07	7.17E-07	7.41E-07	6.55E-07	6.36E-07
18	5868	CA	2.78E-04	9.51E-04	1.31E-03	9.39E-04	5.55E-04	2.17E-04	3.32E-04	4.03E-04	2.18E-04	1.30E-04	9.51E-05	1.03E-04	3.17E-05	2.76E-05
19	3049	CO	1.22E-07	6.28E-07	8.32E-07	4.42E-07	1.92E-07	4.79E-08	1.21E-07	1.46E-07	5.46E-08	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
20	1582	CT	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
21	2733	DE	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
22	23905	FL	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
23	22812	GA	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
24	7336	IA	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
25	1064	ID	0.00E+00	1.01E-05	1.81E-05	1.06E-05	4.48E-06	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
26	34175	IL	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
27	50457	IN	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
28	3366	KS	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
29	30579	KY	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
30	12485	LA	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
31	5897	MA	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
32	10442	MD	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
33	2301	ME	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
34	15045	MI	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
35	4114	MN	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
36	2359	MO	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
37	6731	MS	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
38	2359	MT	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
39	15419	NC	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
40	6156	ND	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
41	2014	NE	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
42	1841	NH	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
43	4689	NJ	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
44	7594	NM	3.05E-05	7.43E-05	9.64E-05	7.43E-05	5.06E-05	2.71E-05	3.72E-05	4.26E-05	2.82E-05	2.00E-05	1.57E-05	1.66E-05	1.18E-05	1.09E-05
45	1812	NV	1.06E-05	2.21E-05	3.17E-05	3.08E-05	2.55E-05	1.67E-05	1.54E-05	1.87E-05	1.49E-05	1.23E-05	9.19E-06	1.04E-05	8.28E-06	8.03E-06
46	14383	NY	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
47	69271	OH	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
48	4200	OK	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
49	1582	OR	3.21E-06	2.09E-05	2.64E-05	1.46E-05	7.13E-06	2.90E-06	3.86E-06	4.79E-06	2.69E-06	1.69E-06	1.16E-06	1.29E-06	7.69E-07	6.94E-07
50	38202	PA	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
51	173	RI	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
52	7968	SC	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
53	26609	TN	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
54	37397	TX	2.87E-05	3.74E-05	4.23E-05	4.04E-05	3.69E-05	3.06E-05	3.20E-05	3.40E-05	3.04E-05	2.76E-05	2.54E-05	2.61E-05	2.34E-05	2.28E-05
55	2244	UT	3.34E-07	1.64E-06	2.58E-06	1.83E-06	1.03E-06	3.28E-07	4.35E-07	5.88E-07	2.81E-07	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
56	10212	VA	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
57	144	VT	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
58	4948	WA	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
59	9234	WI	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
60	3797	WY	0.00E+00	7.37E-07	1.66E-06	9.83E-07	3.76E-07	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
61	23	ACA	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
62	25	AGU	1.19E-10	1.45E-10	1.58E-10	1.55E-10	1.45E-10	1.27E-10	1.30E-10	1.36E-10	1.26E-10	1.17E-10	1.10E-10	1.13E-10	1.04E-10	1.02E-10
63	3446	CAM	2.06E-11	2.37E-11	2.49E-11	2.42E-11	2.27E-11	2.05E-11	2.16E-11	2.21E-11	2.06E-11	1.94E-11	1.88E-11	1.90E-11	1.77E-11	1.75E-11
64	662.7	CEL	6.22E-15	1.19E-14	1.39E-14	1.11E-14	7.89E-15	4.59E-15	6.69E-15	7.29E-15	4.99E-15	3.57E-15	3.23E-15	3.24E-15	2.32E-15	2.11E-15
65	23	CHE	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
66	0	CHI	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
67	117	COC	0.00E+00	1.36E-10	6.13E-10	2.70E-09	3.23E-09	7.32E-10	9.99E-11	1.71E-10	1.67E-10	1.90E-10	6.89E-11	1.16E-10	1.04E-10	1.17E-10
68	23	CUE	8.95E-16	1.10E-15	1.20E-15	1.17E-15	1.09E-15	9.43E-16	9.75E-16	1.02E-15	9.36E-16	8.72E-16	8.14E-16	8.33E-16	7.71E-16	7.59E-16
69	4379	CUL	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
70	18	DUR	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
71	359	GUA	6.68E-10	8.19E-10	5.67E-09	1.00E-08	1.11E-08	5.49E-09	7.18E-10	7.50E-10	6.86E-10	6.35E-10	6.00E-10	6.10E-10	5.60E-10	5.53E-10
72	6246	HER	2.73E-03	1.12E-02	2.29E-02	2.17E-02	1.47E-02	6.13E-03	5.93E-03	8.49E-03	5.30E-03	3.52E-03	2.08E-03	2.59E-03	1.57E-03	1.43E-03
73	67.4	IRA	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
74	206	JUA	7.35E-08	1.01E-07	1.15E-07	6.32E-05	9.39E-05	8.08E-08	8.43E-08	9.00E-08	7.95E-08	7.15E-08	6.47E-08	6.67E-08	5.89E-08	5.75E-08
75	0	LAP	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
76	0	MAT	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
77	6479	MAZ	4.70E-05	1.76E-04	2.46E-04	1.60E-04	8.55E-05	2.93E-05	5.64E-05	6.74E-05	3.27E-05	1.74E-05	1.34E-05	2.05E-04	1.24E-03	3.30E-04
78	940	MER	8.54E-14	9.68E-14	1.02E-13	9.78E-14	9.36E-14	8.53E-14	8.88E-14	9.07E-14	8.58E-14	8.09E-14	7.91E-14	7.94E-14	7.57E-14	7.43E-14
79	26449	MEX	4.95E-06	6.35E-06	7.01E-06	6.74E-06	9.07E-03	9.73E-02	5.42E-06	5.71E-06	2.48E-02	5.08E-01	4.34E-06	6.88E-02	5.32E-01	6.98E-01
80	0	MIN	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
81	10321	MON	6.42E-08	1.05E-07	1.21E-07	1.04E-07	8.27E-08	5.58E-08	7.08E-08	7.61E-08	5.84E-08	4.61E-08	4.21E-08	4.26E-08	3.35E-08	3.14E-08
82	29.9	MOR	3.29E-10	3.92E-10	4.16E-10	3.98E-10	3.71E-10	3.23E-10	3.47E-10	3.56E-10	3.26E-10	3.02E-10	2.92E-10	2.93E-10	2.71E-10	2.65E-10
83	0	NUE	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
84	279	OAX	0.00E+00	0.00E+00	0.00E+00	0.00E+00	6.08E-05	1.54E-02	0.00E+00	0.00E+00	0.00E+00	1.43E-02	0.00E+00	0.00E+00	9.46E-03	4.21E-02
85	801	PAZ	0.00E+00	2.15E-13	1.31E-04	8.81E-04	8.17E-04	4.93E-13	1.41E-13	2.44E-13	2.26E-13	2.45E-13	8.73E-14	1.47E-13	1.27E-13	1.42E-13
86	131	PUE	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	6.37E-04	0.00E+00	0.00E+00	0.00E+00	1.52E-03	0.00E+00	2.45E-05	6.71E-04	2.49E-03
87	9643.1	SAC	6.62E-09	1.04E-08	1.23E-08	1.13E-08	9.47E-09	6.83E-09	7.68E-09	8.37E-09	6.82E-09	5.69E-09	5.04E-09	5.23E-09	4.31E-09	4.12E-09
88	23	SAL	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
89	216	SAN	2.81E-12	3.72E-12	4.02E-12	3.72E-12	3.24E-12	2.59E-12	2.96E-12	3.09E-12	2.66E-12	1.85E-05	2.20E-12	1.70E-04	1.53E-04	1.51E-05
90	124	SLU	4.03E-09	5.06E-09	5.37E-09	7.58E-09	9.03E-09	3.74E-09	4.19E-09	4.33E-09	3.82E-09	3.43E-09	3.29E-09	3.30E-09	2.99E-09	2.89E-09
91	119	TAL	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	5.56E-05	0.00E+00	0.00E+00	0.00E+00	1.80E-04	0.00E+00	3.93E-06	1.55E-04	6.31E-04
92	7413	TAM	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
93	23	TEP	3.31E-14	5.20E-14	5.77E-14	4.90E-14	3.86E-14	2.65E-14	3.47E-14	3.66E-14	2.81E-14	2.21E-14	2.07E-14	2.07E-14	1.64E-14	1.54E-14
94	3429	TIJ	8.71E-06	2.92E-05	1.17E-04	2.61E-04	2.62E-04	9.02E-05	1.04E-05	3.87E-05	3.00E-05	2.70E-05	2.99E-06	3.25E-06	1.55E-06	1.39E-06
95	581.9	TOL	1.36E-13	1.68E-13	1.83E-13	1.78E-13	2.52E-05	2.61E-04	1.48E-13	1.55E-13	1.96E-04	2.26E-03	1.24E-13	5.16E-04	2.69E-03	2.91E-03
96	25	TUX	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
97	3033.6	URU	2.40E-05	9.34E-05	1.37E-04	8.95E-05	4.67E-05	1.56E-05	3.09E-05	3.76E-05	1.64E-04	6.98E-04	5.25E-05	3.09E-03	5.95E-03	1.99E-03
98	12582	VER	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
99	17	ZAC	1.92E-10	2.38E-10	2.62E-10	2.58E-10	2.41E-10	2.11E-10	2.14E-10	2.24E-10	2.09E-10	1.94E-10	1.80E-10	1.87E-10	1.72E-10	1.69E-10
Total Wet SO_4Loading-No Backg.			3.17E-03	1.27E-02	2.50E-02	2.42E-02	2.58E-02	1.20E-01	6.46E-03	9.15E-03	3.09E-02	5.31E-01	2.30E-03	7.55E-02	5.54E-01	7.50E-01
Total Wet SO_4Loading-With Backg.			**4.80**	**4.81**	**4.83**	**4.82**	**4.83**	**4.92**	**4.81**	**4.81**	**4.83**	**5.33**	**4.80**	**4.88**	**5.35**	**5.55**
			1	**2**	**3**	**4**	**5**	**6**	**7**	**8**	**9**	**10**	**11**	**12**	**13**	**14**

Table A.1 (cont'd)

Wet SO_4 Deposition - (kg/ha/year)

15	16	17	18	19	20	21	22	23	24	25	26	27	28	29	30	31	32
0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	3.66E-04
0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
6.16E-07	5.23E-07	5.11E-07	5.75E-07	5.07E-07	5.03E-07	4.22E-07	4.60E-07	4.54E-07	4.28E-07	3.33E-07	3.33E-07	2.50E-07	3.24E-07	3.80E-07	3.08E-07	3.16E-07	7.07E-07
2.77E-05	1.50E-05	1.29E-05	1.83E-05	1.16E-05	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	4.89E-04
0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	2.86E-07
0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	5.37E-06
0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
1.05E-05	6.90E-06	6.35E-06	8.28E-06	6.01E-06	5.76E-06	3.77E-06	4.67E-06	4.43E-06	3.71E-06	1.97E-06	1.87E-06	6.83E-09	1.72E-06	2.60E-06	1.50E-06	2.66E-06	2.40E-05
6.83E-06	4.91E-06	4.89E-06	6.69E-06	5.09E-06	5.11E-06	3.42E-06	4.20E-06	4.17E-06	3.88E-06	2.32E-06	2.46E-06	1.40E-06	2.48E-06	3.55E-06	2.34E-06	1.32E-06	8.50E-06
0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	6.26E-07	1.10E-05
0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
2.22E-05	1.92E-05	1.88E-05	2.09E-05	1.87E-05	1.85E-05	1.59E-05	1.72E-05	1.69E-05	1.61E-05	1.29E-05	1.29E-05	1.01E-05	1.26E-05	1.45E-05	1.21E-05	1.22E-05	2.50E-05
0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	1.01E-06
0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	3.38E-05
0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
9.97E-11	8.98E-11	8.85E-11	9.57E-11	8.85E-11	8.75E-11	7.80E-11	8.27E-11	8.20E-11	7.90E-11	6.73E-11	6.70E-11	5.58E-11	6.57E-11	7.27E-11	6.37E-11	5.98E-11	1.03E-10
1.74E-11	1.61E-11	1.59E-11	1.66E-11	1.56E-11	1.55E-11	1.44E-11	1.49E-11	1.48E-11	1.42E-11	1.27E-11	1.25E-11	1.09E-11	1.23E-11	1.33E-11	1.19E-11	1.31E-11	1.91E-11
2.17E-15	1.48E-15	1.32E-15	1.62E-15	1.21E-15	1.14E-15	7.95E-16	9.54E-16	8.88E-16	7.36E-16	4.16E-16	3.76E-16	0.00E+00	3.39E-16	4.93E-16	2.96E-16	1.07E-15	5.57E-15
0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
5.46E-11	0.00E+00	0.00E+00	1.09E-10	7.17E-11	8.86E-11	0.00E+00	6.04E-11	7.29E-11	9.32E-11	0.00E+00	9.45E-11	5.65E-11	1.58E-10	5.98E-10	2.08E-10	0.00E+00	1.18E-10
7.41E-16	6.62E-16	6.53E-16	7.06E-16	6.51E-16	6.42E-16	5.70E-16	6.07E-16	6.00E-16	5.75E-16	4.90E-16	4.88E-16	4.05E-16	4.81E-16	5.34E-16	4.65E-16	4.60E-16	8.00E-16
0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
5.39E-10	4.85E-10	4.74E-10	5.13E-10	4.70E-10	4.67E-10	4.16E-10	4.42E-10	4.38E-10	4.20E-10	3.55E-10	3.52E-10	2.92E-10	3.46E-10	3.88E-10	3.36E-10	3.59E-10	6.10E-10
1.11E-03	2.84E-06	2.53E-06	9.37E-04	2.37E-06	2.24E-06	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	1.02E-03
0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
5.52E-08	4.70E-08	4.61E-08	5.19E-08	4.55E-08	4.53E-08	3.79E-08	4.14E-08	4.08E-08	3.83E-08	2.99E-08	2.99E-08	2.25E-08	2.90E-08	3.44E-08	2.78E-08	2.68E-08	6.18E-08
0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
2.05E-03	9.07E-03	2.70E-02	0.00E+00	1.40E-03	0.00E+00	8.75E-03	8.23E-04	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	4.68E-05
7.37E-14	6.89E-14	6.81E-14	7.11E-14	6.75E-14	6.71E-14	6.24E-14	6.50E-14	6.44E-14	6.20E-14	5.60E-14	5.52E-14	4.90E-14	5.46E-14	5.83E-14	5.34E-14	5.68E-14	7.96E-14
3.81E-06	3.33E-06	3.25E+00	2.88E-01	5.98E-01	3.36E-02	4.18E-01	1.74E-01	2.94E-06	2.80E-06	2.28E-06	2.26E-06	1.79E-06	2.22E-06	2.53E-06	2.13E-06	2.28E-06	4.42E-06
0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
3.15E-08	2.37E-08	2.21E-08	2.58E-08	2.08E-08	2.00E-08	1.53E-08	1.75E-08	1.67E-08	1.46E-08	9.58E-09	8.98E-09	5.44E-09	8.36E-09	1.10E-08	7.55E-09	1.53E-08	5.49E-08
2.63E-10	2.40E-10	2.34E-10	2.48E-10	2.31E-10	2.28E-10	2.08E-10	2.16E-10	2.14E-10	2.05E-10	1.77E-10	1.73E-10	1.47E-10	1.70E-10	1.86E-10	1.64E-10	1.91E-10	3.02E-10
0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
0.00E+00	0.00E+00	0.00E+00	1.09E-01	6.05E-02	1.40E-01	1.00E-02	6.14E-02	1.11E-01	4.27E-02	1.05E-02	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
0.00E+00	0.00E+00	0.00E+00	1.27E-13	8.01E-14	9.93E-14	0.00E+00	0.00E+00	6.04E-14	9.93E-14	0.00E+00	9.45E-14	0.00E+00	1.59E-13	3.24E-13	2.08E-13	0.00E+00	2.02E-13
0.00E+00	0.00E+00	3.39E-05	2.63E-03	8.28E-03	6.01E-01	3.97E-03	2.71E-03	1.09E-03	7.69E-06	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
3.98E-09	3.11E-09	2.98E-09	3.53E-09	2.92E-09	2.85E-09	2.21E-09	2.52E-09	2.45E-09	2.23E-09	1.54E-09	1.50E-09	9.84E-10	1.46E-09	1.86E-09	1.35E-09	1.79E-09	5.71E-09
0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
1.84E-05	1.44E-04	1.79E-03	1.68E-12	1.48E-12	1.46E-12	1.24E-12	1.34E-12	1.31E-12	1.22E-12	9.55E-13	9.24E-13	6.93E-13	8.86E-13	1.04E-12	8.42E-13	1.25E-12	2.57E-12
2.89E-09	2.54E-09	2.46E-09	2.64E-09	2.41E-09	2.36E-09	2.08E-09	2.21E-09	2.16E-09	2.05E-09	1.69E-09	1.65E-09	1.31E-09	1.61E-09	1.84E-09	1.55E-09	2.28E-09	3.98E-09
0.00E+00	0.00E+00	7.22E-04	1.20E-03	3.03E-03	8.63E-04	2.73E-03	2.00E-03	4.19E-04	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
1.56E-14	1.19E-14	1.11E-14	1.27E-14	1.04E-14	9.98E-15	7.77E-15	8.79E-15	8.40E-15	7.38E-15	4.97E-15	4.65E-15	2.90E-15	4.35E-15	5.61E-15	3.96E-15	1.00E-14	3.13E-14
1.20E-06	4.01E-07	0.00E+00	6.55E-07	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	3.60E-07	9.70E-06
1.12E-13	9.95E-14	1.56E-02	1.07E-03	2.41E-03	1.50E-04	2.32E-03	8.55E-04	1.12E-05	8.67E-14	7.33E-14	7.27E-14	6.05E-14	7.22E-14	8.09E-14	6.98E-14	6.92E-14	1.21E-13
0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
1.50E-03	1.66E-03	1.45E-02	3.25E-04	1.13E-03	0.00E+00	4.00E-03	3.67E-04	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	1.60E-05
0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
1.64E-10	1.48E-10	1.46E-10	1.59E-10	1.45E-10	1.44E-10	1.29E-10	1.37E-10	1.35E-10	1.30E-10	1.10E-10	1.09E-10	9.09E-11	1.08E-10	1.20E-10	1.04E-10	9.15E-11	1.63E-10
4.75E-03	1.09E-02	3.31E+00	4.03E-01	6.75E-01	7.75E-01	4.50E-01	2.42E-01	1.13E-01	4.27E-02	1.05E-02	1.98E-05	1.35E-05	1.94E-05	2.37E-05	1.84E-05	1.98E-05	2.06E-03
4.80	4.81	8.11	5.20	5.47	5.58	5.25	5.04	4.91	4.84	4.81	4.80	4.80	4.80	4.80	4.80	4.80	4.80
15	16	17	18	19	20	21	22	23	24	25	26	27	28	29	30	31	32

Table A.1 (cont'd)

Wet SO_4 Deposition - (kg/ha/year)

69	70	71	72	73	74	75	76	77	78	79	80	81	82	83	84	85	86
0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	1.07E-03	0.00E+00	3.42E-01	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	6.24E-01	1.76E-01	4.94E-03
8.18E-06	5.74E-06	0.00E+00	3.33E-06	0.00E+00	8.71E-05	7.85E-04	2.25E-04	1.74E-02	0.00E+00	0.00E+00	0.00E+00	1.25E-03	0.00E+00	0.00E+00	4.98E-02	1.29E-02	3.76E-02
0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	2.95E-07	0.00E+00	3.45E-02	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	1.43E+00	5.19E-01	2.72E-05
0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	4.95E-04	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	1.56E-04	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	4.87E-04	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	1.19E-01	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	5.64E+00
0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	4.04E-01	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	1.32E-01	0.00E+00
0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	8.19E-06	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	5.02E-08	0.00E+00	3.68E-03	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	2.61E-02	1.25E-02	4.63E-06
0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	4.99E-02	0.00E+00	0.00E+00	0.00E+00	5.27E-04	1.15E-02	0.00E+00	3.06E-05	7.06E-03	1.43E-03
0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	3.83E-07	0.00E+00	2.53E-02	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	7.24E-07	5.26E-06	3.53E-05
2.56E-05	3.46E-05	1.29E-05	2.70E-05	0.00E+00	1.22E-06	2.99E-05	1.13E-05	2.33E-04	0.00E+00	0.00E+00	0.00E+00	1.98E-05	0.00E+00	1.02E-06	1.73E-03	1.53E-04	4.61E-04
2.95E-04	1.51E-02	3.57E-03	1.64E-02	0.00E+00	0.00E+00	5.59E-08	0.00E+00	3.70E-03	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	1.06E-07	7.67E-07	5.16E-06
1.98E-02	9.33E-02	5.00E-02	1.01E-01	3.83E-07	3.58E-02	2.35E-03	1.21E-03	9.09E-04	0.00E+00	7.86E-08	1.84E-07	2.55E-07	2.94E-08	2.64E-07	2.05E-01	0.00E+00	3.21E-02
2.61E-03	3.59E-03	7.98E-04	7.69E-04	0.00E+00	2.42E-03	7.92E-04	5.93E-05	2.14E-03	0.00E+00	0.00E+00	0.00E+00	7.16E-05	0.00E+00	0.00E+00	6.22E-03	1.06E-03	4.69E-03
0.00E+00	1.50E-07	2.56E-07	2.35E-07	0.00E+00	1.55E-06	5.49E-08	0.00E+00	4.94E-03	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	1.57E-02	5.27E-04	5.06E-02
0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	2.85E-08	0.00E+00	1.88E-03	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	5.38E-08	3.91E-07	2.63E-06
0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	4.92E-08	0.00E+00	3.25E-03	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	9.29E-08	6.75E-07	4.54E-06
0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	4.30E-07	0.00E+00	2.84E-02	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	8.13E-07	5.90E-06	3.97E-05
0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	4.11E-07	0.00E+00	2.71E-02	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	7.76E-07	5.63E-06	3.79E-05
0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	1.32E-07	0.00E+00	8.80E-03	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	2.49E-07	1.81E-06	8.44E-02
5.16E-08	7.59E-06	1.18E-05	1.17E-05	0.00E+00	6.28E-05	2.14E-07	9.88E-08	6.01E-04	0.00E+00	0.00E+00	0.00E+00	4.12E-04	0.00E+00	0.00E+00	3.00E-03	2.04E-03	6.52E-03
0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	6.15E-07	0.00E+00	4.07E-02	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	1.05E-02	8.44E-06	3.90E-01
0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	9.08E-07	0.00E+00	6.00E-02	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	1.72E-06	1.25E-05	1.33E+00
0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	6.06E-08	0.00E+00	4.01E-03	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	1.73E-02	8.31E-07	6.93E-03
0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	5.50E-07	0.00E+00	3.64E-02	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	1.04E-06	7.55E-06	5.08E-05
0.00E+00	7.67E-04	9.35E-03	5.23E-02	0.00E+00	0.00E+00	2.25E-07	0.00E+00	1.49E-02	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	4.24E-07	3.08E-06	2.07E-05
0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	1.06E-07	0.00E+00	7.02E-03	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	2.00E-07	1.46E-06	9.79E-06
0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	9.00E-04	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	4.14E-08	0.00E+00	2.74E-03	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	7.82E-08	5.68E-07	3.82E-06
0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	1.02E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	1.65E-05	1.82E+00
0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	7.41E-08	0.00E+00	1.25E-01	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	1.40E-07	1.02E-06	1.62E-01
0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	2.03E-04	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	1.07E-02	0.00E+00	0.00E+00
0.00E+00	5.59E-02	7.88E-03	6.87E-02	0.00E+00	0.00E+00	0.00E+00	0.00E+00	5.80E-04	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	1.12E-03	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	7.29E-03	9.06E-04	4.95E-03
0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	1.33E-03	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	9.73E-03	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	4.36E-04	1.31E-02	1.76E-03
0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	9.85E-06	0.00E+00	2.60E-04	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	3.89E-03	0.00E+00	8.04E-03
0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	1.59E-04	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	4.04E-04	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
7.15E-03	6.70E-02	5.10E-02	8.66E-02	2.64E-06	4.12E-02	2.67E-06	3.43E-06	1.03E-03	0.00E+00	2.30E-09	5.37E-09	7.31E-07	8.35E-10	1.57E-06	2.16E-01	0.00E+00	3.73E-04
3.12E-05	7.27E-05	7.25E-05	8.59E-05	4.02E-06	8.68E-05	3.84E-06	4.71E-06	3.79E-04	0.00E+00	0.00E+00	1.43E-07	1.14E-06	0.00E+00	1.11E-06	1.13E-02	1.07E-04	3.42E-03
0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	1.24E-03	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	5.97E-03	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
1.60E-03	1.80E-02	6.26E-03	1.92E-02	0.00E+00	0.00E+00	0.00E+00	0.00E+00	3.62E-04	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	7.73E-02	0.00E+00	1.64E-03
4.14E-06	9.29E-06	1.19E-05	1.18E-05	0.00E+00	4.86E-05	1.98E-05	3.62E-06	3.45E-03	0.00E+00	0.00E+00	0.00E+00	4.62E-05	0.00E+00	7.18E-08	2.77E-03	1.71E-03	8.02E-03
0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	3.29E-03	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	1.49E-05	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	6.87E-04	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	2.29E-03	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
2.70E-05	9.05E-03	2.50E-01	9.95E-02	1.46E-05	1.36E+00	1.44E-05	1.57E-05	3.44E-03	0.00E+00	3.57E-06	7.59E-06	1.02E-05	1.49E-06	1.05E-05	1.18E-05	7.07E-07	2.58E-02
5.70E-07	1.59E-06	2.15E-06	2.15E-06	0.00E+00	6.57E-06	7.99E-04	2.19E-04	2.49E-03	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	1.77E-03	8.48E-04	1.30E-02
0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	8.80E-04	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	2.76E-03	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	1.19E-04	3.66E-05	1.62E-06	8.86E-03	0.00E+00	0.00E+00	0.00E+00	5.54E-04	0.00E+00	0.00E+00	1.58E-02	3.89E-03	3.12E-02
0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	2.40E-01	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	6.55E-01
4.29E-04	7.97E-07	1.32E-06	1.31E-06	9.68E-05	8.24E-06	1.35E-02	5.73E-03	1.14E-02	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	2.79E-02	1.66E-06	1.26E-02
0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	1.98E-06	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
1.13E-10	1.34E-10	1.45E-10	1.42E-10	7.20E-11	7.65E-05	7.00E-11	7.52E-11	2.16E-06	3.62E-12	2.34E-11	4.12E-11	5.17E-11	1.20E-11	5.28E-11	5.85E-11	7.00E-12	2.19E-11
1.86E-11	2.12E-11	2.25E-11	2.22E-11	1.31E-11	2.63E-11	1.31E-11	1.37E-11	2.97E-04	1.78E-12	6.72E-12	9.92E-12	1.13E-11	4.07E-12	1.18E-11	1.21E-11	2.52E-12	5.82E-12
2.98E-15	5.61E-15	7.49E-15	6.96E-15	5.08E-16	2.09E-02	2.23E-09	9.34E-10	4.17E-08	0.00E+00	0.00E+00	3.02E-16	4.67E-16	0.00E+00	5.86E-16	4.15E-09	1.65E-08	2.56E-08
0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	1.98E-06	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
6.97E-08	7.31E-08	4.82E-08	6.46E-08	1.91E-08	1.73E-08	4.57E-08	4.49E-08	2.95E-08	0.00E+00	0.00E+00	0.00E+00	4.45E-10	0.00E+00	9.93E-11	8.87E-09	8.33E-09	1.26E-08
8.56E-16	1.01E-15	1.10E-15	1.07E-15	5.36E-16	1.32E-15	5.29E-16	5.66E-16	1.98E-06	2.71E-17	1.80E-16	3.22E-16	4.00E-16	9.18E-17	4.14E-16	4.53E-16	5.29E-17	1.67E-16
0.00E+00	6.17E-03	2.29E-02	1.71E-02	0.00E+00	5.17E-02	0.00E+00	0.00E+00	3.77E-04	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
0.00E+00	3.49E-05	1.82E-04	1.50E-04	0.00E+00	7.04E-04	0.00E+00	0.00E+00	1.55E-06	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
2.06E-07	2.32E-07	1.59E-07	2.11E-07	4.52E-08	6.64E-08	1.14E-07	1.14E-07	1.18E-10	2.34E-11	1.45E-10	2.52E-10	3.08E-10	7.50E-11	3.19E-10	2.54E-08	4.16E-11	1.29E-10
3.44E-03	1.79E-02	1.77E-02	2.28E-02	0.00E+00	4.84E-02	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	5.25E-02	0.00E+00	4.24E-04
1.45E-07	2.21E-07	7.28E-08	1.70E-07	0.00E+00	1.06E-04	2.33E-08	2.26E-08	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
5.07E-05	5.34E-04	1.30E-03	1.02E-03	3.44E-08	1.26E-03	3.36E-08	3.73E-08	5.29E-09	0.00E+00	6.47E-09	1.55E-08	2.20E-08	2.39E-09	2.27E-08	3.03E-03	0.00E+00	5.85E-09
0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
1.59E-05	5.27E-05	8.88E-05	7.84E-05	0.00E+00	3.91E-04	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
7.87E-14	8.80E-14	9.29E-14	9.15E-14	5.78E-14	1.06E-13	4.82E-08	1.07E-10	3.28E-06	9.68E-15	3.16E-14	4.49E-14	5.03E-14	2.04E-14	5.15E-14	5.50E-08	6.35E-07	1.07E-06
4.58E-06	5.16E-03	6.22E-06	1.92E-02	2.57E-06	7.86E-06	2.54E-06	2.75E-06	5.79E-07	3.54E-09	7.30E-07	1.46E-06	1.90E-06	3.20E-07	1.97E-06	2.17E-06	1.56E-07	6.40E-07
0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
1.37E-04	1.26E-04	7.96E-08	5.85E-05	1.10E-08	4.74E-02	4.24E-05	3.20E-05	2.17E-04	0.00E+00	0.00E+00	5.83E-09	8.51E-09	0.00E+00	9.92E-09	1.51E-08	3.29E-06	9.06E-06
2.87E-10	3.38E-10	3.62E-10	3.56E-10	1.85E-10	4.96E-04	1.84E-10	1.96E-10	2.58E-06	1.64E-11	8.49E-11	1.37E-10	1.59E-10	4.54E-11	1.67E-10	1.71E-10	2.42E-11	6.88E-11
0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	3.74E-09	1.55E-09	7.37E-08	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	4.32E-09	2.39E-08	3.46E-08
1.54E-03	9.53E-03	1.52E-02	1.53E-02	1.83E-12	1.91E-02	3.23E-12	3.24E-12	3.19E-13	0.00E+00	0.00E+00	0.00E+00	3.61E-13	0.00E+00	1.51E-13	1.31E-12	0.00E+00	2.44E-13
0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
2.12E-05	3.17E-05	1.01E-05	2.40E-05	1.94E-09	4.32E-01	3.54E-06	3.42E-06	0.00E+00	0.00E+00	2.38E-10	8.13E-10	1.27E-09	0.00E+00	1.37E-09	1.58E-09	0.00E+00	0.00E+00
0.00E+00	1.05E-05	8.49E-05	4.85E-05	0.00E+00	3.15E-04	0.00E+00	0.00E+00	1.98E-06	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
1.19E-06	1.76E-06	5.88E-07	1.35E-06	2.53E-08	7.47E-03	2.44E-07	2.38E-07	1.90E-13	0.00E+00	3.39E-13	7.32E-13	9.09E-13	1.25E-13	9.85E-13	1.04E-08	0.00E+00	2.27E-13
2.46E-07	2.80E-07	1.15E-04	4.25E-05	4.15E-08	1.48E-03	1.26E-07	1.25E-07	5.28E-10	1.15E-10	8.52E-10	1.54E-09	1.84E-09	3.99E-10	1.95E-09	2.23E-08	1.70E-10	6.22E-10
0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
5.53E-05	7.01E-05	1.18E-05	4.22E-05	0.00E+00	0.00E+00	1.08E-05	9.04E-06	1.86E-04	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	1.26E-04	1.12E-06	7.78E-04
1.97E-14	3.06E-14	3.75E-14	3.56E-14	5.80E-15	6.58E-14	6.16E-15	7.13E-15	1.98E-06	0.00E+00	1.25E-15	4.19E-15	5.61E-15	0.00E+00	6.60E-15	6.26E-15	0.00E+00	0.00E+00
2.65E-04	7.24E-04	9.98E-04	9.50E-04	0.00E+00	1.39E-03	1.39E-05	1.19E-05	2.17E-03	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	8.23E-03	9.40E-04	1.39E-02
1.30E-13	1.54E-13	1.67E-13	1.64E-13	8.09E-14	2.02E-13	7.91E-14	8.55E-14	2.29E-14	3.90E-15	2.67E-14	4.78E-14	5.99E-14	1.35E-14	6.17E-14	6.75E-14	7.56E-15	2.46E-14
0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	2.16E-06	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
6.67E-06	2.32E-05	4.19E-05	3.58E-05	9.95E-08	2.09E-04	2.61E-07	2.59E-07	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	5.31E-08	0.00E+00	0.00E+00
0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	1.08E-03	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
1.87E-10	2.21E-10	3.96E-05	1.49E-05	1.17E-10	8.47E-04	1.13E-10	1.22E-10	1.47E-06	4.73E-12	3.37E-11	6.15E-11	7.87E-11	1.67E-11	8.04E-11	9.06E-11	9.93E-12	3.28E-11
3.75E-02	3.03E-01	4.37E-01	5.21E-01	1.21E-04	2.08E+00	1.95E-02	7.55E-03	2.69E+00	3.71E-09	4.39E-06	9.41E-06	2.90E-03	1.15E-02	1.65E-05	2.83E+00	8.85E-01	1.04E+01
4.84	**5.10**	**5.24**	**5.32**	**4.80**	**6.88**	**4.82**	**4.81**	**7.49**	**4.80**	**4.80**	**4.80**	**4.80**	**4.81**	**4.80**	**7.63**	**5.68**	**15.15**
69	**70**	**71**	**72**	**73**	**74**	**75**	**76**	**77**	**78**	**79**	**80**	**81**	**82**	**83**	**84**	**85**	**86**

Table A.1 (cont'd)

Wet SO_4 Deposition - (kg/ha/year)

87	88	89	90	91	92	93	94	95	96	97	98	99	100
4.14E+00	4.26E-01	2.21E-02	4.42E-06	8.74E-01	1.39E-06	2.01E-01	7.23E-01	0.00E+00	1.24E-01	2.71E-06	0.00E+00	0.00E+00	3.49E-01
3.16E-02	1.78E-02	6.23E-03	2.29E-03	1.67E-01	4.25E-04	3.53E-02	3.12E-02	0.00E+00	1.81E-02	3.56E-03	0.00E+00	0.00E+00	4.98E-02
0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
4.12E-02	1.24E-02	8.00E-03	5.99E-02	6.66E-01	1.19E-01	9.31E+00	4.27E-02	0.00E+00	1.63E+00	2.47E-06	0.00E+00	0.00E+00	6.35E+00
3.38E-04	3.65E-04	2.29E-04	8.32E-03	0.00E+00	8.38E-02	1.22E-04	3.75E-05	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	4.08E-05
1.06E-04	1.15E-04	7.21E-05	0.00E+00	0.00E+00	5.11E-01	3.83E-05	1.18E-05	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	1.28E-05
3.32E-04	3.59E-04	2.25E-04	0.00E+00	0.00E+00	0.00E+00	1.20E-04	3.69E-05	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	4.01E-05
7.37E-02	1.16E-03	7.26E-04	0.00E+00	9.59E-03	0.00E+00	2.73E+01	1.19E-04	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	2.00E+01
8.19E+00	2.08E-01	7.82E-04	0.00E+00	0.00E+00	0.00E+00	1.33E+00	1.84E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	3.00E+00
5.59E-06	6.04E-06	3.79E-06	7.90E-04	0.00E+00	1.87E-04	2.01E-06	6.21E-07	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	6.74E-07
4.49E-01	0.00E+00	0.00E+00	6.00E-02	0.00E+00	8.18E-02	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
7.87E-01	1.18E-02	1.36E-03	6.86E-07	1.14E-01	2.15E-07	1.77E-02	1.46E-01	0.00E+00	5.11E-03	4.21E-07	0.00E+00	0.00E+00	3.01E-02
4.88E-02	8.95E-03	1.33E-04	5.39E-04	0.00E+00	1.25E-03	2.45E-02	2.37E-03	0.00E+00	8.19E-04	3.20E-05	0.00E+00	0.00E+00	4.88E-03
1.50E-02	1.61E-02	1.04E-02	5.24E-06	8.66E-01	1.64E-06	1.31E-01	1.34E-03	0.00E+00	1.84E-07	3.21E-06	0.00E+00	0.00E+00	2.26E-01
3.96E-04	1.83E-04	5.28E-05	2.12E-05	9.57E-04	6.55E-06	5.32E-04	3.43E-04	2.47E-06	6.09E-04	3.11E-05	9.92E-07	0.00E+00	6.90E-04
2.18E-03	2.35E-03	1.52E-03	7.64E-07	1.26E-01	2.40E-07	1.91E-02	1.95E-04	0.00E+00	2.68E-08	4.69E-07	0.00E+00	0.00E+00	3.29E-02
3.66E-03	1.71E-04	2.89E-08	0.00E+00	4.73E-01	0.00E+00	1.22E-02	1.13E-03	1.35E-07	5.55E-02	0.00E+00	3.42E-07	2.86E-07	2.61E-02
2.68E-03	1.14E-03	6.16E-04	1.65E-04	1.87E-02	2.97E-05	8.04E-03	1.88E-03	0.00E+00	2.70E-03	1.81E-04	5.02E-05	0.00E+00	1.24E-02
6.83E-03	2.87E-03	1.49E-03	7.50E-07	1.93E-01	2.35E-07	2.27E-02	2.27E-03	0.00E+00	9.63E-04	4.60E-07	0.00E+00	0.00E+00	3.57E-02
1.11E-03	1.19E-03	7.74E-04	3.89E-07	6.44E-02	1.22E-07	9.73E-03	9.93E-05	0.00E+00	1.37E-08	2.39E-07	0.00E+00	0.00E+00	1.68E-02
1.92E-03	2.06E-03	1.34E-03	6.72E-07	1.11E-01	2.11E-07	1.68E-02	1.72E-04	0.00E+00	2.36E-08	4.13E-07	0.00E+00	0.00E+00	2.90E-02
1.68E-02	1.80E-02	1.17E-02	5.88E-06	9.73E-01	1.85E-06	1.47E-01	1.50E-03	0.00E+00	2.07E-07	3.61E-06	0.00E+00	0.00E+00	2.53E-01
1.60E-02	1.72E-02	1.12E-02	5.61E-06	9.28E-01	1.76E-06	1.40E-01	1.43E-03	0.00E+00	1.97E-07	3.44E-06	0.00E+00	0.00E+00	2.42E-01
5.16E-03	5.54E-03	3.59E-03	1.80E-06	2.99E-01	5.66E-07	1.06E-01	4.61E-04	0.00E+00	6.34E-08	1.11E-06	0.00E+00	0.00E+00	4.09E-01
1.56E-03	7.02E-04	1.77E-04	4.40E-05	3.34E-03	9.42E-07	7.60E-04	2.43E-03	3.81E-05	1.80E-03	2.44E-04	0.00E+00	0.00E+00	6.98E-04
2.40E-02	2.58E-02	1.67E-02	8.41E-06	3.16E+00	2.64E-06	2.20E-01	2.15E-03	0.00E+00	2.95E-07	5.16E-06	0.00E+00	0.00E+00	3.76E-01
3.55E-02	3.81E-02	2.47E-02	1.24E-05	9.89E+00	3.90E-06	3.22E-01	3.17E-03	0.00E+00	4.36E-07	7.62E-06	0.00E+00	0.00E+00	7.01E-01
2.37E-03	2.54E-03	1.65E-03	8.28E-07	2.46E-01	2.60E-07	2.13E-02	2.11E-04	0.00E+00	3.57E-04	5.08E-07	0.00E+00	0.00E+00	3.67E-02
2.15E-02	2.31E-02	1.50E-02	7.52E-06	1.29E+00	2.36E-06	1.88E-01	1.92E-03	0.00E+00	2.64E-07	4.62E-06	0.00E+00	0.00E+00	3.24E-01
8.78E-03	9.43E-03	6.11E-03	3.07E-06	5.08E-01	9.64E-07	7.68E-02	7.84E-04	0.00E+00	1.08E-07	1.89E-06	0.00E+00	0.00E+00	1.32E-01
4.15E-03	4.45E-03	2.88E-03	1.45E-06	2.40E-01	4.55E-07	3.63E-02	3.70E-04	0.00E+00	5.10E-08	8.90E-07	0.00E+00	0.00E+00	6.25E-02
6.14E-04	6.64E-04	4.17E-04	0.00E+00	0.00E+00	0.00E+00	2.21E-04	6.83E-05	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	7.41E-05
1.62E-03	1.74E-03	1.13E-03	5.66E-07	9.37E-02	1.78E-07	1.42E-02	1.45E-04	0.00E+00	1.99E-08	3.47E-07	0.00E+00	0.00E+00	2.44E-02
1.70E-01	1.29E-01	6.11E-02	1.45E-05	4.86E+00	0.00E+00	5.16E+00	1.36E-03	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	2.14E+00
1.16E+00	1.79E-01	2.01E-03	1.01E-06	1.67E-01	3.18E-07	8.89E-01	8.60E-02	0.00E+00	3.55E-08	6.21E-07	0.00E+00	0.00E+00	1.01E+00
1.39E-04	1.50E-04	9.41E-05	0.00E+00	9.03E-02	0.00E+00	5.00E-05	1.54E-05	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	1.67E-05
3.96E-04	4.28E-04	2.69E-04	0.00E+00	0.00E+00	0.00E+00	1.43E-04	4.40E-05	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	4.78E-05
3.00E-04	1.50E-04	9.41E-05	0.00E+00	7.95E-03	0.00E+00	3.26E-03	9.03E-05	0.00E+00	1.49E-03	0.00E+00	0.00E+00	0.00E+00	3.94E-03
9.07E-04	9.81E-04	6.15E-04	0.00E+00	0.00E+00	0.00E+00	3.27E-04	1.01E-04	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	1.09E-04
7.26E-01	3.05E-02	2.46E-04	0.00E+00	0.00E+00	0.00E+00	2.71E-02	2.22E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	4.78E-02
1.18E-04	1.28E-04	8.04E-05	0.00E+00	2.03E-02	0.00E+00	5.40E-04	1.32E-05	0.00E+00	1.49E-04	0.00E+00	0.00E+00	0.00E+00	6.65E-04
1.08E-04	1.17E-04	7.35E-05	0.00E+00	0.00E+00	0.00E+00	3.90E-05	1.20E-05	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	1.31E-05
2.76E-04	2.98E-04	1.87E-04	0.00E+00	0.00E+00	0.00E+00	9.94E-05	3.07E-05	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	3.33E-05
8.13E-04	5.69E-04	3.03E-04	0.00E+00	6.38E-01	0.00E+00	6.97E-03	4.97E-05	3.92E-09	4.39E-02	0.00E+00	2.98E-06	2.02E-06	2.36E-02
8.44E-04	2.84E-04	7.16E-05	4.93E-06	2.10E-02	0.00E+00	1.37E-03	5.45E-04	9.59E-09	4.06E-03	2.37E-06	1.87E-06	1.14E-06	2.01E-03
8.46E-04	9.15E-04	5.74E-04	0.00E+00	0.00E+00	0.00E+00	3.62E-03	9.41E-05	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	9.10E-03
4.07E-03	4.41E-03	2.76E-03	0.00E+00	0.00E+00	0.00E+00	1.47E-03	4.53E-04	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	2.85E-03
2.47E-04	2.67E-04	1.68E-04	0.00E+00	1.76E-01	0.00E+00	1.12E-04	2.75E-05	0.00E+00	2.52E-03	0.00E+00	0.00E+00	0.00E+00	1.26E-04
5.24E-03	3.10E-03	1.37E-03	3.45E-04	7.28E-03	2.91E-05	4.46E-03	4.29E-03	1.58E-07	1.09E-03	4.08E-04	1.15E-06	0.00E+00	4.24E-03
2.25E-03	2.43E-03	1.52E-03	0.00E+00	0.00E+00	0.00E+00	8.10E-04	2.50E-04	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	2.71E-04
1.02E-05	1.10E-05	6.90E-06	0.00E+00	0.00E+00	0.00E+00	3.67E-06	1.13E-06	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	1.23E-06
4.69E-04	5.07E-04	3.18E-04	0.00E+00	0.00E+00	0.00E+00	1.69E-04	5.21E-05	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	5.66E-05
1.56E-03	1.69E-03	1.06E-03	0.00E+00	0.00E+00	0.00E+00	5.64E-04	1.74E-04	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	1.89E-04
5.27E-03	2.73E-03	2.12E-03	0.00E+00	1.52E-01	0.00E+00	9.54E-03	5.46E-04	5.80E-06	1.37E-05	0.00E+00	1.31E-05	1.12E-05	1.02E-02
2.85E-03	1.27E-03	5.95E-04	0.00E+00	7.54E-03	0.00E+00	4.69E-03	1.99E-03	0.00E+00	3.41E-04	0.00E+00	0.00E+00	0.00E+00	5.07E-03
6.00E-04	6.49E-04	4.07E-04	0.00E+00	0.00E+00	0.00E+00	2.16E-04	6.68E-05	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	7.25E-05
8.48E-04	7.65E-04	1.50E-04	0.00E+00	0.00E+00	0.00E+00	5.03E-05	1.12E-05	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	1.33E-06
1.29E-02	7.47E-03	3.40E-03	7.57E-04	3.02E-02	7.82E-05	1.24E-02	9.95E-03	1.04E-05	7.17E-03	7.92E-04	0.00E+00	0.00E+00	1.26E-02
1.04E+00	5.87E-04	3.68E-04	0.00E+00	0.00E+00	0.00E+00	3.43E+00	6.04E-05	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	2.60E+00
5.77E-03	3.91E-03	3.69E-03	1.17E-06	4.56E-02	1.82E-07	2.32E-02	6.91E-04	0.00E+00	3.99E-03	6.91E-07	0.00E+00	0.00E+00	2.72E-02
1.35E-06	1.46E-06	9.18E-07	0.00E+00	0.00E+00	0.00E+00	4.88E-07	1.50E-07	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	1.63E-07
1.47E-06	1.59E-06	9.97E-07	4.13E-12	4.85E-11	2.17E-12	5.30E-07	1.64E-07	3.37E-11	6.55E-11	3.72E-12	6.28E-11	5.58E-11	1.77E-07
2.03E-04	2.19E-04	1.37E-04	1.67E-12	1.04E-11	1.04E-12	7.31E-05	2.25E-05	8.44E-12	1.32E-11	1.57E-12	1.33E-11	1.24E-11	2.45E-05
4.29E-08	4.07E-08	3.42E-08	1.46E-08	1.61E-08	5.98E-09	3.64E-08	3.39E-08	0.00E+00	1.71E-09	1.11E-08	1.04E-15	8.15E-16	3.18E-08
1.35E-06	1.46E-06	9.18E-07	0.00E+00	0.00E+00	0.00E+00	4.88E-07	1.50E-07	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	1.63E-07
0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
2.77E-08	2.80E-08	2.59E-08	9.79E-09	1.67E-08	4.14E-09	2.88E-08	2.00E-08	5.19E-11	7.32E-09	6.27E-09	1.37E-10	0.00E+00	2.87E-08
1.35E-06	1.46E-06	9.18E-07	3.08E-17	3.73E-16	1.60E-17	4.88E-07	1.50E-07	2.62E-16	5.06E-16	2.78E-17	4.90E-16	4.32E-16	1.63E-07
2.57E-04	2.79E-04	1.75E-04	0.00E+00	0.00E+00	0.00E+00	9.28E-05	2.86E-05	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	3.11E-05
1.06E-06	1.14E-06	7.18E-07	0.00E+00	0.00E+00	0.00E+00	3.82E-07	1.18E-07	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	1.28E-07
1.12E-10	8.47E-11	7.07E-11	2.44E-11	2.85E-08	5.85E-12	1.35E-08	7.29E-11	2.06E-10	2.46E-08	2.22E-11	3.77E-10	3.36E-10	2.27E-08
0.00E+00	0.00E+00	0.00E+00	0.00E+00	1.37E-01	0.00E+00	4.49E-03	0.00E+00	0.00E+00	1.61E-02	0.00E+00	0.00E+00	0.00E+00	1.30E-02
0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
4.84E-09	3.17E-09	2.43E-09	0.00E+00	1.51E-02	0.00E+00	1.31E-05	2.47E-09	1.14E-08	4.72E-04	0.00E+00	2.95E-08	2.43E-08	1.92E-04
0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
3.08E-06	3.14E-06	2.76E-06	7.53E-07	5.17E-07	2.29E-07	2.44E-06	2.04E-06	3.85E-14	1.55E-10	4.21E-07	5.79E-14	5.44E-14	1.95E-06
5.40E-07	3.84E-07	3.07E-07	4.76E-09	1.71E-06	0.00E+00	1.02E-06	3.15E-07	1.13E-06	2.50E-06	4.21E-09	2.44E-06	2.10E-06	1.25E-06
0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
1.36E-04	1.72E-04	1.78E-04	6.98E-06	2.37E-06	8.66E-07	9.78E-05	3.93E-05	3.14E-09	1.58E-08	1.93E-06	1.54E-08	1.25E-08	5.72E-05
1.76E-06	1.90E-06	1.19E-06	1.46E-11	1.41E-10	8.10E-12	6.34E-07	1.96E-07	1.10E-10	1.91E-10	1.35E-11	1.93E-10	1.79E-10	2.12E-07
0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
7.09E-08	7.14E-08	6.50E-08	2.66E-08	2.05E-08	1.15E-08	6.00E-08	5.38E-08	0.00E+00	1.61E-09	1.79E-08	0.00E+00	0.00E+00	5.11E-08
2.56E-13	1.43E-13	1.03E-13	0.00E+00	1.39E-12	0.00E+00	8.41E-13	8.17E-14	0.00E+00	1.30E-12	0.00E+00	2.17E-13	0.00E+00	1.18E-12
0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
0.00E+00	0.00E+00	0.00E+00	0.00E+00	1.01E-09	0.00E+00	3.93E-10	0.00E+00	5.13E-10	2.04E-09	0.00E+00	2.00E-09	1.55E-09	5.64E-10
1.35E-06	1.46E-06	9.18E-07	0.00E+00	0.00E+00	0.00E+00	4.88E-07	1.50E-07	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	1.63E-07
1.77E-13	1.17E-13	8.83E-14	0.00E+00	1.47E-08	0.00E+00	7.50E-09	9.53E-14	5.12E-13	8.96E-09	0.00E+00	1.26E-12	1.12E-12	1.37E-08
5.08E-10	3.63E-10	2.94E-10	8.75E-11	2.54E-08	1.55E-12	1.08E-08	3.15E-10	1.19E-09	2.16E-08	8.04E-11	2.33E-09	2.11E-09	1.96E-08
0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
3.40E-04	1.02E-04	3.40E-05	1.49E-06	1.44E-03	0.00E+00	5.31E-04	9.79E-05	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	5.83E-04
1.35E-06	1.46E-06	9.18E-07	0.00E+00	3.61E-15	0.00E+00	4.88E-07	1.50E-07	2.30E-15	8.53E-15	0.00E+00	9.84E-15	8.33E-15	1.63E-07
5.11E-03	1.80E-03	3.60E-04	7.82E-06	1.91E-02	1.24E-06	4.11E-03	4.05E-03	0.00E+00	3.19E-03	4.11E-06	1.77E-07	0.00E+00	4.39E-03
2.15E-14	1.62E-14	1.36E-14	4.39E-15	5.56E-14	0.00E+00	3.67E-14	1.37E-14	3.88E-14	7.62E-14	3.95E-15	7.33E-14	6.46E-14	4.34E-14
1.47E-06	1.59E-06	9.97E-07	0.00E+00	0.00E+00	0.00E+00	5.30E-07	1.64E-07	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	1.77E-07
0.00E+00	0.00E+00	0.00E+00	0.00E+00	6.10E-08	0.00E+00	2.87E-08	0.00E+00	0.00E+00	5.07E-08	0.00E+00	0.00E+00	0.00E+00	4.94E-08
7.40E-04	8.00E-04	5.02E-04	0.00E+00	0.00E+00	0.00E+00	2.67E-04	8.23E-05	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	8.93E-05
1.00E-06	1.08E-06	6.78E-07	5.80E-12	7.50E-11	0.00E+00	3.60E-07	1.11E-07	5.00E-11	1.02E-10	5.13E-12	9.66E-11	8.43E-11	1.21E-07
1.71E+01	1.24E+00	2.36E-01	1.33E-01	2.77E+01	7.98E-01	4.93E+01	5.14E+00	5.83E-05	1.93E+00	5.30E-03	7.33E-05	1.68E-05	3.87E+01
21.88	**6.04**	**5.04**	**4.93**	**32.51**	**5.60**	**54.11**	**9.94**	**4.80**	**6.73**	**4.81**	**4.80**	**4.80**	**43.46**
87	**88**	**89**	**90**	**91**	**92**	**93**	**94**	**95**	**96**	**97**	**98**	**99**	**100**

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1. INTRODUCTION

Various pollutants, alone or in combination, reduce plant growth, especially near sources of emissions. Important air pollutants that affect woody plants include sulfur dioxide (SO_2), ozone (O_3), fluoride, oxides of nitrogen (NOx), peroxyacetylnitrate (PAN), and particulates (e.g. cement kiln dusts, fluorides, soot, and heavy metals) (Kozlowski et al. 1991). Pollutants decrease leaf area by inhibiting leaf formation and expansion, injuring leaves and hastening leaf fall or leaf shedding. They also decrease height growth, diameter growth, and reproductive growth of woody plants sometimes without leaf damage. Reproductive organs may also be injured resulting in reduced yield of fruits and seeds.

2. EFFECTS OF AIR POLLUTANTS ON PLANT PHYSIOLOGY

Effects of pollutants on plant growth result from changes in physiological processes such as photosynthesis, enzyme activity and ease with which leaf pores (stomata) open and close in response to stress. Responses to pollution vary with plant species and genotype, pollutant dosage, types and combination of pollutants, plant response measured, developmental stages of plants, environmental conditions, and interactions of pollutants with plant disease and insects.

Air pollutants may alter the rate of photosynthesis by several mechanisms, including (1) clogging of leaf pores (stomata); (2) influencing stomatal aperture; (3) altering optical properties of leaves by changing reflectance and decreasing light intensity; (4) altering the heat balance of leaves; and (5) inducing changes in leaf anatomy. In the long term, total photosynthesis per plant is lowered by decreased leaf formation and expansion, necrosis and leaf fall (Kozlowski and Constantinidou, 1986a).

3. LEAF INJURY

Several forms of leaf injury in trees is believed to result from air pollutants. Injury has been assessed by different investigators as leaf necrosis (dark spots on leaves indicating death of tissue), shoot dieback, tree mortality, or destruction of forest stands. On the basis of injury, plants have simply been classified by some investigators as sensitive, intermediate, or tolerant to pollution, and divided by other investigators into many classes of pollution tolerance. Injury to leaves has been reported as extent of leaf damage (necrosis) in broad-leaved trees, amount of tip necrosis in conifers and leaf shedding.

Effects of injury have been classified as acute, chronic or nonvisible. Acute injury is relatively rare, whereas nonvisible effects of pollution occur commonly. Acute injury is severe and follows rapid absorption of enough pollutant to kill tissues. During or soon after exposure, leaf cells collapse and dark spot patterns subsequently appear. Chronic injury caused by rapid absorption of sublethal amounts of pollutants over a long time is characterized by appearance of yellow spots on leaves

4.5 Interactions of air pollutants with disease and insects

The effects of air pollutants on host-disease and host-insect interactions are rather complex. Air pollutants may decrease or increase the severity of diseases depending on whether the effect of the pollutants are exerted primarily on the pathogen or on the host (Treshow, 1975). Disease severity commonly is influenced by changes in metabolism of the host plant.

The effects of pollutants on insect injury are exerted largely on insects attacking weak or injured trees. Increased infestations of primary insect pests (those attacking vigorous trees) also have been reported in polluted areas. Such attacks have been attributed to suppression of insect predators of pests (Kozlowski et al. 1991).

5. CONCLUSION

Because of our increasing needs, we constantly modify our environment. We must therefore learn to better manage ourselves, our interactions with the environment and our use of forests and forest resources, if we want to preserve the environment for generations to come (Mauffette, 1996). Many studies worldwide have demonstrated direct and indirect effects of air pollutants on forests. However, our understanding of these effects are limited and does not allow us to create management programs (Winner 1994). The following **research goals** should become a priority in the development of such programs:

1) Monitor and map pathways of air pollutants and link them to receptor forests.

2) Clarify critical relationships between air pollutants and plants, from the cellular to the ecosystem level.

3) Construct predictive models of the reaction of plants to air pollutants to enhance our ability to predict the impact of air pollutants and future emission scenarios on forest ecosystems(Winner, 1994).

The knowledge gained by such a program would be of the utmost importance if we are to protect our forests from the negative impacts of air pollution.

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Case Study

Persistent Organic Contaminants in the Canadian Arctic: Implications for Indigenous Peoples

Authors:

Derek Muir-Freshwater Institute, Winnipeg, Canada

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1. ABSTRACT

It is now recognized that there are major contamination problems in the North American Arctic due to long range transport of pollutants from the south as well as to local factors. This paper briefly reviews the pathways of persistent pollutants and their presence in Arctic marine and freshwater biota as well as the implications for indigenous peoples. Pollutants of concern in the Arctic include PCBs, chlorinated pesticides and mercury. PCBs, toxaphene and mercury are the major contaminants in lake trout and charr muscle from lakes in the Northwest Territories,the Yukon and northern Alaska. In small-toothed arctic whales (beluga and narwhal) toxaphene is the most prominent organochlorine while in ringed seals and polar bears PCBs and chlordane-related compounds predominate. Mercury, in the form of toxic methyl mercury, is elevated in muscle and other whale tissues. Human exposure assessments based on calculations of dietary intakes have shown that, average daily intakes of chlordane, toxaphene, and occasionally PCBs from the traditional Inuit diets in Arctic Canada and Greenland may exceed the accepted tolerable daily intakes (TDIs) for these compounds. However, the TDIs are established with large safety factors and the nutritional benefits of the traditional diet of the Inuit are very high. Thus current advice to indigenous people is to continue to consume traditional foods which, apart from their nutritional value, also provide significant cultural benefits for the communities.

2. INTRODUCTION

Traditionally, the North American Arctic (defined here as northern Alaska, Arctic Canada and Greenland) has been regarded as a pristine environment without the pollution problems associated with heavily populated and industrialized temperate latitudes. It is now recognized that there are major contamination problems in the Arctic due to long range transport of pollutants from the south as well as to local factors. Interest in the presence of persistent, toxic contaminants in Arctic aquatic and terrestrial ecosystems arises from concerns that indigenous peoples depending upon "traditional food" for most of their diet (Kinloch *et al.* 1992) may be adversely affected by chronic exposure to these pollutants (Dewailly *et al.* 1989; 1993). There are also concerns that the health of top predators such as small-toothed whales and polar bears (*Ursus maritimus*) may be affected because of chronic exposure to polychlorinated biphenyls (PCBs) and other organochlorines (OCs), as well as heavy metals such as mercury and cadmium, in their diet. This case study provides a brief review of the pathways of persistent pollutants *(contribution to come from T. Bidleman*) and their presence in Arctic food chains as well as the implications for indigenous peoples .

3. PATHWAYS OF CONTAMINANTS TO THE NORTH AMERICAN ARCTIC

(this could be replaced by T. Bidleman's contribution which may be more up to date)

Winds and ocean currents combine to move pollutants emitted in temperate and sub-arctic regions of Canada and Eurasia, to the Arctic (Fig. 1). Well known examples of long range transport are the emission of sulfur dioxide from coal-fired generator plants and metal smelters and the deposition acid rain in Canadian shield lakes, and the movement of radionuclides, following the 1986

Chernobyl nuclear accident in the former Soviet Union, to Scandinavia and around the globe. The circulation of the lower troposphere in winter is dominated by high pressures over the continents and low pressures over the northern Pacific and Atlantic Oceans. The intense Siberian high pressure cell tends to force air northward into the Arctic while the high pressure ridge over North America drives air out of the Arctic and over the North Atlantic. In summer northward transport of air borne pollution decreases because continental highs and oceanic low pressure cells weaken.

Ocean currents deliver pollutants to the Arctic mainly via northward flow of the Gulf stream (Fig. 1) and to a lesser extent via Bering Strait. Water and ice exits via Fram Strait and through the Canadian archipelago. Water borne contaminants also enter Arctic ecosystems from northward flowing rivers which in Canada and Russia drain some relatively industrialized areas.

Air-borne contaminants are removed from the atmosphere by gas absorption, precipitation and dry deposition . Many chlorinated organics and mercury are present as gases even at low temperatures and are absorbed from the gas phase by water, snow and plant surfaces. Precipitation scavenging of gas and particles from the air also deposits particle-associated OCs and metals in snow and rain. Dry deposition is a third pathway of input of aerosol-bound contaminants to terrestrial and aquatic ecosystems.

In the water column, more highly lipophilic organochlorines such as DDT and PCBs (Cl_4-Cl_{10}) are associated with particles while others which are more water soluble (HCH and toxaphene) are mainly in the dissolved phase. Chemicals are accumulated by fish and invertebrates via three major route of exposure: gills, diet, and dermal. Dissolved chemicals enter the organism by diffusion across the gill membrane or other respiratory surfaces into the blood (Connell 1988). Absorption from food also involves diffusive transfer across the gut membrane. Transfer across gill, gut or internal membranes is influenced by the hydrophobicity and polarity of the molecule as well as by molecular size and shape and biodegradability. Hydrophobic organics which are not metabolized, and have molecular weight less than about 600 Daltons are highly bioaccumulative. This group includes PCBs, and organochlorine pesticides such as DDT, chlordane and toxaphene. Mercury in the form of methyl mercury is also relatively resistance to biodegradation and is accumulated by aquatic organisms.

Transfer within the food web through food ingestion is the dominant pathway for uptake of persistent organic chemicals in larger marine and freshwater organisms, and in terrestrial food webs. These pathways coupled with the slow rate of excretion and metabolism, lead to biomagnification (the successive increase in concentration of a chemical with increasing trophic levels). Most Arctic food chains are neither longer nor shorter than they are in temperate regions. Species that are first level carnivores are common in both climes, but the Arctic is distinguished by having major populations of marine mammals such as ringed seals, beluga and narwhal, and by a true third-level carnivore, the polar bear, which feeds on ringed seals. Food traditionally consumed by humans in the Arctic is often higher in the food chain than food consumed in southern latitudes.

4. SPATIAL TRENDS OF ORGANOCHLORINE CONTAMINANTS IN ARCTIC FRESHWATER FISHES

Salvelinus species (lake trout and arctic charr) are among the most widely distributed species of fishes in Canada and in the Canadian Arctic. Fig. 2 presents concentrations of PCBs and toxaphene, the two major organochlorine contaminants in lake trout and charr muscle for lakes in the Northwest Territories, the Yukon and northern Alaska. Wide variation in toxaphene and PCB concentrations is evident. Even in lakes of similar size at the same latitude (Peter, Belot and Colville; Travaillant, Raddi, Fish) there were differences in concentrations of toxaphene and to a lesser extent of PCBs. This variation is due in part to fish age and weight which are significantly correlated with toxaphene levels in lake trout at several locations. Older large lake trout are mainly fish-eating and accumulate elevated levels via biomagnification.

Highest concentrations observed so far are found in lake trout from Lake Laberge in the Yukon. These elevated levels resulted in a health advisory from Health Canada and the closure of an important commercial, sport and native subsistence fishery in 1991. Levels of toxaphene and other OCs such as DDT and PCBs in Laberge fishes were up to 125 times higher than those found in the same species from other regional lakes (Kidd *et al.* 1995). The fish community structure in Lake Laberge is known to be different from other lakes with few lake trout and lake whitefish, and many burbot and longnose sucker. The lake trout are also high in lipid and are solely piscivorous unlike trout populations from other regional lakes. Concentrations of toxaphene, PCBs and total DDT (ng. g^{-1} wet wt.) were found to correlate significantly with the ^{15}N of organisms through the food webs of Laberge, Fox and Kusawa Lakes (Kidd *et al.* 1995). The heavier isotope of nitrogen (^{15}N) is enriched 3 to 5 parts per thousand in an organism compared to its diet (Peterson and Fry, 1987), and is used as continuous measure of trophic level. The top predators in Laberge feed at a higher trophic level than those from the reference lakes, a factor which has been previously implicated in higher contaminant concentrations in lake trout (Rasmussen *et al.* 1990). Rasmussen *et al.* (1990) found that lake trout from lakes in Ontario with longer food webs had higher concentrations of persistent contaminants. This trophic difference between lakes provides an explanation for the higher concentrations of toxaphene, DDT and PCBs in the fishes from Lake Laberge. These results suggest that the variation in concentrations of organochlorines in lake trout and charr from other Arctic and subarctic lakes (Fig. 2) may also be due to the extent of feeding on fish rather than invertebrates i.e. the trophic level of the fish.

Mercury levels are high in lake trout throughout NWT and northern Québec, i.e. levels frequently exceed 0.5 $\mu g.g^{-1}$, the Canadian and FDA? guideline limit for commercial fish and 0.2 $\mu g.g^{-1}$ the guideline for subsistence fish consumption. By contrast uniformly low levels of mercury are found in sea run Arctic charr.

5. SPATIAL TRENDS OF ORGANOCHLORINES IN MARINE MAMMALS

Ringed seals, beluga and polar bears are found throughout the North American Arctic marine environment. These animals are top predators, their population distributions are well documented, and samples of fat and other tissues are available as a result of the Inuit hunt. Therefore, all three species are useful as indicators of spatial trends in contamination of the Arctic marine food web. Analysis can also provide information on levels of contaminants in traditional foods consumed by the Inuit (such as beluga muktuk and blubber)for use in human health assessments. The most extensive measurements of OCs especially of PCBs and DDT-related compounds have been on blubber samples from beluga whales and ringed seals (Fig. 3) as well as adipose tissue from polar bears (Norstrom *et al.* 1997). In small-toothed Arctic whales (beluga and narwhal) toxaphene is the most prominent OC while in ringed seals and polar bears PCBs and chlordane-related compounds predominate. Seals and polar bear appear to have the capability to metabolize toxaphene. Polar bear have the highest concentrations of PCBs of any Arctic marine animal due to their diet which consists almost exclusively of seals.

Few statistically significant differences in the mean concentrations of the major OCs have been found in the five beluga stocks sampled in the period 1983-89 (Muir *et al.* 1990) and again in 1992-95 (Fig. 3). Toxaphene levels showed no geographical trends however PCBs and DDT were significantly higher ($p < 0.05$) in male beluga from Cumberland Sound than in samples from Hudson Bay or the Beaufort Sea. No significant differences were found among mean levels in females within the five stocks. These levels are about 10 to 25-times lower (in males) than in blubber of dead beluga from the St. Lawrence estuary (Muir *et al.* 1990).

Measurements of metal contaminants in tissues of Arctic marine mammals collected in the 1970's and ✇80's indicated that mercury, cadmium, and lead were relatively high in some marine mammals but information on spatial trends was limited (Muir *et al.* 1992). Recent work has shown that ringed seals in the western Arctic had higher concentrations of total mercury in the liver than those in the eastern and central Arctic and a lower concentration of cadmium in the liver and kidney, than those in the eastern Arctic and Hudson Bay (Figure 4). Most mercury in the liver was inorganic mercury, and was positively correlated with selenium. The presence of selenium is regarded as providing a protective effect against mercury poisoning through the formation of mercury selenides. Mercury concentration in beluga from the western Arctic averaged (1.33 $\mu g.g^{-1}$ wet wt) in muscle and 0.77 $\mu g.g^{-1}$ wet wt in muktuk (skin). These levels are higher than the Health Canada guideline for mercury in fish muscle (0.5 $\mu g.g^{-1}$ wet wt). Approximately, 100 % of the total mercury in beluga muscle was in the form of toxic methylmercury (1.32 $\mu g.g^{-1}$ wet wt).

6. IMPLICATIONS FOR INDIGENOUS PEOPLES

Given the widespread presence of persistent OCs and mercury in fish and marine mammals in the Arctic it is not surprising that the extent of contamination of the traditional foods of the Inuit, and to a lesser extent the Dene people of inland NWT and Yukon, has been the subject of a number of studies. Women and their infants are generally considered at special risk for exposure to PCBs because of the transfer of these contaminants via the placenta and through breast milk (Skaare *et al.* 1988). Adverse effects on infants, such as shortened gestation, reduced head circumference and birth weight, compromised neuromuscular development and later visual recognition memory have been report in studies of children born to consumers of significant amounts of Great Lakes fish (Rogan *et al.* 1985; Jacobson *et al.* 1990). In a study of the community of Broughton Island (NWT), Kinloch *et al.* (1992) found that PCB levels in blood exceeded tolerable levels in 63% of females and males under 15 years of age and in 39% of females aged 15 to 44. Blood mercury levels also exceeded at risk levels in 3 of 203 individuals. Estimated dietary intakes of 12% of community residents exceeded tolerable daily intakes (TDIs) for PCBs and toxaphene set by Health Canada. On the benefits side, however, the major foods which also contributed contaminants such as narwhal, walrus, ringed seal, caribou and char, were excellent sources of nutrients including omega-3- fatty acids, retinol, iron, zinc, copper, calcium and phosphorus.

A study of the Dene people in a Mackenzie River valley community found much lower exposure to PCBs and other OC contaminants than for the Inuit, because of a traditional diet consisting mainly of freshwater fish, plants and caribou (Kuhnlein *et al.* 1995). However, average daily intakes of chlordane and toxaphene (but not PCBs) in the Dene community exceeded TDIs set by Health Canada. The nutritional benefits of the traditional diet of the Dene were also very high. There are also significant cultural benefits to the use of traditional foods because of the involvement of the family and the community in hunting, and preparation of foods and garments as well as because of physical fitness (Kuhnlein *et al.* 1995).

The presence of contaminants in traditional diets thus poses a dilemma for regulators and for indigenous people. Average daily or weekly intakes of several organochlorine contaminants, particularly toxaphene and chlordane, exceed TDIs. However these TDIs are established by applying large safety factors - 1000 in the case of chlordane - to results for the no observed adverse effect level (NOAEL) in laboratory animal studies. Exposures generally do not approach levels for NOAELs. A side effect of the investigation of contaminants in the diet has been increased knowledge of the nutritional benefits of traditional diets.

There has been international action on the issue of contaminants in the Arctic. The eight circumpolar countries agreed in June 1991 to develop an Arctic Environmental Protection Strategy (AEPS). One of the activities under AEPS is the Arctic Monitoring and Assessment Program (AMAP) which has the objective of throughly measuring levels of anthropogenic pollutants and assessing their effects on the Arctic environment including humans. Indigenous peoples organizations from the circumpolar countries are participating in AEPS and AMAP. A ministerial report on the state of the Arctic environment is being prepared by AMAP for release 1997.

At the regulatory level there has been action by the United Nations Economic Commission for Europe to develop protocols for elimination of certain persistent organic pollutants and for reduction in emissions of heavy metals including mercury. Nevertheless, the problem of contaminants in the traditional foods of Arctic peoples will not disappear quickly because of the persistence of many of the pollutants in the environment. There is evidence that levels of PCBs and OC pesticides were higher during the 1970's than they are at present. Mercury concentrations

OC pesticides were higher during the 1970's than they are at present. Mercury concentrations appear to be increasing in marine mammals over this 20 year period. Yet little is know about the long term biological consequences for human populations from this relatively elevated exposure compared to most residents of temporal climates. Continued effort is needed to monitor temporal trends and to study possible effects on aboriginal peoples and on the wildlife populations which they depend for traditional food.

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To be added:

Figures 1 - 5

Captions for Figures:

Fig. 1. Pathways of transport of pollutants from temperate to the North American arctic via the atmosphere and ocean. *(provided by T. Bidleman?)*

Fig. 2. Pathways of transport and accumulation of persistent organics and metals to Arctic terrestrial, freshwater and marine/estuarine ecosystems.

Fig. 3. PCB and toxaphene levels (ng/g wet wt) in muscle of arctic char and lake trout from northern Alaska, Arctic Canada and western Greenland. Shown as bars on a map.

Fig. 4. ΣPCB and ΣDDT concentrations in ringed seal blubber from northern Alaska, Arctic Canada and western Greenland. Shown as bars on a map.

Fig. 5. Mercury and cadmium concentrations in liver of ringed seals from northern Alaska, Arctic Canada and western Greenland. Shown as bars on a map.

Case Study

The Water Perspective

Author:

Marisa Mazari-Hiriart

Instituto de Ecología, Universidad Nacional Autónoma de México, México

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1. LOCAL BINATIONAL CASES

1.1 New River, Baja California-California

The New River originates from agricultural runoff south of Mexicali, Mexico, passes through Mexicali acquiring sewage and industrial waste, crosses the Mexico-US border, flows through the Imperial Valley, California, to a final destination at the Salton Sea, a saline lake in Imperial County. The New River flows into the southern most region of the Salton Sea where there is a National Wildlife Refuge.

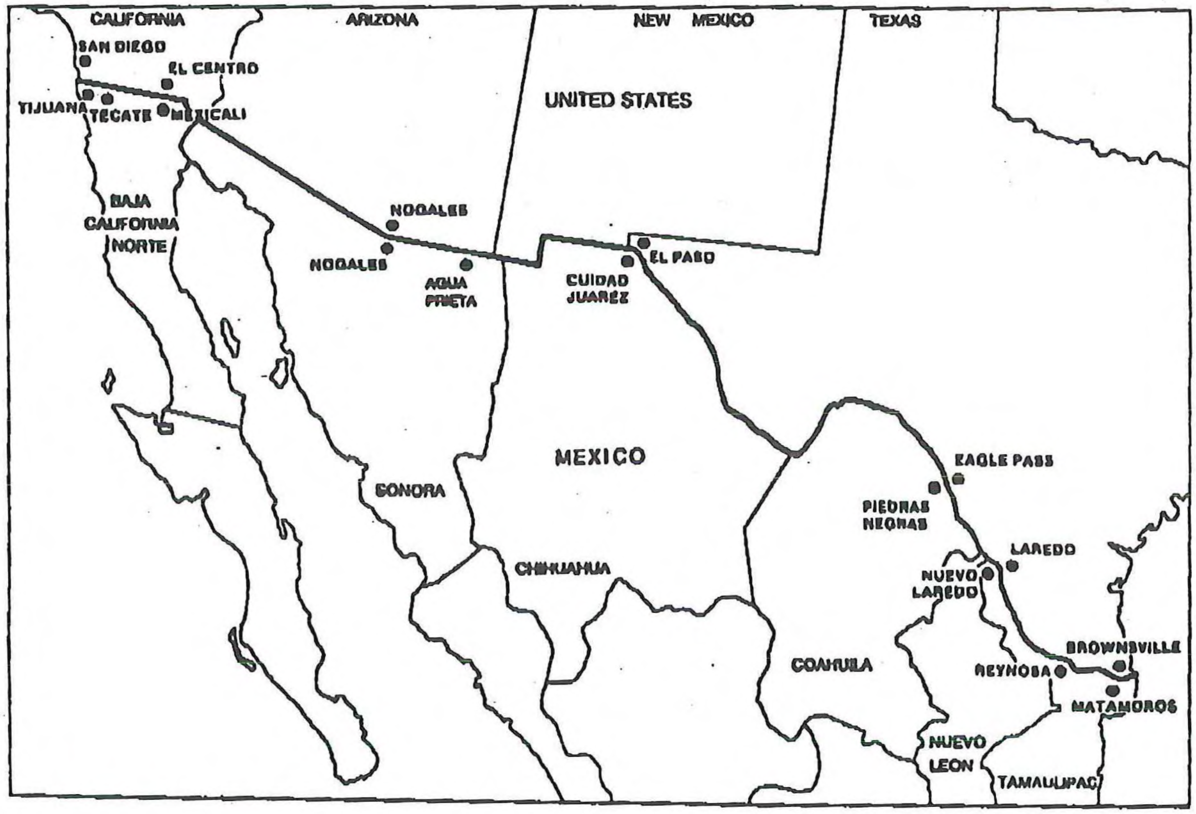
Volatile Organic Compounds (VOCs) known to be used in the printed circuit board assembly process (National Institute for Occupational Safety and Health, 1985. Hazard assessment of the electronic component manufacturing industry. Atlanta, Georgia) were matched with VOCs detectable by EPA analytical methods. Composite samples were composed of hourly samples collected during either a 10 or 24 hour basis. Eighty one percent (13) of the 16 volatiles known to be used in the printed circuit board industry were detected repeatedly in water samples collected in the New River at the international boundary (Table 1/ Figure 1). Many of the detected VOCs also have been measured in water samples taken up-river in Mexico at locations closer to the industrial sources. Overall, 90 different VOCs have been detected in the New River, 30% of them on three or more occasions from a combination of 26 composite and grab samples collected from 1982 to 1987.

Table 1. **Volatile organic compounds (VOCs) used in the printed circuit board assembly process (NIOSH, 1985) and detected by US Environmental Protection Agency methods in composite samples collected in the New River at the Mexico-US border (California Regional Water Quality Control Board, 1987. Analytical results for VOCs for water samples collected in the New and Alamo Rivers).**

		New River water samples frequency of detection				
VOCs printed circuit board assembly	Detection limit (µg/liter)	1982 N = 1	1983 N = 2	1984 N = 3	1987 N = 8	Total
Acetone	0.5	x	xx	xxx	xxx	9
Benzene	0.5	x	x	xx		4
Chloroform	0.5	x	xx	xx	xxxxxxxx	13
Chloromethane	0.5					0
Dichlorobenzene	0.5	x	xx			3
Ethyl benzene	0.5	x	xx			3
Ethyl xylene	0.5	x	x			2
Hexane	0.5	x	x	xx		4
2-Methoxyethyl acetate	0.5					0
Methylene chloride	1.0		x	xx	x	4
Methylene isobutyl ketone	1.0					0
Tetrachloroethylene	0.5				xx	2
1,1,1-Trichloroethane	0.5	x	xx	xxx	xxxxx	11
Trichloroethylene	0.5			x	x	2
Toluene	0.9	x	xx	xxx	xxx	9
Xylene	0.5	x	xx	x	xxx	7

[a]Detection limit for each compound is listed. N = number of samples.

Figure 1 . Map of the Mexico - US border (Perry et.al., 1990)



The detected VOCs have a diverse range of known adverse health effects, which vary from being carcinogenic (i.e. benzene and tetrachloroethylene), teratogenic (i.e. chloroform and ethyl benzene), mutagenic (i.e. hexane and dichlorobenzene) to causing respiratory tract damage (i.e. methylene chloride and hexane) and hepatic damage (i.e. acetone and chloroform) (Verschueren, 1983; Sitting, 1985; Sax. and Lewis, 1989). Although the New River is not utilized for drinking water, people in the Imperial Valley fish at various locations along the river.

The growth and diversification of the industry in the last decades has resulted in a substantial increase in the use of hazardous chemicals and, consequently, generation of hazardous waste.

This represents an example of multinational corporations, located in Mexican territory, that contribute to the flux of both raw hazardous materials and hazardous wastes used and generated by industrial processes. The difference between material and waste is very important as part of shipment of the finished products to the place of origin, there is generally the doubt of who is responsible for the waste generated and its adequate final disposal, and the procedure to ship it back when appropriate.

The actual risk to human health or to natural resources from the hazardous waste generated by the industry cannot be assessed until one can characterize the types and quantities of chemicals used and waste generated by each industry and track the disposition of these wastes on both sides of the border. The example of VOCs discharged into the New River likely represents a small fraction of the chemical and thus only a limited view of the potential for risk to human health and the environment (Perry et al. 1990).

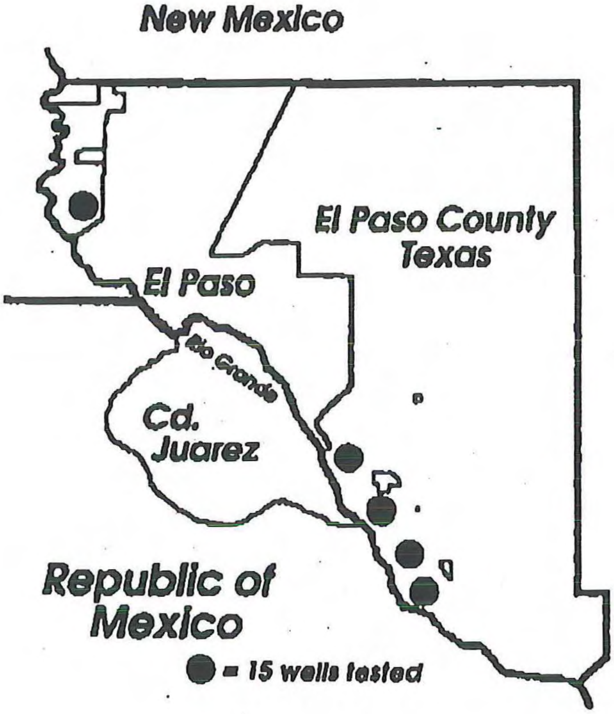
1.2 Rio Grande Valley, Ciudad Juárez-El Paso

Numerous population center in both the United States and Mexico rely on groundwater as a source of potable water supplies. Rapidly increasing population and industrialization along the US-Mexico border has resulted in large numbers of people on both sides of the border, living in poor conditions.

The Ciudad Juárez-El Paso region is one of the most heavily populated areas on the Mexico-US border, with a combined population of nearly 2 million people. Urban growth along the border has been primarily due to development of manufacturing plants known as *maquiladoras*. Many workers in the *maquiladoras* live in underdevelopment communities referred as *colonias*, lacking protected drinking water or wastewater treatment facilities. Of the several hundred *colonias* in El Paso, County, Texas, less than 20% have any form of sewer systems or wastewater treatment facilities.

The study was carried out in the Rio Grande Valley in El Paso County, an agricultural and industrial zone located in a region with a water table at only 2 to 3 meters below the surface for most of the year (Figure 2). The shallow transboundary groundwater reservoirs are suspected to be contaminated by agricultural runoff, raw sewage, and possibly industrial effluents.

Figure 2. Location of sampling sites in El Paso County (Mroz & Pillai, 1994)



At present there is no comprehensive database of safe and unsafe drinking water sources at the Juárez-El Paso border region. More significantly, the existing data on the quality of the groundwater in the region border is incomplete. The North American Free trade Agreement (NAFTA) will promote increasing industrialization and the concomitant urbanization and population growth in this region. This will have a direct impact on environmental quality at the Mexico-US border. There is an ongoing project to assess the extent of microbial contamination of groundwater in the Upper Rio Grande in El Paso County, Texas (Mroz, and. Pillai, 1994), but there is not a complete assessment on the situation in the border region.

As part of this study groundwater samples were collected from 73 wells in *colonias* between July and August 1992. Samples from 47 wells (64%) were contaminated and contained aerobic heterotrophic bacterial populations. One bacteria genus was found in 37 wells, two different bacterial genera was found in 37 wells, two different bacterial genera were found in 9 wells, and three genera were found in 1 well. A total of 58 isolates were obtained from these wells (Table 2).

Table 2. **Isolates identified from groundwater samples based on API (Analytical Profile Index) results in El Paso County, Texas** (Mroz y Pillai, 1994).

Isolate Identification	No. of Isolates
Acinetobacter species	3
Aeromonas hydrophila	4
Bacillus revens	4
Bacillus species	2
Escherichia coli	11
Enterobacter cloarne	1
Enterococcus species	0
Klebsiella pneumoniae	1
Unidentified oxidase-negative gram-positive rods	1
Unidentified oxidase-negative gram-negative rods	
Unidentified oxidase-positive gram-negative rods	11
Pseudomonas aeroginosa	14
Pseudomonas stutzeri	4
Serratia species	1

The results of this study indicate some problems with the routine water testing methods for microbial water quality, resulting in false-positive results or false-negative results. On the basis of the results, 18% of the 73 wells would be considered to be unsuitable for human use because of fecal coliform contamination. However, the selective isolation method showed that other potentially pathogenic bacteria were present in 34 wells.

All of the wells tested in the study were relatively shallow, close to the Rio Grande, and located amidst agricultural lands extensively irrigated by river water. The International Boundary and Water Commission reports that the Rio Grande at El Paso contains, on average, 250 fecal coliform per 100 mL year round. The characteristic fine soil texture along the Rio Grande could aid in the downward movement of bacteria and viruses movement through unsaturated soil zones. The actual mode of contamination , movement and survival of bacterial pathogens in the soils in this region is practically unknown and is currently under investigation (Mroz and. Pillai, 1994).

2. LONG DISTANCE CASE

2.1 Agricultural Area Mexicali Valley, Baja California - Imperial Valley, California

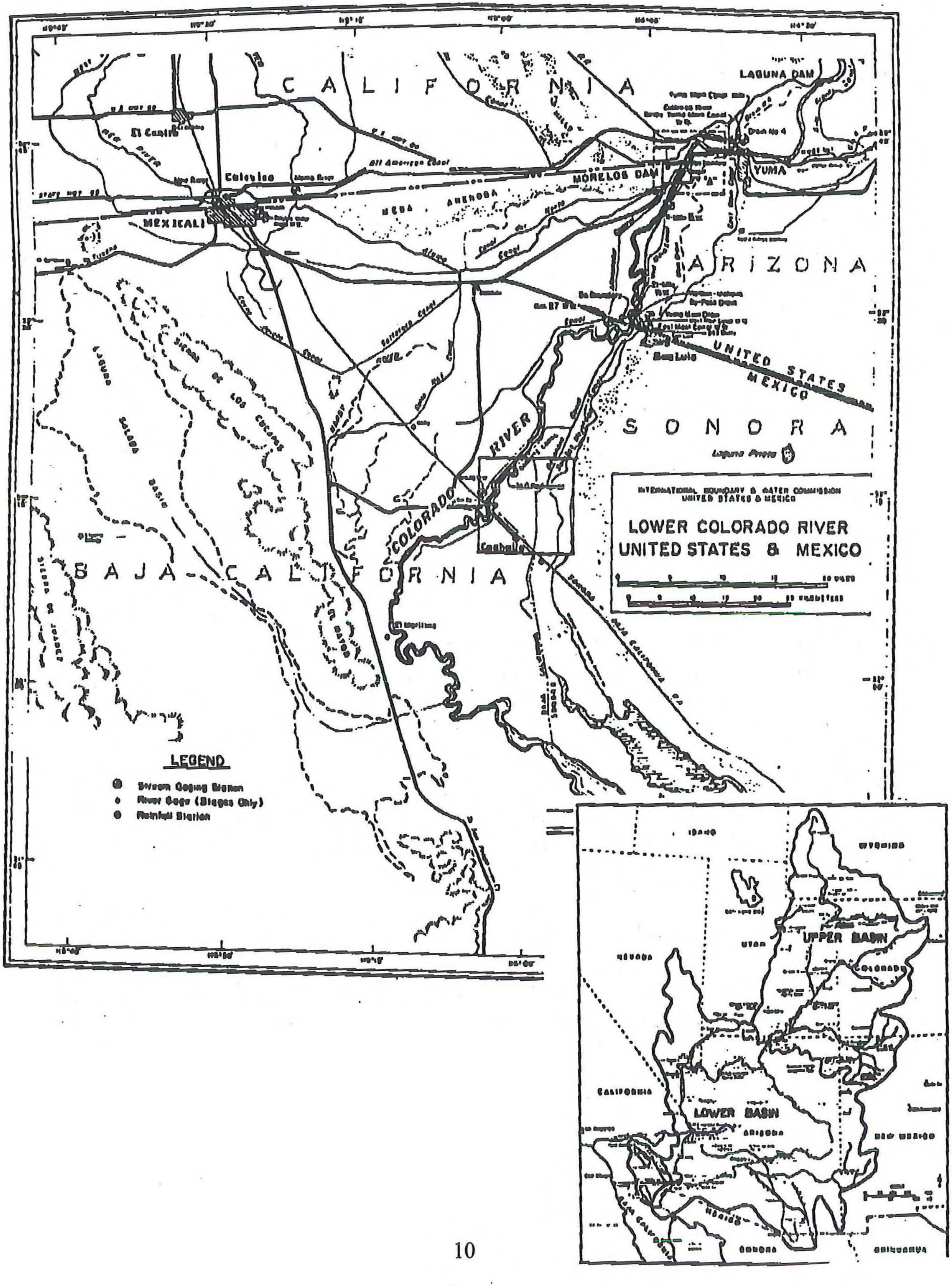
A larger scale case is represented by the Colorado River Basin, with an important agricultural area, the Mexicali-Imperial Valley locate in the lower part of the basin (Figure 3). It is a productive agricultural region due to the management of the waters from the Colorado River. Also, a dynamic region due to modern technology available for the agricultural activities.

The population in the eighties was reported as more than 800,000, and although projections are not available, this is expected to increase to more than 1'000,000 by the year 2000 (Skogerboe, 1982).

The region lies primarily on the western side of the Colorado River, which drains 242,00 square miles in the southwestern United States and 2,000 square miles in northwestern Mexico (U.S. Department of the Interior, 1983). The entire basin includes parts of seven states in the US (Wyoming, Colorado, Utah, Nevada, New Mexico, Arizona and California), and two states in Mexico (Baja California and Sonora), flowing to the Gulf of California or Sea of Cortez.

The Lower Colorado River Basin probably comes closer to using the last drop of available waters for human needs than any other area. These waters are also among the most regulated and metered. The irrigated agricultural area is virtually flat with an average altitude of about 5 m above sea level, ranging from 43 m above sea level where the Colorado enters the region to 2 m below sea level 10 km west of Mexicali on the border. The alluvial soils are principally clay, in about 30% of the area with sandy soils that help soil drainage.

Figure 3. **The Colorado River Basin** (Weatherford & Brown, 1986)



Case Study

The International Joint Commission and its Role in Transboundary Air Quality Issues

Author:

Andrew L. Hamilton

Secretariat, Commission for Environmental Cooperation, Montréal, Canada

Reviewers:

Gary Foley-US Environmental Protection Agency, Research Triangle Park, USA

James W. S. Young, Senes Consultants Limited- Richmond Hill, Canada

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This provision and the application of Article IX of the Treaty have combined to enable the Commission to play extremely important roles, on behalf of the Parties, in addressing water and air issues as well as in furthering a cooperative ecosystem approach to managing shared ecosystems. Article IX of the Treaty provides that:

> *"The High Contracting Parties further Agree that any other questions or matters of difference arising between them involving the rights, obligations, or interests of either in relation to the other or to the inhabitants of the other, along the common frontier between the United States and the Dominion of Canada, shall be referred from time to time to the International Joint Commission for examination and report, whenever either the Government of the United States or the Government of the Dominion of Canada shall request that such questions or matters of difference be so referred."*

Implementation of Commission recommendations made under such references is at the discretion of the two Governments. This article has been used extensively by the two governments. In practice the references have been joint references from both governments rather than from one government.

The use of joint references by governments is illustrative of the way governments have chosen to limit the range of options provided by the Treaty. They have cooperated to avoid adversarial approaches - especially adversarial approaches that would place the Commission in a position where it would be obliged to operate in situations where at least one Party would likely be hostile to the Commission. The joint use of Article IX references is fully consistent with the way some of the other provisions of the Treaty have been implemented. For example the governments have never used Article X of the Treaty which is a provision which would, if the Parties so agree, call upon the Commission to play an arbitration role in resolving differences between the two governments.

The question of reciprocal access to the other countries courts was contemplated - within limits - under Article II of the Treaty. The Parties agreed that parties injured as a result of interference with, or diversion of, waters from natural channels which would flow across the boundary or into boundary waters are entitled to the same legal remedies as if such injury took place in the country where such diversion or interference occurs. Again this provision has not been used.

While there is no obligation on the part of governments to implement the Commission's recommendations made in response to Article IX references, the negotiation of joint references and the approaches that have been developed to *"examine and report"* have resulted in the implementation of a very high proportion of the Commission's recommendations. An important feature of the mode of operation of the Commission is that the Commissioners act as a single body seeking common solutions rather than as separate national delegates. In an analogous fashion the engineers, scientists and others appointed by the Commission to serve on the Control Boards, Advisory Boards and other bodies that it establishes to help it address Article IX references are expected to serve in a personal and professional capacity and not as representatives of their agencies and jurisdictions.

Historically such appointees, including those appointed from government agencies, have taken the responsibility to serve in a personal and professional capacity very seriously which has, in the opinion of most observers, been an important factor contributing to the Commission's long history of success. Another important feature that has facilitated a spirit of cooperation is that the membership of Boards, Committees and other subgroups established by the Commission almost invariably consist of an equal number of members from each country.

3. THE GREAT LAKES WATER QUALITY AGREEMENTS OF 1972, 1978 AND THE REVISED GREAT LAKES WATER QUALITY AGREEMENT OF 1978 AS AMENDED BY PROTOCOL IN 1987

The Great Lakes Water Quality Agreement of 1972 grew out of an important joint reference to the Commission on the state of the lower Great Lakes. The 1972 Agreement, which focused primarily on eutrophication and the need to reduce pollution from municipal sewage treatment plants also included two very important references. These references, one on pollution from land use activities and one on water quality in the upper Great Lakes combined to demonstrate the importance of toxic substances and need for an ecosystem approach. Both references also clearly documented the importance of atmospheric pathways as a source of pollutants to the Great lakes.

The purpose of the Great Lakes Water Quality Agreement of 1978 as expressed in Article II of that Agreement is stated as:

> *"The purpose of the Parties is to restore and maintain the chemical, physical, and biological integrity of the waters of the Great lakes Basin Ecosystem. In order to achieve this purpose, the Parties agree to make a maximum effort to develop programs, practices and technology necessary for a better understanding of the Great Lakes Basin Ecosystem and to eliminate or reduce to the maximum extent practicable the discharge of pollutants into the Great Lakes System."*

The Article also states amongst other things that:

> *"Consistent with the provisions of this Agreement, it is the policy of the Parties that: (a) The discharge of toxic substances in toxic amounts be prohibited and the discharge of any or all persistent toxic substances be virtually eliminated".*

Persistent Toxic Substances are addressed in detail in Annex 12 of that Agreement and under the "General Principles" of that annex the Parties agreed that:

> *"The philosophy adopted for control of inputs of persistent toxic substances shall be zero discharge".*

The concluding paragraph of the preamble to the Agreement spells out the basic rational and concludes with a statement that is central to the workings of the Agreement:

> *"Concluding that the best means to preserve the aquatic ecosystem and achieve improved water quality throughout the Great Lakes System is by adopting common objectives, developing and implementing cooperative programs and other measures, and assigning special responsibilities and functions to the International Joint Commission."*

The Parties further agreed under Article V(1) that:

> *"Water Quality Standards and other regulatory requirements shall be consistent with the achievement of the General and Specific Objectives. The Parties shall use their best efforts to ensure that water quality standards and other regulatory requirements of the State and Provincial Governments shall similarly be consistent with the achievement of these objectives."*

The 1978 Agreement was amended by protocol in 1987. The amendments aim to strengthen the programs, practices and technology described in the 1978 Agreement and to increase accountability for the implementation of specific programs. The Parties meet biennially to discuss progress and report periodically to the Commission. New annexes were added which address airborne toxic substances, contaminated sediments, groundwater and non point sources of pollution. Annexes also incorporate the development and implementation of Regional Action Plans for Areas of Concern and Lakewide Management Plans to control critical pollutants. From an air quality perspective Annex 15, on airborne toxic substances, is particularly relevant and provides an additional focus for the Commission's work on air quality.

The Powers, Responsibilities and Functions of the International Joint Commission are addressed in Article VII of the Agreement. Those of its Great Lakes Institutions are outlined in Article VIII of the Agreement and in more detail in an appended Terms of Reference for the Great Lakes Institutions. The Agreement is, in effect , a standing reference to the Commission to assist the

Parties in the implementation of the Agreement. A major portion of the Commissions current work load is related to the Agreement. Amongst the Commission's many responsibilities is a responsibility to:

> *"Make a full report to the Parties and to the State and Provincial Governments no less frequently than biennially concerning progress toward the achievement of the General and Specific objectives including, as appropriate, matters related to Annexes to this Agreement."*

The Great Lakes Water Quality Board, the Great Lakes Science Advisory Board and the Great Lakes Regional Office are the "Great Lakes Institutions" established to assist the International Joint Commission in the exercise of the powers and responsibilities assigned to it in the Agreement. These three Institutions as well as the International Air Quality Advisory Board, established under a 1966 air reference, and the Council of Great Lakes Research Managers, established by the Commission under the Great lakes water Quality Agreement, have all played active roles in addressing airborne pollutants as these pollutants relate to the Great Lakes Basin Ecosystem.;

4. THE AIR REFERENCES

Under the 1909 Boundary Waters Treaty, the Commission, in response to specific requests from Governments, has addressed matters related to transboundary air pollution since 1928. It was then that the first air quality reference dealing with emissions from a smelter at Trail, British Columbia was received by the Commission. The significance of the Trail smelter case extended well beyond the two countries and provided an important international precedent for addressing transboundary air pollution issues. Several other references related to air pollution, notably in the Detroit-Windsor area, have since been received by the Commission.

A principle source of the Commission's authority with respect to air quality matters is derived from a 1966 reference from governments. The focus of the reference was on air pollution in the Detroit-Windsor and Port Huron-Sarnia areas, but the reference also contained a paragraph which reads as follows:

> *"The Commission is also requested to take note of air pollution problems in boundary areas other than those referred to in Question 1 which may come to its attention from any source. If at any time the Commission considers it appropriate to do so, the Commission is invited to draw such problems to the attention of both Governments."*

This paragraph has been used as the basis of a continuing advisory role on transboundary air quality even though the investigation of the Detroit-Windsor and Port Huron-Sarnia areas was completed and reported on in 1972. It has generally been considered that the Commission's continuing responsibility under this reference is limited to alerting Governments to transboundary issues or concerns, as opposed to undertaking investigations or studies.

The International Air Quality Advisory Board was established to assist the Commission in this alerting role and this Board, particularly in the last decade, has been very active in bringing important air quality issues to the attention of the Commission. The Board currently includes five members from Canada and five from the United States who have expertise in various aspects of air pollution sources, effects and control. They are appointed by the Commission and serve as advisors in their personal and professional capacities. The role of the Board is entirely advisory in nature.

In 1975 the Commission was further requested, in a separate reference, to continue to examine into and report on air quality in the Detroit-Windsor and Port Huron-Sarnia areas and on measures being undertaken for its improvement. In 1984, the Commission submitted its *"final report"* notifying the Governments of *the "effective completion of the Reference"* and reminding them of emerging problems *related "to a wider range of air pollutants particularly toxic and hazardous substances."*

In 1988, the Governments requested the Commission to re-commence work under the 1975 Reference and to examine and report upon the actual and potential hazards posed to human health and the environment from airborne emissions in the Detroit-Windsor area.

The Commission reported to Governments under this Reference in February 1992. In its report it made a number of recommendations and indicated that it would undertake further studies and report routinely to the Governments. This Reference allows the Commission to undertake a broad range of activities related to the environmental and human health effects of air pollutants. The region is one of the most heavily impacted by transboundary pollution, including ozone, particulates and toxic chemicals. It is geographically located within the Great Lakes basin, thus allowing the Commission to link specific reference activities with the requirements of the Great Lakes Water Quality Agreement.

The Revised 1978 Great Lakes Water Quality Agreement (GLWQA) provides the Commission with a reference, to examine, report and advise on, among other things, the Governments' commitments under Annex 15 with regard to airborne toxic substances. Under that Annex, the Parties, in cooperation with the State and Provincial Governments, shall conduct research, surveillance and monitoring, and implement pollution control measures for the purpose of reducing atmospheric deposition of toxic substances, particularly persistent toxic substances, to the Great Lakes Basin Ecosystem. These control measures are to include those which control or eliminate sources of emissions of persistent toxic substances. The Parties are required by the Agreement to report their progress in implementing this Annex to the Commission biennially. In practice the Governments have not lived up to the Annex 15 commitments in recent years.

The responsibilities assigned to the Commission under the Great Lakes Water Quality Agreement enable the Commission to pursue a broad range of activities in relation to atmospheric transport and deposition, and in particular the impact of airborne pollutants on the Great Lakes. The Commission's assessment and evaluation function enables it to request specific information, to review programs and initiatives, and to make recommendations pertaining to atmospheric transport and deposition of air contaminants. Government commitments pertaining to airborne toxic substances are laid out in Annex 15 of the Agreement. These provisions have allowed the International Joint Commission to make a number of significant comments and recommendations concerning the long range transport of air pollutants.

The Commission has also been given a reference under the 1991 Canada-United States Air Quality Agreement. Specifically, the Commission was asked: (a) to invite comments, including through public hearings as appropriate, on each progress report prepared by a bilateral Air Quality Committee composed of members representing each of the Parties, (b) to submit to the Parties a synthesis of the views received, and (c) to release a synthesis to the public after its submission to the Parties. This Reference is fairly restrictive with respect to the Commission's responsibilities, and has been implemented strictly as written.

5. PUBLIC INFORMATION AND PUBLIC INVOLVEMENT

The Boundary Waters Treaty requires that the Commission give all interested parties a *"convenient opportunity to be heard"* on matters under consideration. In practice this has meant that the Commission has developed a variety of means of relating to the public. Public hearings have often played an important role in the work of the Commission.

The Commission receives reports from its Boards at semi annual meetings. In most instances these reports are available to the public. As an example the International Air Quality Advisory Board provides advice to the Commission primarily through semi-annual progress reports which are based on workshops, technical analyses and published reports on the many aspects of transboundary air pollution.

The Commission invites public participation and comment when it prepares reports to Governments. In many instances, citizens, both specialists and non specialists, also serve on Commission boards and task forces. Informational materials and Commission reports are available from Commission offices. The meetings of its Boards are increasingly open to the public. For example once a year the International Air Quality Advisory Board invites public comment on its work through public meetings held in various locations along the Canada/United States boundary.

The Commission has held public hearings to assist it in meeting its responsibilities to invite public comment on progress reports prepared by the Government's Air Quality Committee under the 1991 United States-Canada Air Quality Agreement and to provide a synthesis of the comments to the Governments and the public.

Under the Great Lakes Water Quality Agreement of 1978, the Commission takes an ecosystem approach to advising the Governments on matters related to water quality. This has enabled the Commission and its Boards to consider and advise on airborne pollutants generated within the basin as well as those transported into the Great Lakes Basin Ecosystem from outside the basin. It has also provided a strong rationale for increased public involvement and for a strong public information program.

The Commission has, throughout the history of the Great lakes Agreements, typically held at least one major general meeting every one or two years as well as numerous other public meetings in the Great Lakes Basin. Many of the Commission's general meetings held under the Great Lakes Water Quality Agreement attract a great deal of public and media interest. Most observers consider that the Commission's activist public involvement stance has been a major factor in helping to create a strong and well-informed binational Great Lakes public constituency that helps maintain support for the Great Lakes Water Quality Agreement.

Case Study

Solving Air Pollution Problems in Paso del Norte

Author:

Peter M. Emerson, Christine L. Shaver and Carlos A. Rincon, Environmental Defense Fund
Carlos Angulo, Baker and McKenzie
Francisco Nunez, Director de Saneamiento de la Junta Municipal de Agua de Cd. Juarez
L. Roberto Fernandez Reyes, Fernandez, Espino y Asociados
Jesus J. Reynoso, El Paso City-County Health & Environmental District
Archie C. Clouse, Texas Natural Resource Conservation Commission
Danny L. Vickers, EDM International
Wesley Leonard, The University of Texas at El Paso
J. Clayton Heskett, Los Alamos National Laboratory
Jose Antonio Gonzalez Barreda, Confederacion Patronal de la Republica Mexicana
Oscar Ibañez, Director de Desarrollo Urbano y Ecologia de Cd. Juarez
Beatriz Vera, Physicians for Social Responsibility
Octavio Chavez, Chair Person of the Paso del Norte Air Quality Task Force

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1. INTRODUCTION

Along the U.S.-Mexico border, environmental conditions reflect the strains of heavy use and poverty. Fortunately, there is also a persistent spirit of self-reliance and many examples of community leaders and citizens from both countries working together to achieve a better future. One of the most interesting community-led efforts for environmental improvement is the development of a cooperative strategy for transboundary management of air quality in the Paso del Norte region.

After serving as an important but remote crossroads of trade for centuries, the Paso del Norte region today hosts an international metropolitan complex of about 1.8 million people -- most of whom live and work in the sister cities of Cd. Juarez, Chihuahua; El Paso, Texas; and Sunland Park, New Mexico.[1] Population and industrial activity continue to grow. Occupying a wide valley surrounded by mountains, Paso del Norteans share a single air basin in which airborne pollutants are readily transported back and forth across the international border. For many years, the air basin has recorded unhealthy levels of ozone, carbon monoxide, and particulate matter in excess of national standards in both the United States and Mexico. Exposure to these pollutants can cause adverse health effects, such as: asthma attacks, bronchitis, chest pains, headaches, shortness of breath, irritation of the eyes and lungs, reduced attention span, diminished mental skills, and greater susceptibility to communicable diseases.

Local, state and federal agencies in both countries have worked diligently to find ways to address the regional air pollution problem. Under the 1983 U.S.-Mexico Border Environment Agreement, significant progress has been made in holding binational planning meetings and in conducting several air pollution studies.[2] For many years, however, incomplete scientific information, huge economic disparities, and national sovereignty considerations thwarted the design and implementation of a transboundary air quality management program.

In late 1992, the debate over the likely environmental consequences of the North American Free Trade Agreement created a new opportunity to focus public attention on the long-standing need to reduce air pollution in the Paso del Norte region. In response to this opportunity, the Paso del Norte Air Quality Task Force -- a binational group of business leaders, regulators, scientists, environmentalists and elected officials -- was organized to work for cleaner air.[3] Through its regularly scheduled meetings (the first of which was held on May 20, 1993) and specific pollution reduction projects, the Task Force has helped the citizens of the region reach agreement on the value of cleaner air and take responsibility for achieving it.

But local cooperation could not achieve its potential so long as national air quality officials continued to define air quality goals and programs strictly on a national basis, ending at the international border. On May 7, 1996, the U.S. and Mexican governments signed a pioneering agreement empowering Paso del Norteans to develop cooperative transboundary strategies to improve air quality throughout the air basin and committing to implement those strategies through national law. As a result, private citizens living in the region now have a legally recognized mechanism to engage government regulators and to work as a binational community on joint management of the shared air basin.

This paper has four major sections. The first section describes the air pollution problem in the Paso del Norte's region. The second section presents the relevant environmental statutes, international agreements, and administrative authorities of Mexico and the United States that relate to transboundary air quality management. The third section describes the steps that were taken to create an international air quality management basin. The fourth section offers recommendations for air pollution control in the Paso del Norte region in particular, and for transboundary resource management in general.

2. THE REGIONAL AIR POLLUTION PROBLEM

The sister cities of Cd. Juarez, El Paso, and Sunland Park are located in a mountain pass formed where the Rio Grande/Rio Bravo intersects the Rocky Mountains in the Chihuahuan desert. The cities share a single air basin that has a base elevation approximately 3,700 feet above sea level. The air basin is defined by the Hueco Mountains in Texas to the east, the Juarez Mountains in Chihuahua to the south, the Franklin Mountains in Texas to the west, and tablelands that gradually rise to the Organ Mountains in New Mexico to the north.[4] Wind rose data indicate that Cd. Juarez is downwind from El Paso and Sunland Park roughly 20 percent of the time, while the reverse is true roughly 15 percent of the time.[5]

In this complex mountain terrain, stable air masses and radiation inversions occur regularly during the fall and winter months. Under such conditions, cooler pollution-laden air is trapped near the ground beneath a layer of warm air and forms a "brown cloud" that can be seen in the valley. During other times of the year, hot, sunny days are quite common, creating ideal conditions for photo-chemical reactions that turn certain air contaminants into ozone pollution. Semi-arid conditions of no more than 7 or 8 inches of rainfall per year and frequent winds are responsible for the entrainment of much particulate matter.

Air quality monitoring programs have been in place in the Paso del Norte region since the 1970s. El Paso air quality exceeds several U.S. national air quality standards; different parts of the region have been designated as a serious nonattainment area for ozone, and moderate nonattainment areas for carbon monoxide and particulate matter.[6] Air quality monitoring in Cd. Juarez indicates a pollution problem at least as severe as in El Paso. It is likely that ambient concentrations in Cd.

Juarez regularly exceed Mexican air quality goals for ozone, carbon monoxide and particulate matter.[7]

Ozone is a major component of the haze, or smog, that troubles many large cities. It is not discharged directly into the atmosphere by polluters, but is formed in the air. Volatile organic compounds and nitrogen oxides that are emitted from many sources -- although, primarily from vehicles in the Paso del Norte air basin -- undergo photo-chemical reactions in the presence of sunlight causing oxygen to become ozone. In El Paso, the federal ozone standard is typically exceeded 5 to 7 days per year in the summer or early fall.[8] Statistical analysis shows that the trend in peak ozone concentration level was upward before 1989, but has decreased slightly since that time.[9]

Monitoring data for El Paso and Cd. Juarez reveal elevated levels of carbon monoxide in the fall and winter months, with the highest levels often occurring in December on days of low wind speed. From 1981 to 1990, the federal carbon monoxide standard was exceeded, on average, 12 days each year.[10] El Paso is currently the only area in Texas to exceed this standard. Data for the winter months of 1992 through 1995 show a marked downward trend in the magnitude and frequency of carbon monoxide exceedences in El Paso. However, data on the highest readings for carbon monoxide from an El Paso monitoring site for the time period, 1982 to 1993, do not indicate a downward trend.[11] Carbon monoxide measurements -- which have also been high in June and July -- are known to vary widely depending on atmospheric conditions.

High levels of inhalable particulate matter (called PM-10 to denote a particle size of 10 microns or less) are generally recorded on stagnant weather days during the fall and winter months when particles are trapped near the ground. They tend to occur in the downtown business areas of El Paso and Cd. Juarez, and are often associated with visible air pollution and the region's brown cloud. El Paso is the only area in Texas that exceeds both the federal 24-hour standard and the annual standard for particulate matter.[12] Exceedences above the 24-hour standard are regularly observed at monitoring sites on both sides of the border, although, the highest readings of particulate matter in El Paso have shown a decreasing trend in recent years.[13] Several years ago, it was determined that levels of benzene, a known carcinogen that is found in particulate matter, were 3.5 ppm in El Paso and 28.8 ppm in Juarez.[14] The American Lung Association has warned residents of both cities that they face serious health threats from respiratory disease as a result of particulate matter pollution.

Air pollution in the Paso del Norte region comes from many sources, manmade and natural, and from both sides of the border. Its accumulation, movement back-and-forth across the border, and adverse impacts are intensified by the unique topography and meteorology of the natural air basin. Using information from the emissions inventory for El Paso and Sunland Park and from studies that have been carried out in Cd. Juarez, air quality experts believe that major sources of pollutants in the Paso del Norte air basin include: motor vehicles; open-burning of trash; home-fuel consumption; fuel transport and storage; dust from highway traffic, construction materials and equipment; brick ovens and small-scale industrial sources; and fugitive solvents from painting, architectural coatings and manufacturing processes.[15] Heavy industry in the region -- a copper smelter, two refineries and a large cement plant -- also contributes pollutants.

Over the past decade, the number of sources emitting pollutants in the Paso del Norte air basin has increased rapidly as the populations of Cd. Juarez and El Paso grew 40 percent and 23 percent, respectively.[16] However, the important question of which political jurisdiction contributes the most air pollution in the air basin cannot be answered precisely. It is known that Cd. Juarez has fewer pollution controls, a much larger population, a significantly older vehicle fleet, many more miles of unpaved roads, and more open-burning than El Paso and Sunland Park. Certainly, each of these factors contributes to large amounts of air pollution originating in Cd. Juarez. On the other hand, resource utilization tends to be greater in higher income El Paso and Sunland Park which, in turn, causes large amounts of air pollution. Some environmental benefits of newer, cleaner-running vehicles and paved streets in the U.S. cities are partially, or completely, offset by the fact that El Paso and Sunland Park residents have more cars and their vehicle mileage per capita is about six times higher than in Cd. Juarez.[17]

Industrial growth in the Paso del Norte region has been driven by a boom in the number of maquiladora plants located in Cd. Juarez and northern Mexico. Serving customers in the U.S. and other markets, production processes at these plants -- especially in automotive parts and electrical parts manufacturing that use large amounts of solvents -- contribute to air pollution. They have also triggered a huge increase in industrial traffic. For example, "in 1988 about 500 trucks crossed the international boundary (in the Paso del Norte region) each day; in 1993 it was about 2,000 trucks daily...."[18] In 1995, there were 1.2 million northbound international bridge crossings by commercial vehicles in the region.[19] Heavy transport vehicles and stalled traffic at the border crossings are an important source of air pollution emissions.

Scientists agree that the air pollution problem in the Paso del Norte region is transboundary both with respect to its origin and adverse impacts on human health and the environment. To fully understand the magnitude and causes of the problem, U.S. and Mexican scientists and government officials need to complete an air quality monitoring network and an emissions inventory for the air basin. They also need to continue working together on pollution characterization studies and modeling efforts. Development of an effective transboundary management strategy to reduce air pollution will require both countries to adopt policies and implement mitigation tools that meet the special needs of the international air basin.

3. CURRENT EFFORTS TO REGULATE AIR QUALITY

Management of the environment along the U.S.-Mexico border is shaped by the domestic policies and priorities of the two countries and by their international commitments. This section describes the existing environmental statutes, international agreements, and administrative authorities of Mexico and the United States that regulate transboundary air quality in the Paso del Norte region.

3.1 Mexican Air Quality Regulation

In Mexico, the development and implementation of environmental policy is generally highly centralized, with virtually all powers falling under the federal government's jurisdiction. However, in recent years, efforts have been made to decentralize environmental policy responsibility to states and municipalities.

Article 27 of the Mexican Constitution provides the main legal framework related to natural resources. Its first paragraph deals with matters related to private property regulations and restrictions to protect the public interest and provides that the federal government will adopt the necessary measures designed to regulate land use, and to preserve and protect the ecological balance. "Ecological Balance" is defined by the General Law of Ecological Equilibrium and the Protection of the Environment (the "Environmental Law") as "the relationship of inter-dependence between the elements that compromise the environment, which makes possible the existence, transformation and development of mankind, and other living beings."[20]

Section XXIX-G of Article 73 of the Mexican Constitution states that the Mexican Congress has the power to issue laws related to environmental protection and the preservation of the ecological balance. Congress has exercised this authority in recent years, enacting a number of important laws, regulations and standards, the most important of which is the 1988 Environmental Law. Currently, the administration of President Zedillo is continuing a thorough review of the Environmental Law, with the intent of proposing a comprehensive set of amendments to that law. Although the administration has released several proposed amendments, it is difficult to predict what changes may ultimately be adopted. While the provisions on air quality addressing allocation of jurisdiction may be modified, no significant change in the general objectives of the law is expected.

The Environmental Law was the result of the first serious effort on the part of the Mexican government to regulate a number of environmental topics, such as natural protected lands, environmental impact, hazardous activities and hazardous substances, prevention and control of soil, water and air pollution. The Environmental Law also establishes administrative and criminal sanctions. The federal law also outlines which areas of environmental legislation fall within the jurisdiction of federal, state or municipal regulating agencies.

The Environmental Law provides that the federal government has jurisdiction over natural protected areas, the rendering of activities considered as highly hazardous, all matters related to hazardous materials and hazardous waste, prevention and control of water pollution when wastewater is discharged to federal bodies of water, and the prevention and control of air pollution when air emissions affect two or more states or another country. The Environment Law also has a blanket provision stating that the federal authority has jurisdiction over "those matters which due to their nature and complexity require the federal government's participation."

Since 1988, several regulations have been promulgated dealing with environmental matters, most of which derive from the Environmental Law. On November 25, 1988, the regulations in the area of prevention and control of air pollution were published. These regulations set forth that the authority in air quality issues shall be concurrently exercised by the federal, state and municipal authorities and basically repeats the provisions of the Environmental Law related to air emissions affecting two or more states or another country.

Finally, the federal government has issued a great number of environmental standards, known as "Official Mexican Standards" (*Normas Oficiales Mexicanas* or "NOMs"), including NOMs on permissible levels of air pollution. These NOMs play an important role in the actual application and enforcement of federal laws and regulations because their purpose is to establish technical and scientific guidelines to be applied in the protection of the environment. In the area of air quality, among the most important NOMs are ones setting maximum air emission levels for solid particles from fixed sources; maximum air emission levels of pollutant gases from vehicles that use gasoline as fuel; maximum allowable levels for particles, carbon monoxide, nitrogen oxide, sulfur bioxide, sulfuric acid fog, and suspended particles, as well as conditions and requirements for the operation of equipment that use direct heating through combustion, used by fixed sources that utilize liquid or gas fossil fuels.

The majority of Mexico's 31 states have also enacted their own environmental laws since 1988. Many of these laws are quite similar to their federal counterpart, and contain provisions dealing with state environmental policy, including air pollution. In the case of state environmental laws and regulations, federal NOMs are also the applicable technical standards used by state authorities in enforcing such laws and regulations. The State of Chihuahua published its Environmental Law ("Ecological Law of the State of Chihuahua") on October 26, 1991. This Law contains a specific chapter dealing with prevention and control of air emissions originating from fixed sources under state or municipal jurisdiction and private motor vehicles.

Mexico's Environmental Law and regulations provide two means for enforcing air pollution control requirements: sanctions and operating licenses. Sanctions include: plant closures (temporary or permanent; partial or full); administrative arrest; imposition of fines; and revocation or cancellation of a source's license. Criminal sanctions are also authorized for flagrant and criminal violations.

The environmental regulations include a licensing requirement and process for stationary sources. Licenses generally set maximum permissible emission levels consistent with the NOMs, but authority exists to establish source-specific limits. In addition, allowable emissions specified in an operating license may be modified if the source is located in a "critical zone", like the Northern Border Area (defined as the 100 kilometer wide strip along the border). Licenses may also be modified if a more efficient technique exists to control air pollution or if production processes change at the source. Therefore, Mexican law and regulations provide ample authority for establishing, modifying and enforcing emission limits on stationary sources.

3.2 Air Quality Regulation

In the United States, air pollution is regulated under the federal Clean Air Act, which was most recently amended in 1990. The Clean Air Act is one of the most complex environmental laws in the U.S., and there are numerous federal regulations and guidelines that go into even greater detail on many aspects of its implementation.[21] The following summary is intended only to give broad outlines; therefore, it necessarily oversimplifies details that may be important for individual sources or programs.

Since 1970, the U.S. has followed an air pollution control strategy that focuses on the universal attainment of National Ambient Air Quality Standards (NAAQS) for six common air pollutants: particulate matter; sulfur dioxide; carbon monoxide; nitrogen oxide; ozone; and lead. The NAAQS are set at the federal level according to scientific criteria in order to protect the public health with a margin of safety. They typically include standards for both short-term peak concentrations and long-term average concentrations. Geographical regions that have not yet attained the NAAQS for a pollutant are termed nonattainment areas. As noted earlier, El Paso is a nonattainment area for ozone, carbon monoxide, and particulate matter. Although many U.S. cities exceed the ozone standard, El Paso is one of only a few cities that is nonattainment for three separate pollutants. All statutory deadlines for attainment of standards have already passed or will expire at the end of 1996 except for cities with serious or severe ozone, carbon monoxide, or particulate matter problems.

Under U.S. law, a nonattainment area is subject to stringent cleanup requirements and may be penalized for failure to meet the requirements. However, a border city like El Paso cannot be penalized if it shows that it has taken all necessary measures within its jurisdiction and its failure to meet the requirements is only due to emissions outside the United States (section 179B of the Clean Air Act providing this exemption was added in 1990).

To attain acceptable air quality requires, of course, reduction of pollutant emissions into the atmosphere. The federal Act divides sources of air pollution into two broad groups: mobile sources and stationary sources. For mobile sources, the emissions standards for motor vehicles and the fuel quality requirements for motor gasoline (e.g., no lead) are set at the federal level. In addition, the federal law calls upon the states and localities that are nonattainment for ozone to adopt various "transportation control plan" measures to reduce the use of cars and trucks, with the exact measures required depending in part on the severity of the local ozone problem. Finally, ozone nonattainment areas may be required to establish federally-approved programs for vehicle emissions inspection and maintenance programs, but the exact design of such programs is left to the states and localities. Because El Paso's ozone problem is classified as "serious," its extended deadline for attaining that standard is November 15, 1999. Toward that goal, the area must achieve a 15 percent reduction in volatile organic compound emissions by the end of 1996 and a further 3 percent per year reduction through the various strategies specified in the statute.

Under the Clean Air Act, the stationary source group is subdivided into two classes. For new sources or substantially renovated sources in certain major industrial classes, the U.S. Environmental Protection Agency (EPA) has established nationwide "new source performance standards" (NSPS) that set minimum pollution control requirements based on performance of the best control technologies. State and local circumstances may, however, require additional pollution controls beyond the NSPS requirements for some sources. Requirements for control of certain listed hazardous air pollutants are also set at the national level for existing as well as new sources under a similar technology-based approach and evaluation of any residual health risks.

All existing stationary sources of air pollution and new sources in industry classes for which there are no NSPS are governed by a more complex array of controls implemented at the state level, but under federal guidelines and minimum requirements. In general, it is up to each state to develop a "state implementation plan" (SIP) that will result in attainment of all air quality standards. The federal requirements for SIPs are set out in Section 110 of the Act. The SIP will set minimum emissions control requirements for sources in many different industry classes and may set different requirements for different geographic areas depending on current air quality and the degree of improvement that is needed. The state laws and rules must meet minimum federal requirements, but the states are free to adopt laws or rules that are more stringent or more comprehensive. Thus, for example, the state of Texas regulates many small sources not covered by the federal Act, and regulates pollution problems such as odors that are also not covered under federal law.

The state clean air laws and regulations of Texas and New Mexico generally track the federal scheme, and provide detailed rules designed to implement the federal requirements at the state level. The Texas Clean Air Act contains the general legal authority for matters such as design of transportation control plans and inspection and maintenance programs for vehicle emissions control, and for the review and permitting of new sources and application of reasonably available control technology for existing sources. The Texas Natural Resource Conservation Commission (TNRCC) is given the authority for implementation of the Act's provisions through promulgation and administration of detailed administrative regulations that appear in the Texas Administrative Code. Notably, some aspects of the Texas statute and rules go beyond the federal Act's minimum requirements. These provisions, such as control of outdoor burning or requirements for paving of parking lots and roadways in El Paso, represent the state's choices about how best to attain the national ambient air quality standards. It is also notable that the Texas Clean Air Act specifically instructs the TNRCC to fully implement all the federal requirements for stationary source control in El Paso so that the state qualifies for the special exemption from sanctions for not meeting clean air deadlines, provided in section 179B of the Clean Air Act.

With respect to the process of emissions offsets and emissions banking, the Texas law and regulations provide for emission reduction credits, not only for stationary sources, but for mobile sources as well. Thus, accelerated implementation of programs such as switching El Paso's bus fleet to alternative fuels can result in emission reduction credits usable in transactions with other contributors to the ozone nonattainment problem. The Texas rules also specify the use of oxygenated fuels in El Paso in the winter season to help reduce carbon monoxide emissions, and ban the use of wood-burning stoves and similar devices during inversion episodes to reduce particulate emissions.

In New Mexico, the controlling state statute is the Air Quality Control Act, administered by the state's Department of the Environment. The state's regulations appear in the New Mexico Administrative Code as a compendium of Air Quality Control Regulations. As with Texas, the basic provisions implement the federal Act's requirements with respect to such matters as permits, review of new sources, transportation control planning in ozone nonattainment areas, and emission offsets.

For many years under Texas law, and federally-required since 1990, each air pollution source must have a permit that specifies the pollution control requirements for that particular source. The permit is the basic mechanism for identifying sources, setting limits, monitoring performance, and enforcing the requirements for the source.

In nonattainment areas, the federal Act specifies that the states shall ensure that existing sources apply at least “reasonably available control technology” (RACT) as determined for each source individually applying EPA technology guidelines. For inhalable particulates, RACT guidelines are supplemented by “reasonably available control measures” for area sources of particulates such as roads and wastewater treatment ponds. For nonattainment areas, the federal Clean Air Act also requires that SIP provisions result in “reasonable further progress” toward attainment and that they include a procedure for reviewing the air quality effects and pollution control systems on any new facility or expansion of an existing facility emitting more than minimal amounts of the nonattainment pollutants in the area. (For ozone nonattainment areas, stationary sources of volatile organic compounds are regulated.)

Three federal Clean Air Act requirements must be met before a new or expanded facility can get a permit to operate in a nonattainment area. First, the permit applicant must show that other already existing sources of the same pollutant in the area will "offset" the new emissions with even greater reductions in current emissions (i.e., more than one ton per year of reductions for each ton per year of new permitted emissions). Second, the permit applicant must adopt the most stringent pollution controls at the new or expanded facility -- referred to by the term "lowest achievable emissions rate." Third, the applicant must certify that all other facilities under its control in the state are complying with their air pollution control requirements.

To promote progress towards attainment by reduction of emissions from existing sources and to facilitate the emissions trades that may allow new or expanded industrial activity in nonattainment areas, EPA rules allow sources to "bank" their credits for emissions reductions. The source can then draw on its "bank account" for offsets to its own increases in emissions, or it can sell those credits to a third party that may need offsets. Government agencies that generate air emissions or that can show reductions of emissions through government programs can also contribute their emission reductions to private projects. For example, control of emissions from garbage incineration can reduce particulate matter, and changes in the specifications for road paving materials may reduce emissions of volatile organic compounds.

In the Paso del Norte region, the availability of emission offsets as both an opportunity for new facilities to locate in the region and as an incentive for further reductions in emissions from existing facilities has been constrained by the fact that Clean Air Act jurisdiction, and thus the opportunity to trade emissions, ends at the Rio Bravo/Rio Grande even though the air passes freely back and forth across the border. An important objective in creating the Paso del Norte Air Quality Management Basin is to allow the benefits of offsets and emission trading to be available to sources throughout the common airshed.

3.3 Binational Cooperation on Air Quality

For many decades, both the U.S. and Mexican governments have recognized that environmental problems in the border region cannot be addressed effectively by different and uncoordinated approaches on each side of the border. In 1983, after several largely unsatisfactory ad hoc arrangements dealing with specific problems, the two countries entered into an "Agreement Between the United States of America and the United Mexican States on Cooperation for the Protection and Improvement of the Environment in the Border Area," known simply as the La Paz Agreement after the city of La Paz, Baja California, in which it was signed. Air pollution was noted as one of the transborder problems that could be addressed under the agreement, which gives the two federal environmental agencies primary responsibility for environmental policy within 100 kilometers of either side of the border.

In 1989, recognizing the serious nature of the air pollution problem in the Paso del Norte region and the need for binational cooperation to address the problem, the countries added Annex V to the La Paz Agreement. Annex V seeks a better understanding of the problem through a binational inventory of emissions sources, air quality monitoring, and modeling. The studies conducted under Annex V have created a solid foundation for the next step -- a coordinated, cooperative transboundary management strategy to improve air quality in the region by reducing emissions of air pollutants. Furthermore, when Congress amended the Clean Air Act in 1990, it included specific authority (in section 815 of the amendments bill) for the executive branch to negotiate with Mexico on programs to monitor and improve air quality in the region.

Additional binational cooperation on air quality has also occurred through the U.S.-Mexico Integrated Border Environmental Plan, a broad program of cooperation between the Mexican and U.S. environmental agencies.[22] The border plan expanded the Annex V program of monitoring and modeling to include more emphasis on reducing vehicle emissions and pollution control strategy development. Recently, the governments have developed the elements of a second-generation plan called the US/Mexico Border XXI Program.[23]

Some further international support for air quality management in the Paso del Norte region may come from the assessments, policy analyses, and factual reports of the North American Commission for Environmental Cooperation. The commission was established in 1994 by the NAFTA partners to address environmental issues of common concern, including matters of transboundary pollution. Finally, another NAFTA-related institution, the Border Environment Cooperation Commission (BECC), may have a role to play in helping to develop and arrange financial support for specific environmental infrastructure projects such as new solid waste disposal facilities that could help reduce air pollution. BECC-certified projects become eligible for financing through the North American Development Bank.

3.4 Summary

Current efforts to reduce air pollution in the Paso del Norte region depend on air quality policies, standards and enforcement procedures promulgated in Mexico and the United States and on binational efforts authorized under the 1983 La Paz Agreement and other bilateral or continent-wide arrangements. Though the region's air pollution is shared by everyone living in the air basin, there are important differences between the Mexican and U.S. pollution control regimes. Statutes, regulations, and enforcement procedures in each country differ significantly because of different constitutional structures, legal traditions and stages of economic development. Furthermore, the legal jurisdiction of each country ceases at the international border. Together, these factors lead to a "governance failure" -- a gap between each government's regulations controlling air pollution emissions and the transboundary character of the polluted air.

Because of the governance failure, air pollution control in the shared Paso del Norte air basin is costly and ineffective. However, the community's interest in better air quality led local leaders to advocate a transboundary management strategy that emphasizes binational cooperation, innovation and local responsibility in achieving cleaner air. The positive working relationships that exist in the Paso del Norte region can promote evolution of existing U.S. and Mexican regulations and future economic growth to benefit the entire air basin.

4. TAKING THE NEXT STEP TO TRANSBOUNDARY AIR QUALITY MANAGEMENT

Twenty-two years ago, John C. Ross, then El Paso City Attorney, speaking at the First Binational Symposium on Air Pollution Along the United States-Mexico Border, observed that: "Air pollution by its very nature transcends the international boundary without regard to jurisdiction areas, and .... we must seek legal solutions which likewise can bridge the jurisdictional lines between our two countries."[24] At the same symposium, another U.S. attorney proposed creation of an International Environmental Air Pollution Commission[25] and a highly-regarded Mexican attorney proposed a bilateral treaty to deal with Paso del Norte air pollution, noting, however, that: "It would be necessary to protect the sovereignty of each State to insure that it had the right to enforce standards within its own boundaries."[26]

In 1989, three professors from the University of Texas at El Paso (UTEP) made the case for a locally-controlled, binational program to manage the international air basin. They were the first to point out that economic instruments, such as emissions trading, could be used in the air basin to lower the cost of air pollution clean up below that of a command-and-control system.[27] Recognizing that it was not realistic for the two countries to immediately establish uniform ambient standards for the air basin, the professors recommended that the countries take three steps: (1) implement an air quality accounting system; (2) allocate initial control responsibility based on existing standards through operating permits; and (3) develop emissions trading procedures for uniformly mixed assimilative pollutants, such as volatile organic compounds. They reasoned that such a program would insure progress by each country toward its own air quality goals, although pollution control costs would fall disproportionately on the country with the most stringent standards. To a large extent, the 1989 UTEP proposal has served as the motivating framework for the most recent efforts to achieve transboundary management of air quality in the Paso del Norte region.

The likelihood of achieving this goal has been greatly increased by the important work of the Binational Air Working Group. Created under the auspices of the U.S.-Mexico Border Environmental Agreement, the Binational Air Working Group has met annually since 1984. It is currently co-chaired by Dr. Adrian Fernandez, Instituto Nacional de Ecología, Secretaría de Medio Ambiente, Recursos Naturales y Pesca, and David Howekamp, Director of the Region 9 Air and Toxics Division, U.S. Environmental Protection Agency. This group of U.S. and Mexican scientists and air pollution control administrators, along with officials from Cd. Juarez and El Paso, have planned and carried out projects on air quality monitoring, emissions inventories, particulate matter transport, vehicle emissions, air toxics, and modeling of pollution control strategies.

4.1 Obtaining Authority for Transboundary Management

In 1992, the NAFTA debate focused attention on environmental issues along the U.S.-Mexico border, creating a new opportunity to make headway on the Paso del Norte air pollution problem. In meetings with the late Luis Donaldo Colosio, then Mexico's top environmental officer, Environmental Defense Fund (EDF) staff members discussed several ways that new transboundary institutions and economic instruments could be used to solve border region problems.[28] Mr. Colosio was interested in ideas that would help decentralize and increase the efficiency of environmental regulation in Mexico. He was especially supportive of the EDF's recommendation to create an international air quality management district in the Paso del Norte region.[29] As a result, Mr. Colosio directed Dr. Miguel Angel Orozco Deza, who then served as Delegado de la Procuraduría Federal de Protecion Ambientale in Chihuahua, to work with the EDF staff in carrying out this recommendation.

With the encouragement of Mexican and U.S. officials, the EDF staff next pursued a series of meetings in Cd. Juarez and El Paso. Important activities included a presentation to the Mayor's Environmental Roundtable in El Paso, work with air quality experts at the Direccion Municipal de Ecología and the City-County Health Environmental District, and meetings with local business leaders. In April 1993, with the support of El Paso-Cd. Juarez environmental officials and business leaders, the EDF staff appeared before the Texas Air Control Board (now reorganized into the Texas Natural Resource Conservation Commission) to recommend creation of a regional task force to help solve the Paso del Norte air pollution problem. The Board accepted the recommendation and appointed Texas members to the Task Force.

The first meeting of the Paso del Norte Air Quality Task Force was held on May 20, 1993. Chaired by Dr. Elaine Barron, an El Paso physician and a member of the Texas Air Control Board, the meeting was attended by about 15 people. Today, the Task Force meets five or six times a year, with 40 or more business leaders, regulators, scientists, environmentalists and elected officials from both countries in attendance. The Task Force has worked very closely with key government officials and both U.S. and Mexican members have asked for more local responsibility in air quality regulation. From the beginning, the effectiveness of the Task Force was enhanced by the active participation of Francisco Nuñez, Cd. Juarez's chief environmental officer, and Kirk P. Watson, chairman of the Texas Air Control Board. After creating the Task Force, Mr. Watson took the lead in publicly supporting an innovative approach to transboundary air quality management.[30]

Throughout 1993 and 1994, Task Force members worked to understand the regional air pollution problem and to gain the confidence of the EPA, Mexico's federal environmental agency and other government agencies. The Task Force also debated and prepared -- with the assistance of attorneys from EDF, Cd. Juarez, El Paso, TNRCC, Chihuahua, and the Texas General Land Office -- a proposed Annex VI to the La Paz Agreement to create an international air quality management district (IAQMD) for the Paso del Norte region (see Appendix A).

The Task Force envisioned that the IAQMD would not supersede regulatory authority of the two countries, but that it would act at the local level to conduct joint activities in data gathering, pollution prevention, technology transfer, compliance review, public education, and the development of transborder pollution control strategies including the use of economic instruments such as an emissions trading program. The Task Force also recommended a governing board drawn from the responsible government agencies and interested citizens living in the region. After preparing its proposed Annex VI, the Task Force succeeded in winning recognition and support from many elected officials at all levels of government, including Presidents Clinton and Salinas, and later, President Zedillo.

As the proposed Annex VI was being reviewed by federal authorities, the Task Force continued to meet on a regular basis and was active in community outreach concerning the region's air quality and in carrying out specific pollution reduction projects. For example, EDF and the Task Force have worked with officials in Cd. Juarez to improve the vehicle inspection and maintenance program by helping to set up vehicle emission diagnostic centers and a training program for automobile mechanics. The Task Force has also worked with government agencies on initiatives related to extending the U.S. Department of Energy's Clean Cities Program to Cd. Juarez, speeding the introduction of alternative fuel vehicles, and reducing traffic congestion on international bridge crossings. A project was also started to quantify and address paint and solvent shop emissions in Cd. Juarez, including preparation of a handbook on reducing emissions.

The work on these pollution reduction projects has benefited from a successful project aimed at helping the owners of high polluting brick kilns in Cd. Juarez adopt cleaner fuels and more energy-efficient kilns. Administered by the Mexican Federation of Private Associations for Health and Community Development, the brickmakers project has involved scientists from El Paso Natural Gas, and Los Alamos and Sandia National Laboratories. The brickmakers project is a model of effective binational cooperation.[31]

In June 1995, at a La Paz Agreement meeting in Mexico City, the two governments agreed to enter into formal negotiations regarding the establishment of an IAQMD and a joint committee on air quality improvement for the Paso del Norte air basin. In August 1995, U.S. and Mexican officials met in Washington, D.C. for their first negotiating session. During the remainder of 1995, the federal negotiators continued to exchange information among themselves and with their respective state governments. Also, a delegation of Task Force members from Cd. Juarez and the EDF staff met with environmental and foreign relations officials in Mexico City to be briefed on the negotiation process and to make specific recommendations to the negotiators.

On May 7, 1996, Secretary of State Warren Christopher and Secretary of Foreign Relations Angel Gurria signed an agreement adding a new Appendix 1 to Annex V, creating a binational committee on air quality improvement for an international air basin defined as those parts of El

Paso County, Texas, Doña Ana County, New Mexico and the metropolitan area of Cd. Juarez, Chihuahua that are within 100 km of the border (see Appendix B). The official title of the committee is the Joint Advisory Committee on Air Quality Improvement for the El Paso - Cd. Juarez - Doña Ana County Air Quality Management Basin (hereafter referred to as the "Joint Committee"). Christopher praised the agreement as a unique, cooperative approach to "enable business and government leaders from Texas, New Mexico, and Cd. Juarez to reduce some of the region's worst air pollution."[32]

The Joint Committee will develop and present recommendations to the Binational Air Working Group on the prevention and control of air pollution in the international air quality management basin (IAQMB). According to a "guidance document" prepared by the negotiators, the Joint Committee's work is expected to include recommendations on:

- the integration of air quality monitoring networks and reporting of air quality pollution indices for the IAQMB;
- the exchange of information, technical training, and technology transfer to benefit air quality;
- the development and implementation of public education and outreach programs for the IAQMB;
- the use of air quality modeling to evaluate specific pollution abatement and prevention strategies;
- the development and implementation of economic instruments including emissions trading programs (with emissions budgets, loading allowances, and/or caps) for the IAQMB; and
- specific steps to improve the effectiveness of air quality programs in the IAQMB.

Membership of the Joint Committee will consist of 20 persons, ten selected by each country. The representatives from each country will include five government officials from federal, state and municipal jurisdictions and five non-government members from businesses, civic and environmental organizations, and academia. The non-government members of the Joint Committee must reside in the jurisdiction of the IAQMB. Members will be appointed by the EPA and the Secretaría de Medio Ambiente, Recursos Naturales y Pesca and all meetings must be open to the public. Otherwise, the Joint Committee will establish its own rules of procedures and meeting schedule.

5. IMPLEMENTING THE JOINT ADVISORY COMMITTEE ON AIR QUALITY IMPROVEMENT

With the international agreement now in place, the next step toward transboundary management of air quality in the Paso del Norte region is implementation of the new Joint Advisory Committee. Implementation needs to be carried out in a way that fully involves the local community and contributes to efficient and open operating procedures. It is also important that the Joint Committee achieve early improvements in air quality and its management throughout the basin.

5.1 a. Joint Committee Procedures

Involving the Local Community. Both the U.S. and Mexico have agreed that non-government representation is very important to ensure that the Joint Committee reflects and responds to local concerns, needs, and priorities. Although, recommendations made by the Joint Committee will be forwarded to the Binational Air Working Group for a final decision and many proposed actions will require federal or state government involvement. Those who live and work in the Paso del Norte community must have a significant role in developing cost-effective solutions to the air quality problems that will be implemented cooperatively at a local level. Therefore, commitment from the business community and local governments will determine whether strategies are successfully implemented.

The Joint Committee should become a primary forum for receiving public input on air quality problems and solutions. It must also be accountable to the public and assume responsibility for disseminating information and reporting on actions taken and results achieved. In order to carry out these functions effectively, the Joint Committee will need to establish and maintain a good working relationship with the Binational Air Working Group so that there is a routine method for acquiring necessary information and holding the federal and state governments accountable for actions taken in response to the Joint Committee's recommendations. The Joint Committee will also need to interact regularly with the Paso del Norte Air Quality Task Force. The Task Force members are not only knowledgeable about local air quality problems, but they are also action-oriented and could play a significant role in tapping financial resources and skills needed to implement strategies recommended by the Joint Committee.

Operating Efficiently and Openly. In order to maintain credibility, the activities of the Joint Committee should be carried out in an open and transparent manner. Therefore, the Joint Committee's administrative procedures need to specify the frequency, time and place (preferably alternating among the affected communities) of meetings; indicate that all meetings will be open to the public, with adequate notice provided and time set aside for public input; and explain where and how records may be accessed by the public.

Because formal meetings of the Joint Committee may only occur a few times each year, a method for routinely receiving public input and disseminating information will need to be established. The Joint Committee should have a continuous and known presence in the region. Although federal government officials will chair the Joint Committee, a local liaison office or person should be established. The role of the liaison could include maintaining records, responding to inquiries, and monitoring relevant activities of state and local governments as well as the Paso del Norte Air Quality Task Force. Similarly, because the Binational Air Working Group only meets a few times a year, it should establish a point of contact for the local Joint Committee liaison.

The operating procedures of the Committee should also explain how the Joint Committee will interact with the Binational Air Working Group. The procedures should outline the Joint Committee's expectations regarding timely decisions on recommendations, as well as a process for appealing negative decisions, including the option of pursuing alternative means for implementation if no federal action is needed. Joint Committee meetings should be held in conjunction with Binational Air Working Group meetings to facilitate timely response to recommendations. The operating procedures could outline methods for streamlining decision-making and strategy implementation, particularly if the Joint Committee meets more frequently than the Binational Air Working Group. The Joint Committee could develop criteria for categorizing recommendations and propose alternative decision-making paths, so that routine recommendations could be implemented more expeditiously. For example, if a particular recommendation can be implemented by local governments or private interests and no additional federal funding is needed, the Joint Committee should be authorized to proceed without formal action by the Binational Air Working Group.

Successful implementation of the Joint Committee's recommendations will depend upon commitments from various governmental officials, as well as the public and business community. Therefore, decision-making within the Joint Committee should be done by consensus, with equal weight given to the views of all representatives. In addition, the Joint Committee should have responsibility for estimating the financial, human and technological resources needed to implement its recommendations, as well as identifying the means for obtaining necessary resources. This will help expedite decision-making and stimulate a positive response from both governments.

5.2 Achieving Air Quality Objectives

The credibility of the Joint Committee will depend on community participation and a fair and open process for conducting business, but its effectiveness will be judged by what it achieves. Therefore, an important first step for the Joint Committee will be establishing objectives and deciding how to achieve them. One of the highest priorities should be agreeing on short and long term air quality objectives for the international air basin, consistent with attaining the respective national ambient air quality standards. Air quality and emissions based targets and timelines should be established as measures of progress.

Information Acquisition. Establishing and achieving air quality objectives will require reliable information. The Joint Committee will need to devote immediate attention to assessing available data, identifying its information needs, and developing plans for acquiring information needed to characterize air quality conditions, document sources of pollution, and compare the effectiveness of pollution control strategies. The following activities should be undertaken:

- ***Expansion and integration of air quality monitoring network.*** Reliable and regular information on air quality conditions throughout the international air basin is needed to understand the nature, extent and magnitude of pollution problems. Publicizing air quality conditions on a daily basis, along with information about associated health effects and measures that can be taken to reduce pollution, promotes public awareness. Monitoring is also needed to identify the sources of the pollution problem. The Joint Committee will need to evaluate the adequacy of the existing air quality monitoring network; recommend additional sites or types of monitoring, including methods for tying together the respective governments monitoring efforts; and ensure daily, public access to monitoring data.

- ***Completion of comprehensive emissions inventory.*** The Joint Committee will need to examine existing emissions inventories and identify inventory needs. Although existing information may be adequate to begin developing pollution control and prevention strategies for known pollution sources, a comprehensive emissions inventory will be needed to guide long-term planning. The Joint Committee will need to assert a strong role to ensure that data are collected as quickly as possible.

- ***Development of analytical tools.*** Air quality models will be needed to assess the effectiveness of various strategies. The Committee should obtain information about the current state-of-the-art models, including data needed to drive them and associated uncertainties, so they can make reasonable decisions regarding their use and the highest priority data needs. The Joint Committee should seek expert advice on how existing models could be refined or adapted.

- ***Transferring technology and skills.*** The Joint Committee could also play a key role in continuing and expanding technology exchanges and technical training programs so that local entities will have the technical hardware and expertise to operate and maintain their air monitoring networks and emissions inventories at comparable levels of efficiency.

Although a substantial base of data already exists to provide a basis for initial emission reduction plans, acquiring comprehensive air quality and emissions information will require significant resources and time. The Joint Committee should take a leadership role in obtaining the necessary commitments and financial resources from government and private interests. In the interim, actions can and should be taken based on what is already known about the air pollution problem.

Pollution Abatement Projects. In order to demonstrate results in the short term, the Joint Committee should develop a list of pollution abatement projects to address known problems and strive to ensure implementation of a few specific projects within its first year of operation. As a starting point, projects identified by the Paso del Norte Air Quality Task Force could be reviewed and supported or supplemented, if appropriate. In particular, the following projects merit attention:

(1) Continuing and expanded training programs for automobile mechanics;
(2) A mobile source management plan for the air basin, including investment in public transit and cleaner fuels;
(3) Workshops for paint and solvent shop businesses, including methods for reducing emissions through process changes (e.g., spray painting booths) and material substitution (e.g., use of less reactive solvents);
(4) Brick oven conversions to natural gas;
(5) Installation of vapor recovery equipment at gas stations;
(6) Industry-sponsored automobile tune-ups for employees;
(7) Increased transborder sale of electricity and cleaner fuels;
(8) Use of waste materials to pave roads; and
(9) Increased staffing and operational changes at international bridge crossings to reduce traffic congestion.

Most of these projects could be implemented through local cooperative efforts between the business community and state and local government agencies. An initial emphasis on pollution abatement projects that do not require federal government approval or action will help achieve short-term results and create a favorable track record for more complex projects.

Use of Economic Incentives. In selecting projects for implementation, the Joint Committee should place a high priority on cost-effectiveness and promoting transboundary investment in pollution control technology and strategies. The international agreement gives the Joint Committee a specific role in developing and implementing economic instruments, including studying the feasibility of a transboundary emissions trading program. The emphasis on economic instruments is consistent with the shift that is occurring in environmental policy generally in the U.S. and the specific encouragement of market-based programs in the U.S. Clean Air Act. Market-based programs promote innovation and the most cost-effective solutions to air pollution by allowing industry greater flexibility and rewarding efforts that result in greater emission reductions than otherwise required by law or regulation.[33]

There are many ways in which economic instruments could be used in the Paso del Norte transboundary air basin.[34] One of the most promising and intriguing options is to allow U.S. companies to earn emission reduction credits under the U.S. Clean Air Act for pollution control projects in Cd. Juarez. The U.S. Clean Air Act requires that new industries wishing to locate in the El Paso nonattainment area must obtain emission reductions elsewhere in the area to offset pollution increases from the proposed facility. Furthermore, EPA and TNRCC have developed regulations for emission reduction credits, offsets, banking and Area Emissions Reduction Credit Organizations.[35] Many cost-effective emission reductions opportunities exist in Cd. Juarez. U.S. companies would have a powerful incentive to seek out and implement the most cost-effective "offsets" if they could earn credit for pollution control projects in Cd. Juarez. The following examples illustrate the type of opportunities that exist:

- A new El Paso-based manufacturer of electric motors emitting 200 tons of volatile organic compounds per year might opt to meet its offset requirement by investing in Stage I vapor recovery systems in Cd. Juarez rather than a more costly alternative on the U.S. side of the border.
- A new or expanding cement plant could choose to offset increasing particulate matter emissions by agreeing to pave roads in Cd. Juarez.
- A U.S.-based gasoline distributor could sell cleaner fuel in Cd. Juarez or implement programs to retire high-emitting vehicles to offset emission increases associated with higher sales.
- Local government agencies could earn credit toward their annual emission reduction requirements by convincing federal authorities to modify operations of the international bridges to reduce emissions from vehicle idling.
- Electric utilities could earn credit to offset emissions resulting from construction or expansion of generating plants by selling electricity or investing in renewable energy development in Cd. Juarez to replace existing high-polluting diesel generators.

The Environmental Defense Fund has begun work on guidelines for a transboundary emission reduction credit program that could be refined and adapted for use. The paramount concern is insuring that reductions achieved comply with the fundamental requirements of EPA's emissions reduction credit program. The Paso del Norte Air Quality Task Force is already working to help identify industries seeking to expand or locate in El Paso that might be good candidates for demonstration projects. Identifying and conducting prototype projects would not only achieve pollution reductions but also help solve many of the legal, administrative, political and scientific issues associated with transboundary emissions trading. In particular, in order to ensure the credibility and integrity of an emission reduction credit program, mechanisms will need to be developed (and possibly tailored to each case) for ensuring that reductions achieved meet the following criteria:

(1) Quantifiable -- through development of pre- and post-construction/implementation monitoring, reporting, and recordkeeping requirements that would outline how to quantify emission reductions obtained and demonstrate air quality improvement;

(2) Surplus -- through development of procedures for defining baseline emissions level from which emission reductions may be credited, using each country and states existing laws and regulations as a benchmark. For example, the following types of activities may produce surplus reductions:

  a. a permanent shutdown of a facility, coupled with a demonstration that emission reductions will not be negated by construction of a similar emission-producing activity
  b. installation of a level of control which is greater than required by regulations or state implementation plan provisions
  c. installation of different processes or equipment which emit less than previous processes or equipment
  d. more effective operation and maintenance of abatement and process equipment
  e. verifiable and permanent reduction in production rates or hours of operations
  f. utilization of alternative fuel vehicles beyond what is required by law

(3) Permanent -- through development of procedures for demonstrating that emission reductions will be contemporaneous with and assured for the life of the corresponding increase. This may include requirements for participation in training programs to acquire skills needed to continuously maintain and monitor the efficiency of pollution control equipment or process changes.

(4) Enforceable -- through requirements that emission reduction strategies be set forth in permit conditions or regulation changes, memorandum of understanding between government agencies, or contracts between private parties. Legal enforceability must be coupled with an assurance of practical enforceability -- i.e., resources must be available to devote to inspection and enforcement activities. Emission fees or transaction fees could be established for sources wishing to engage in the transboundary emission reduction credit program, with the fees being used to augment existing enforcement resources.

Alternatively, if a contractual arrangement is used, the contract between affected parties could incorporate certain incentives to ensure the permanence and enforceability of the emission reductions. For example, if a U.S. firm has financed the cost of pollution control equipment and periodic loan payments are established, the payment schedule could be accelerated in the event of noncompliance; or the sponsoring company could agree to pay their partner periodically for operation and maintenance costs so long as compliance occurs. The contract could also include a liquidated damages and arbitration provision to avoid potential litigation. Finally, the contract could designate the Joint Committee as a third party beneficiary, so that independent enforcement action could be taken by governmental entities in the event of noncompliance.

As mentioned above, specific requirements will likely need to be developed on a case-by-case basis as transboundary emission reduction opportunities arise. The Joint Committee should conduct an outreach program to help solicit companies to participate in demonstration projects so this promising approach to reducing pollution can be tested and implemented more broadly in the air basin. The Joint Committee could serve as the "certifying" agency by helping to negotiate appropriate conditions, develop suitable instruments, review and verify the validity of emission reduction credit transactions, and monitor compliance. In addition, any application for obtaining transboundary emission reduction credits would need to be subjected to public review, and credit could not be awarded until enforceable mechanisms are in place.

Public Education. The Joint Committee should have a central role in educating the public about air pollution problems and disseminating information about how individuals can reduce pollution through their own actions and behavior. To increase public awareness, the Joint Committee's work plan should include preparing regular (at least annual) reports to the public on air quality conditions and trends throughout the international air basin. Once monitoring networks are integrated and automated, daily reports on air quality conditions should be issued. The Joint Committee may also sponsor workshops and prepare informational materials for businesses and schools outlining pollution prevention and reduction strategies.

Public education efforts should also be used to demonstrate accountability of the Joint Committee. The annual report should summarize recommendations made by the Joint Committee and action taken on the recommendations. Pollution abatement projects implemented should be described and their effectiveness assessed. The report should also disclose financial aspects of the Joint Committee's activities and projects, including an accounting of funding received and how it was used. Through these reports, the public will gain confidence that the Joint Committee is an effective and efficient institution -- not just another layer of bureaucracy.

6. CONCLUSIONS

The agreement signed by the U.S. and Mexican governments on May 7, 1996, represents a step toward better regional management of transboundary air pollution. This result came about through the effective work of the Paso del Norte Air Quality Task Force which put forth a strong proposal and got most of what it asked for. Although the federal governments necessarily remain major players with important authorizing and oversight roles, the international air quality management basin is a reality and the Joint Committee will promote greater community involvement in decisionmaking and pollution abatement activities.

In particular, the Joint Committee will be representative of the local binational community and will have broad authority to establish its own operating and administrative procedures; to engage in technology transfer and training programs; to generate and disseminate information for the public; to serve as a forum for hearing public concerns about air pollution problems and potential solutions; to conduct joint studies and analyses of air pollution problems, including the integration and automation of monitoring networks; and to develop, and in many cases, implement, pollution control and prevention projects. The Joint Committee will also be accountable to the public and serve to hold the federal governments accountable for acting on recommendations. Perhaps one of the Joint Committee's most important functions will be to marshall the financial and human resources needed to bring about real air quality improvement, including, creating economic incentives for transboundary investment in pollution control.

As an evolutionary -- and unprecedented -- community-led effort toward institutionalizing shared responsibility for managing transboundary air pollution problems, the progress made by the Joint Committee will need to be carefully monitored and periodically evaluated. As barriers are identified, creative approaches to overcoming them will need to be developed. Perhaps the greatest potential lies in carrying out pollution reduction projects that help achieve cleaner air and support economic growth without getting bogged down in conflicts that arise because of the international border or differing stages of economic development. At some point in the future, it may be helpful to develop a time schedule and procedure for moving from transboundary governance based on differing domestic standards to a set of uniform and consistent ambient air quality standards, coupled with the establishment of emission budgets for the various pollutants and source sectors that contribute to air pollution problems in the international air basin.

Most importantly, the Joint Committee and the Paso del Norte Air Quality Task Force will help the citizens of the region take responsibility for achieving cleaner air. With strong community support, the Joint Committee can implement a transboundary air quality management program that promotes cooperation among the parties, minimizes the cost of pollution control, and delivers better air quality. If the new governance structure proves to be effective, it will serve as a good model for other border communities struggling with similar transboundary air pollution problems or other shared environmental problems.

Endnotes

[1] City of El Paso Department of Planning, El Paso Metropolitan Planning, (El Paso, Texas: 1996).

Cd. Juarez Planning Department, Plan Director de Desarrollo Urbano, (Cd. Juarez, Chihuahua.: 1995).

[2] U.S. Environmental Protection Agency, Compendium of EPA Binational and Domestic U.S./Mexico Activities, EPA 160-B-95-001, (Washington, D.C.: 1995).

[3] Laura Margarita Uribarri, The Paso del Norte Air Quality Task Force: A Case Study in Binational Cooperation, Senor Honors Thesis, International Relations, Stanford University, (Stanford, California: 1996).

[4] James W. Yarbrough, An Overview of U.S.-Mexico Border Air Activities Schedule Through The Integrated Border Air Activities Schedule Through The Integrated Border Environmental Plan, Unpublished Report, U.S. Environmental Protection Agency, (Dallas, Texas.: 1996).

[5] John C. Ross, Jr., "Do the Existing Air Pollution Statutes and Regulations Provide the Required Protection in the El Paso-Juarez Area?" in Air Pollution Along The United States-Mexico Border edited by Howard G. Applegate and C. Richard Bath, Texas Western Press, The University of Texas at El Paso, (El Paso, Texas: 1974).

[6] U.S. Environmental Protection Agency and Secretaria De Desarrollo Urbano y Ecologia, Integrated Environmental Plan For The Mexican-U.S. Border Area. EPA A92-171, (Washington, D.C.: 1992).

[7] *Ibid.*

[8] Larry Butts, Air Quality Assessment Program, Texas Natural Resource Conservation Commission, (Austin, Texas: 1996).

[9] Maria M. Aponte - Pons and Alison A. Miller, Air Pollution and the Texas-Mexico Border: Recent Research Initiatives. Air and Waste Management Association, 87th Annual Meeting, (Cincinnati, Ohio: 1994).

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[11] W. Einfeld and H. W. Church, Winter Season Air Pollution in El Paso-Ciudad Juarez: A Review of Air Pollution Studies in an International Airshed, SAND95-0273, Sandia National Laboratories, (Albuquerque, New Mexico: 1995).

[12] Archie Clouse, El Paso Air Quality Fact Sheet. Texas Natural Resource Conservation Commission, (El Paso, Texas: 1995).

[13] W. Einfeld and H.W. Church, *op. cit.*

Appendix A: Annex VI Proposed by the Paso del Norte Air Quality Task Force

Appendix A: Annex VI Proposed by the Paso del Norte Air Quality Task Force

ANNEX VI TO THE AGREEMENT
BETWEEN
THE GOVERNMENT OF THE UNITED STATES OF AMERICA
AND
THE GOVERNMENT OF THE UNITED MEXICAN STATES
ON COOPERATION FOR THE PROTECTION AND IMPROVEMENT OF THE ENVIRONMENT IN THE BORDER AREA

AGREEMENT OF COOPERATION
BETWEEN
THE GOVERNMENT OF THE UNITED STATES OF AMERICA
AND
THE GOVERNMENT OF THE UNITED MEXICAN STATES
REGARDING THE FORMATION AND PURPOSES OF AN INTERNATIONAL AIR QUALITY MANAGEMENT DISTRICT

The Government of the United States of America ("the United States") and the Government of the United Mexican States ("Mexico") ("the Parties"),

Recognizing that substantial improvement of air quality is needed in their common border zone;

Realizing that such air quality improvement can occur most effectively and efficiently with increased cooperation;

Recognizing that the formation of a joint district to work for such air quality improvement is desirable in the El Paso, Texas - Ciudad Juarez, Chihuahua - Doña Ana County, New Mexico area;

Reaffirming Principle 21 of the 1972 Declaration of the United Nations Conference on the Human Environment, adopted at Stockholm, which provides the Nations have, in accordance with the Charter of the United Nations and the principles of international law, the sovereign right to exploit their own resources pursuant to their own environmental policies and the responsibility to ensure that activities within their jurisdiction or control do not cause damage to the environment of other Nations or areas beyond the limits of national jurisdiction;

Recognizing that Article 3 of the Agreement between the Parties on Cooperation for the Protection and Improvement of the Environment in the Border Area of 1983 ("the 1983 Agreement") provides that the Parties may conclude specific arrangements for the solution of common problems in the border areas as annexes to that Agreement,

Have agreed as follows:

ARTICLE 1
GENERAL PURPOSE

In cooperation with existing National, State, and Local authorities, the Parties agree to hereby establish an International Air Quality Management District ("IAQMD") on their common border.

ARTICLE II
APPLICABILITY

1. The IAQMD shall encompass the following geographic area of jurisdiction: El Paso County, Texas; Doña Ana County, New Mexico; and the greater Metropolitan Area of Ciudad Juárez, Chihuahua.

2. Nothing in this agreement will supersede the obligations of affected United States State and Local authorities or affected Mexico State and Local authorities to meet all existing applicable rules and regulations of the United States and Mexico, respectively.

ARTICLE III
SPECIFIC PURPOSES

The purposes of the IAQMD will be as follows:

(1) to develop a consolidated air emissions inventory and ambient air quality data base

and, using environmental targets established by the Parties, including those based on health considerations,

(2) to develop options for both short-term and long-term air pollution control and prevention measures, including an emergency action plan

(3) to identify optimum, joint control strategies, including an economic incentives-based program to encourage investment in pollution control projects and similar innovative mechanisms of pollution control

(4) to recommend joint control strategies to the respective National Governments

(5) to work with the respective National, State, and Local Governments to implement joint control strategies and to promote transboundary transfer of technology and skills to facilitate the optimization of such strategies

(6) to educate the general public regarding air quality control issues

(7) to establish procedures to allow effective and efficient communication among the parties

(8) to establish cooperative compliance efforts between the respective governments, provide a forum for citizen participation, and jointly develop recommendations for appropriate remedial actions by the respective governments.

ARTICLE IV
STRUCTURE OF GOVERNING BOARD

The Parties agree to establish a Governing Board ("the Board") for the IAQMD. The Board shall be established according to the following guidelines:

(1) The Board shall be appointed by the Parties. The Board shall consist of representatives from the Federal, State (Texas, New Mexico, Chihuahua) and Local (El Paso, Ciudad Juárez, Doña Ana County) Governments. The parties shall each appoint two members-at-large representing the public, including at least one each representing local industry. All Board members shall serve a three-year term.

(2) The Board shall have the authority to appoint an Executive Director and technical staff to enable it to carry out its responsibilities. The size and composition of the staff shall be determined by the Board, consistent with operational needs.

(3) The Parties agree to provide sufficient funding and personnel to ensure the IAQMD's functions can be adequately carried out.

ARTICLE V
RELEASE OF INFORMATION TO THIRD PARTIES

The Parties shall follow the guidelines set forth in Article 16 of the 1983 Agreement related to the procedure for sharing technical information with third parties and Article VI of Annex V of the 1983 Agreement for establishing procedures to protect the confidentiality of proprietary or sensitive information conveyed between the Parties pursuant to this Annex.

ARTICLE VI
EFFECT ON OTHER INSTRUMENTS

Nothing in this Annex or its appendices shall be construed to prejudice other existing or future agreements concluded between the Parties or affect the rights or obligations of the Parties under international agreements to which they are party.

ARTICLE VII
IMPLEMENTATION

Implementation of this Annex is dependent upon the availability of sufficient funding.

ARTICLE VIII
APPENDICES

Appendices to this Annex may be added through an exchange of diplomatic notes and shall form an integral part of this Annex.

ARTICLE IX
AMENDMENT

This Annex, and any appendices added thereto, may be amended by mutual agreement of the Parties through an exchange of diplomatic notes.

ARTICLE X
REVIEW

The National Coordinators under the 1983 Agreement or their designees shall meet at least every year from the date of entry into force of this Annex, at a time and place to be mutually agreed upon, in order to review the effectiveness of its implementation and to agree on whatever individual and joint measures are necessary to improve such effectiveness.

ARTICLE XI
ENTRY INTO FORCE

This Annex shall enter into force after signature when each Party has informed the other through diplomatic notes that it has completed the internal procedures necessary for the Annex to enter into force.

ARTICLE XII
TERMINATION

This Annex shall remain in force indefinitely, unless one of the Parties notifies the other in writing through diplomatic channels of its desire to terminate it, in which case the Annex shall terminate six months after the date of such written notification.

IN WITNESS WHEREOF the undersigned, being duly authorized by their respective Governments, have signed this Annex.

Done at ___(location)_________, in duplicate, this ____(date)______ in the English and Spanish languages, both texts being equally authentic.

FOR THE GOVERNMENT OF THE UNITED STATES OF AMERICA:	FOR THE GOVERNMENT OF THE UNITED MEXICAN STATES:
____________________________	____________________________

Appendix B: Appendix 1 to Annex V

Endnotes

[1] City of El Paso Department of Planning, El Paso Metropolitan Planning, (El Paso, Texas: 1996).

Cd. Juarez Planning Department, Plan Director de Desarrollo Urbano, (Cd. Juarez, Chihuahua.: 1995).

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[3] Laura Margarita Uribarri, The Paso del Norte Air Quality Task Force: A Case Study in Binational Cooperation, Senor Honors Thesis, International Relations, Stanford University, (Stanford, California: 1996).

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[9] Maria M. Aponte - Pons and Alison A. Miller, Air Pollution and the Texas-Mexico Border: Recent Research Initiatives. Air and Waste Management Association, 87th Annual Meeting, (Cincinnati, Ohio: 1994).

[10] Larry Butts, *op cit.*

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[12] Archie Clouse, El Paso Air Quality Fact Sheet. Texas Natural Resource Conservation Commission, (El Paso, Texas: 1995).

[13] W. Einfeld and H.W. Church, *op. cit.*

[14] Howard G. Applegate, C. Richard Bath and Jeffrey T. Brannon, "Binational Emissions Trading in an International Air Shed: The Case of El Paso and Ciudad Juarez," Journal of Borderland Studies Vol. IV, No. 2, New Mexico State University, (Las Cruces, New Mexico: 1989).

[15] Archie Clouse, *op. cit.*

[16] Christopher J. Kennedy, Innovations in Binational Management of the Border Environment: The Case of Air Quality in El Paso del Norte. Air and Waste Management Association, 87th Annual Meeting, (Cincinnati, Ohio: 1994).

[17] W. Einfeld and H. W. Church, *op.cit.*

APPENDIX 1

ANNEX V TO THE AGREEMENT BETWEEN THE UNITED MEXICAN STATES AND THE UNITED STATES OF AMERICA ON THE COOPERATION FOR THE PROTECTION AND IMPROVEMENT OF THE ENVIRONMENT IN THE BORDER AREA

AGREEMENT OF COOPERATION BETWEEN THE UNITED MEXICAN STATES AND THE UNITED STATES OF AMERICA REGARDING INTERNATIONAL TRANSPORT OF URBAN AIR POLLUTION

Recalling that in the preamble to Annex V the Parties affirm their intention to ensure a reduction in air pollution concentrations for the benefit of their citizens living in the urban areas along the United States-Mexico border; and

Recognizing the importance of the participation of the local communities in carrying out the efforts to achieve this objective;

The Parties, having decided to establish a Joint Advisory Committee for the Improvement of Air Quality (hereinafter "the Committee") in the Ciudad Juarez, Chihuahua/El Paso, Texas/Dona Ana County, New Mexico air basin (hereinafter "air basin"),

Have agreed as follows:

DEFINITION

The air basin is defined as the geographic area that includes El Paso County, Texas and those parts of Dona Ana County, New Mexico and the metropolitan area of Ciudad Juarez, Chihuahua that are within 100 km. of the border.

OBJECTIVE

The Committee is established for the purpose of developing and presenting recommendations to the Air Work Group established under the La Paz Agreement regarding strategies for the prevention and control of air pollution in the air basin.

SCOPE OF ACTIVITIES

The Committee may develop recommendations for the Air Work Group on:

a) The joint development of studies and analyses on air quality monitoring and modeling, and air pollution prevention and abatement strategies in the air basin;

Draft Interim Report to the Secretariat fo the Commission for Environmental Cooperation

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Case Study

Monitoring Air Toxics: The Integrated Atmospheric Deposition Network of the Great Lakes

Authors:

Angela Bandemehr- Environmental Protection Agency, Chicago, USA
Richard Hoff- Environmental Protection Agency, Chicago, USA

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1.0 BACKGROUND

During the 1980s, it was found that in the Great Lakes atmospheric deposition was the major route of introduction of a number of pollutants to the Great Lakes. The reason for this thinking was that the downward trend of contaminant concentration achieved in the late 70s and early 80s due to control actions seemed to be levelling out.

As a result of these concerns, it was decided at a 1986 International Joint Commission Workshop in Toronto, Ontario, that atmospheric deposition of pollutants to the Great Lakes was of importance. Participants agreed that insufficient data existed to determine the relative importance of atmospheric deposition of toxic contaminants; sampling and analytical techniques were not standardized or adequate to quantify concentrations of contaminants in deposition; there was insufficient knowledge of the physio-chemical behavior of these compounds; and accuracy of the physio-chemical parameters that are required for mass balance calculations was limited. The workshop results showed that up to 96 per cent of polychlorinated biphenyls (PCBs) in the Great Lakes were from atmospheric deposition (Eisenreich and Strachan, 1988). The question was whether atmospheric deposition was contributing a significant amount of pollutants to cause continued elevated levels in biota.

2.0 ESTABLISHMENT OF THE INTERNATIONAL AIR DEPOSITION NETWORK (IADN)

Soon after the IJC meeting, the establishment of the Integrated Atmospheric Deposition Network (IADN), a joint U.S./Canadian monitoring network, was proposed. In 1990 an implementation plan for the IADN was signed by Canada and the U.S.

The IADN fulfills legislative mandates in Canada and the U.S. that address the monitoring of air toxics. An international Great Lakes deposition network is mandated by Annex 15 of the Great Lakes Water Quality Agreement (GLWQA) between the U.S. and Canada. In the U.S., the 1990 Clean Air Act Amendments also require a Great Lakes deposition network that establishes one measurement site on each of the Great Lakes. The IADN has provided the necessary standardized methods, monitoring data and loadings estimates to assess the relative importance of atmospheric deposition compared to other inputs to the Great Lakes; determined temporal trends and geographic variations in deposition; and ultimately provided information about sources of these atmospheric pollutants.

The management questions IADN was originally designed to answer are:

1. Is atmospheric deposition causing the continued elevated levels of pollutants in biota in the Great Lakes?
2. What is the spatial and temporal variation of atmospheric deposition in the Great Lakes Basin?
3. How can the data from IADN contribute to management decisions as part of Lakewide Management Plans and Remedial Action Plans to reduce the deposition of these contaminants to the Great Lakes?

Target compounds were chosen for the study based on: 1) whether detection methods existed for the compounds; 2) their potential to bioaccumulate; 3) their tendency to be transported atmospherically; and 4) their importance in the Great Lakes. An initial list of target compounds was obtained from the Annex 1 of the GLWQA. The pollutants were then separated into prioritized groups. The highest priority includes PCBs, lindane (which is the gamma isomer of hexachlorocyclohexane, g-HCH), polyaromatic hydrocarbons (PAHs, particularly benzo(a)pyrene) and lead (Pb). This first group was chosen for the first phase of the IADN (1990-1992) to demonstrate the feasibility and accuracy of sampling and analytical methods.

The second priority group included the chlorinated pesticides hexachlorobenzene (HCB), 1,1,1-trichloro-2,2-bis(4-chlorophenyl)ethane (p,p'-DDT) and its metabolites (p,p'-DDE and p,p'-DDD), trans-nonachlor, methoxychlor, mirex, dieldrin and aldrin. The additional trace metals included are arsenic, selenium, cadmium, and mercury. Except for mercury, the sampling and analysis methods for most of these species had been implemented by 1993.

A third priority group contains compounds, which have an important atmospheric component but require additional methods development for measurement of atmospheric samples. These include toxaphene (a mixture of polychlorinated camphenes), co-planar PCBs, dioxins/furans, and agrochemicals such as triazines and alachlor/metolachlor.

The network consists of master (research-grade) stations on each lake, with additional satellite stations. There are two master stations in Canada and three in the U.S. that were chosen to be representative of regional deposition patterns (see figure 1). Siting criteria determine the location of IADN sites so that they are remote from urban plumes and less than one kilometer away from the shoreline. The IADN implementation design allows for periodic re-evaluation of the existing sites to determine whether other sites are needed.

The IADN does not specify samplers or sampling protocols as long as comparability among all participants can be established. At each site concentrations of target chemicals are measured in rain and snow (wet deposition), airborne particles (dry deposition), and airborne organic vapors. In addition, precipitation rate, temperature, relative humidity, wind speed and direction and solar radiation are measured at each site.

The Quality Control/Quality Assurance (QA/QC) program development was a major achievement of the IADN program. The IADN Quality Assurance Program Plan and Standard Operating Procedures have been used as models in other atmospheric toxics monitoring efforts, such as the St. Lawrence Action Plan, the Lake Michigan Mass Balance Project and the European Monitoring of the Environment Project (EMEP).

The use of QA/QC audits and QA/QC procedures, such as the Research Data Management and Quality Assurance System (RDMQ), have provided assurance that high quality data is coming out of the IADN program. Station audits have continued on a quarterly basis throughout the program and these ensure that on-site contractors maintain protocols.

3.0 IADN LOADINGS ESTIMATES

The IADN measures concentrations of target chemicals in the atmosphere. In order to calculate loadings to bodies of water, as series of equations must be used. The model that is being used to determine atmospheric loadings contains many different terms to describe the movement of atmospheric contaminants from air to water. In general, the equation determines the wet, dry and gas phase inputs to the water surface minus the volatilization output.

Loadings of pollutants to the lake are a balance of input and output. For some pollutants, there is a net output from the lakes, i.e. the lake is a net source of these pollutants to the atmosphere. If input and output are roughly equal, the atmosphere is said to be in equilibrium with the lakes.

IADN is a combination of a monitoring network and a research program, having as its goals source attribution, process identification, and assessment of the environmental impact on environmental systems. Because these goals are not completely compatible, it was necessary to compromise certain network design parameters. One of these was high temporal resolution, which allows for better determination of sources, assessment of public health and ecosystem effects and trends analysis. Instead, the primary goal has been to determine annual and seasonal averages. This has been achieved over the four years of IADN operation. This is important because it enables IADN to answer the original management questions posed by its founders. Understanding the relative importance of atmospheric deposition for specific pollutants allows environmental managers to begin to target programs at cost effective reduction of appropriate pollutants.

To date it is difficult to draw conclusions about trends, due to the high variation in data. It may require 7 to 10 years of IADN monitoring data to detect a trend with a high degree of statistical confidence and that this length of time is also dependent on the number of stations used in the analysis (Blanchard et al. 1996).

Today, after six years of operation, many of the sampling and analysis issues of the IADN have been resolved. Comparability between sampling and analysis was achieved through extensive laboratory intercomparison studies. Toxics at extremely low concentrations in air, such as PCBs and some agrochemicals, have the highest uncertainties (over 40 per cent). Much higher than the analytical uncertainty, however, is the variation due to seasonal and annual fluctuations in air concentration (up to 90 per cent) (Hoff et. al. 1996)

The uncertainties in some of the parameters (called forcing functions, i.e. deposition velocities) for the equation used to calculate loadings are the largest (up to 300 per cent) and far outweigh analytical uncertainties and variations caused by seasonal fluctuations of air concentrations. There are large uncertainties in physio-chemical parameters (i.e. Henry's Law Constant, mass transfer coefficients) used to predict the atmospheric loadings terms in the flux equation. Also, sufficient water concentration data for all the lakes has been lacking, though recently more data has been collected. This data is important in the calculation of gas transfer of pollutants across the lake surface.

4.0 RESULTS

The first consensus report on atmospheric loadings to the Great Lakes occurred before IADN data was available and was based on very broad estimates compiled by Strachan and Eisenreich (1988). The data were revised and the numbers were substantially improved in a subsequent document (Eisenreich and Strachan, 1992). The most recent data is incorporated in a 1994 estimate and compared to the earlier results (Hoff, et. al., 1996).

As discussed above, there are many problems with obtaining reliable data. In producing these data, scientists from both the US and Canada agree that a substantial error is involved in these estimates. Further, the estimates are partly based on old or very limited data viewed as rough approximations only. As a result, data based on different sources generally show different results. Although leading edge technology is used in monitoring studies, many factors are still beyond our control. These include the effect on data of climatic variation, background noise, differences in the instrumentation and the scope and objectives of the various jurisdictions and agencies involved.

4.1 1994 Loadings Estimates using IADN data

Summarized by Hoff et. al. (1996), the most recent data indicates that PCBs, dieldrin, HCB, DDE, phenanthrene, and pyrene show a net loss via volatilization from the lakes and that gas flux is the dominant transport mechanism. Both a-HCH and g-HCH are in near equilibrium with the lakes, showing some seasonal variation. But the estimated variation in the net loading for HCH values ranges from 70 to 3,700 per cent. Errors greater than 100 per cent in the flux indicate that the direction of the net flux can be in either direction. Indeed this difference in direction is seen in the data. It has been shown that for most compounds gas flux is strongly seasonally dependent and may be out of the lake in the summer and into the lake in the winter (Achman et al, 1993, McConnell et al, 1992, Dolan et al, 1993).

Overall wet and dry deposition of HCHs appears to be fairly uniform across all lakes and is less important than gas transfer.

Estimates of wet deposition of PCBs made in 1994 are nearly the same for Lakes Superior, Michigan, and Ontario (ranging from 52 to 58 kilograms per year) with Lake Erie showing a low wet deposition rate (21 kilograms per year). Dry deposition rates are similar in Lakes Superior, Michigan and Erie (16 to 27 kilograms per year) with Lake Ontario showing a low dry deposition rate (5.7 kilograms per year). These values are low compared to gas transfer rates ranging from 1,700 to 5,000 kilograms per year.

Trace metals show wet deposition being the dominant transport mechanism. For mercury, wet and dry deposition to Lake Superior are about 5 times higher than volatilization to the atmosphere. Thus, mercury shows a net input to Lake Superior of about 645 kilograms per year.

4.2 Regional Loading Comparisons

An intercomparison of loading estimates done in 1988, 1992 and 1994 is difficult to interpret, because earlier estimates were based on sparse and uncertain data. Any trends may be an artifact of better monitoring and analysis methods today.

The most consistent trend is the reduction in 1994 lead deposition versus 1988 values for all the lakes. This is not surprising given the ban of leaded gas in the U.S. Arsenic deposition has also decreased. The reasons for this are not as clear but it has been hypothesized that process changes in a major emitter of arsenic in Canada may have led to this result.

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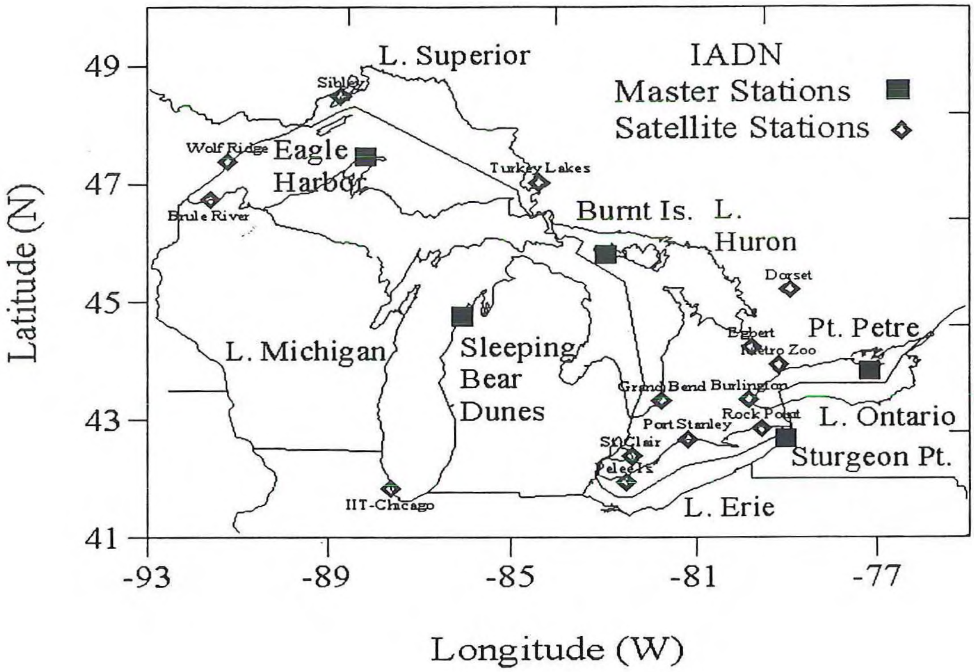
L. Superior
IADN
Master Stations ■
Satellite Stations ◇
Sibley
Wolf Ridge
Eagle Harbor
Brule River
Turkey Lakes
Burnt Is.
L. Huron
Dorset
L. Michigan
Sleeping Bear Dunes
Egbert
Metro Zoo
Pt. Petre
Grand Bend
Burlington
Rock Point
Port Stanley
St. Clair
Pelee Is.
L. Ontario
Sturgeon Pt.
L. Erie
IIT-Chicago
Latitude (N)
49
47
45
43
41
-93
-89
-85
-81
-77
Longitude (W)

Case Study

International Cooperation: The Arctic Environmental Protection Strategy and The Arctic Council

Author:
David Stone
Department of Indian Affairs and Northern Development, Hull, Canada

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1. INTRODUCTION

The following paper provides a brief overview of the formation of a "non-binding" multi-lateral agreement, the Arctic Environmental Protection Strategy, traces its evolution into the more formal (but still not binding) Arctic Council, and illustrates the scope of activities which have been addressed under this umbrella. This experience may be of relevance with respect to the CEC "Continental Pathways" project.

2. THE DEVELOPMENT OF THE ARCTIC ENVIRONMENTAL PROTECTION STRATEGY AND THE ARCTIC COUNCIL

In response to concerns regarding the Arctic environment, the government of Finland initiated discussions among the circumpolar nations in 1989 and proposed that in order to protect the integrity of the Arctic ecosystem, co-operation at the regional level is imperative.

The eight countries (Canada, Denmark/Greenland, Finland, Iceland, Norway, Russia, Sweden and the United States) agreed that collective action was needed to safeguard the Arctic ecosystem. They reviewed international legal instruments in place to protect the Arctic environment and studied a number of reports describing the state of the Arctic. Two years later in 1991 in (Rovaniemi Finland), the ministers signed the "Declaration on Protection of the Arctic Environment" and officially adopted its accompanying plan of action, the "Arctic Environmental Protection Strategy" (AEPS).

The Declaration laid out a framework of fundamental principles and objectives, while the AEPS constructed a "living" action plan designed to evolve over time. Working groups of international experts were established to carry out the tasks agreed to in the AEPS. These working groups concentrated on specific program areas: Protection of the Arctic Marine Environment (PAME); Emergency Prevention, Preparedness, and Response (EPPR); Conservation of Arctic Flora and Fauna (CAFF); and the Arctic Monitoring and Assessment Program (AMAP).

An important aspect of the AEPS is its continuity. Ministers agreed to meet regularly and further develop the Strategy. At Nuuk, Greenland in 1993, they expanded their commitments by signing "The Nuuk Declaration on Environment and Development in the Arctic". The Nuuk Declaration emphasized the adoption of the principles of the Rio Declaration. The ministers also agreed at Nuuk to establish a Task Force on Sustainable Development and Utilization to propose steps that governments should take to meet their commitment to sustainable development in the Arctic. In 11996 in Inuvik (Canada), the ministers further elaborated the AEPS and stated their resolve for an early establishment of the Arctic Council. This occurred in Ottawa in September 1996.

The AEPS was an important milestone in the history of international Arctic co-operation which will maintain its momentum beyond 1996 as a component of the work being conducted under the Arctic Council. However, the Arctic Council will be able to address a scope of activities which will move beyond the "environmental protection field occupied by the AEPS, and has potential to foster Arctic cooperation in the broad area of sustainable development..

Thus the broad aims of the Arctic Council are to:

> promote co-operation and co-ordination of action on common Arctic issues, particularly sustainable development and environmental protection;
>
> oversee and co-ordinate the established programs of the AEPS;
>
> oversee and co-ordinate a sustainable development program; and
>
> disseminate information, encourage education and promote interest in Arctic-related issues.

The Arctic Council Declaration named each of the eight former AEPS Arctic States as Members and three Permanent Participants: the Inuit Circumpolar Conference, the Saami Council, and the Association of Indigenous Minorities of the North, Siberia and the Far East of the Russian Federation. The Declaration allowed for further Permanent Participant status for other Arctic indigenous groups and Observer status for other non-Arctic countries, and intergovernmental and interparliamentary and non-governmental organizations.

3. WORK PROGRAMS CONDUCTED UNDER THE AEPS/ARCTIC COUNCIL

A good appreciation of the scope of work being conducted under this initiative can be obtained by examining both the four original programme descriptions as prepared for the Roveniemi 1991 Declaration, and the corresponding "Future Work" sections for each programme as identified in the 1996 Inuvik adjustments to the AEPS. The developing plans for the work related to sustainable development are not summarized in this paper.

3.1 The Arctic Monitoring and Assessment Programme (AMAP).

3.1.1 Rovaniemi 1991

The programme will have as its primary objective measurement of the levels of pollutants and assessment of their effects in the Arctic environment. Priority shall be given to persistent organic pollutants, radionuclides and heavy metals, and to the biological impact and effects of contaminants on human health. From the outset, the Arctic Environmental Protection Strategy Programme will coordinate and review the existing national programmes and develop these programmes within the international framework.

3.1.2 Future Work - Inuvik 1996

Following their review of the work being undertaken by AMAP, the Ministers determined that the priority for AMAP is the publication in 1997 of its report "Arctic Pollution Issues - A State of the Arctic Environment Report" (SOAER) and its accompanying technical document, "Arctic Pollution Issues - The AMAP Assessment Report" (AAR). These two documents will present a comprehensive assessment of the situation concerning POPs, heavy metals, radionuclides, acidification, hydrocarbons, climate change and UV-b radiation in the Arctic. Special attention has been given to the implications regarding human health.

In addition, the Ministers asked AMAP to provide them with advice relating to:

i. the feasibility of developing sub-regional cooperative oil-related monitoring and assessment activities;

ii. the state of knowledge concerning organotins in the marine environment; and,

iii. options and priorities for a second phase of AMAP's work.

The Ministers will meet again in June 1997 to consider the two AMAP reports and to formulate appropriate actions.

The work of AMAP is a good example of the AEPS in action. Backed by scientific evidence and combined political strength, the Arctic countries have brought the issue of persistent organic pollutants to the attention of the international community. It was primarily AMAP data which was used at the eighteenth session of UNEP Governing Council to substantiate the need for global action on these substances. With respect to the CEC Contaminants Pathways project, the experience of AMAP is of interest since it illustrates the potential of a relatively informal multi-lateral agreement in fostering scientific cooperation. AMAP did not initiate many new programmes, but instead harmonized existing national activities in a way which enabled a circumpolar assessment to be realized.

3.2 Conservation of Arctic Fauna and Flora (CAFF)

3.2.1 Rovaniemi 1991:

The intention is to cooperate in the conservation of Arctic fauna and flora, protecting their diversity and habitats. Such cooperation includes exchanges in research and management, information and data, and coordination of research. Mechanisms will be established for furthering conservation strategies and regularly compiling and disseminating information on activities regarding the protection of Arctic fauna and flora.

3.2.2 Future Work - Inuvik 1996:

Following their review of the work that had been carried out by CAFF, the Ministers agreed that the priority for CAFF was to continue with its work on the development of the Circumpolar Protected Area Network (CPAN) and to assist the member countries with the implementation of the CPAN Strategy and Action Plan.

The Ministers asked CAFF to assist countries with the implementation of the Circumpolar Murre Conservation Strategy and Action Plan, and acknowledged that there may be a need, in the future, to develop strategies and action plans for other species of common conservation concern.

In response to CAFF's preliminary report identifying threats to Arctic biodiversity, the Ministers directed CAFF to advance its work on the development of a draft Arctic strategy relating to the goals of the Convention on Biological Diversity, for the next meeting of Ministers.

3.3 Protection of the Arctic Marine Environment (PAME).

3.3.1 Rovaniemi 1991

The Arctic countries will emphasize measures to prevent marine pollution irrespective of its origin. The countries place particularly high priority on adopting the strictest international standards regarding various discharges. Joint action will be taken to heighten recognition of the particularly sensitive character of ice-covered parts of the Arctic Ocean. The initiatives in international organizations promoting preparedness and response to accidental pollution and to ensuring proper application of such standards will be supported.

3.3.2 Future Work - Inuvik 1996:

The Ministers reiterated that the activities of PAME are focussed upon legal and other similar arrangements related to protection of the Arctic marine environment. In view of the information provided by the PAME Working Group, the Ministers agreed upon the following priorities for action:

i. To request the development of an Arctic Regional Programme of Action to address marine pollution resulting from land-based activities.

ii. To request the development of guidelines for offshore petroleum activities in the Arctic, in particular guidelines for timely and effective measures for protection of the Arctic environment.

iii. To request working toward the development of a coordinated system for collection and sharing of data on shipping activities in the Arctic and to provide a description of current and potential shipping activities to AMAP so as to enable a scientific assessment. Based on this work, PAME will report to Ministers.

iv. To request PAME to maintain an overview of the effectiveness of existing international arrangements relevant to the objectives of the PAME Programme area, taking into consideration the AMAP assessment reports (the AAR and SOAER), and to report to the Ministers on a regular basis.

3.4 Emergency Prevention, Preparedness and Response in the Arctic (EPPR).

3.4.1 Rovaniemi 1991

It is suggested that action be taken to improve international cooperation with the aim of minimizing pollution of the environment as a result of the accidental release of oil or other harmful substances. This calls for the establishment of a system for early notification in the event or threat of major accidental pollution and coordination of preventive strategies, research and assessments. Mutual assistance for an emergency response in the event or imminent threat of accidental pollution is of great importance in the Arctic region.

3.4.2 Future Work - Inuvik 1996

Following consideration of the work completed by EPPR, the Ministers agreed that their priority was to complete the "Arctic Guide for Emergency Prevention, Preparedness and Response" and an in-depth analysis of the adequacy and effectiveness of existing agreements or arrangements in the Arctic. The Ministers requested EPPR to make every effort to complete the Guide for consideration at the next Ministerial Conference. The Ministers also requested EPPR to continue their work on contributing to the development of preventative, mitigating and response measures for oil and gas accidental releases in the Arctic.

4. CONCLUSIONS

This paper has not attempted to evaluate the effectiveness of the AEPS/Arctic Council or of its component programmes. Obviously, some aspects have been more effective than others. However the AEPS experience has illustrated the potential scope of non-binding agreements as a mechanism to address certain issues, particularly of a scientific and environmental nature. Countries are often more willing to attempt to participate in a programme if it has a non-binding status. The rewards can be substantial, as was demonstrated by the impact of the AMAP generated data with respect to the initiation of global action on POPs. The aspects summarized above may be of relevance to the CEC "Contaminants Pathways project.

J. CASE STUDY ON OPPORTUNITIES

Draft Interim Report to the Secrtariat of the Commission for Environmental Cooperation

Case Study

Pollution Prevention and Increased Regional Competitiveness through Technology Innovation

Authors:

Carlos Santos Burgoa- Instituto de Salud, Ambiente y Trabajo, Mexico City, Mexico
Maria Isabel Ibarrola- Instituto de Salud, Ambiente y Trabajo, Mexico City, Mexico
Mark Cohen- Queen College- Flushing, USA
Jon MacDonagh- Michigan State University- East Lansing, USA
Jim McTaggart-Cowan- Royal Road University, Victoria, Canada

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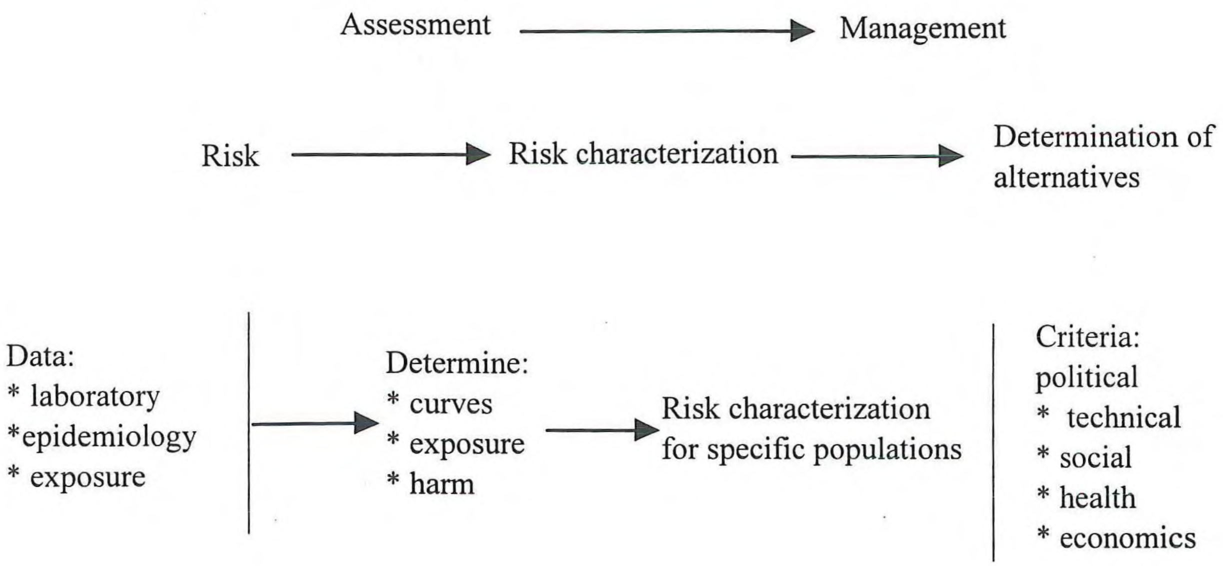
Figure 1. Risk Assessment and Management Paradigm.

RISK ASSESSMENT AND MANAGEMENT PARADIGM



Graphically the dose-response line that is derived from exposure-impact is calculated based on a model to evaluate and determine risks, that considers epidemiological and laboratory knowledge. It may start at zero level, and from there a maximum permissible level for a given product is determined as:

> "the exposure level to a toxic pollutant, below of which it is considered that the health risk is acceptable" Note that it is not pointed an nonexistence of risk, but an acceptable level. (Santos Burgoa et al., 1995)

Risk levels can be calculated to serve as the bases for at least three distinctly different types of decisions:

1. 'acceptable risk' which determines the actions to be taken if the risk level exceeds a certain number;

2. 'cost-benefit' which determines a) the benefit of reducing risk by a proposed action, usually expressed in dollar amounts, and compares this to b) the estimate of cost to implement the proposed action, and c) a decision rule chosen to evaluate the acceptability for achieving a given level of benefit, and

3. 'cost-effectiveness' which determines the maximum level of risk reduction per unit cost. (Carnegie,1993)

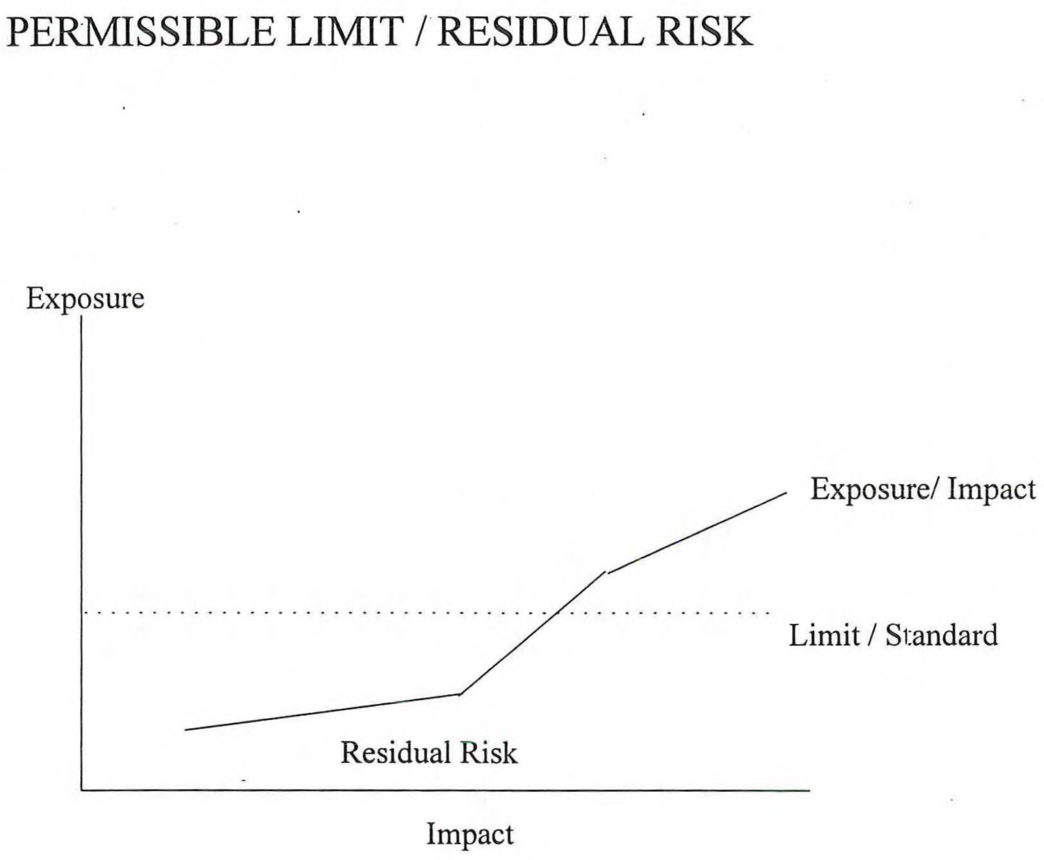
Risk level here means 'acceptable risk' or 'the (maximum) exposure level to a toxic pollutant below which it is considered that the health risk is acceptable.' Note that this does not mean that no risk exists but that there is an acceptable level of risk.

Acceptable risk levels are a controversial issue with both kind of approaches: environmental restoration or command-and-control regulations. For example: how much ecosystem damage should be permitted? how much and what kind of adverse health effects in the population should be allowed? Each society defines what is an acceptable level of risk based on their social perceptions, culture and values, and also considering political issues.

Frequently, different ideas of 'acceptable risk' cause controversies and require negotiations among producers, government and the people affected, or potentially affected, by pollutant exposure. Although frequently the public do not believe in the numbers used by scientists. If there is no compensation, when they do not feel they should accept the risk, it is difficult to achieve public agreement.

An attempt to determine an acceptable level of risk based on relatively objective parameters is given by the line exposure/impact, shown in Figure 2.

Figure 2 . Permissible Limit / Residual Risk.



1.2 A Third alternative: technology innovation.

Another point of view to the controversy of acceptable risk levels can be considered:

"if there is damage, exposure must be eliminated".

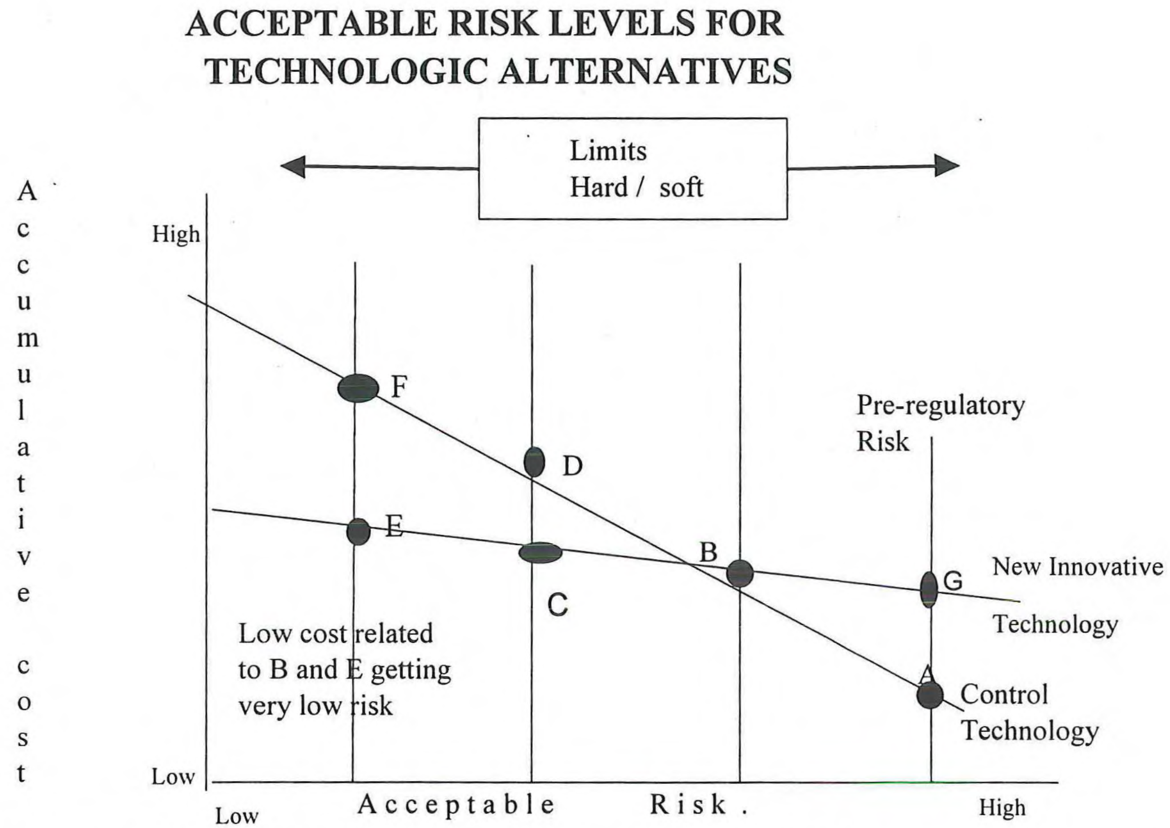
If society or decision makers tolerate a high population risk (e.g., accept a high rate of diarrhea), the cost to permit or allow such levels through control of pollutant emissions may be low. On the other hand, if social toleration to risk is low (e.g., not to accept any cancer or reproductive effects), emissions control costs rise predictably and may become cost prohibitive (i.e., product is no longer profitable).

As the emissions control regulations become more restrictive the producer will have an incentive to search for new technology. This proposal shows that risk intolerance may encourage innovation in pollution control technology; that is, the desire of society to improve environmental protection promotes newer, cleaner and perhaps more efficient technology.

Search for new technology, may generate undesirable by-products. Then it is also required to reassess the need for the product, to reassess the methods by which the products are generated, the design of the product, the materials used to generate it and the nature of the by-product. With such a review, often new products, processes and uses for by-products are found, most often resulting in improvements in all regards.

For such an alternative, the national economy benefits because capital that would be required for environmental restoration is now free for investment in other sectors. Moreover, investment in technology innovation by the producer may save significant costs over the medium term and thus add value to the product or generate a better one. As a consequence, one may often develop new businesses either in technology or in services. As seen in Figure 3, costs rise exponentially to achieve an incremental low acceptable risk when using emissions control technology.

Figure 3. Technology alternatives and acceptable risk levels



A: High accepted risk don't promote management; B: Risk with equal cost of control vs new technology
C: Lower risk achievable with lower costs of innovated technology versus D control technology
E: Lowest risk achieved at much lower costs of new technology versus F the highest cost of controls

Figure 3 shows how, lax norms that tolerate high acceptable risk makes cheaper the control technology, but its accumulative costs makes it really inefficient. By contrast, strict norms that accept low risk, makes more expensive the control technology, standing as a more attractive solution the change of production process and products through technology innovation.

This analysis suggests that strong economic development in the long term requires technology innovation more than conventional emissions control.

1.3 Costs versus Opportunities

Traditionally when many economists and business executives speak of the environment, they do so in terms of costs. They see investments in measures to "protect" the environment or "to clean it up " as "externalities to the normal costs of business". Such money must be carried as a line of debt, something that is costing them money without any return. In the same vein , they see government environmental protection legislation as something that must be minimized, neutered if at all possible.

Lawyers are hired to "interface" with politicians, to lobby so as to prevent "onerous and unwanted interference" from impeding the "normal course of business". When this view was almost universal when governments first launched the environmental issues back in the late 60´s, currently there are some large and small companies that no longer see the world in the same light.

These corporations are seen by executives who view the environment as the framework within which they must work if they are to achieve long-term success. They see pressures to change, within generated by governments as by the feedback received from the environment itself, as opportunities to investigate new ways of doing things. They see the potential for new lines of business evolving from these pressures, within it be new products or services, new processes or new technologies. They see each "cost" as a reason to question "traditional practices"; they see each expenditure as a way of generating more jobs, putting money back into the market place, and possibly generating new partnerships and economic diversity.

Such executives look to learn from the environment. They see the strength generated by diversity in nature. They observe the weakness of monocultures. They recognize the infractions of different parts of the environment, how the residue of one segment becomes the resource for another and mimic such system in "integrated industrial parks". They see how the abuse of common resources can cause unnecessary expense for all, and thus back to government for assistance in ensuring all respect The "global commons". In addition such companies executives work with scientists and politicians to maximize the potential benefits that come from increased knowledge and recognition of constraints. They embrace change and welcome it.

Such executives are not alone, non traditional economists are viewing the "environment as a constraint" model as being outdated. They recognize that their "indicators of progress" are not that at all; that there is something fundamentally wrong when stress on the environment shows up as a positive.

They note that the "expenses" are nothing more than a productive reinvestment in the global corporate community. That job and wealth creation come from the generation of business opportunities, not the simple transfer or accumulative of funds. They see new employment and corporate capitalization as generating new taxes as well, improving government revenue as well as political support.

From this "different" perspective, political leaders can be encouraged to see the environment as a win-win situation. A win for the public-cleaner air, water and land leading to reduced health costs and greater enjoyment of natural amenities, a win for industry and a win for job creation and the real economy that which truly operates sustainable within the constraints of the environment.

3.2.2 Medical waste incineration

Much infectious waste can be prevented by using reusable materials rather than disposable materials. An alternative to medical waste incineration is to sterilized by autoclaving infectious waste, and then disposed of in a landfill.

When expensive pollution controls are required for medical waste incinerators, most hospitals and commercial establishments quickly realize that autoclaves are a cheaper way to meet the regulations. Nevertheless, is important to be very careful and do it right, given that a poorly designed and/or improperly operated autoclave can pose health risks to workers and the surrounding community. For example, when strict dioxin regulations were promulgated for emissions from medical waste incinerators in California in the early 1990's, almost all on-site medical waste incinerators closed down. Today, there are almost no medical waste incinerators in California; most hospitals dispose o their waste through on site or off site (commercial) autoclaves. At least a thousand hospitals in the US have installed on site autoclaves.

3.2.3 Hazardous waste incineration

Most hazardous waste does not have to be produced in the first place. Many companies have changed production processes so that less or no hazardous waste is produced. This is often found to be cheaper than waste incineration. (U.S. Congress, Office of Technology Assessment, 1986) (Huisingh D, Martin H, Hilger H., Seldman N, 1986)

3.2.4 Sewage sludge incineration

Sewage sludge incineration is a source of dioxin, mercury, and other toxic compounds. There are other ways to handling sludge: Prevention approaches need to be taken to sharply reduce or eliminate discharge of toxic compounds to the sewage system (specially heavy metals). Sewage sludge can be composted, and, the composted sludge can be safely used as a soil amendment product.(Tenenbaum, 1997)

3.2.5 Emissions of mercury from battery life cycles

Mercury content in many types of batteries has been sharply reduced and in some cases eliminated in the US. The technology of batteries has been improved so that they can work without using as much, or in some cases, any, mercury. (U.S. EPA,1996)

3.2.6 Emissions from mobile sources

Elimination of leaded gasoline is a good example, use of alternative fuels can sharply reduce emissions and be cost competitive with heavily controlled conventionally fueled vehicles. This may be particularly true with heavy duty diesel fueled vehicles. Use of compressed natural gas produces far less environmental impacts and, can be a cheaper way to meet strict emissions controls than add-on pollution control equipment on such trucks. The use of electric motors with battery or fuel coal power, however is the pollution prevention approach available before the year 2000. It will rapidly replace the inefficient internal combustion engine. The newly proposed regulations on fine-particle pollution in the US might ultimately force this change onto a resistant US industry. (Cohen M &Commoner B,1993)

3.2.7 Emissions from burning coal and oil for heat and electricity

Solar and wind energy can be cheaper than conventional energy sources, especially if it were used at the same levels of production (Eils H & Commoner B, 1993).

These and other cases show that technology innovation can overcome the backward sectors, and also it open for them the international market for their products and their technology thanks to the regulations push. So, the question is not arguing about acceptable risk but eliminate the risk whenever is possible.

Considering the experiences where in order to promote industrial growth, pollutant industry was favored with mechanisms as energy subsides, contrasting with the cases where innovation was encouraged, the propose is to regulate strictly in order to encourage creation of innovative products. In this way, the latecomer sectors would be in conditions to compete in the international market. .

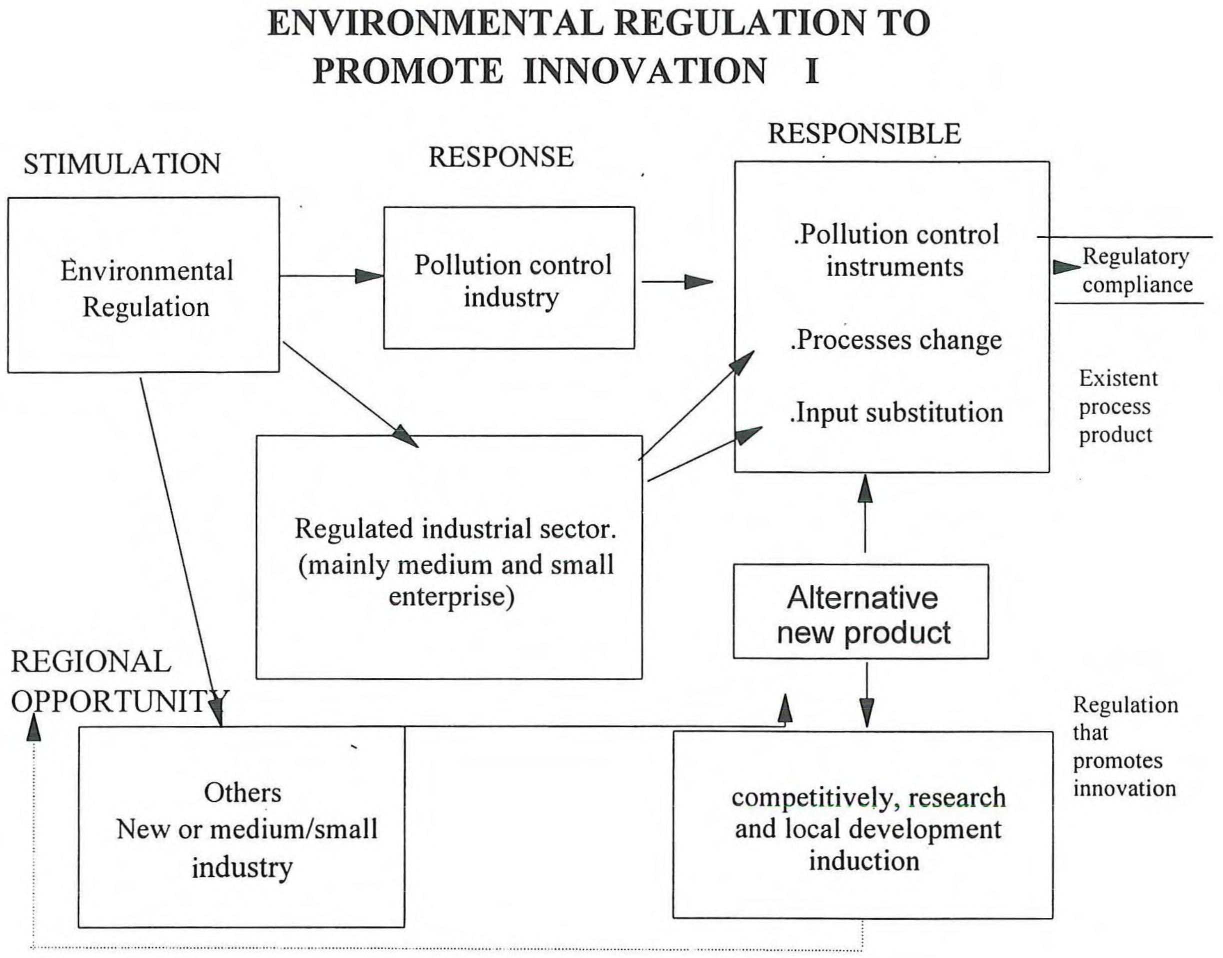
4. IDENTIFYING ECONOMIC AND SOCIAL OPPORTUNITIES: REGULATION OR INNOVATION?

Sustainability, which includes labor and environmental health among other factors, has become a criteria of global competitiveness. Sustainability may be understood to mean that every technological change requires a cultural change. For example, it has been impossible to establish environmental control of waste production on a global scale; control of emissions is possible but with high operating costs. The alternative is to change product and production process technologies which obviously implies technology innovation. In the North American case, are need new competitive and environmental friendly products. Within this process, it is important to maintain balanced conditions during the process of innovation. Innovation should occur so that market competitiveness is maintained. This requires that the society in each country understand the long term costs and benefits and agree on acceptable levels of risk.

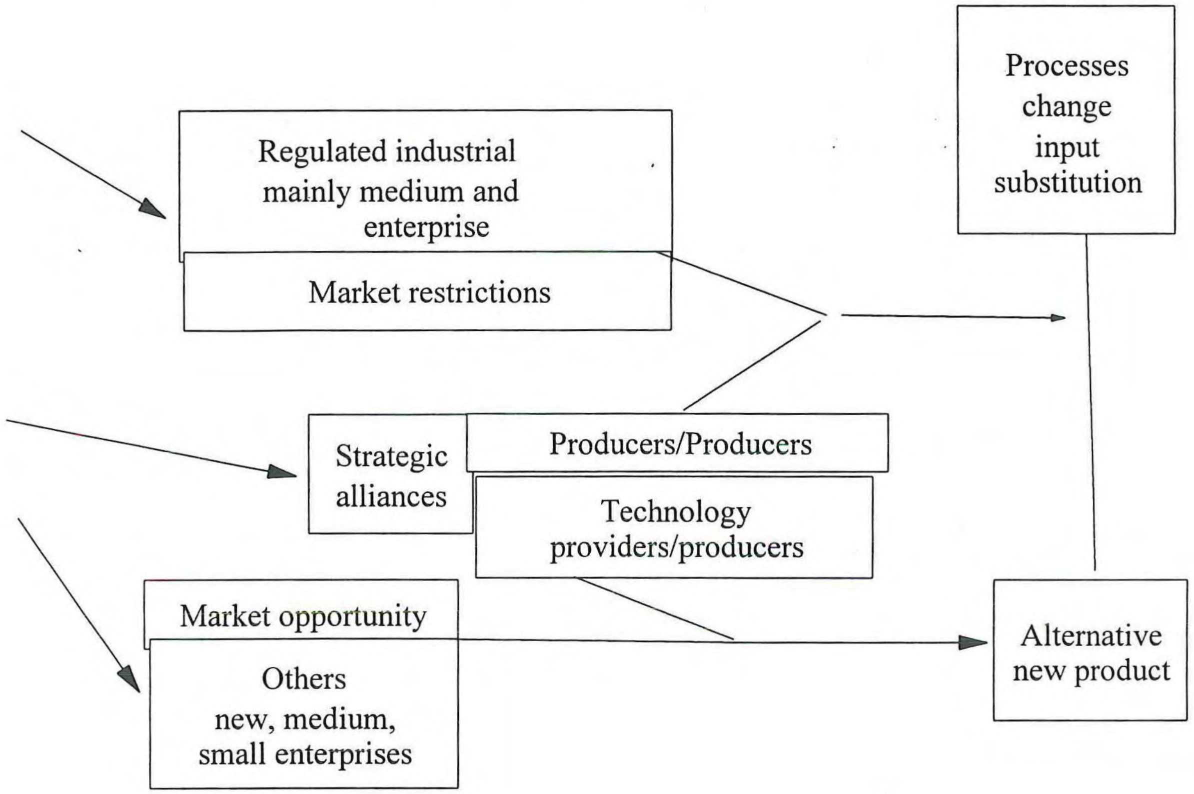
Efficient emission controls would require balancing the additional social and industrial opportunities arising from environmental restoration with the marginal social benefit(s) produced. Sectorial and intergovernmental coordination and cooperation would be required to implement these policies.

Generally the pollution prevention that occurs is the result of industry response to control-based regulations. Although large corporations are resistant to change products or technology, given the investment that hey have already made, generally the industry realizes that they can meet the standards cheaper by changing their production process than by adding on even more pollution control equipment. If investment decisions were made with pollution prevention in mind in the first place, corporations would reach the "virtual elimination" situation faster, with less cost and not waste a lot of money on pollution control equipment along the way.

Figure 4 Environmental regulation to promote innovation



ENVIRONMENTAL REGULATION TO PROMOTE INNOVATION II



5. INTERGOVERNMENTAL POLICY: CONTROL OR DEVELOPMENT?.

A case for collaboration.

In order to address the continental pollutant pathways problem, the dilemma is to choose between a regulatory policy that promotes control and a policy that promotes development through innovation.

Consistent with the model presented above, strong regulations encourage innovations by the emissions control industry as well as the regulated sector - the regulation effect is exponential. In addition, it is crucial to change the priority of government expenditures from permanent command-and-control regulations to investment in innovative and environmental friendly technologies. An intergovernmental body such as the Commission on Environmental Cooperation (CEC) of North America has a unique role to play. Promoting and generating this investment will produce more competitive products and add efficiency to regional economies in Mexico, Canada and USA. A policy is needed to promote strategic alliances between New technology providers with producers.

International cooperation also has a key role in promoting strategic alliances within economic sectors that favor technological innovation and in promoting regulation to encourage innovation rather than relying only on environmental controls.

Achieving development through innovation is a process. A temporal windows strategy with a long term vision, is recommended. Temporal objectives will favor research, technology development and innovation, as well as optimizing environmental policy.

Regarding the direction of government actions there are five groups of governmental functions to involve in this task:

1. Technology Innovation: assess, encourage and establish bases for their organization, development, transfer and marketing.

2. Support for economic sectors seeking technology innovation: financial incentives and promoting consumer pattern changes.

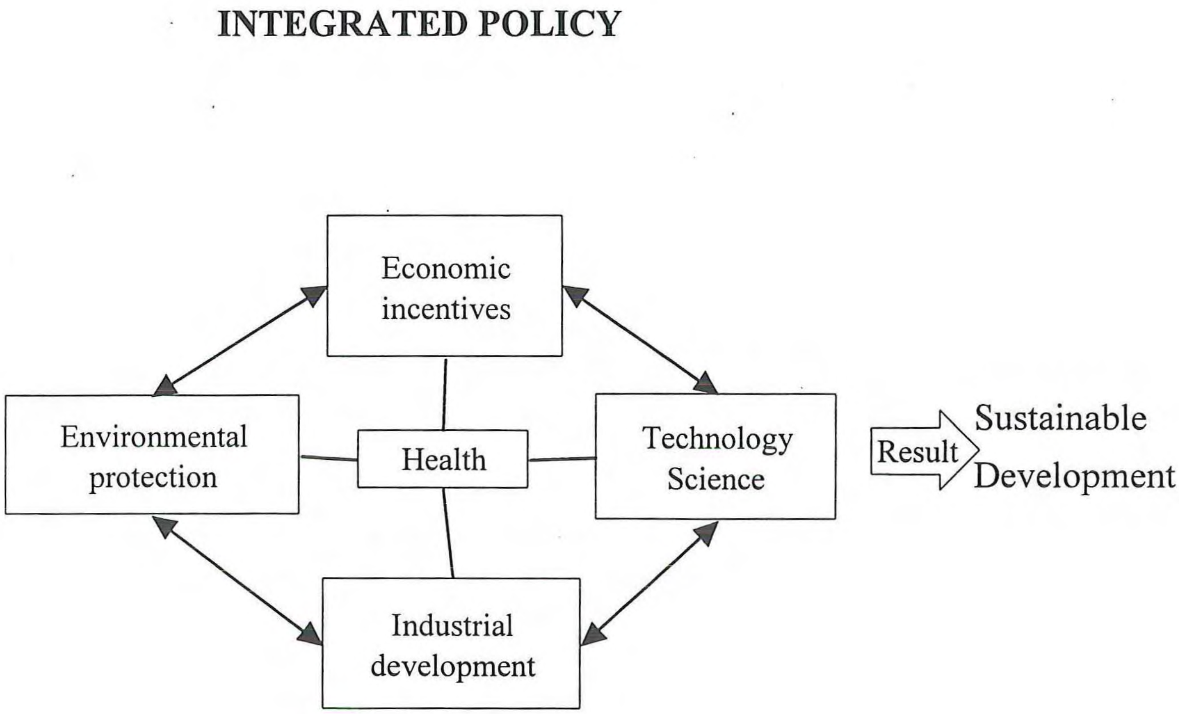
3. Promote strategic alliances within and among technology suppliers and producers.

4. Sectorial and international coordination.

5. Support research and technology diffusion.

An integrated policy as can be seen in the next figure is required.

Figure 5 Integrated Policy



The functions of the CEC (related to continental pollutants pathways) play a key role on intergovernmental coordination and in supporting the governments developing these specific functions. The CEC can facilitate the processes of international cooperation linking economical and environmental needs.

At the present time NAFTA allows the transfer of knowledge regarding maximum levels of pollutants permitted. The environmental cooperation agreement seeks to avoid distortions and barriers to trade and to sustainable development. It recognizes innovation based on competition but there are no mechanisms for technology integration or policies to promote technology innovation of existing control technologies.

6. TECHNOLOGY INNOVATION INITIATIVES.

Well suited for the less capital intensive industry (Erosa, 1995), an approach consistent with this idea has been pursued by Canada, through the Canadian Environmental Technology Advancement Centers. (CCA, 1996a). Furthermore, for 1995 the US Environmental Protection Agency started the Environmental Technology Initiative , (EPA 1994) and in its presentation the ETI states: "Technology for America's Economic Growth: A New Direction to Build Economic Strength", mentioning as the second objective to "strengthen the capacity of technology developers and users to succeed in environmental technology innovation".

Mexico has given first steps by stating its policy in this area within its National 1995-2000 government environmental program, and reflected it in its recent reforms to the law. Also, new experiences related to technology innovation are currently developing in Mexico, supported by international cooperation.

UNEP and UNIDO established in Mexico the Mexican Center for clean production as part of their global project "National Centers for a cleaner production". This center is also supported by the Instituto Politecnico Nacional (Polytechnic National Institute), and CANACINTRA (National Chamber of the transformation industry). (CMPL 1996)

Other effort is the Research and environmental training national center (CENICA) established by the National Environmental Institute (INE) with support from the Japanese International Cooperation Agency (JICA). The aim of this center is to empower the environmental action capacity of all levels of government, private and social sectors through research and training.(INE,SEMARNAP 1996)

7. RECOMMENDATIONS

Regulation to promote innovation must be strict and integrated into consistent standards within countries and strategic regional programs. They must establish a progressive timetable of objectives for technology development.

The following policy instruments could provide coordinated intergovernmental direction:

A. Establish initiatives to promote technology innovation for environmental and occupational risk reduction through coordination of policies as: low interest loans, tax incentives, financial risk sharing mechanisms and regulations to discourage noxious products consumption and use.

B. Assess the current situation with strategic programs based on risk assessment methods, and development of strategic goals.

C. Review legislation in order to harmonize requirements and standards and incorporate into the financing of economic development and technology innovation through different mechanisms.

D. Within the three countries, create associations of producers using environmental friendly technology and the suppliers of these technologies.

E. Increase support of research for innovation, trying to achieve balance within the countries.

F. Increase diffusion of environmental friendly technology throughout the region.

G. Define a strict policy that will support governments implementing regulations to improve the development of competitive industries.

H. Promote, through environmental education programs, a culture of innovation within consumers and producers.

This proposal goes beyond the minimum levels of protection for the environment and the work place, but it is needed to address transboundary pollutant effects.

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