The Transport and Deposition of Dioxin to Lake Michigan: A Case Study

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ABSTRACT

In this White Paper, data on the atmospheric deposition of dioxin to Lake Michigan are presented and discussed. Included is information on the role of atmospheric deposition relative to other dioxin loading pathways, the amount of dioxin deposited, and the relative contributions from different dioxin sources. The essential steps used in developing estimates and the policy implications of this research are presented.

In addition, the degree of certainty for which each of the information elements are known is discussed. The reasons and relative importance of different sources of uncertainty are outlined, and potential steps for reducing key uncertainties is recommended. It is hoped that this White Paper will be useful to both policy-makers and scientists in discussing "why we need to know", "what we think we know", "how well we know what we think we know", “what good is what we know”, and "what we don't know" about atmospheric dioxin deposition to Lake Michigan.

1. CONTEXT

In order to design effective policies regarding toxics reduction in the Great Lakes (or any other receptor) for any given pollutant, the following information is needed:

A. EFFECTS: To what extent are harmful effects caused by the pollutant in the Lake, e.g., to wildlife, to public health? This question is often divided into two parts: what is the exposure? and what are the consequences of this exposure?

An analysis and discussion of the effects of dioxin contamination in Lake Michigan is far beyond the scope of this White Paper. However, there are a wide range of potential concerns due to dioxin contamination in Lake Michigan. There is concern, for example, regarding human consumption of Lake Michigan fish due to contamination by dioxin and other toxic substances. There appears to be some uncertainty in the relative contribution of dioxin in the suite of human and ecosystem effects. The IJC Science Advisory Board has recommended that “the IJC advise the Parties to collaborate on the preparation of a comprehensive statement, for the entire Great Lakes basin, of the threat to human health posed by critical pollutants...”[1].

B. CAUSES: What is the relative contribution of different loadings pathways contributing to the harmful effects, e.g., what is the relative importance of atmospheric deposition, tributary loading, contaminated sediments, or other pathways in causing harmful effects? And, for any given significant loading pathway, what are the relative contributions of different sources? For example, if the atmospheric deposition loading pathway is significant for a given lake, what is the relative importance of different air emissions sources of the pollutant?

Presuming there is some adverse effect of concern, one must determine the significant contributing sources. A basic but crucial question is the relative importance of different loading

pathways – liquid effluent discharges, atmospheric deposition, contaminated sediment, etc. One must have some idea of this, because it guides the rest of the analysis – which gets more and more involved. This is the seemingly "easy" question at the beginning of, but it is important to get the answer approximately right, so that attention can be focused on the most significant pathways and resources are not wasted on insignificant pathways.

Table 1 shows two available relative estimates of dioxin pathway loadings to Lake Michigan that could be found in the literature.[2,3] Cohen *et al*.[3] looked only at atmospheric deposition and liquid effluent discharge. Other pathways, such as contaminated sediments and groundwater were not included in the analysis. Pearson *et al.*[2] compared dioxin homologue profiles in sediment cores with estimates of profiles in atmospheric deposition, to estimate the proportion of the material found in the sediments that arrived via atmospheric deposition. While these studies were carried out with very different methodologies, the estimates are relatively consistent. *However, it must be noted that neither of these two studies fully answered the overall question of the relative contribution of different loading pathways to the exposure of wildlife and humans.* That is, the question, for example, of the relative contributions of different pathways to the observed dioxin concentrations in consumed fish was not addressed. Additional research will likely be necessary to provide further insight into this question.

Table 1. Estimates of the Percent of Lake Michigan Dioxin Loadings Attributable to the Atmospheric Deposition Pathway

Study	Fraction of Current Loadings Contributed Through Atmospheric Pathway
Cohen *et al.*[3]	PCDD/F TEQ: 50-100 (central estimate: 88)
Pearson *et al.*[2]	PCDD: 50-100 PCDF: 5-35

The relative contributions of the various air emissions sources of dioxin have been further studied – providing additional detail on the contribution of the atmospheric deposition pathway – and details of this analysis are presented below (Section 3).

C. COSTS: What are the technical, economic, and social aspects involved in reducing or eliminating the contributions from major sources?

The third area of crucially needed information involves the costs and opportunities for reducing emissions from significant sources. There has been relatively little work done exploring the technical options and the costs of reducing the impacts from major sources of dioxin to Lake Michigan. One study[4] looked at economic aspects of reducing and/or eliminating dioxin contributions to the Great Lakes from the top contributors. In this study, detailed estimates of costs were made for remedial action to reduce and/or eliminate dioxin contributions from municipal waste incinerators, medical waste incinerators, cement kilns burning hazardous waste, iron sintering plants, and pulp and paper mills. This kind of information is crucial to the development of rational toxics reduction plans but unfortunately is relatively scarce.

Furthermore, the information that does exist is typically very uncertain and generally controversial.

2. THE ROLE AND POTENTIAL VALUE OF MODELS

"Models", in the context of this paper, are defined as mathematical/conceptual descriptions of one or more real-world phenomena. A model is necessarily a simplification; the real world will generally be more complicated than the level of detail that can be handled in a model. In order for a model to be successful, enough of the key processes and/or interdependencies in the system must be adequately characterized to allow a sufficient degree of accuracy in the simulation. One of the principal uses of models is that they can be used to examine different large-scale scenarios, scenarios that cannot be easily tested in the real world (e.g., different emissions reduction scenarios).

Another valuable feature of models is that they provide a synergistic complement to measurements. By themselves, measurements do not generally provide detailed information regarding the processes and contributing factors influencing the observed levels. A model can assist in interpreting and understanding the measurements that have been made. At the same time, models without measurements are of limited use – measurements must be used to "ground truth" any model. Finally, models can help fill in the spatial and temporal gaps between measurements, in order to provide a more complete description of a given system. Thus, it can be argued that models and measurements are of greatest value when they are undertaken together.

Models attempt to put together everything important that is known about a given system. If a model fails to provide a reasonable simulation, then this generally means that we don't understand enough about the system. Thus, in a very real sense, models provide a "test" of our knowledge.

It can be noted that models of various types are used in essentially all approaches in developing approximate answers to each of the above three fundamental question areas (i.e., effects, causes, and costs). Adequate knowledge regarding *all three* of the these areas is necessary in order to make the most well-informed decision; decision-making with insufficient information in any one area is obviously far from ideal.

3. MODELED ATMOSPHERIC DIOXIN DEPOSITION TO LAKE MICHIGAN

A. Introduction

This paper focuses primarily on the relative contribution of different air emissions sources to the overall atmospheric deposition of dioxin to Lake Michigan. This analysis builds on earlier work analyzing the transport and deposition of dioxin to the Great Lakes[3,5,6,7,8]. The analysis include an emissions inventory and a model that simulates the atmospheric fate and transport of emitted dioxin, including its potential transport to and deposition to Lake Michigan (and the other Great Lakes). The modeling system used here is somewhat unique, in that comprehensive source-receptor information is developed in the analysis.

This case study is for 1996, and attempts to describe the impacts of dioxin air emissions sources in the United States and Canada on Lake Michigan in that year. Emissions from some sources may have changed since 1996; thus, obviously, the results presented here do not necessarily represent the current situation. It is noted that the only substantial impediment to carrying out the analysis for the current situation (in addition to a lack of funding) is the lack of current, accurate, geographically resolved emissions inventories. All other elements of the analysis – the model, the requisite meteorological data, and ambient measurements for model evaluation – are readily available for carrying out a more up-to-date analysis once a more recent, accurate inventory is made available.

B. Emissions Inventories

A U.S. dioxin emissions inventory[6] for 1996 has been utilized consistent with a U.S. EPA inventory[9], except for the addition of several source categories (e.g., backyard burning and iron sintering). For Canada, a dioxin emissions inventory for 1995 was prepared by Environment Canada and the Canadian Federal-Provincial Task Force on Dioxins and Furans.[10] It has been assumed that these 1995 emissions are representative of 1996 emissions from Canada. Estimated emissions from backyard burning were added to the Canadian inventory. Speciation information was added to the Canadian inventory using congener profiles derived from the U.S. inventory. WHO-proposed mammalian toxic equivalency factors[11] were used throughout this analysis. An overall summary of the emissions inventories for the U.S. and Canada is shown in Figures 1 and 2. The inventory contains over 5700 point sources. Area sources -- e.g., mobile sources and backyard burning -- were estimated at the county level in the U.S. Canadian area sources were estimated on a 50-km grid near the Great Lakes and a 100-km grid elsewhere.

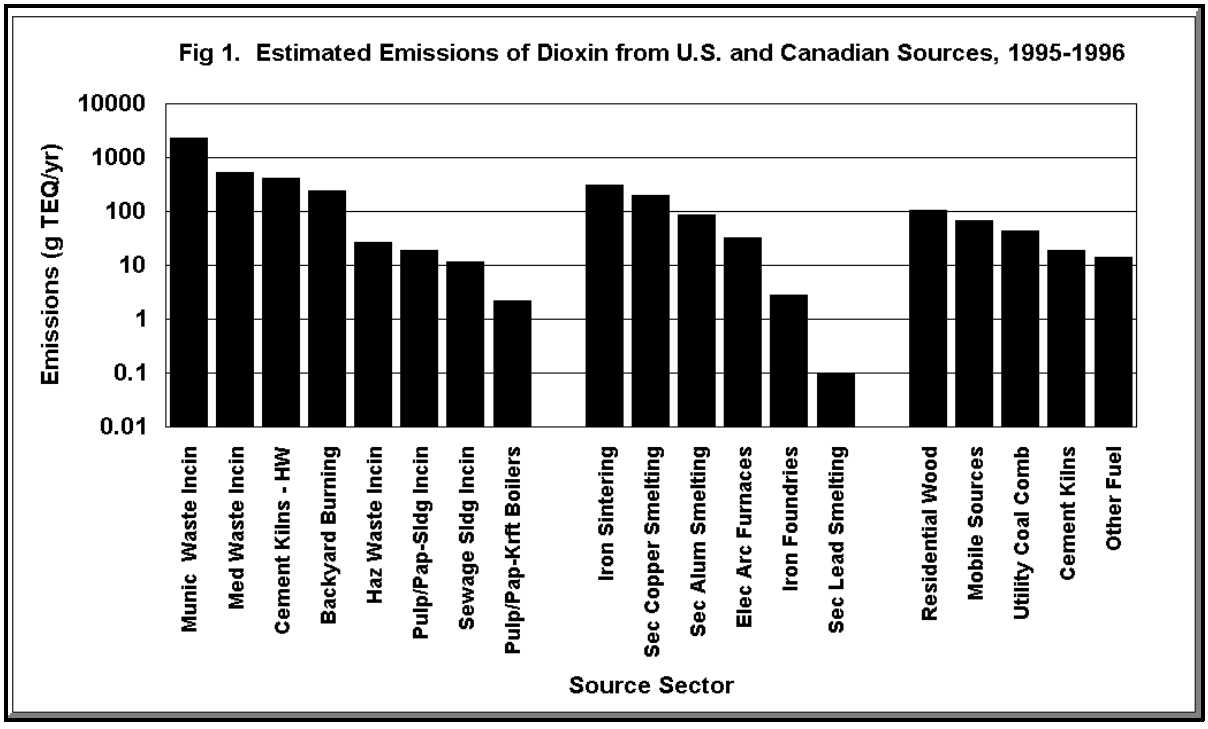
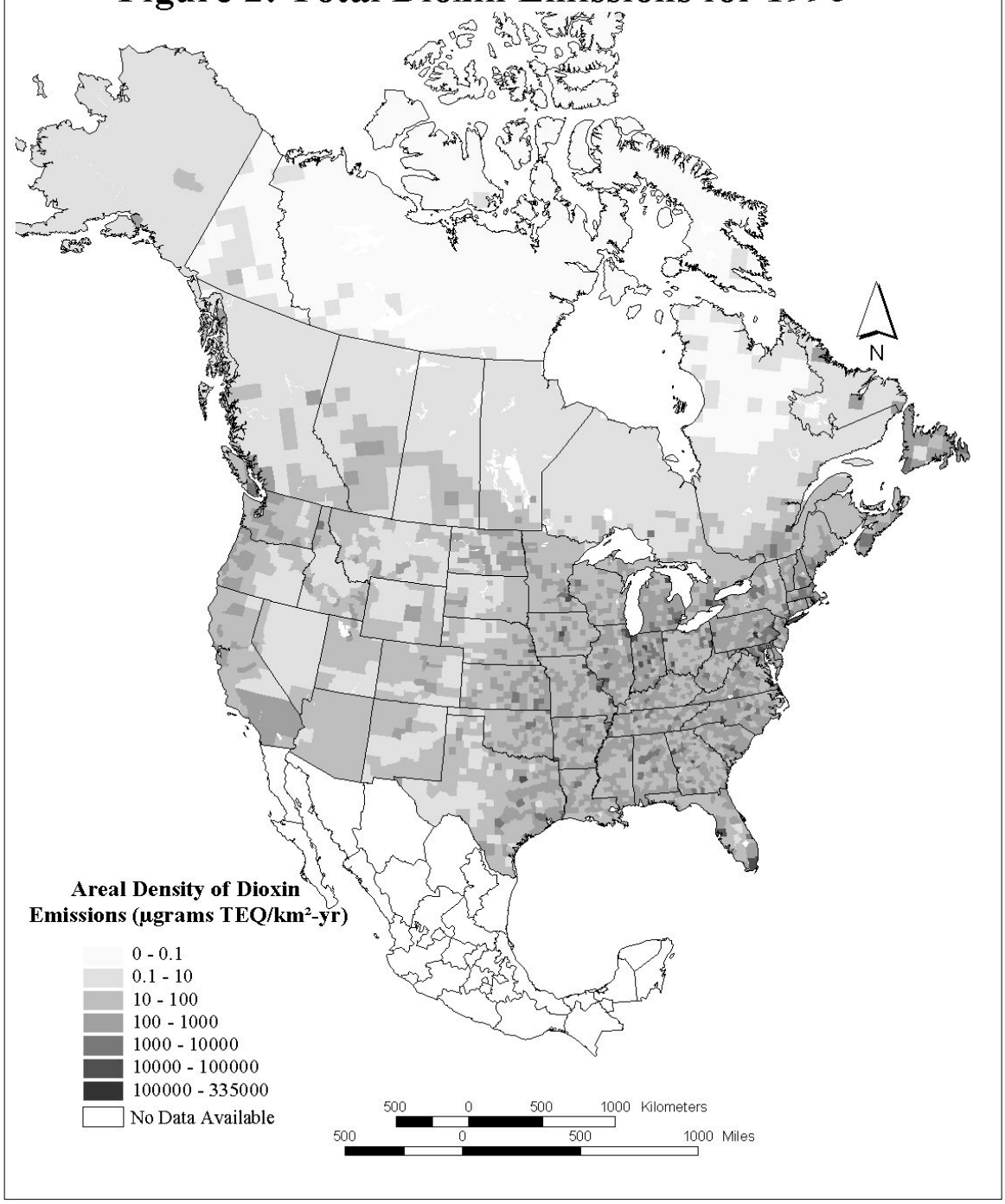


Figure 2: Total Dioxin Emissions for 1996



The uncertainties in the estimated overall dioxin emissions in the U.S. and Canada are significant -- on the order of a factor of three on either side of the mid-range estimates for each source category shown in Fig. 1. Uncertainties in the estimated emissions from any given individual source in the inventory is generally even greater than this. Few sources have been tested for dioxin emissions. Even for facilities that have been tested, there have generally been very few tests; given that dioxin emissions often appear to be highly variable -- frequently depending intimately on even small changes in feedstocks and process variables -- it cannot be said that a small number of stack tests will necessarily serve to adequately characterize the emissions from a given facility.

For essentially all the emissions inventory used in this analysis (except for many municipal waste incinerators), an emissions factor approach has been used to estimate emissions. In such an approach, *emissions factors* (e.g., grams of dioxin emitted per metric ton of material processed by a given facility) are multiplied by *activity factors* (the amount of material processed by a given facility, e.g., metric tons per year) to obtain estimates of the facility's emissions. The emissions factors used are based on a critical review of existing emissions data for a given type of source, and where data allow, attempt to include information regarding differences in emissions due to differences in the type(s) of air pollution control equipment present, key process factors, and the nature of the material processed by the source. There is effectively no choice available in the matter of whether this approach should be used or not, as most individual sources have never been measured. It can be argued, however, that emissions factors may provide a better "average" emissions picture for a given facility than the use of one or more sporadic stack tests (even if they were available), given the potential variability in emissions discussed above. The accuracy of the emissions factor approach used here could be greatly increased in the following ways: (1) testing at source types for which few or no source tests have ever been conducted; (2) additional stack tests at facilities to provide a more robust database for developing emissions factors and to better understand the variability in emissions from individual facilities; (3) regular, accurate updates on basic source information from significant sources, such as data on processes, air pollution control equipment, and activity factors. Only modest resources would be required to collect and disseminate such data (to better quantify the "causes"), especially when considered in relation to the potential costs of reducing emissions and the potential scale of adverse effects if no action is taken.

In addition, the inventories used in this analysis have at least the following omissions: (a) the U.S. inventory does not contain estimated emissions from residential or commercial coal combustion, magnesium manufacturing, or small commercial incinerators; (b) neither the U.S. nor the Canadian inventories include emissions from open-burning of PVC-coated wires (e.g., structure and vehicle fires), asphalt production, landfill fires and landfill gas combustion, coke production, leaded gasoline combustion, or petroleum refining. The accuracy of the analysis is directly and inextricably linked to the accuracy of the emissions inventory. While the emissions information used in this analysis appears adequate to generate a useful estimate of source/receptor linkages (see below), improvement of the inventory is clearly necessary.

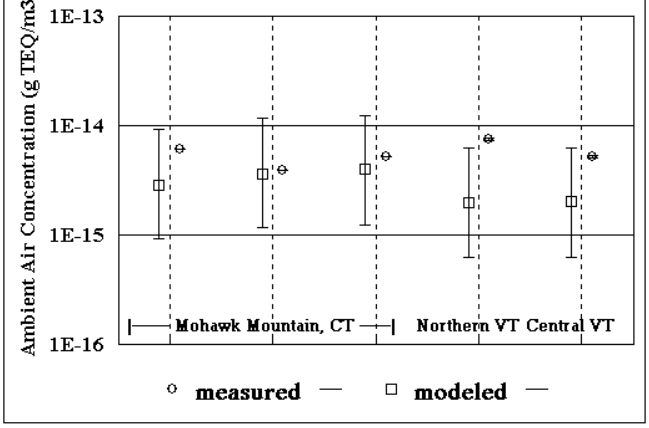
C. Atmospheric Fate and Transport Modeling

A modified version of the NOAA HYSPLIT[12] (Hybrid Single Particle Lagrangian Integrated Trajectory) model was used to simulate the atmospheric fate and transport of dioxin from sources in the United States and Canada to the Great Lakes. HYSPLIT is a Lagrangian model, in which puffs of pollutant are emitted from user-specified locations, and are then advected, dispersed, and subjected to destruction and deposition phenomena throughout the model domain. Similar to many atmospheric fate and transport models, HYSPLIT uses gridded meteorological data obtained from other sources. For these simulations, archived output from NOAA's Nested Grid Model (NGM), a sophisticate meteorological simulation model, was used. The modeling of the atmospheric fate of a dioxin performed here includes simulation of vapor/particle partitioning, wet and dry deposition, reaction with the hydroxyl radical, and photolysis. Addition details regarding the modeling can be found in the previously cited references.

D. Model Evaluation

Suitable 30-day rural ambient air measurements at two sites in Vermont and one site in Connecticut are available for dioxin in 1996[6]. A comparison of the modeling predictions with these ambient measurements is presented in Fig. 3. The model predictions are consistent with the ambient measurements, within the uncertainty of each. The uncertainty range in the modeling results was derived solely from an estimate of the source-by-source uncertainty in the emissions inventory; the overall range would be somewhat greater than this if it included other aspects of the modeling uncertainty. Obviously, it would be better if more data were available for model evaluation. Unfortunately, dioxin measurements are not included in the Integrated Atmospheric Deposition Network (IADN), and, there have been few measurements made in the Great Lakes region. However, the EPA's newly instituted National Dioxin Ambient Measurement Network (NDAMN) contains a few sites in the Great Lakes region. When data from these sites becomes available, NDAMN will be able to provide valuable model evaluation data for new studies.

Fig 3. Comparison of Model Predictions with Ambient Measurements at Month-Long Sample Sites



E. Source-Receptor Results

An example of the detailed source-receptor linkage from each U.S. county and Canadian grid square to dioxin deposition to Lake Michigan is presented in Fig. 4. An overall summary of the relative contributions to each of the Great Lakes from different distances is presented in Fig. 5. It can be seen that a significant fraction – on the order of 40% -- of the deposition to Lake

Figure 4. Estimated Contributions to the 1996 Atmospheric Deposition of Dioxin to Lake Michigan (µgrams TEQ/km²-yr)

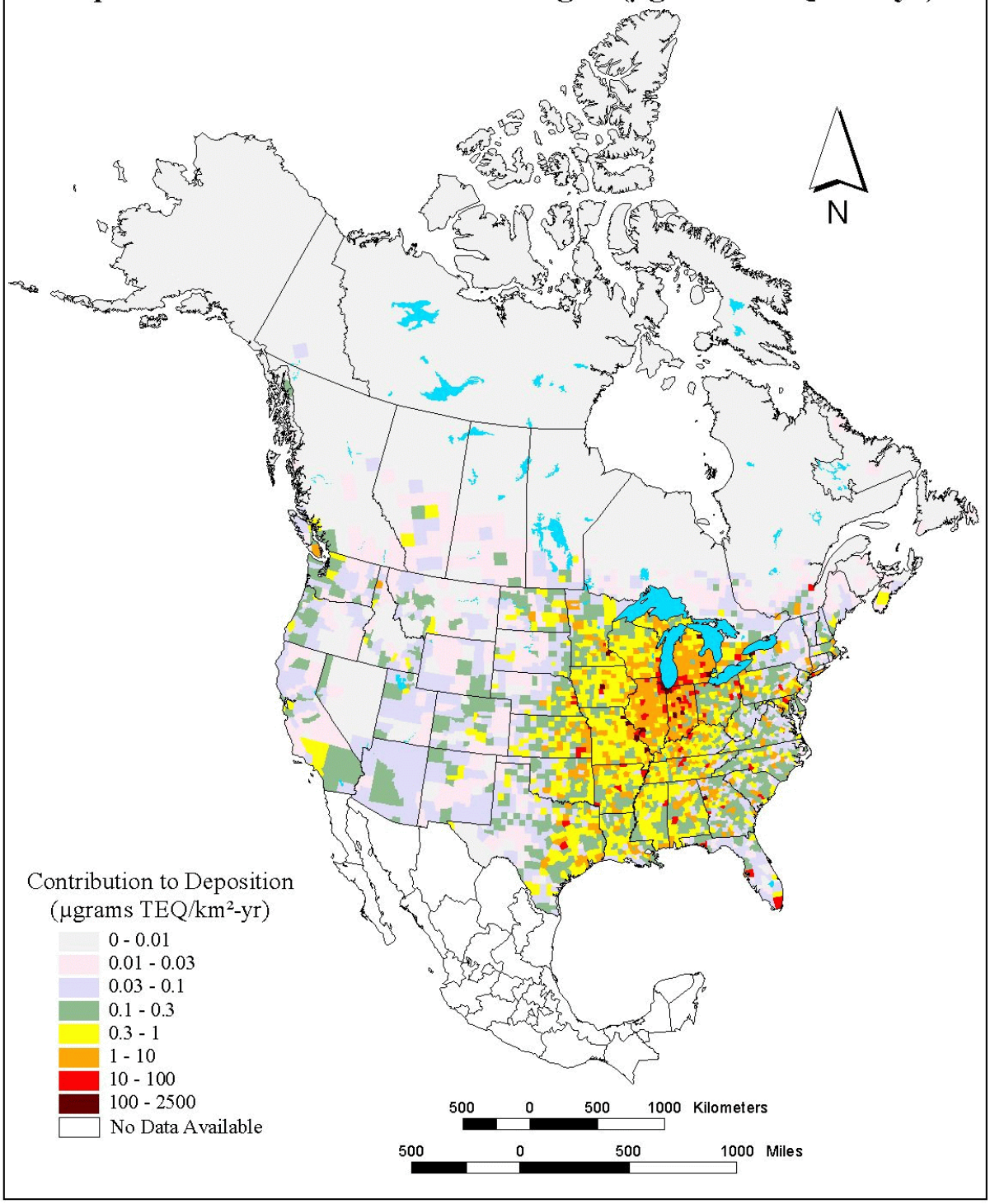
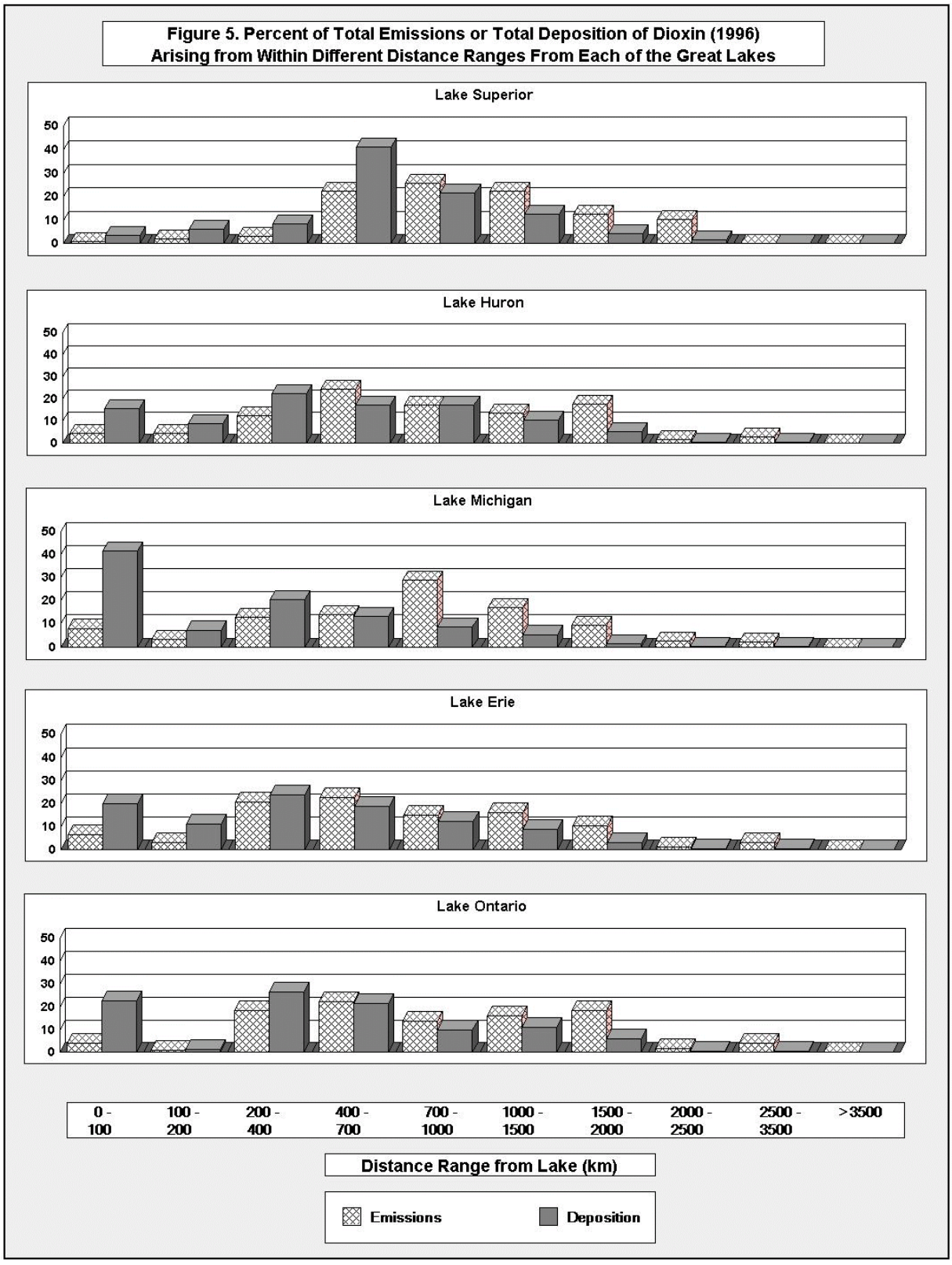


Figure 5. Percent of Total Emissions or Total Deposition of Dioxin (1996) Arising from Within Different Distance Ranges From Each of the Great Lakes



Michigan originates from within 100 km of the lake. For all of the other lakes, the fraction of the deposition originating from sources within 100 km is less than this. The estimated total dioxin deposition flux to Lake Michigan for 1996 is on the order of 17 grams TEQ/year and the uncertainty range due solely to the estimated uncertainties in the emissions is 5 - 53 (approximately a factor of 3 on each side of the central estimate).

F. Further Discussion of Uncertainties and Limitations in the Modeling

The largest source of uncertainty in the analysis is the uncertainty in the emissions inventory. However, there is also, obviously, uncertainty in the modeling methodology, including the meteorological data used, the characterization of atmospheric dispersion, and algorithms describing chemical transformation and deposition processes. The largest such uncertainty may be the choice of algorithm used to estimate dry deposition of small atmospheric particles to water bodies. The approach used in this analysis is that proposed by Slinn and Slinn[13], with a correction for humidity-induced particle growth near the water surface. Future work could attempt to characterize this and other non-emissions-related modeling uncertainties. In addition, this analysis included only sources in the United States and Canada. Sources in other regions will not likely add significantly to the loading of dioxin to the Great Lakes, but this could be tested, as well, in future work.

4. POLICY IMPLICATIONS

This White Paper began by suggesting that there were three fundamental, interdependent information areas necessary to develop toxics reduction strategies -- (a) effects; (b) causes, and (c) remediation options and costs. However, there is one additional, critical question: *How precise must one know the answers to these basic questions in order to make rational policy decisions?*

There are inherent uncertainties in each of the three areas, and so, there will always be some imprecision in our knowledge. For example, it will never be possible to precisely quantify human health effects, due to the complexities introduced by exposure to varying complex mixtures, physiological differences between individuals, complex and varying patterns of individual exposure, extrapolation of toxicological data across species and across large dosages, and other inherent difficulties. Nevertheless, important information can be assembled regarding human and ecosystem health effects, which allow at least a basis for rational decision making.

For example, the collection and use of carcinogenic risk factors, Lowest-Observed-Effect-Levels, and other similar “benchmark” data allow exposures to put in some context. Are the exposures far, far, below the level at which effects might be expected? Are the exposures far above these levels, so that effects must surely be happening? Even though precise information may be lacking (and is unlikely ever to be obtained), this type of qualitative, approximate data allows the policy-making process to go forward on some reasonable basis.

Following the same line of argument, the precise contribution of a given source to the exposure of a given population will never be able to be known precisely. Inherent variability in emissions and the inability to precisely model environmental fate and transport will always create

uncertainty in such estimates. However, to consider policy options, it can be argued that the exact contributions of individual sources to a given problem do not need to be known. Indeed, it may be enough to know about a few key issues:

- What is the geographical extent of the problem, i.e., is the problem predominantly local, regional, continental, etc., or some complex mixture of these different scales? While every effort should be made to develop the most accurate possible answer to this question, even rough approximations to the answer are useful for policy considerations. For example, it does not particularly matter, in the development of policy, if 40% or 30% of the contributing air sources arise from within 100 km of the Lake – the policy response will be similar in either case. *Only if the estimates are grossly incorrect will policy deliberations be seriously affected.*

- Which source categories are the most significant contributors? Again, while every effort to develop the most accurate estimates should be made, approximate answers are still useful in the development of policy. For example, it does not matter that much whether municipal solid waste incinerators contribute 20% or 40% to the deposition – the policy response will likely be very similar. *Again, the estimates will be of little or no use only if they are extremely inaccurate.*

5. CONCLUSIONS AND RECOMMENDATIONS

There are three basic areas of knowledge necessary for developing toxics reduction strategies for dioxin in Lake Michigan -- effects, causes, and the costs of addressing the causes. The degree of knowledge in each of these areas is limited, and that this uncertainty plays an integral role in the discussion of toxics reduction strategies. Uncertainties in the assessment of atmospheric deposition should be considered along with uncertainties in other aspects of the overall policy analysis.

More “exact” answers to questions regarding atmospheric loading can and should be pursued. However, the rough approximations that are available now are a useful starting point for policy deliberations. To the extent that additional accuracy is desired, there are some relatively straightforward actions that can be taken to decrease many of the uncertainties in the analysis of the atmospheric deposition pathway for dioxin loading. These include, but are not necessarily limited to, the following:

- Ambient monitoring for dioxin must be increased in the Great Lakes region. This will allow model evaluation and independent semi-empirical estimates of atmospheric deposition to be made.

- Additional efforts to improve the accuracy of emissions inventories – including timely updates – must be made. Timely (e.g., annual) updates for at least the largest sources in the inventory would be extremely helpful, because often, these largest sources tend to drive the analysis. If they can be better characterized, the accuracy of the overall analysis can be greatly (and relatively easily) improved.

- More information on other non-atmospheric loading pathways needs to be collected in order to more accurately place the atmospheric contributions in their proper context.

These recommendations are technically and administratively very manageable. They are small but necessary steps toward providing better answers for the development of toxics reduction strategies.

REFERENCES

1. International Joint Commission (2000). **1997-1999 Priorities and Progress Under the Great Lakes Water Quality Agreement**, Windsor, Ontario.

2. Pearson, R.F., D.L. Swackhamer, S.J. Eisenreich, and D.T. Long (1998). "Atmospheric Inputs of Polychlorinated Dibenzo-p-dioxins and Dibenzofurans to the Great Lakes: Compositional Comparison of PCDD and PCDF in Sediments." J. Great Lakes Research 24(1): 65-82.

3. Cohen, M., *et al.*, 1995. **Quantitative Estimation of the Entry of Dioxins, Furans, and Hexachlorobenzene into the Great Lakes from Airborne and Waterborne Sources.** Flushing, NY: CBNS. Final Report to the Joyce Foundation.

4. Commoner, B., M. Cohen, *et al.*, 1996. **Zeroing Out Dioxin in the Great Lakes: Within Our Reach**. Flushing, NY: CBNS. Excerpts reprinted in New Solutions 7(2):98-103 (Winter 1997), Univ. of Mass., Lowell.

5. Cohen, M., *et al.*, 1997. **Development and Application of an Air Transport Model for Dioxins and Furans**, Organohalogen Compounds **33**:214-219.

6. Commoner, B., Richardson, J. and Cohen, M., *et al.*, 1998. **Dioxin Sources, Air Transport, and Contamination in Dairy Feed Crops and Milk**. Flushing NY: CBNS.

7. Cohen, M., D. Meyer, L. Mathewson, J. McDonald, and D. Ratte, 1999. **Sources and Source Regions Contributing to Atmospheric Deposition of Toxic Pollutants to the Great Lakes – A Case Study for Dioxin**, Proceedings of the Air and Waste Management Association's Conference – The Emissions Inventory: Regional Strategies for the Future, Raleigh, NC, October 26-28.

8. Cohen, M., Mathewson, L., Artz, R., and Draxler, R., 2000. **The Atmospheric Transport and Deposition Of Dioxin to the Great Lakes**. Organohalogen Compounds **45**: 252-255.

9. US EPA (1998), The Inventory of Sources of Dioxin in the United States. External Review Draft. EPA/600/P-98/002Aa. Office of Research and Development, Washington D.C.

10. Environment Canada and the Federal/Provincial Task Force on Dioxins and Furans (1999), Dioxins and Furans and Hexachlorobenzene Inventory of Releases.

11. Van den Berg *et al.* (1998) Environmental Health Perspectives 106(12): 775-792

12. Draxler, R., and G.D. Hess (1998) Australian Meteorological Magazine. 47(4): 295-308.

13. Slinn, S.A. and W.G.N. Slinn (1980) Atmospheric Environment 14: 1013-1016.