EPA/600/A-92/234

92-84.17

To be presented at the 85th Annual Meeting
of the Air & Waste Management Association,
Kansas City, MO, June 23-27, 1992

Model Calculations of the Annual Atmospheric Deposition of Toxic Metals to Lake Michigan

Terry L. Clark*
Atmospheric Sciences Modeling Division
Air Resources Laboratory
National Oceanic and Atmospheric Administration
Research Triangle Park, North Carolina 27711

Pamela Blakley
Air and Radiation Division
U.S. Environmental Protection Agency
77 West Jackson Street
Chicago, Illinois 60604

George Mapp
Computer Sciences Corporation
3210 Chapel Hill-Nelson Highway
Research Triangle Park, North Carolina 27711

* On assignment to the Atmospheric Research and Exposure Assessment Laboratory, U.S. Environmental Protection Agency

INTRODUCTION

The presence of toxic substances in the Great Lakes creates significant environmental risks for both human and wildlife populations. Originally, concern was focused on water quality degradation due to point source discharges into waterways as the primary source of toxic input. However, for many toxic substances the atmosphere is considered to be a substantial contributor of loadings.

The 1990 Clean Air Act Amendments include specific provisions to study significant sources of atmospheric deposition of toxic substances and their impacts on the health and welfare of the Great Lakes and other major water bodies. This act also requires an assessment of the atmospheric loadings to the Great Lakes and other major water bodies. However, because of cost and technological limitations, spatially-integrated atmospheric deposition to a body of water can not be directly determined. Until recently, the total annual atmospheric deposition was estimated from the product of (1) spatially-limited, land-based, rural air concentrations, (2) constant theoretical or empirical atmospheric removal rates, and (3) the surface area of the water body[1].

Although it is expedient, this approach, with its inherent assumptions, has several serious drawbacks that cast doubts on the spatially-integrated atmospheric loadings. First, this approach assumes that both air concentrations and removal rates are constant everywhere over the water body. Secondly, the approach assumes that the land-based rural air concentrations at one or several remote sites are representative of those over the entire water body.

These assumptions may not be valid. In reality air concentrations and removal rates across large areas the size of the Great Lakes are both spatially and temporally inhomogeneous. Moreover, since the sources of airborne toxic emissions of many pollutants are concentrated within or near large urban areas, air concentrations near urban areas are greater than in remote areas. This must be considered when calculating total atmospheric deposition to the lake. Finally, because of the frequent shallow tropospheric marine layer and its resulting atmospheric stability, air pollutants may never reach the water surface[2]. Thus, the air concentrations at the air/water interface may be considerably lower than those at the nearby air/land interface.

Comprehensive atmospheric deposition models based on future enhanced versions of currently-available regional air pollution models offer a more accurate alternative to this traditional approach. However, since a better understanding of the emission rates and atmospheric processes governing airborne toxic deposition is a prerequisite for the enhanced model versions, these comprehensive models will not be operational within the next several years. In the interim, the Regional Lagrangian Model of Air Pollution (RELMAP)[3], a

simple atmospheric deposition model, was applied to quantify the expected range of annual deposition amounts of toxic trace metals to Lake Michigan. These metals are arsenic (As), cadmium (Cd), chromium (Cr), lead (Pb), and nickel (Ni).

INTERIM MODELING APPROACH

RELMAP is a four-layer Lagrangian puff model that calculates, for each 3-hour time step, the pollutant mass within and the amount of pollutant mass deposited from each of a series of puffs. The heights of the top 3 layers are 200 m, 700 m and the monthly mean height of the daytime mixed layer. Only at night a fourth surface-based layer exists, the depth of which ranges from 30 m to 50 m, depending on the time of year. Pollutant puffs are released within the 200-700-m layer every 3 hours from virtual sources located at the emission-weighted center of each unit-degree source cell (Figure 1).

The model input data consist of hourly precipitation amounts from approximately 2000 sites, surface and 850-mb wind velocities, climatological atmospheric stability categories and mixing heights, and spatial distributions of 11 land-use categories. As puffs are transported across the model domain, both dry and wet deposition amounts of As, Cd, Cr, Pb, and Ni are calculated for the land and water areas of each of the receptor cells depicted by Figure 1. The model was applied for the entire year of 1985, the year when U.S. and Canadian emissions data were available.

Dry deposition of particles is calculated for each 3-h time step as the product of a surface-layer air concentration (C_a) and a time-dependent dry deposition velocity (v_d). Since this latter term varies with season, land-use, and atmospheric stability, the simulated dry deposition rates exhibit considerable diurnal, seasonal and spatial variabilities. Currently the model assumes that the water surface is always in a liquid phase. Since it also assumes a uniform particle size, the model was applied twice using two different particle sizes: 0.5 microns and 5.0 microns, the range of particle sizes of importance to regional deposition. Wet deposition of particles is based on the product of a dimensionless washout ratio and the hourly precipitation rate (mm/h) raised to the power of 0.622^4.

In this study annual air emissions of the toxic metals were calculated from point and area source parameters in the 1985 National Acid Precipitation Assessment Program (NAPAP) emissions inventory and published pollutant emission factors. Emissions from both Canadian and United States sources were then gridded to the unit-degree RELMAP configuration. Although emissions from all states and provinces within the Figure 1 domain were used in the model applications, Table 1 provides the annual emission rates for each state within Region V, the states nearest Lake Michigan. By virtue of the proximity of the sources to the lake, emissions from these states will have a significant contribution to the deposition to Lake Michigan.

MODEL CALCULATIONS OF ANNUAL DEPOSITION

The annual deposition amounts of any pollutant to Lake Michigan depend on the emission rate, proximity of sources, meteorological factors, and removal efficiency. Since the emissions rate of Pb was an order of magnitude greater than the emission rates of the other four metals, its annual deposition to Lake Michigan was also an order of magnitude greater, as Table 2 demonstrates. Pb deposition, approaching 700,000 kg/yr, dominated the total deposition of the remaining metals, 18,000 kg/yr to 58,000 kg/yr.

Table 2 also shows that the relative contribution of dry deposition to total deposition is highly dependent on the assumed particle diameter. For smaller particle sizes (e.g., 0.5 micron) dry deposition accounted for 10% or less of the total deposition. On the other hand, dry deposition from larger particles (e.g., 5.0 microns) accounted for nearly 40% of the total deposition. The total deposition for the two particle sizes, however, differs by less than 20% for As and by approximately 10% for the other metals.

The slight difference in total deposition can be explained by the fact that when dry deposition is small more mass exists in the atmosphere for wet deposition. The greater difference in total As deposition amounts appears to be linked to the spatial distribution of As sources; As emissions from sources near the lake were much lower than the emissions of the other metals. That is, long range transport appears to play a more important role for As.

An independent estimate of atmospheric Pb deposition to Lake Michigan was presented by Strachan and Eisenreich[1]. They based their annual estimate on a typical precipitation rate, washout rate, dry deposition velocity, and air concentration. From this mass balance exercise, they estimated an annual atmospheric deposition rate of 543,000 kg/yr, approximately 20% lower than the RELMAP calculation. It is not surprising for the model calculation to be greater than the mass balance estimate, since the latter approach ignores the urban contribution the former approach considers. That is, typical air concentrations used in the mass balance approach reflect only the lower rural/remote air concentrations only.

CONCLUSIONS AND FUTURE PLANS

Although toxic emission inventories and current regional toxic models are in states of infancy, annual atmospheric deposition to Lake Michigan was calculated to provide a preliminary assessment. As the quality and completeness of emission inventories improve and as modeled processes are refined, these annual deposition amounts will be recalculated and uncertainties will be reduced.

This preliminary modeling exercise for five toxic metals indicated that Cd deposition was lowest of the five toxic metals, approximately 20,000 kg/yr. Cr and Ni deposition ranged approximately from 26,000 kg/yr to 35,000 kg/yr. As, showing a wide range of deposition amounts based on the range of assumed particle sizes, varied from approximately 40,000 kg/yr to 58,000 kg/yr. Pb deposition by far was the greatest, ranging from 675,000 kg/yr to nearly 690,000 kg/yr. The 1985-calculated Pb deposition amount is likely greater than that for subsequent years, since Pb emissions from motor vehicles decreased dramatically after the mid-1980's.

In the future, using source-classification-code (SCC) specific emission grids for each pollutant, RELMAP will determine the relative contribution of various source types to the total deposition to Lake Michigan. The modeling exercise will also be extended to Lakes Superior, Huron, Erie, and Ontario, in that order. Efforts will continue to compare published monitoring values and deposition estimates with modeled air concentrations and deposition amounts.

ACKNOWLEDGEMENTS

The authors wish to recognize the valuable air toxics inventory efforts of Bill Benjey, Dale Coventry, and Alfrieda Rankins of the Atmospheric Research and Exposure Assessment Laboratory at Research Triangle Park, North Carolina. Without their assistance this modeling effort would not have been possible.

NOTICE

This paper has been reviewed in accordance with U.S. Environmental Protection Agency's peer review and administrative review policies and approved for presentation and publication. Mention of trade names or commercial products does not constitute endorsement or recommendation for use.

REFERENCES

1. W.M.J. Strachan and S.J. Eisenreich, *Mass Balancing of Toxic Chemicals in the Great Lakes: The Role of Atmospheric Deposition*, Report to the International Joint Commission, Windsor, Ontario, 1988, pp. 50-52.

2. W.A. Lyons, R.A. Pielke, J.L. Eastman, *et al.*, "Mesoscale numerical model evaluations of the meteorological factors associated with elevated ozone levels in the southern Lake Michigan region", in Proceedings of the 1991 AMS/AWMA Conference on Applications of Air Pollution Meteorology, P7.10, American Meteorological Society, Boston, MA, 1991, pp. 164-167.

3. B.K. Eder, D.H. Coventry, T.L. Clark, *et al.*, *RELMAP: A Regional Lagrangian Model of Air Pollution User's Guide*, EPA-600/8-86/013, U.S. Environmental Protection Agency, Research Triangle Park, NC, 1986.

4. T.L. Clark, R.L. Dennis, E.C. Voldner, *et al.*, *The International Sulfur Deposition Model Evaluation*, EPA/600/3-87/008, U.S. Environmental Protection Agency, Research Triangle Park, NC, 1987, p. 20.

TABLE 1. Annual emissions of toxic metals from states within REGION V

Based on 1985 NAPAP Emissions Data
and Emission/Speciation Factors from Various Years

Pollutant	Emissions Rate (kg/yr)	State
Arsenic	**1976**	**Total**
	798	Minnesota
	653	Illinois
	205	Ohio
	141	Indiana
	106	Michigan
	73	Wisconsin
Cadmium	**656**	**Total**
	234	Minnesota
	188	Illinois
	79	Ohio
	69	Indiana
	61	Michigan
	25	Wisconsin
Chromium	**1183**	**Total**
	318	Indiana
	275	Illinois
	234	Michigan
	219	Ohio
	99	Minnesota
	38	Wisconsin

TABLE 1. Continued

Pollutant	Emissions Rate (kg/yr)	State
Lead	**28518**	**Total**
	6287	Illinois
	6174	Ohio
	5182	Michigan
	4111	Indiana
	4084	Minnesota
	2680	Wisconsin
Nickel	**983**	**Total**
	384	Illinois
	154	Indiana
	141	Ohio
	122	Michigan
	115	Minnesota
	66	Wisconsin

TABLE 2. RELMAP calculations of the 1985 deposition (kg/yr) of toxic metals to Lake Michigan

Pollutant	Particle Size (μm)	Dry Deposition	Wet Deposition	Total
Arsenic	0.5	5,870 (10.0%)	52,552 (90.0%)	58,422
	5.0	19,033 (39.2%)	29,484 (60.8%)	48,517
Cadmium	0.5	2,053 (10.0%)	18,575 (90.0%)	20,628
	5.0	7,179 (39.2%)	11,122 (60.8%)	18,301
Chromium	0.5	2,908 (8.7%)	30,678 (91.3%)	33,586
	5.0	13,129 (37.1%)	22,275 (62.9%)	35,404
Lead	0.5	62,456 (9.1%)	627,306 (90.9%)	689,762
	5.0	254,201 (37.7%)	420,818 (62.3%)	675,019
Nickel	0.5	2,483 (8.3%)	27,489 (91.7%)	29,972
	5.0	9,539 (35.9%)	17,069 (64.1%)	26,608

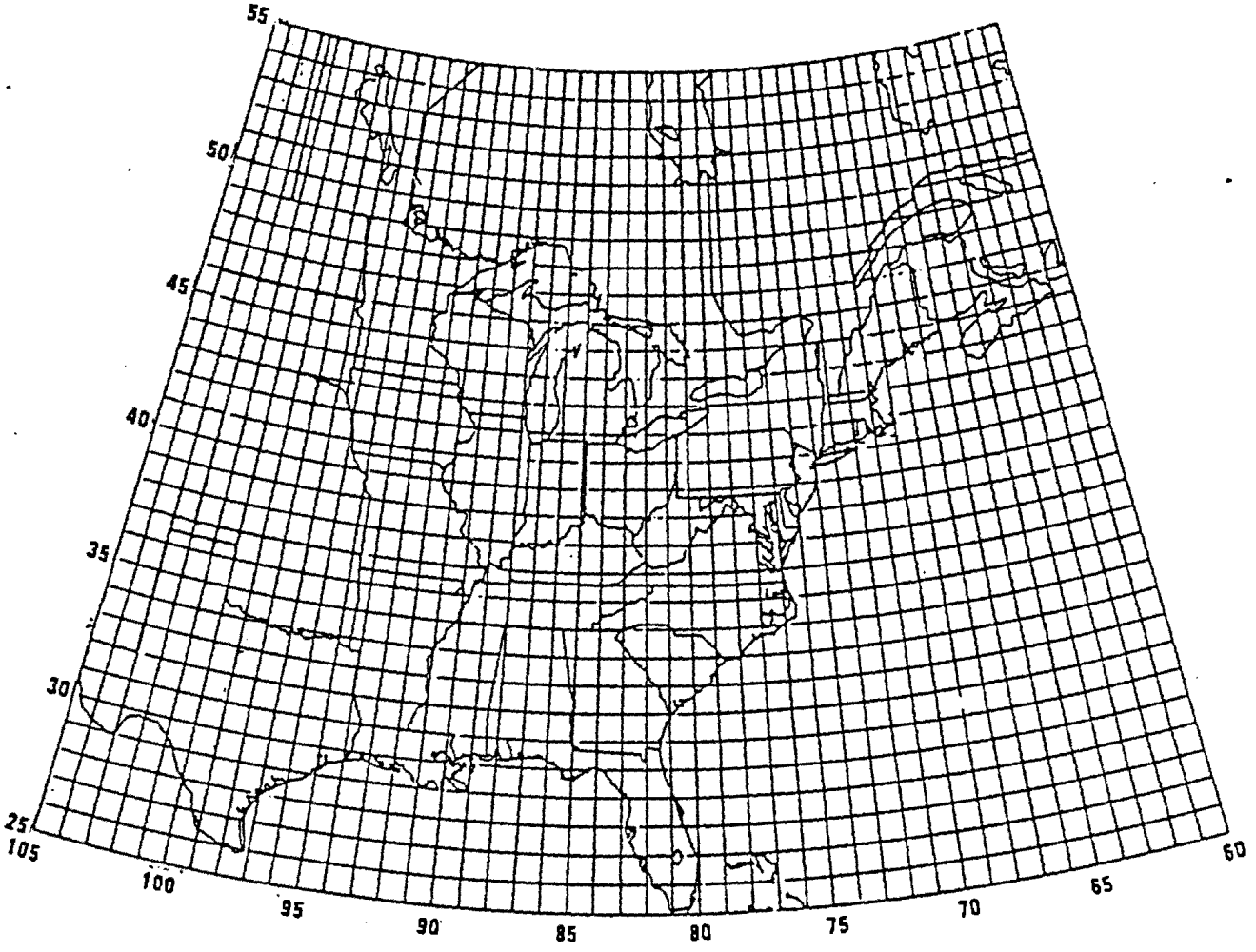

FIGURE 1. The unit-degree grid configuration of the Regional Lagrangian Model of Air Pollution (RELMAP).

TECHNICAL REPORT DATA (Please read Instructions on the reverse before comple)		
1. REPORT NO. EPA/600/A-92/234	2.	3.
4. TITLE AND SUBTITLE MODEL CALCULATIONS OF THE ANNUAL ATMOSPHERIC DEPOSITION OF TOXIC METALS TO LAKE MICHIGAN		5. REPORT DATE 6. PERFORMING ORGANIZATION CODE
7. AUTHOR(S) Terry L. Clark		8. PERFORMING ORGANIZATION REPORT NO.
9. PERFORMING ORGANIZATION NAME AND ADDRESS Same as 12.		10. PROGRAM ELEMENT NO. A101G/C/72/01 11. CONTRACT/GRANT NO. Inhouse
12. SPONSORING AGENCY NAME AND ADDRESS Atmospheric Research & Exposure Assessment Laboratory Office of Research and Development U.S. Environmental Protection Agency Research Triangle Park, North Carolina 27711		13. TYPE OF REPORT AND PERIOD COVERED Interim 4/91-5/92 14. SPONSORING AGENCY CODE EPA/600/09
15. SUPPLEMENTARY NOTES Presented at the 85th Annual Meeting of the Air & Waste Management Association, Kansas City, MO., June 23-27, 1992		

16. ABSTRACT

The 1990 Clean Air Act Amendments include specific provisions to study significant sources of atmospheric deposition of toxic substances and their impacts on the health and welfare of the Great Lakes and other major water bodies. This act also requires an assessment of the atmospheric loadings to the Great Lakes and other major water bodies. However, because of cost and technological limitations, spatially-integrated atmospheric deposition to a body of water can not be directly determined. Until recently, the total annual atmospheric deposition was estimated from the product of (1) spatially-limited, land-based, rural air concentrations, (2) constant theoretical or empirical atmospheric removal rates, and (3) the surface area of the water body. Although it is expedient, this approach, with its inherent assumptions, has several serious drawbacks that cast doubts on the spatially-integrated atmospheric loadings.

Comprehensive atmospheric deposition models based on future enhanced versions of currently-available regional air pollution models offer a more accurate alternative to this traditional approach. However, since a better understanding of the emission rates and atmospheric processes governing airborne toxic deposition is a prerequisite for the enhanced model versions, these comprehensive models will not be operational within the next several years. In the interim, the Regional Lagrangian Model of Air Pollution (RELMAP), a simple atmospheric deposition model, was applied to quantify the expected range of annual deposition amounts of toxic trace metals to Lake Michigan. These metals are arsenic (As), cadmium (Cd), chromium (Cr), lead (Pb), and nickel (Ni).

17. KEY WORDS AND DOCUMENT ANALYSIS		
a. DESCRIPTORS	b. IDENTIFIERS/OPEN ENDED TERMS	c. COSATI Field/Group

18. DISTRIBUTION STATEMENT	19. SECURITY CLASS (This Report)	21. NO. OF PAGES
RELEASE TO PUBLIC	UNCLASSIFIED	12
	20. SECURITY CLASS (This page)	22. PRICE
	UNCLASSIFIED	