

# ATMOSPHERIC DEPOSITION OF TOXIC SUBSTANCES TO THE GREAT LAKES: IADN RESULTS THROUGH 1998



<b>Stephanie Buehler</b>	School of Public and Environmental Affairs and Department of Chemistry, Indiana University
<b>William Hafner</b>	School of Public and Environmental Affairs and Department of Chemistry, Indiana University
<b>Ilori Basu</b>	School of Public and Environmental Affairs and Department of Chemistry, Indiana University
<b>Céline V. Audette</b>	Center for Atmospheric Research Experiments, Meteorological Service of Canada, Environment Canada
<b>Kenneth A. Brice</b>	Air Quality Research Branch, Meteorological Service of Canada, Environment Canada
<b>C.H. Chan</b>	Ecosystem Health Division, Ontario Region, Environment Canada
<b>Frank Froude</b>	Center for Atmospheric Research Experiments, Meteorological Service of Canada, Environment Canada
<b>Elisabeth Galarneau</b>	Air Quality Research Branch, Meteorological Service of Canada Environment Canada
<b>Melissa L. Hulting</b>	Great Lakes National Program Office, U.S. Environmental Protection Agency
<b>Liisa Jantunen</b>	Air Quality Research Branch, Meteorological Service of Canada, Environment Canada
<b>Melanie Neilson</b>	Ecosystem Health Division, Ontario Region, Environment Canada
<b>Keith Puckett</b>	Air Quality Research Branch, Meteorological Service of Canada Environment Canada
<b>Ronald A. Hites</b>	School of Public and Environmental Affairs and Department of Chemistry, Indiana University

Atmospheric Deposition of Toxic Substances to the Great Lakes: IADN Results through 1998

Published by  
Environment Canada and the United States Environmental Protection Agency

ISBN: 0-662-31219-8

Public Works and Government Services Canada Catalogue Number: En56-156/1998E

US EPA Report Number: 905-R-01-007

Report available in printed form from

Air Quality Research Branch  
Environment Canada  
4905 Dufferin Street  
Toronto ON  
M3H 5T4  
Canada

Great Lakes National Program Office  
U.S. Environmental Protection Agency  
77 West Jackson Boulevard (G17-J)  
Chicago IL  
60604  
U.S.A.

and in electronic form at

[www.msc.ec.gc.ca/IADN/](http://www.msc.ec.gc.ca/IADN/)

[www.epa.gov/glnpo/iadn/](http://www.epa.gov/glnpo/iadn/)

# ATMOSPHERIC DEPOSITION OF TOXIC SUBSTANCES TO THE GREAT LAKES: IADN RESULTS THROUGH 1998

## Executive Summary

The Integrated Atmospheric Deposition Network (IADN) was established in 1990 to determine the magnitude and trends of atmospheric loadings of toxic contaminants to the Great Lakes. By maintaining a master station on each of the Great Lakes, IADN is able to monitor the atmospheric deposition of selected pollutants. These data have been used to calculate loadings estimates to the Great Lakes from 1992 to 1996. IADN incorporates three atmospheric deposition processes into its loadings estimates: wet deposition, dry particle deposition, and net gas exchange which combines the processes of gas absorption (air to water) and volatilization (water to air). This document reports the biennial loadings to the Great Lakes for 1997 and 1998.

A subset of the substances measured at the IADN master stations are used in the loadings calculations. These substances are the pesticides  $\alpha$ - and  $\gamma$ -hexachlorocyclohexane (HCH), dieldrin, *p,p'*-DDE, *p,p'*-DDD, *p,p'*-DDT, *trans*-nonachlor, *trans*- and *cis*-chlordane,  $\alpha$ -endosulfan, and endosulfan sulfate; hexachlorobenzene (HCB) and polychlorinated biphenyl (PCB) congeners 18, 44, 52, and 101, as well as a sum of 56 PCB congeners and coeluting congener groups expressed as "suite-PCB"; the individual polycyclic aromatic hydrocarbons (PAHs) phenanthrene, pyrene, benzo[*k*]fluoranthene, benzo[*b*]fluoranthene, indeno[1,2,3-*cd*]pyrene and benzo[*a*]pyrene as well as a the total of four of these PAHs expressed as sum-PAH; and the trace metals lead, arsenic, selenium, and cadmium.

The 1997-1998 loadings presented in this report were calculated in a manner consistent with the 1995-1996 loadings. The estimates are presented here as flows (kg/yr) to better understand the amount of the substances being deposited to the lakes, and as fluxes (ng/m<sup>2</sup>/day) to account for differences in lake areas and to facilitate spatial trend analysis.

Downward fluxes for pesticides in 1997 and 1998 ranged from 0.01 ng/m<sup>2</sup>/day to 40 ng/m<sup>2</sup>/day, with in-use pesticides such as  $\gamma$ -HCH accounting for the highest fluxes. Volatilization fluxes for those pesticides banned from use were almost 10 times greater than those for currently used pesticides, reaching -37 ng/m<sup>2</sup>/day at their highest. PCB and HCB downward fluxes ranged from 0.02 ng/m<sup>2</sup>/day to 11 ng/m<sup>2</sup>/day across the basin. Volatilization fluxes for these banned commercial chemicals were on the same order as those for banned pesticides. Suite-PCB volatilization fluxes increased from west to east across the basin. Downward fluxes for PAHs ranged from 0.3 ng/m<sup>2</sup>/day to 530 ng/m<sup>2</sup>/day with volatilization fluxes ranging from -0.00001 to -240 ng/m<sup>2</sup>/day. Where water concentration data are available, volatilization of PAHs was almost always less than net inputs. Fluxes for metals ranged from 13 to 840 ng/m<sup>2</sup>/day for dry deposition and from 130 to 5400 ng/m<sup>2</sup>/day for wet deposition. Since the metals analyzed by IADN are nonvolatile, they are not measured in the gas phase. The PAHs and metals measured by IADN are currently emitted through anthropogenic means into the atmosphere and

thus have downward (air to water) fluxes much greater than those of the pesticides and PCBs that have been banned from use.

Current (1997-1998) fluxes ( $\text{ng}/\text{m}^2/\text{day}$ ) were compared across time and space to better understand loadings trends in the Great Lakes. Pesticide wet deposition fluxes seem to be generally decreasing over time except for  $\gamma$ -HCH at Lakes Huron and Ontario. Since  $\gamma$ -HCH is still in use, this trend is expected. Volatilization of dieldrin from Lake Ontario is the largest pesticide flux observed. The magnitude of PCB wet deposition fluxes is similar for Lakes Superior, Erie, and Michigan. Lake Erie, however, seems unique in that all PCBs measured there reached peak fluxes around 1994 and 1995 and then decreased in the following three years. Gas exchange of PCBs has been, for the most part, in the direction of net volatilization consistently over time with only Lake Michigan showing signs of nearing air-water equilibrium. Wet and dry deposition of PAHs shows no real temporal trend, but spatial analysis indicates that fluxes have increased from west to east across the basin. Gas exchange fluxes for Lakes Superior and Erie for all PAHs show net absorption over time. Metal fluxes for Lakes Huron and Ontario are similar over the years with dry deposition showing no real trend and wet deposition decreasing from 1992-1996 for Cd and Pb, then increasing in 1997 and 1998.

All of the flows and fluxes mentioned above are based on IADN master station data. These stations are remote sites, one on each lake, which measure what are considered to be Great Lakes background contaminant levels. However, spatial differences exist across each lake for many of the compounds we monitor, particularly near urban areas, where atmospheric deposition from cities can be much greater than that from remote sites. In an attempt to assess the impact of urban areas on lakewide loadings, and in accordance with the 1995-1996 loadings report, depositional data from IADN's Chicago site were extrapolated onto Lake Michigan loadings. The impact of Chicago pollution on a small sub-area of Lake Michigan was then compared to loadings calculated at the remote master station. Results demonstrate that urban inputs have a minor lakewide effect for most pesticides. There does, however, seem to be a large effect on *cis*- and *trans*-chlordane, drastically changing lakewide volatilization and markedly increasing total mass loadings. Urban inputs also have a strong effect on the net gas exchange of PCBs. PAHs, currently emitted urban pollutants, show consistently large urban effects in all deposition categories.

In an attempt to explore a more tangible means of examining the loadings results, estimates were investigated on a Great Lakes basin-wide basis by summing the total deposition flow ( $\text{kg}/\text{yr}$ ) of each substance over all five lakes for each year. These sums give a good approximation of the larger, regional atmospheric deposition to the Great Lakes. Total deposition for  $\alpha$ -HCH showed a decreasing trend, going from 950  $\text{kg}/\text{yr}$  in 1992 to -210  $\text{kg}/\text{yr}$  in 1998. Dieldrin and *p,p'*-DDE, two organochlorine pesticides banned from use, had negative total deposition across time, indicating that the lakes are acting as a source of these chemicals to the atmosphere. Sum-PCB total deposition across the basin also showed net volatilization for all years. Even so, PCB flows out of the Great Lakes have decreased dramatically over time, with the largest drop occurring between 1994 and 1995 when total PCB flows went from -3100  $\text{kg}/\text{yr}$  to -940  $\text{kg}/\text{yr}$ . PAHs and metals had

the largest regional deposition. PAH flows have, for the most part, remained stable across time. While total loads of metals to the Great Lakes basin have decreased over time, the region was still receiving 78000 kg of lead in 1998.

# DÉPÔT ATMOSPHÉRIQUE DE SUBSTANCES TOXIQUES DANS LES GRANDS LACS : RÉSULTATS DU RMDA JUS- QU'EN 1998

## Résumé

Créé en 1990, le Réseau de mesure des dépôts atmosphériques (RMDA) détermine l'ampleur et les tendances des charges atmosphériques de contaminants toxiques des Grands Lacs. Par le maintien d'une station principale sur chacun des Grands Lacs, le RMDA peut surveiller le dépôt atmosphérique de polluants sélectionnés. Ces données ont servi à estimer les charges des Grands Lacs de 1992 à 1996. Le RMDA incorpore trois processus de dépôt atmosphérique dans ses estimations de charges : dépôt humides, dépôts de particules sèches et échange net de gaz qui combine les phénomènes d'absorption des gaz (de l'air à l'eau) et de volatilisation (de l'eau à l'air). Ce document rend compte des charges biennales des Grands Lacs de 1997 et de 1998.

Un sous-ensemble des substances mesurées aux stations principales du RMDA sert au calcul des charges. Il s'agit des pesticides  $\alpha$ - et  $\gamma$ -hexachlorocyclohexane (HCH), de la dieldrine, *p,p'*-DDE, *p,p'*-DDD, *p,p'*-DDT, de l'endosulfane *trans*-nonachlor, *trans*- et *cis*-chlordane, du  $\alpha$ -endosulfane et du sulfate d'endosulfane; des substances organoleptiques 18, 44, 52 et 101 de l'hexachlorobenzène (HCB) et du biphenyle polychloré (BPC), ainsi que d'une somme de 56 substances organoleptiques et groupes de substances organoleptiques coéluantes exprimées sous la forme de «suite-BPC» des hydrocarbures aromatiques polycycliques (HAP); du phénanthrène, du pyrène, du benzo[*k*]fluoranthène, du benzo[*b*]fluoranthène, de l'indeno[1,2,3-*cd*]pyrène et du benzo[*a*]pyrène, ainsi que du total de quatre de ces HAP exprimés sous forme de somme-HAP; et des métaux à l'état de traces que sont le plomb, l'arsenic, le sélénium et le cadmium.

Les charges de 1997-1998 présentées dans ce rapport ont été calculées d'une façon compatible avec les charges de 1995-1996. Les estimations sont présentées ici sous forme de débit (kg/an) pour mieux comprendre la quantité des substances qui se déposent dans les lacs et sous forme de débit (ng/m<sup>2</sup>/jour) pour rendre compte des différences existant dans les zones lacustres et faciliter l'analyse des tendances spatiales.

Les débits descendants des pesticides en 1997 et en 1998 se sont situés entre 0,01 ng/m<sup>2</sup>/jour et 40 ng/m<sup>2</sup>/jour, les pesticides en usage comme le  $\gamma$ -HCH causant les débits les plus élevés. Les débits de volatilisation pour les pesticides dont l'utilisation est interdite ont été au moins dix fois plus grands que ceux des pesticides utilisés à l'heure actuelle, atteignant -37 ng/m<sup>2</sup>/jour à leur maximum. Les débits descendants de BPC et de HCB se sont situés entre 0,02 ng/m<sup>2</sup>/jour et 11 ng/m<sup>2</sup>/jour dans tout le bassin. Les débits de volatilisation, pour ces produits chimiques commerciaux interdits, étaient analogues à ceux des pesticides interdits. Les débits de volatilisation du Suite-BPC ont augmenté de l'ouest à l'est dans tout le bassin. Les débits descendants des HAP sont allés de 0,3 ng/m<sup>2</sup>/jour à 530 ng/m<sup>2</sup>/jour, les débits de volatilisation se situant entre -0,00001 et -240 ng/m<sup>2</sup>/jour. Dans les cas où l'on disposait des données de concentration de l'eau, la vola-

tilisation des HAP a été presque toujours inférieure aux apports nets. Les débits des métaux ont oscillé entre 13 et 840 ng/m<sup>2</sup>/jour pour les dépôts secs et entre 130 et 5400 ng/m<sup>2</sup>/jour pour les dépôts humides. Comme les métaux analysés par le RMDA ne sont pas volatils, on ne les mesure pas dans la phase gazeuse. Les HAP et les métaux mesurés par le RMDA sont actuellement émis par des moyens anthropiques dans l'atmosphère et ont de ce fait des débits descendants (de l'air à l'eau) nettement supérieurs à ceux des pesticides et des BPC dont l'utilisation a été interdite.

On a comparé les débits actuels (1997-1998) (ng/m<sup>2</sup>/jour) dans le temps et l'espace pour mieux comprendre les tendances des charges dans les Grands Lacs. Les débits des dépôts humides de pesticides semblent, en général, baisser dans le temps, sauf pour le  $\gamma$ -HCH aux lacs Huron et Ontario. Comme le  $\gamma$ -HCH est encore utilisé, cette tendance est attendue. La volatilisation de la dieldrine du lac Ontario est le débit de pesticide le plus important qu'on ait observé. L'ampleur des débits de dépôts humides de BPC est analogue à celle des lacs Supérieur, Érié et Michigan. Le lac Érié semble toutefois unique en ce que tous les BPC qui y sont mesurés ont atteint leur débit maximal vers 1994 et 1995, débit qui a baissé au cours des trois années suivantes. L'échange gazeux de BPC s'est surtout manifesté par le renforcement régulier d'une volatilisation nette dans le temps, seul le lac Michigan montrant des signes de se rapprocher de l'équilibre. Les dépôts humides et secs de HAP n'indiquent aucune tendance temporelle réelle, mais une analyse spatiale révèle que les débits se sont accrus d'ouest en est dans tout le bassin. Les débits d'échanges gazeux du lac Supérieur et du lac Érié pour tous les HAP montrent une absorption nette dans le temps. Les débits de métal pour les lacs Huron et Ontario sont analogues au cours des ans avec les dépôts secs : aucune tendance véritable; dépôt humide baissant de 1992-1996 pour le Cd et le Pb, puis augmentant en 1997 et 1998.

Tous les flux et les débits mentionnés reposent sur les données de la station principale du RMDA. Il s'agit de stations reculées, une par lac, qui mesurent ce qu'on considère être les niveaux de contaminants de fond des Grands Lacs. Il existe toutefois des différences spatiales dans chaque lac pour nombre de composés que nous surveillons, en particulier près des zones urbaines, où les dépôts atmosphériques des villes peuvent nettement dépasser ceux des stations reculées. Pour tenter d'évaluer l'effet des zones urbaines sur les charges panlacustres et suivant le rapport des charges de 1995-1996, les données de dépôt de la station de Chicago du RMDA ont été extrapolées aux charges du lac Michigan. L'effet de la pollution de Chicago sur une petite zone secondaire du lac Michigan a été comparé aux charges calculées à la station principale reculée. Les résultats révèlent que les apports urbains exercent un effet panlacustre mineur pour la plupart des pesticides. Il semble bien y avoir un gros effet, toutefois, sur le cis- et le trans-chlordane, effet qui modifie énormément la volatilisation panlacustre et accroît nettement les charges totales de masse. Les apports urbains produisent aussi un grand effet sur l'échange gazeux net des BPC. Les HAP, polluants urbains rejetés à l'heure actuelle, produisent des effets régulièrement importants dans toutes les catégories de dépôts.

Pour essayer d'explorer un moyen plus tangible d'examiner les résultats des charges, on a étudié les estimations sur tout le bassin des Grands Lacs, en calculant la somme du flux total de dépôts (kg/an) de chaque substance sur les cinq lacs à la fois pour chaque année.

Ces sommes donnent une bonne approximation du dépôt atmosphérique régional, plus grand, des Grands Lacs. Les dépôts totaux de  $\alpha$ -HCH ont accusé une tendance à la baisse, en passant de 950 kg/an en 1992 à -210 kg/an en 1998. La dieldrine et p,p'-DDE, deux pesticides organochlorés dont l'utilisation est interdite, ont eu des dépôts totaux négatifs dans le temps, ce qui signale que les lacs servent de source de ces produits chimiques pour l'atmosphère. Les dépôts totaux de somme-PPC dans tout le bassin ont aussi enregistré une volatilisation nette pour toutes les années. N'empêche, les flux de BPC sortant des Grands Lacs ont énormément baissé dans le temps, la plus forte baisse survenant entre 1994 et 1995, période où les flux totaux de BPC sont tombés de -3100 kg/an à -940 kg/an. Les HAP et les métaux ont engendré le plus fort dépôt régional. Les flux de HAP sont, en grande partie, restés stables dans le temps. Le total des charges de métaux du bassin des Grands Lacs a baissé dans le temps, mais la région recevait encore 78000 kg de plomb en 1998.



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## 1. Introduction

Over the past few decades, the Great Lakes have been the recipient of many different toxic organic pollutants from a variety of sources. As the uses of many of these substances were banned, concentration levels in biota decreased and then seemed to level off in the mid-80's. One explanation for this lack of continued reduction was that the contaminants were coming from the atmosphere. In an attempt to better understand the sources of this contamination, the International Joint Commission (IJC) held a workshop in Scarborough, Canada in 1986 and requested that a paper be prepared outlining the concentration and flux data for several contaminants. The product of this workshop was a report (Strachan and Eisenreich, 1988) identifying atmospheric deposition as a significant contributor to contamination levels in the Great Lakes. This report provided enough justification to include a separate Annex (Annex 15) within the 1987 revision of the Great Lakes Water Quality Agreement of 1978.

Among other things, Annex 15 called for the determination of atmospheric loadings of toxic substances to the Great Lakes as well as continuing study of temporal and spatial trends of these substances, both to be reported to the IJC on a biennial basis. Most importantly, the Annex called for the creation of the Integrated Atmospheric Deposition Network (IADN) to carry out surveillance and monitoring of the toxic contaminants. In 1990, the first Implementation Plan (Egar and Adamkus, 1990) was signed. It laid out the chemicals to be measured, siting and sampling methods, and the Quality Assurance/Quality Control program that would ensure that data was consistent over time and between sampling agencies.

In accordance with Annex 15, IADN has produced biennial loadings estimates on data from 1992 through 1996 (Hoff, 1996; Hillery et al., 1998; Galarneau et al., 2000). This report details the 1997 and 1998 loadings to the Great Lakes and follows closely the methodology and format of the previous reports. As with the previous documents, the centerpiece of this report is atmospheric loadings. Other pathways besides atmospheric deposition do exist that may contribute to the total loadings, such as point sources, tributary input, and bubble spray production. Because of this, the atmospheric loadings presented here should not be seen to represent the only contributions of toxic contaminants to the Great Lakes.

## 2. Methods

### ***2.a. Substances Considered***

Previous IADN loadings results (Hoff, 1994; Hoff et al., 1996; Hillery et al., 1998) have reported on 20 substances. These substances are  $\alpha$ - and  $\gamma$ -hexachlorocyclohexane (HCH), dieldrin, *p,p'*-DDE, *p,p'*-DDD, *p,p'*-DDT, hexachlorobenzene (HCB), polychlorinated biphenyl (PCB) congeners 18, 44, 52, and

101, as well as total PCBs ( $\Sigma$ PCB); individual polycyclic aromatic hydrocarbons (PAHs) phenanthrene, pyrene, benzo[*k*]fluoranthene, and benzo[*a*]pyrene; and the trace metals lead, arsenic, selenium, and cadmium. The 1995-1996 loadings report (Galarneau et al., 2000) added the species *trans*-nonachlor, *trans*- and *cis*-chlordane,  $\alpha$ - and  $\beta$ -endosulfan, endosulfan sulfate, benzo[*b*]fluoranthene, and indeno[1,2,3-*cd*]pyrene. The PAHs benzo[*a*]pyrene, benzo[*k*]fluoranthene, benzo[*b*]fluoranthene, and indeno[1,2,3-*cd*]pyrene comprise a list of PAHs suggested for reporting under the United Nations Economic Commission for Europe Long-Range Transboundary Air Pollution (UNECE LRTAP) Convention's 1998 Aarhus Protocol on Persistent Organic Pollutants (POPs) (UNECE, 1998). These individual PAHs were then summed to give sum-PAH loading results.

In accordance with all previous loadings calculations, the 1997-1998 results presented here will report on the same list of 28 substances described above, with two exceptions. First,  $\beta$ -endosulfan will not be reported because no reliable Henry's Law constant could be found, putting previous results for this pesticide into question. Second,  $\Sigma$ PCB will be presented as suite-PCB for 1997 and 1998 results.  $\Sigma$ PCB represents a sum of congeners determined by each individual analytical laboratory that participates in the IADN project. In an attempt to report one consistent sum of PCB congeners across all agencies, a suite of PCBs was developed. This suite encompasses those PCB congeners that contribute an important percentage to the overall total PCB mass in gas and precipitation phases. The congeners in the suite are also considered to be toxicologically important. The resulting suite-PCB contains 56 congeners and congener groups that are representative of the entire range of PCBs (Neilson, 2000). While the number of congeners used in suite-PCB is approximately half of what was used by some agencies in previous  $\Sigma$ PCB calculations, these 56 congeners account for approximately 90% of the total PCB mass at each site. Thus, the averages found for suite-PCB are not largely different from  $\Sigma$ PCB averages.

## 2.b. Calculations

Detailed descriptions of the loadings calculations can be found elsewhere (Hoff, 1994; Hoff et al., 1996; Hillery et al., 1998). A brief summary will be presented here. Net atmospheric flows ( $L$ , in kg/yr) are based on three processes: wet deposition, dry deposition, and net gas exchange. They are represented by the equation:

$$L = C_p R_p A + C_a \mathbf{f}_a v_d A + [k_{ol}(1 - \mathbf{f}_a)C_a (RT / H)A - k_{ol}(1 - \mathbf{f}_w)C_w A] \quad (1)$$

Wet deposition is the product of the volume-weighted mean precipitation-phase concentration,  $C_p$  (kg/m<sup>3</sup>), the rate of precipitation,  $R_p$  (m/yr), and the area of the lake,  $A$  (m<sup>2</sup>). In a similar manner, dry deposition is the product of the total atmospheric concentration of the pollutant,  $C_a$  (kg/m<sup>3</sup>), the fraction of the compound in the particle phase,  $\mathbf{f}_a$ , the deposition velocity of the particles,  $\mathbf{n}_d$  (m/yr), and the area of the lake. Gas transfer is divided into two components: volatilization and absorption. The variable,  $k_{ol}$  (m/yr), is the overall air-water mass transfer coefficient,  $R$  (atm m<sup>3</sup> K<sup>-1</sup> mol<sup>-1</sup>)

is the ideal gas constant,  $T$  (K) is the temperature at the air-water interface,  $H$  ( $\text{mol atm}^{-1} \text{m}^{-3}$ ) is the Henry's Law constant, and  $C_w$  ( $\text{kg/m}^3$ ) is the concentration of the compound in the water. For absorption,  $(1 - f_a)C_a$  is the concentration of the compound in the gas phase. Absorption is the transfer of the compound in the gas phase from air to water. For volatilization,  $f_w$  is the fraction of the compound on the particle phase in the water, thus making  $(1 - f_w)C_w$  the dissolved concentration of the compound of interest. Volatilization is then the transfer of the compound from water to air. Net gas exchange is the sum of the absorption and volatilization estimates. Positive net gas exchange indicates absorption, while negative net gas exchange indicates volatilization.

The gas exchange calculations are made based on the two-film air-water exchange model (Schwarzenbach et al., 1993). Typically, this model uses relationships involving the mass transfer coefficients of  $\text{CO}_2$  and  $\text{H}_2\text{O}$  to determine, respectively, the water-side and air-side mass transfer coefficients for the chemical of interest, which are then used to determine  $k_{ol}$  (Schwarzenbach et al., 1993; Hornbuckle et al., 1994; Hoff et al., 1996). Galarneau et al. (2000) simplified these mass transfer calculations in the 1995-1996 loadings report by replacing Schmidt number ratios with diffusivity ratios and then further reducing diffusivity calculations to simpler terms involving molar and diffusion volumes and masses. These same simplifications are used in the calculation of the results presented here.

The loadings estimates are presented in this report as annual values and as both flows and fluxes. Fluxes ( $\text{ng/m}^2/\text{day}$ ) are simply the flows ( $\text{kg/yr}$ ) converted to  $\text{ng/day}$  and divided by the appropriate lake area. These areas are  $82,100 \text{ km}^2$  for Lake Superior,  $57,800 \text{ km}^2$  for Lake Michigan,  $59,600 \text{ km}^2$  for Lake Huron,  $25,700 \text{ km}^2$  for Lake Erie, and  $18,960 \text{ km}^2$  for Lake Ontario. Fluxes allow for comparisons between the lakes by removing the variation due to differing lake areas. Flows and fluxes were calculated seasonally and then summed to give annual loads and averaged to give annual fluxes.

For the 1997-1998 annual fluxes, errors are presented for each term as a coefficient of variation (COV). These COVs were calculated in accordance with the error propagation analysis by Hoff (1994). The major sources of the errors are associated with the spatial and temporal variability of the samples as well as the large uncertainties involved in the physical parameters used, such as the Henry's Law constants (Hoff, 1994; Hillery et al., 1998). For dry deposition and gas exchange, a COV of  $< 267\%$  indicates that the annual fluxes are significantly different from zero at the 95% confidence interval (Galarneau et al., 2000). Similarly, for wet deposition, fluxes are significant if the COV is  $< 165\%$ . Because of a different precipitation phase sampling schedule on Lake Huron, wet deposition there is significant if the COV is  $< 248\%$  (Galarneau et al., 2000).

All results reported here are only estimates. In fact, the error inherent in individual results, such as gas exchange and volatilization, can be quite large. As estimates are aggregated, however, results become more reliable. For instance, examining the

loadings over time or in terms of regional deposition imparts more reliability to the results. Even total deposition is a more reliable way to interpret the results than individual depositional components are. Thus, individual loadings terms should not be regarded as precise numbers, but should be viewed more as a means to understand the behavior of the chemicals across time and space in the Great Lakes basin.

### ***2.c. 1997-1998 Model Refinements***

A few refinements were made to the loadings model for the 1997-1998 calculations. The Henry's Law constant (HLC) for  $\alpha$ -endosulfan was updated. In order to compare the loadings across time, the 1995 and 1996 loadings for  $\alpha$ -endosulfan were recalculated using this new HLC. Previous years loadings did not have to be updated since  $\alpha$ -endosulfan was only added to the list of substances considered in 1995. This year,  $\beta$ -endosulfan was removed from the list of species because no reliable HLC could be found.

This discovery called into question the 1995-1996 results, so no  $\beta$ -endosulfan loadings are presented here. A list of all the HLCs used is given in Table 1.

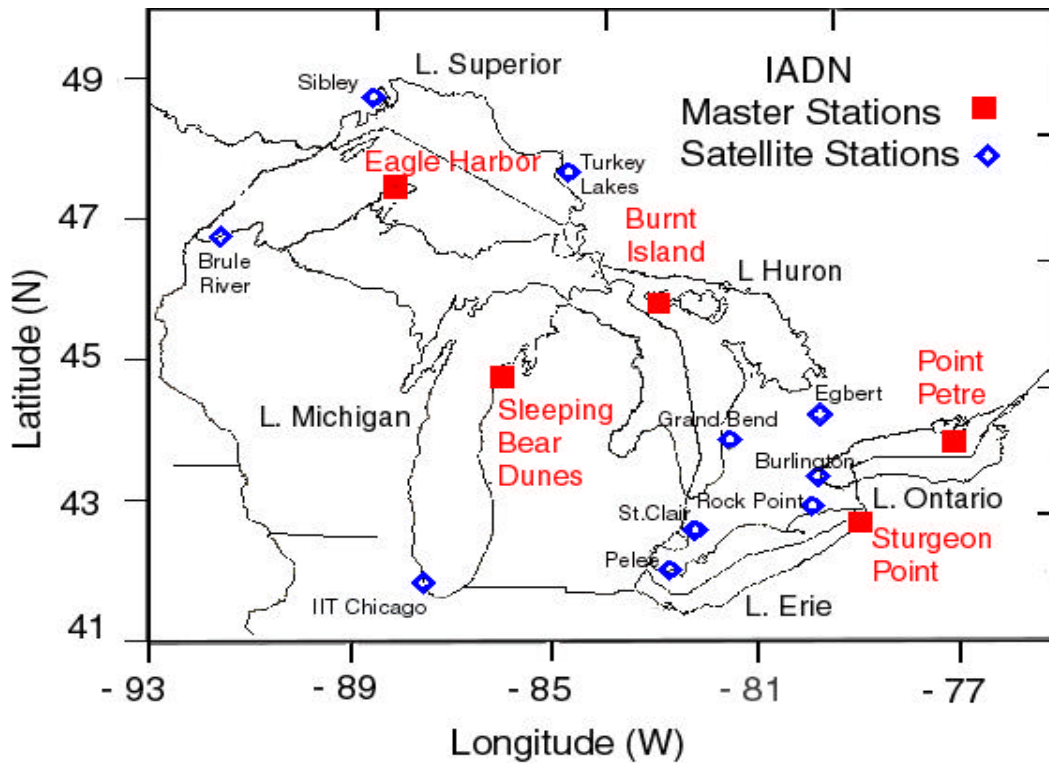
**Table 1. HLCs used in IADN calculations of gas exchange**

Substance	Parameters <i>m</i> and <i>b</i> for Henry's Law Constant, <i>H</i> (Pa•m <sup>3</sup> /mol), $\log_{10}H=m/T+b$		Source
	<i>m</i>	<i>b</i>	
$\alpha$ -HCH	-3054	10.1	Cotham and Bidleman (1991); Jantunen and Bidleman (2000)
dieldrin	-3416	12.2	Cotham and Bidleman (1991)
<i>cis</i> -chlordane	-3416	12.5	Iwata et al. (1995)
<i>trans</i> -chlordane	-3416	12.7	Iwata et al. (1995)
<i>trans</i> -nonachlor	-3416	13.2	Iwata et al. (1995)
<i>p,p'</i> -DDD	-3416	11.3	Suntio et al. (1987); Tateya et al. (1988); as per Hoff et al. (1996)
<i>p,p'</i> -DDE	-3416	12.6	Iwata et al. (1995)
<i>p,p'</i> -DDT	-3416	11.7	Cotham and Bidleman (1991)
$\gamma$ -HCH	-2694	8.54	Cotham and Bidleman (1991); Jantunen and Bidleman (2000)
$\alpha$ -endosulfan	-1001	4.26	Rice et al. (1997)
HCB	-2559	10.4	Ten Hulscher et al. (1992)
PCB 18 (tri)	-2611	10.4	Murphy et al. (1987); Ten Hulscher et al. (1992)
PCB 44 (tetra)	-2716	10.5	Murphy et al. (1987); Ten Hulscher et al. (1992)
PCB 52 (tetra)	-2716	10.4	Mackay et al. (1992); Ten Hulscher et al. (1992)
PCB 101 (penta)	-3416	12.9	Murphy et al. (1987); Ten Hulscher et al. (1992)
phenanthrene	-2469	8.89	Bamford et al. (1999)
pyrene	-2239	7.59	Bamford et al. (1999)
benzo[ <i>b</i> ]fluoranthene	-3416	10.4	Ten Hulscher et al. (1992)
benzo[ <i>k</i> ]fluoranthene	-3416	10.7	Ten Hulscher et al. (1992)
benzo[ <i>a</i> ]pyrene	-3416	10.8	Ten Hulscher et al. (1992)
indeno[1,2,3- <i>cd</i> ]pyrene	-3416	6.95	Ten Hulscher et al. (1992)



## 2.d. Data Sources

IADN collects gas, particle, and precipitation phase samples at each of its master stations, one on each of the Great Lakes (see Figure 1). Sampling details can be found elsewhere (Environment Canada, 1994; Basu, 1996). Vapor and particle phase samples were collected every 12 days for 24 hours at a time. At Lakes Erie, Michigan, and Superior, approximately 815 m<sup>3</sup> of air were collected using XAD-2 resin to trap the gaseous contaminants. At Lakes Huron and Ontario, the average volume of air collected was 350 m<sup>3</sup> using polyurethane foam (PUF) to trap the gaseous contaminants. Canadian sites used glass fiber filters for particle phase collection, while U.S. sites used quartz fiber filters (QFF). Precipitation-phase samples were collected as 28-day composites at the U.S. sites and Lake Ontario. Precipitation samples were collected as 14-day composites at Lake Huron.



**Figure 1. IADN master and satellite sampling stations**

Meteorological data, including wind speed and precipitation measurements necessary for the loadings calculations, were collected on-site at each master station. Annual and seasonal averages were then determined from these measurements to use in the calculations. Lake water surface temperatures were obtained from the National Oceanic Atmospheric Administration's (NOAA) Great Lakes Environmental Research

Laboratory (GLERL) satellite data (NOAA, 2000). Annual meteorological data can be found in Table A1 of Appendix A.

Lake water concentrations are crucial to the loadings calculations. However, IADN does not make these measurements. In order to perform the calculations, water concentration data for 1997 and 1998 from available sources were used. If more than one source was available for a given lake for one year, the water concentration data were pooled by the method of weighting by inverse variance (Taylor, 1990). The lake water concentrations that were used and their sources are listed in Table A2 of Appendix A.

This report will explore temporal and spatial trends in contaminant fluxes and flows from 1992-1998. Over this time, there have been some missing contaminant concentration data as well as lake water concentration data. These missing data mean that results cannot be reported for certain substances. For example, without any water concentration data for endosulfan sulfate, no volatilization estimates can be made. We have constructed data availability charts for air and water concentration data over the years in Tables A3 through A5 in Appendix A. These charts serve as a guide to understanding when unreported fluxes or flows are a result of missing data.

### ***2.e. Quality Assurance/Quality Control***

IADN follows a strong quality assurance program. The Quality Assurance Program Plan (QAPP) was documented jointly by Environment Canada, the United States Environmental Protection Agency, and the Ontario Ministry of the Environment and Energy (February 1994). Each agency developed their own Quality Assurance Project Plan (QAPjP). In the United States, Don Gatz wrote the QAPjP in 1990 and then revised it (Gatz, 1993) for the IADN project at Illinois State Water Survey. Subsequently, it was modified at Indiana University (IU) (Basu, 1994 and 1995). The Canadian QAPjP was first developed by TRC Environmental Corporation in 1993 and has been updated as needed. Environment Canada laboratories are accredited by the Standards Council of Canada (SCC) through the Canadian Association for Environmental Analytical Laboratories (CAEL) to SCC Guide CAN-P-4D and must meet ISO Guide 17025 requirements. Laboratories undergo regular (biennial) performance tests for organochlorine and PAH parameters and biennial audits of the methods used.

All air and precipitation chemical data and all meteorological data measured by IADN go through a quality control process via the Research Data Management and Quality Control System<sup>TM</sup> (Sukloff et al., 1995) at the Center for Atmospheric Research Experiments of Environmental Canada.

Recently, a laboratory audit of all IADN participants was carried out by Peter Fowlie of Cornerstone Science (Fowlie, 2001). Below is a summary of the findings taken directly from the report:

A laboratory audit program was carried out to ensure that the SOPs were being followed by the individual laboratories. A comparison of the different analytical approaches used by each laboratory was made and findings for each laboratory were presented. The analytical methods are logical and capable of producing defensible data. A few of the SOPs were out of date. Occasionally, lab practice varies from a current SOP, but variances were of a relatively minor nature and do not constitute major errors affecting data quality. The different sampling and analysis methods used in the program have the potential to cause between laboratory bias in the data.

The IADN Quality Assurance Program Plan (QAPP) was reviewed by the IADN Steering Committee in January 2000 and the updated Revision 1.1 of the QAPP was issued to all Steering Committee members in July 2001.

A Common Reference Standard for the IADN suite of PCBs, Pesticides and PAHs was implemented. Although these standards do not replace the existing calibration standards used in participating laboratories, they were used as round robin standards and will be used as independent check standards to verify continuing calibrations.

A split sample interlaboratory round robin was carried out in early 2001 to evaluate equivalence of data for IADN organics. With the exception of a few particular PAHs and pesticides, the round robin demonstrated reasonable agreement for trace organic data. Precipitation data is less precise than PUF data which is, in turn, less precise than GFF data.

For PAHs, IU [Indiana University] reports data about 30% higher than MSC [Meteorological Services of Canada] for PUF samples but there was no such bias for GFF or melted snow. For PCBs, NLET [National Laboratory for Environmental Testing] reports data about 40% higher than IU in melted snow. IU reported data about 30% higher than MSC on PUF. This implies that NLET PCB data is biased quite high compared to MSC. For pesticides, IU reports data about 30% higher than MSC in PUF but comparable to NLET on the melted snow. It is important to note that these results can only be applied to one point in time early in 2001 and should not be applied without caution to the 10 years of the IADN program.

A split sample co-located sampler program was operated at Point Petre during 2000 and 2001. Monthly paired samples showed some disagreement which should be resolved. (Fowle, 2001)

Based on the results of our quality assurance studies to date, differences among sites/lakes, as cited in this loadings report, should be viewed in consideration of the identified measurement differences among agencies involved in IADN. We are continuing to address this issue to be sure that data are compatible. Another question may arise as to the consistency of the procedures as the United States lab switch from the Illinois State Water Survey (ISWS) to IU in 1994. From its inception, Dr. Basu has been with the project. A co-developer of the Standard Operating Procedures at ISWS, Dr. Basu followed the same procedures at IU and had any modifications approved by the EPA.

### 3. Results and Discussion

The results presented here will be discussed in two distinct forms: as flows and as fluxes. Flows are always presented in kg/yr while fluxes are in ng/m<sup>2</sup>/day. The term “loadings” is defined in the IADN Second Implementation Plan as the transfer of the chemical to the “...water or watershed in units of mass per square meter per unit time or...mass per year” (Environment Canada and Great Lakes National Program Office, 1998) and will thus be used generally throughout this report to represent both flows and fluxes. “Loadings” should be considered a generic, all encompassing term in this document, representative of the atmospheric deposition of pollutants to the Great Lakes in any units.

The flows and fluxes, or loadings, will often be referred to as downward. Downward flows and fluxes are those that go from the air to the lakes. Gas absorption, wet deposition, and dry deposition are all downward loadings, as are positive net gas exchange and positive total deposition. An upward loading would thus be the movement of the contaminant from the lake water to the air. This process is referred to as volatilization. Negative flows and fluxes indicate volatilization.

When chemicals are being volatilized out of the water, the lakes are often referred to as sources of these chemicals to the atmosphere. This is not to suggest that the lakes are the original sources of the chemicals to the Great Lakes atmosphere, but merely to indicate that after years of the chemicals being deposited to the lakes through the atmosphere, tributary inputs, direct dumping, and other means, the lakes are now releasing these substances into the overlying air.

#### **3.a. 1997-1998 Master Station Fluxes (ng/m<sup>2</sup>/day)**

The main purpose of this report is to calculate the biennial loadings for 1997 and 1998 and add these results to previous years (1992-1996) results in order to explore spatial and temporal trends in the flows and fluxes. However, it is also important to analyze the current results (1997-1998) to gain a better understanding of how these chemicals are now behaving in the Great Lakes basin.

The 1997-1998 results will be presented as annual and relative fluxes. Tables B1 to B10 in Appendix B give the annual daily fluxes (ng/m<sup>2</sup>/day) for each substance for each lake. Each table represents one lake and gives wet and dry deposition fluxes as well as gas exchange fluxes, which include gas absorption, volatilization, and net gas exchange. Seasonal fluxes are not reported here but can be found at [http://www.msc.ec.gc.ca/iadn/results/DEFAULT\\_e.html](http://www.msc.ec.gc.ca/iadn/results/DEFAULT_e.html) and are available upon request. Annual averages from 1992 through 1998 are also available on the website or by request.

Figures C1 through C10 in Appendix C show the relative loadings for each substance for each lake in a given year (1997 and 1998). The sum of the downward inputs, from air to water, for each substance is normalized to 100%. The individual downward loading components (wet deposition, dry deposition, and gas absorption) are then expressed as a positive percentage of this normalized sum. The volatilization loading term in the graphs is expressed as a negative percentage of the summed downward loadings. A substance that has a relative volatilization term greater than -100% is experiencing a net loss from the water to the air. Substances that show net downward and upward relative loadings of the same proportion can be said to be at equilibrium between the lake water and the overlying air.

### 3.a.i. Organochlorine Pesticides

IADN measures both in-use pesticides and those whose uses have been banned (referred to as “banned pesticides”). Tables B1 through B10 list the two groups. Individual downward fluxes (from air to water) for banned organochlorine pesticides range from 0.01 ng/m<sup>2</sup>/day to 22 ng/m<sup>2</sup>/day across the basin, while volatilization fluxes range from -0.002 to -37 ng/m<sup>2</sup>/day. The inputs for these substances on Lakes Superior, Michigan, and Erie are dominated by gas absorption. Over Lake Ontario, gas absorption dominates the inputs of almost all of the banned pesticides in 1998, but in 1997, wet deposition dominates the inputs of *p,p'*-DDT and *p,p'*-DDD. Wet deposition mainly dominates inputs of DDT-related compounds at Lake Huron. However, since pesticides are not measured in the particle phase over this lake, estimates of their dry deposition fluxes cannot be made, thus making it hard to draw any real conclusions.

$\alpha$ -HCH and *cis*-chlordane are near air-water equilibrium in Lakes Huron and Ontario. However, 1998 relative loadings graphs for these lakes (see Figures C6 and C10) show an increase in the volatilization component of these compounds, indicating a slight departure from equilibrium. Lakes Michigan and Erie show net inputs of these two pesticides, with gas absorption being almost twice as much as volatilization. Lake Superior shows net volatilization of  $\alpha$ -HCH and *cis*-chlordane.

*trans*-Chlordane still shows great variability across the basin. Lakes Superior and Erie show net volatilization for this compound, with volatilization exceeding net inputs by almost nine times in Lake Superior in 1998. Lake Huron shows that net inputs exceed net outputs of *trans*-chlordane, while Lakes Michigan and Ontario show the compound is near equilibrium.

Particle phase concentrations of all pesticides (except for *p,p'*-DDE) are only measured at Lakes Superior, Erie, and Michigan. The figures in Appendix C, however, clearly show that dry deposition accounts for a small fraction, 20%-30% at most, of the net input of these compounds to the Great Lakes.

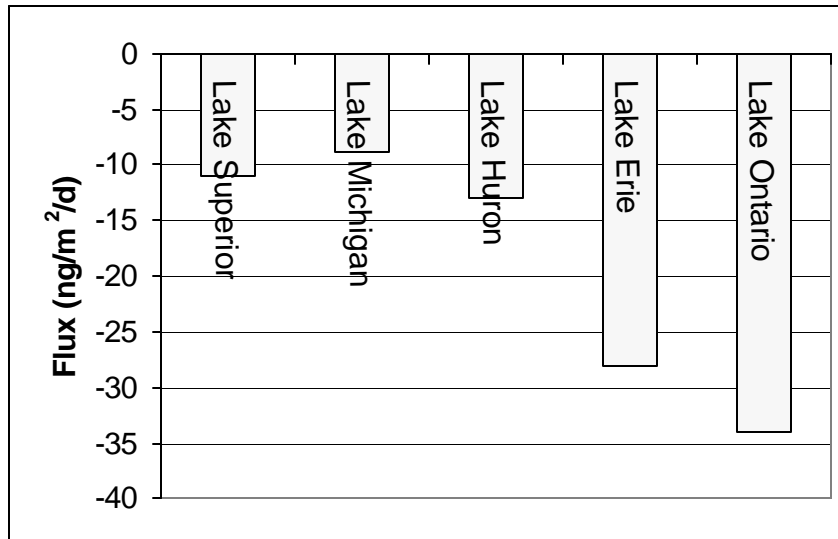
For the most part, 1997 and 1998 loadings show the same results. There are, how-

ever, a few notable differences between the two years. In Lake Superior, relative volatilization fluxes for dieldrin and *trans*-chlordane increased dramatically, as did the dieldrin volatilization from Lake Ontario in 1998. Since volatilization fluxes are similar for the two years, net inputs must have decreased over time in these lakes.

Only three in-use pesticides are reported here:  $\gamma$ -HCH (lindane),  $\alpha$ -endosulfan, and endosulfan sulfate. No HLC could be found for endosulfan sulfate, so there are no volatilization or gas absorption estimates available. Downward fluxes for these pesticides range from 0.05 ng/m<sup>2</sup>/day to 40 ng/m<sup>2</sup>/day, about twice as much as the highest banned pesticide downward flux. Volatilization fluxes for  $\gamma$ -HCH and  $\alpha$ -endosulfan range from -0.004 to -4.3 ng/m<sup>2</sup>/day, almost 10 times lower than some banned pesticide volatilization fluxes and, on average, half as much as total downward fluxes of these compounds. Since these compounds are still in use, we would expect their inputs to the lakes to far exceed their outputs from the lakes. Figures C1 through C10 demonstrate this nicely. Inputs of these chemicals are dominated by gas absorption on Lakes Superior, Michigan, and Erie. This is especially true for  $\alpha$ -endosulfan on Lakes Michigan and Erie, where areas growing crops that are routinely sprayed with this substance are often found near the sites.  $\gamma$ -HCH is mainly dominated by wet deposition on Lakes Huron and Ontario. Conversely,  $\alpha$ -endosulfan inputs are led by gas absorption on these two lakes.

### 3.a.ii. PCBs and HCB

PCBs and HCB are organochlorine commercial chemicals whose uses have been banned. HCB is also banned from use as a pesticide, but it is still released into the atmosphere as a byproduct of some chemical manufacturing processes. Downward fluxes for these chemicals range from 0.02 ng/m<sup>2</sup>/day to 11 ng/m<sup>2</sup>/day across the basin. Volatilization fluxes range from -0.03 ng/m<sup>2</sup>/day to -34 ng/m<sup>2</sup>/day, with suite-PCB fluxes higher than those of HCB. Interestingly, suite-PCB volatilization fluxes tend to increase from west to east across the basin (see Figure 2).



**Figure 2. 1997 annual suite-PCB volatilization fluxes (ng/m<sup>2</sup>/day) for each lake**

Dry deposition is not measured for any of these compounds because concentrations were found to be insignificant in this phase. PCBs were also not measured in the precipitation phase on Lakes Huron and Ontario. Given these omissions, it is hard to make comparisons regarding the fractions of the total deposition across the region, but some trends are evident. Gas absorption is the dominant downward loading in Lakes Superior, Michigan, and Erie. Downward loadings at Lake Erie show a particularly strong dependence on gas absorption, while Lakes Superior and Michigan show a stronger balance between gas absorption and wet deposition.

Suite-PCB shows a net loss from the lakes, except for Lake Michigan in 1998. This means that the lakes are acting as a source of PCBs to the atmosphere. Individual congeners, however, are acting slightly differently. PCB 52 is close to equilibrium in Lake Ontario, but in the other four lakes the water is a net recipient of this congener. In fact, Lakes Superior, Michigan, and Erie show considerably more atmospheric depositions of PCB 52 than volatilization, by up to 30 times.

HCB is near equilibrium in all of the lakes except for Lake Ontario, where it has a net loss from the water. Fluxes for HCB were well above individual PCB congener fluxes, but were up to 5 times lower than suite-PCB fluxes.

### 3.a.iii. PAHs and Metals

The PAHs and metals measured by IADN are currently emitted into the atmosphere of the Great Lakes as byproducts of anthropogenic activity. Since metals are relatively nonvolatile, gas absorption is not measured. As in indicated in Table A3, metals data are not available for Lakes Superior, Michigan, or Erie.

Downward fluxes for PAHs range from 0.3 ng/m<sup>2</sup>/yr to 530 ng/m<sup>2</sup>/day. Volatilization fluxes range from -0.00001 to -240 ng/m<sup>2</sup>/day. The individual PAHs show strikingly different deposition patterns. Deposition of the lighter PAH phenanthrene is dominated by gas absorption, while that of pyrene is more evenly split between wet and dry deposition and gas absorption. The heavier PAH inputs are dominated by wet and dry deposition. Interestingly, dry deposition of the heavier PAHs appears to have increased in Lakes Michigan and Erie from 1997 to 1998. Where water concentration data are available, volatilization of PAHs is almost always less than net inputs. As with the in-use pesticides, this is to be expected.

Fluxes for metals range from 13 to 840 ng/m<sup>2</sup>/day for dry deposition and from 130 to 5400 ng/m<sup>2</sup>/day for wet deposition. Fluxes for these substances are much higher than those for pesticides, PCBs, or HCB. The wet deposition flux of Pb averages 20 times higher than that of Cd. Though Pb still has the highest dry deposition values, its fluxes are closer to those of the other metals than its wet deposition fluxes.

### **3.b. 1992-1998 Flows (kg/yr)**

In an effort to fulfill the goals of IADN as laid out in the introduction, it is important to look at both spatial and temporal trends from all possible angles. Because the units and therefore the results are more tangible, the most telling means of looking at temporal trends is by use of flows in kg/yr. The flow tables can be found in Appendix D, Tables D1-D5. The tables contain loadings data from 1992 through 1998. The columns represent wet and dry deposition, net gas exchange, and total deposition. In most cases, the total deposition is the sum of the three aforementioned components. However, deposition from the particle phase was assumed to be negligible for many species and measurements were not made at the Canadian sites for organochlorine pesticides and PCBs. Analysis of PCBs as well as DDE and HCB in the particle phase was ended in 1996 at the U.S. sites. In addition, dry deposition data for pesticides and PCBs were not included in 1996 estimates for the U.S. sites. Table A3 in Appendix A illustrates these missing data. In the cases mentioned above, total deposition is the sum of all three components when present, otherwise only wet deposition and total gas exchange are summed.

#### **3.b.i. Organochlorine Pesticides**

In a few cases, wet deposition is decreasing over time. In Lake Huron,  $\alpha$ -HCH has dropped by over 50% in the last two years. A decreasing trend is also seen in Lake Erie. Although currently in use,  $\gamma$ -HCH (lindane), shows a decrease in wet deposition in Lakes Michigan and Erie over the sampling period, though the trend over Lake Michigan is somewhat erratic. Annual precipitation totals for these lakes have either remained fairly constant or slightly increased, indicating that precipitation amounts are not playing a significant role in the decreasing trends of these pesticides.



Few distinct trends are discernable for dry deposition flows. However, dieldrin flows show an overall increase from 1992 to 1998 in Lakes Superior and Michigan. Where they are measured, these particle phase flows are small compared to net gas exchange and even wet deposition, justifying the discontinuation of these measurement at the Canadian sites.

In most lakes, net gas exchange is the dominant process. For Lake Superior, volatilization of  $\alpha$ -HCH began between 1994 and 1995 and has been increasing since then, from -230 to -470 kg/yr. Conversely, dieldrin has been showing a decrease in its flow out of the Lake Superior. After a series of slight decreases, lindane showed net volatilization for the first time in 1998 in Lake Superior, but the total deposition is still positive due primarily to wet deposition. Nearly the opposite of Lake Superior, Lake Michigan shows an increase in gas exchange to the lake between 1994 and 1995 for  $\alpha$ -HCH and is joined by Lake Erie as the only lakes still showing net absorption of  $\alpha$ -HCH.

Total deposition typically mirrors the trends seen in net gas exchange. Exceptions to this are the HCHs in Lakes Huron and Ontario as well as *p,p'*-DDT in Lakes Huron, Erie, and Ontario, where wet deposition contributes as much, if not more, to the total.

### 3.b.ii. PCBs and HCB

Lakes Huron, Erie and Ontario continuously depict volatilization for HCB, with Lake Ontario having the most pronounced temporal trend. Flows out of this lake decreased from -160 kg/yr in 1992 to -21 kg/yr in 1998.

For all lakes with available data, 1997 and 1998 showed a decrease for both wet deposition and net gas exchange of total PCBs. The increase in the 1998 wet deposition flux of suite-PCB over Lake Michigan from 1997 values may be due to significantly increased precipitation amounts that year. Net gas exchange flows for total PCBs show net volatilization consistently over time, although all lakes show a trend towards equilibrium for this process. Individual congeners in most lakes also show the same trend towards air-water equilibrium.

### 3.b.iii. PAHs and Metals

Unlike nearly all other species monitored by IADN, PAHs and metals are still emitted into the atmosphere from several anthropogenic sources. Because of this, trends tend to be more stagnant or variable over time, with no clear increasing or decreasing pattern in the flows.

The lakes on which metals are measured, Lakes Huron and Ontario, have seen a decrease in flows for wet deposition of cadmium and lead from 1992 through 1996. In

fact, wet deposition flows of lead on Lake Huron have dropped from 100,000 kg/yr to 28,000 kg/yr. In all cases, this trend was reversed with higher flows for 1997 and 1998. Annual precipitation over all years has remained fairly steady at both lakes. Dry deposition of metals does not follow much of a trend, but rather remains at similar levels throughout the years.

### **3.c. Temporal and Spatial Flux Trends (1992-1998)**

To get a complete picture of the trend comparison, fluxes (ng/m<sup>2</sup>/day) must also be analyzed. Being independent of lake area, they provide an excellent means of doing spatial analysis. Graphs of the annual fluxes can be found in Figures E1 through E15 in Appendix E. The graphs on each page are organized to include one compound group and one deposition component. For gas exchange and total flux, upward bars indicate flow into the lake and bars pointing downward indicate volatilization.

The uncertainties in gas exchange fluxes have proven to be very large, especially near equilibrium. To circumvent this problem, fugacity ratios have been used to examine air-water equilibrium. As mentioned, the direction of gas exchange can change from flux into to flux out of the water. The fugacity is a measure of the tendency of a substance to escape from the phase in which it resides, with phases in equilibrium having equal fugacities (Mackay, 1991). Because the mass transfer coefficient is the same for the diffusive transport in both directions, the ratio of gas exchange mass fluxes as calculated for the loadings report is interchangeable with the ratio of fugacities. Fugacity ratios reported in Table F1 in Appendix F are the ratio of air over water. Ratios greater than one indicate absorption and ratios less than one indicate volatilization. A ratio of 1 indicates that equilibrium has been reached. Table F1 contains fugacity ratios from 1992-1998 for all species reported and is organized by lake.

#### **3.c.i. Organochlorine Pesticides**

Wet deposition fluxes for  $\alpha$ -HCH are decreasing over time in Lakes Ontario and Erie, while dieldrin wet deposition fluxes decline over Lakes Michigan and Erie. Wet deposition fluxes for  $\gamma$ -HCH at Lakes Huron and Ontario, however, do not follow this declining trend. For Lake Ontario, 1997 and 1998 wet deposition fluxes are similar to 1992 and 1993 fluxes.  $\gamma$ -HCH at Lake Huron has been at similar levels for the past five years. Fluxes of both  $\gamma$ - and  $\alpha$ -HCH also seem to be quite higher at these lakes as well. The magnitude of *p,p'*-DDD wet deposition fluxes are comparable across the basin over time while *p,p'*-DDT fluxes show more variability both spatially and temporally. Meteorological data do not indicate that precipitation averages are affecting these trends.

For all three lakes in which the particle phase is collected and measured, dieldrin consistently has the largest dry deposition flux over time. With the exception of *p,p'*-

DDT steadily decreasing at Lake Erie and the variability in annual fluxes of dieldrin and  $\alpha$ -HCH across the basin, the majority of the other pesticide dry deposition fluxes are relatively constant.

Gas exchange of dieldrin at Lake Ontario is consistently the largest flux observed, indicating net volatilization of this pesticide. Also of note is the decreasing trend of  $\gamma$ -HCH absorption in Lake Erie and Michigan. The graphs, and to a greater extent the fugacity ratio of 0.92, suggest that  $\gamma$ -HCH began volatilizing for the first time in Lake Superior in 1998.

While not included in the graphs, the currently in-use pesticide  $\alpha$ -endosulfan is presented in the fugacity table with ratios ranging from 530 to 6,600 for Lakes Erie and Ontario. These fugacity ratios dwarf the values for the banned pesticides, suggesting that gas absorption is still the dominant mechanism of gas exchange for this in-use pesticide. Fugacity ratios for other pesticides show some trends over time. Ratios for dieldrin are much less than one in all lakes and have been fairly constant over the years. *trans*-Nonachlor ratios have also been constant over the past four years for each lake. Total flux trends follow gas exchange trends closely.

### 3.c.ii. PCBs and HCB

Not much is discernible over time for the wet deposition of individual PCB congeners. In fact, wet deposition fluxes are similar over time and in magnitude on Lakes Superior, Michigan, and Erie. The homogeneity of data between these lakes is evidence for the atmospheric ubiquity of PCBs. Suite-PCB fluxes on all lakes except Lake Superior are, however, decreasing over time. Although recent comparisons cannot be made for Lakes Huron and Ontario because of a lack of data, they do show the largest suite-PCB wet deposition fluxes across the basin. Lake Erie seems unique in that all PCBs measured there reached peak fluxes in 1995 and then decreased in the following three years.

Lake Ontario consistently has the largest PCB fluxes and is the only lake that shows volatilization for all congeners for all years. For most years though, PCBs across the basin show net gas exchange to be in the direction of volatilization. In Lakes Superior and Huron, gas exchange fluxes are near equilibrium for some individual congeners. This is also seen in the fugacity table as values are near one. PCB 52 has gone from volatilization to absorption in 1997 for all lakes but Lake Ontario. In the case of Lakes Michigan and Erie, the fugacity ratios for PCB 52 are quite high in 1997 and 1998, ranging from 12 to 32. Total fluxes for PCBs also mirror net gas exchange fluxes both temporally and spatially, but, more importantly, they indicate how significant the process of air-water exchange is in the fate of PCBs in the Great Lakes.

### 3.c.iii. PAHs and Metals

As previously mentioned, there is not much of a temporal trend for PAHs in wet deposition. However, there is a rather strong spatial trend. From west to east, the fluxes of all of the PAHs measured increase. For example, the phenanthrene wet deposition flux in 1993 increases from approximately 6 ng/m<sup>2</sup>/day in Lake Superior to approximately 78 ng/m<sup>2</sup>/day in Lake Ontario. Whether this is merely due to the location of the sites or if it is result of increased urbanization from west to east in the Great Lakes basin remains to be seen. Much the same can be said for the analysis of dry deposition fluxes.

In all lakes but Lake Superior, data are too spotty to look at trends in PAH gas exchange. Phenanthrene from 1992 to 1994 and pyrene in 1994 are the only PAHs to exhibit any volatilization from this lake. Indeno[1,2,3-*cd*]pyrene, a relatively heavy PAH which is not on the flux graphs, has some very high fugacity ratios for Lakes Superior and Erie. The fugacity ratios for the sum-PAH for these lakes are also greater than one, which supports the idea that the bulk of currently-emitted pollutants are still entering the Great Lakes through gas absorption. Because of a lack of gas exchange data, it is also hard to assess any total flux trends.

The wet deposition of Cd and Pb in Lakes Huron and Ontario in 1997 and 1998 reverses the trend of decreasing fluxes from 1992 to 1996. Between the two lakes, the magnitude of wet and dry deposition fluxes are quite similar. Dry deposition seems to follow no trend at all, either through time or between the lakes. These fluxes have remained relatively constant over time for each metal. Total fluxes for metals mirror wet deposition trends, indicating that this is the dominant process for depositing metals to the Great Lakes. This is not surprising since wet deposition fluxes are up to six times greater than dry fluxes.

### **3.d. Urban Impacts**

One of the major sources of error in the loadings calculations is that IADN uses data from one remote sampling site to characterize each lake (Hoff, 1994; Hillery et al., 1998). Research suggests that spatial variability exists for many compounds across any one lake (Achman et al., 1993; Monosmith et al., 1996). In particular, it has been shown that urban areas have increased levels of deposition to the Great Lakes as compared to background levels (Offenberg et al., 1997; Simcik et al., 1997; Zhang et al., 1999; Franz et al., 1998). This implies that loadings calculated using only data from a remote sampling site might grossly underestimate the true deposition to the entire lake. Given the many urban centers that exist in the Great Lakes region, there is the potential for many urban impacts on loadings estimates to the Great Lakes.

IADN operates a satellite station in the urban center of Chicago at the Illinois Institute for Technology (IIT) campus. In the 1995-1996 loadings report, Galarneau et al. (2000) used concentration data from Chicago for 1996 to prepare loadings estimates for the city and then compared those results to estimates from Sleeping Bear Dunes, IADN's master station on Lake Michigan, to determine the impact of the urban center on lakewide loadings.

The purpose of the urban impacts assessment in the 1995-1996 loadings report was to establish a base for estimating urban impacts for all major cities across the entire basin. This work will be carried out in the 1999-2000 loadings report. We have decided, however, to continue with the work of Galarneau et al. (2000) here by performing the same calculations from the 1996 data on the 1997 and 1998 Chicago and Lake Michigan data in an effort better understand urban impacts and trends over time in comparison to loadings trends at a remote site.

Water concentration data was not available for Lake Michigan for dieldrin,  $\alpha$ -endosulfan, and all of the PAHs. In order to have a complete picture of the loadings estimates, lake water concentrations from Lake Erie were used in place of the missing data.

Flux calculations were first made for all substances at both Sleeping Bear Dunes (SBD) and Chicago. These estimates were calculated as if they occurred over adjacent waters, just as the loadings for the master stations were calculated. The results for wet and dry deposition, net gas exchange, and total mass flux are shown in Table 2 for 1996-1998.

We can see from Table 2 that Chicago fluxes for most substances are much greater than fluxes found at Sleeping Bear Dunes, a site that is representative of regional background (rural) concentrations. The total mass flux for PAHs at Chicago is always positive, indicating net inputs of these compounds into the lake from this site. They are also 40 to 190 times greater than Sleeping Bear Dunes PAH total mass fluxes. In fact, Chicago PAH fluxes in all deposition categories far exceed those from Sleeping Bear Dunes.

PCBs follow the same trend, with one striking difference from the master station fluxes. PCB net gas exchange fluxes are negative at Sleeping Bear Dunes, with the exception of PCB 52, indicating the lake is a source of these compounds to the atmosphere. At Chicago, all net gas exchange fluxes are positive, indicating gas absorption is the dominant process here. This is not unexpected since PCBs are mainly an urban pollutant.

Pesticides, on the other hand, show much less disparity in flux values between the two sites. In fact, the dry deposition of  $\alpha$ -HCH and  $\alpha$ -endosulfan is greater at Sleeping Bear Dunes than Chicago. This is because many pesticides are often used for agricultural purposes, which take place in the more rural areas of a state. Dieldrin, *cis*- and *trans*-chlordane, and *p,p'*-DDE show net volatilization fluxes at Sleeping Bear

Dunes but net absorption at Chicago.

While differences in flux magnitudes are interesting between the two sites, it is also interesting to note the trends over the past three years at these sites. Dieldrin fluxes have remained fairly constant across time at Sleeping Bear Dunes, and a similar trend can be seen at Chicago, though 1997 showed an increase in total mass flux. As HCB gas exchange and total mass fluxes have increased over time at Sleeping Bear Dunes, they have decreased at Chicago. Total mass flux values of suite-PCB at Sleeping Bear Dunes have decreased dramatically from 1996 to 1998 and have even gone from the lake being a net source of PCBs to it being a net receiver of the contaminants. Suite-PCB total mass flux values at Chicago have held steady over time. PAH wet deposition has been variable at Chicago with an overall doubling in flux from 1996 to 1998, while net gas exchange has greatly decreased there. Conversely, Sleeping Bear Dunes, while also exhibiting some variability over the years, shows fairly constant wet deposition fluxes from 1996 to 1998, while PAH net gas exchange at this site shows a decrease for phenanthrene but a slight increase for sum-PAH. Though temporal trends for some compounds mirror each other at both sites, more often the trends differ between IADN's urban and remote sites on Lake Michigan.

**Table 2. Mass fluxes (ng/m<sup>2</sup>/day) of atmospheric deposition at Sleeping Bear Dunes (SBD), an IADN master station, and Chicago from 1996-1998**

Species	Year	Wet Deposition		Dry Deposition		Net Gas Exchange		Total Mass Flux	
		SBD	Chicago	SBD	Chicago	SBD	Chicago	SBD	Chicago
$\alpha$ -HCH	1996	0.71	3.2	-	-	14	32	15	35
	1997	1.9	3.1	0.16	0.023	14	18	16	21
	1998	0.8	1.7	0.043	0.019	9.7	7.2	11	8.9
$\gamma$ -HCH	1996	0.31	2.9	-	-	5.1	14	5.4	17
	1997	1.5	4.8	0.11	0.25	7.2	11	8.8	16
	1998	0.61	3.6	0.089	0.093	8.9	11	9.6	15
dieldrin	1996	1.4	4.2	-	-	-10	6.5	-8.6	11
	1997	1.2	7.1	0.73	5.3	-6.9	11	-5	23
	1998	1.5	3.9	0.87	4.4	-9.7	2	-7.3	10
$\alpha$ endosulfan	1996	0.65	1.4	-	-	17	22	18	23
	1997	0.69	2.4	1.7	1.3	39	25	41	29
	1998	0.49	3.3	6.6	1	40	38	47	42
<i>cis</i> -chlordane	1996	0.18	0.11	-	-	-0.15	5.6	0.03	5.7
	1997	0.47	8.4	0.12	2.8	0.1	6.8	0.69	18
	1998	0.52	1.2	0.15	2.7	-0.21	4.6	0.46	8.5
<i>trans</i> -chlordane	1996	0.45	1.5	-	-	-0.14	5.1	0.31	6.6
	1997	0.052	0.78	0.069	0.62	-0.017	5	0.1	6.4
	1998	0.058	1.1	0.085	0.65	-0.23	3.6	-0.087	5.4
<i>trans</i> -nonachlor	1996	0.045	0.46	-	-	-0.7	0.1	-0.66	0.56
	1997	0.037	0.53	0.054	0.22	-0.77	-0.027	-0.68	0.72
	1998	0.042	0.31	0.039	0.14	-0.79	-0.21	-0.71	0.24
<i>p,p'</i> -DDD	1996	0.084	0.38	-	-	0.22	0.78	0.3	1.2
	1997	0.017	0.86	0.036	0.13	0.17	1.3	0.22	2.3
	1998	0.14	0.28	0.047	0.15	0.25	1.4	0.44	1.8
<i>p,p'</i> -DDE	1996	0.19	1.3	-	-	-0.13	4.4	0.06	5.7
	1997	0.12	1	-	-	-0.073	3.2	0.047	4.2
	1998	0.13	1	-	-	-0.57	3	-0.44	4
<i>p,p'</i> -DDT	1996	0.43	3.9	-	-	0.26	7.6	0.69	12
	1997	0.2	3.8	0.052	1.5	0.62	5.1	0.87	10
	1998	0.14	3.1	0.093	0.82	0.66	5	0.89	8.9
HCB	1996	0.043	0.11	-	-	0.39	2.6	0.43	2.7
	1997	0.035	0.17	-	-	0.74	1.9	0.78	2.1
	1998	0.039	0.17	-	-	1.2	1.3	1.2	1.5
PCB18	1996	0.08	0.24	-	-	-1.1	2	-1	2.2
	1997	0.039	0.25	-	-	-0.39	2.3	-0.35	2.6
	1998	0.065	0.59	-	-	-0.26	2.2	-0.2	2.8
PCB44	1996	0.046	0.27	-	-	-1	6.7	-0.95	7
	1997	0.062	0.43	-	-	-0.52	5.7	-0.46	6.1
	1998	0.13	0.89	-	-	-0.52	4	-0.39	4.9
PCB52	1996	0.1	0.38	-	-	0.1	8.3	0.2	8.7
	1997	0.079	0.45	-	-	0.45	8.1	0.53	8.6
	1998	0.091	0.86	-	-	0.52	7.1	0.61	8
PCB101	1996	0.064	0.44	-	-	-0.43	4.4	-0.37	4.8
	1997	0.051	0.52	-	-	-0.24	4.8	-0.19	5.3
	1998	0.082	1.2	-	-	-0.2	4.7	-0.12	5.9
suite-PCB	1996	2.3	13	-	-	-15	97	-13	110
	1997	1.2	11	-	-	-3.2	80	-2	91
	1998	1.8	27	-	-	-1.6	76	0.2	100
phenanthrene	1996	10	330	5	180	-340	17000	-330	18000
	1997	15	860	4.6	220	33	8900	53	10000
	1998	8.5	790	5.8	140	-15	6500	-0.7	7400
pyrene	1996	6.8	400	5.1	340	-45	2100	-33	2800
	1997	11	910	4.7	320	-0.84	1300	15	2500
	1998	6.4	760	7.1	220	0.86	1000	14	2000
B[ <i>b+k</i> ]F	1996	12	390	9.2	340	2.6	70	24	800
	1997	16	930	8.7	330	4.1	84	29	1300
	1998	13	900	14	320	4.8	24	32	1200
B[ <i>a</i> ]P	1996	4	210	1.9	130	-0.12	17	5.8	360
	1997	5.7	540	2.3	160	0.65	25	8.7	730
	1998	4.2	450	4.1	110	0.48	6.9	8.8	570
I[1,2,3- <i>cd</i> ]P	1996	6.3	230	4.8	180	0.96	23	12	430
	1997	9.4	440	4	180	1.3	22	15	640
	1998	6.6	390	8.4	180	1.8	7.7	17	580
sum-PAH	1996	23	840	16	650	3.4	110	42	1600
	1997	31	1900	15	670	6	130	52	2700
	1998	24	1700	26	600	7.1	38	57	2300

To further explore the effect of urban inputs on lakewide loadings, we followed the lead of Galarneau et al. (2000) and assessed whether the larger Chicago fluxes were significant across the entire lake. To do this, the impact Chicago might have over the whole lake was estimated by using an urban plume effect that assumes deposition from Chicago extends only over a small sub-area of Lake Michigan. This sub-area corresponds to 1.7% of the total lake area for wet and dry deposition, and 3.5% of the total lake area for gas exchange (Galarneau et al., 2000, p.14). A temporal component was also added to account for the time that air is flowing from the city over the lake and can thus impact loadings. Then, the increase in lakewide flows as a result of the inclusion of the urban flows with the regional data was calculated as a percent (urban effect). The results of these calculations are shown in Table 3. Negative urban effect percentages indicate that the background (BG) lakewide volatilization flows are decreased by including the urban inputs. Background (BG) flows refer to Sleeping Bear Dunes data.

Urban inputs have a minor lakewide effect for most pesticides. There does seem to be a large effect, however, on *cis*- and *trans*-chlordane net gas exchange and total mass flows. Net gas exchange of *trans*-chlordane over the Chicago lake sub-area will drastically change lakewide volatilization and markedly increase total mass flows. The same is true for *cis*-chlordane, though to a lesser extent. *p,p'*-DDE also shows strong urban effects, with lakewide volatilization being greatly reduced and total mass flows being increased.

PCBs show a varied urban effect. While urban inputs do not impressively affect wet deposition flows, stronger effects are seen in gas exchange and total mass flows. For example, in 1998, inclusion of Chicago flows will decrease suite-PCB volatilization by as much as 53% while they will increase total flows to the entire lake by over 500%. In the same year, wet deposition flows will only increase 8.9% by incorporating Chicago into the lakewide loadings. Similar disparities in depositional effects are seen in 1996 and 1997, though not to the same extent as total flow increases in 1998. Since PCBs had mainly urban uses, it is not surprising that an urban site would affect PCB flows to such a great extent. PAHs, which have many current sources in Chicago, show large urban effects in all deposition categories as compared to other chemicals.

With three years of data for Chicago loadings, we wondered if urban effects were consistent or changed over time. For chemicals with similar urban and background loads, urban effects can change radically over time. For example, *p,p'*-DDE total mass flows for both sites tend to track each other very well, so urban effect values are sensitive to small changes, such as in 1997. Consistently high background flows as compared to urban inputs generally have smaller urban effects over time that are more resilient to change, such as  $\alpha$ -endosulfan urban effects for total mass flows and net gas exchange.



**Table 3. The effect on lakewide loadings of adding flows from Chicago to master station estimates of regional background (BG) flows from Sleeping Bear Dunes.**

Species	Year	Wet Deposition			Dry Deposition			Net Gas Exchange			Total Mass Flows		
		BG	Chicago	Urban Effect	BG	Chicago	Urban Effect	BG	Chicago	Urban Effect	BG	Chicago	Urban Effect
		(kg/yr)	(kg/yr)		(kg/yr)	(kg/yr)		(kg/yr)	(kg/yr)		(kg/yr)		
$\alpha$ -HCH	1996	15	0.41	2.7%	-	-	-	300	8	2.7%	320	8.4	2.6%
	1997	40	0.37	0.9%	3.4	0.0028	0.1%	300	4.2	1.4%	340	4.6	1.4%
	1998	17	0.24	1.4%	0.91	0.0022	0.2%	200	1.5	0.8%	220	1.7	0.8%
$\gamma$ -HCH	1996	6.6	0.34	5.2%	-	-	-	110	3.2	2.9%	120	3.5	2.9%
	1997	31	0.51	1.6%	2.3	0.029	1.3%	150	2.4	1.6%	180	2.9	1.6%
	1998	13	0.47	3.6%	1.9	0.011	0.6%	190	2.5	1.3%	200	3	1.5%
dieldrin	1996	30	0.52	1.7%	-	-	-	-220	1.6	-0.7%	-190	2.1	-1.1%
	1997	24	0.85	3.5%	15	0.63	4.2%	-150	2.4	-1.6%	-110	3.9	-3.5%
	1998	31	0.53	1.7%	18	0.53	2.9%	-210	0.41	-0.2%	-160	1.5	-0.9%
$\alpha$ -endosulfan	1996	14	0.16	1.1%	-	-	-	360	5	1.4%	370	5.2	1.4%
	1997	15	0.28	1.9%	36	0.15	0.4%	830	5.7	0.7%	880	6.1	0.7%
	1998	10	0.45	4.5%	140	0.12	0.1%	830	8.3	1.0%	980	8.9	0.9%
<i>cis</i> -chlordane	1996	3.9	0.012	0.3%	-	-	-	-3.3	1.3	-39.4%	0.6	1.3	216.7%
	1997	9.9	0.88	8.9%	2.5	0.31	12.4%	2.2	1.6	72.7%	15	2.8	18.7%
	1998	11	0.12	1.1%	3.2	0.31	9.7%	-4.5	1	-22.2%	9.7	1.4	14.4%
<i>trans</i> -chlordane	1996	9.6	0.16	1.7%	-	-	-	-3	1.2	-40.0%	6.6	1.4	21.2%
	1997	1.1	0.1	9.1%	1.5	0.075	5.0%	-0.36	1.2	-333.3%	2.2	1.4	63.6%
	1998	1.2	0.17	14.2%	1.8	0.075	4.2%	-4.9	0.85	-17.3%	-1.9	1.1	-57.9%
<i>trans</i> -nonachlor	1996	0.95	0.064	6.7%	-	-	-	-15	0.023	-0.2%	-14	0.087	-0.6%
	1997	0.78	0.068	8.7%	1.1	0.027	2.5%	-16	-0.0048	0.0%	-14	0.09	-0.6%
	1998	0.89	0.04	4.5%	0.82	0.017	2.1%	-17	-0.049	0.3%	-15	0.008	-0.1%
<i>p,p'</i> -DDD	1996	1.8	0.05	2.8%	-	-	-	4.7	0.2	4.3%	6.5	0.25	3.8%
	1997	0.36	0.1	27.8%	0.76	0.015	2.0%	3.5	0.3	8.6%	4.6	0.42	9.1%
	1998	2.9	0.029	1.0%	0.99	0.016	1.6%	5.3	0.31	5.8%	9.2	0.36	3.9%
<i>p,p'</i> -DDE	1996	3.9	0.16	4.1%	-	-	-	-2.7	1	-37.0%	1.2	1.2	100.0%
	1997	2.5	0.12	4.8%	-	-	-	-1.5	0.74	-49.3%	1	0.86	86.0%
	1998	2.7	0.14	5.2%	-	-	-	-12	0.71	-5.9%	-9.3	0.85	-9.1%
<i>p,p'</i> -DDT	1996	9	0.53	5.9%	-	-	-	5.6	1.8	32.1%	15	2.3	15.3%
	1997	4.3	0.42	9.8%	1.1	0.17	15.5%	13	1.2	9.2%	18	1.8	10.0%
	1998	3	0.43	14.3%	2	0.092	4.6%	14	1.1	7.9%	19	1.6	8.4%
HCB	1996	0.91	0.014	1.5%	-	-	-	8.3	0.66	8.0%	9.2	0.67	7.3%
	1997	0.75	0.018	2.4%	-	-	-	16	0.46	2.9%	17	0.48	2.8%
	1998	0.83	0.021	2.5%	-	-	-	26	0.33	1.3%	27	0.35	1.3%

Table 3 (continued)

Species	Year	Wet Deposition			Dry Deposition			Net Gas Exchange			Total Mass Flows		
		BG (kg/yr)	Chicago (kg/yr)	Urban Effect	BG (kg/yr)	Chicago (kg/yr)	Urban Effect	BG (kg/yr)	Chicago (kg/yr)	Urban Effect	BG (kg/yr)	Chicago (kg/yr)	Urban Effect
PCB18	1996	1.7	0.029	1.7%	-	-	-	-24	0.45	-1.9%	-22	0.48	-2.2%
	1997	0.82	0.03	3.7%	-	-	-	-6.2	0.53	-8.5%	-5.4	0.56	-10.4%
	1998	1.4	0.075	5.4%	-	-	-	-5.5	0.52	-9.5%	-4.1	0.6	-14.6%
PCB44	1996	0.97	0.033	3.4%	-	-	-	-22	1.4	-6.4%	-21	1.4	-6.7%
	1997	1.3	0.057	4.4%	-	-	-	-8.20	1.4	-17.1%	-6.9	1.5	-21.7%
	1998	2.8	0.11	3.9%	-	-	-	-11	0.9	-8.2%	-8.2	1	-12.2%
PCB52	1996	2.2	0.047	2.1%	-	-	-	2.2	1.9	86.4%	4.4	1.9	43.2%
	1997	1.7	0.054	3.2%	-	-	-	7.10	1.9	26.8%	8.8	2	22.7%
	1998	1.9	0.11	5.8%	-	-	-	11	1.7	15.5%	13	1.8	13.8%
PCB101	1996	1.4	0.056	4.0%	-	-	-	-9.1	0.98	-10.8%	-7.7	1	-13.0%
	1997	1.1	0.063	5.7%	-	-	-	-3.79	1.1	-29.0%	-2.7	1.2	-44.4%
	1998	1.7	0.15	8.8%	-	-	-	-4.3	1.1	-25.6%	-2.6	1.3	-50.0%
suite-PCB	1996	48	1.6	3.3%	-	-	-	-320	22	-6.9%	-270	24	-8.9%
	1997	25	1.3	5.2%	-	-	-	-51.4	19	-37.0%	-26	20	-76.9%
	1998	38	3.4	8.9%	-	-	-	-34	18	-52.9%	4	21	525.0%
phenanthrene	1996	220	46	20.9%	100	23	23.0%	-7200	3800	-52.8%	-6900	3900	-56.5%
	1997	320	95	29.7%	97	26	26.8%	700	2000	285.7%	1100	2100	190.9%
	1998	180	100	55.6%	120	16	13.3%	-310	1500	-483.9%	-10	1600	-16000.0%
pyrene	1996	140	57	40.7%	110	43	39.1%	-950	500	-52.6%	-700	600	-85.7%
	1997	220	99	45.0%	99	39	39.4%	-18	300	-1666.7%	300	440	146.7%
	1998	130	98	75.4%	150	24	16.0%	18	230	1277.8%	300	350	116.7%
B[b+k]F	1996	258	53	20.5%	197	42	21.3%	55	18	32.7%	510	110	21.6%
	1997	338	99	29.3%	180	39	21.3%	86	19	22.1%	610	160	26.2%
	1998	278	118	42.4%	288	35.8	12.4%	100	5.7	5.7%	670	160	23.9%
B[a]P	1996	84	29	34.5%	41	15	36.6%	-2.4	4.6	-191.7%	120	49	40.8%
	1997	120	57	47.5%	48	18	37.5%	14	5.5	39.3%	180	81	45.0%
	1998	89	58	65.2%	86	12	14.0%	10	1.6	16.0%	190	72	37.9%
I[1,2,3-cd]P	1996	130	32	24.6%	100	22	22.0%	20	6.3	31.5%	250	60	24.0%
	1997	200	47	23.5%	85	21	24.7%	27	4.9	18.1%	310	73	23.5%
	1998	140	51	36.4%	180	21	11.7%	37	1.8	4.9%	360	74	20.6%
sum-PAH	1996	480	110	22.9%	340	79	23.2%	72	29	40.3%	890	220	24.7%
	1997	650	200	30.8%	320	79	24.7%	130	29	22.3%	1100	310	28.2%
	1998	510	230	45.1%	550	68	12.4%	150	9.1	6.1%	1200	310	25.8%

Flow and flux estimates from 1996-1998 show that an urban center can have a significant impact on lakewide loadings for certain chemicals. These results further encourage the use of urban data for future reports and also signal the need for more urban satellite stations in the IADN program. Having precise measurements from urban centers on all lakes will hopefully facilitate the calculation of better, more exact loadings so that a more complete picture of the Great Lakes can be formed.

#### **4. Regional Deposition**

Another property of flows (kg/yr) is that they are additive. From the flow tables in Appendix D, the sum over all 5 lakes for each species for total deposition was calculated for every year and can be seen in Table 4. Chicago flows were not included in the sums because data was not available for all years and this was the only urban area for which we had any data. Data for most urban centers around the Great Lakes basin would need to be included in order to develop a regional picture, and we do not have that information. All sums, except for the metals, represent values taken from three or more lakes. The metal sums include data from Lakes Huron and Ontario only. This table gives an approximation of the background regional flows to the waters of the Great Lakes. Table 4 also shows x-intercept and r-squared values for the linear regression of flows (kg/yr) vs. time (year). The x-intercept values were found by setting the flow to zero for each regression. These intercepts represent the approximate year when equilibrium for a given chemical will be reached in the Great Lakes. X-intercept values for metals approximate when inputs (wet and dry deposition) to the lakes will be zero. These years are only estimates and are taken directly from the regression parameters. Relative errors based on other regression parameters are on average 37% for this term.

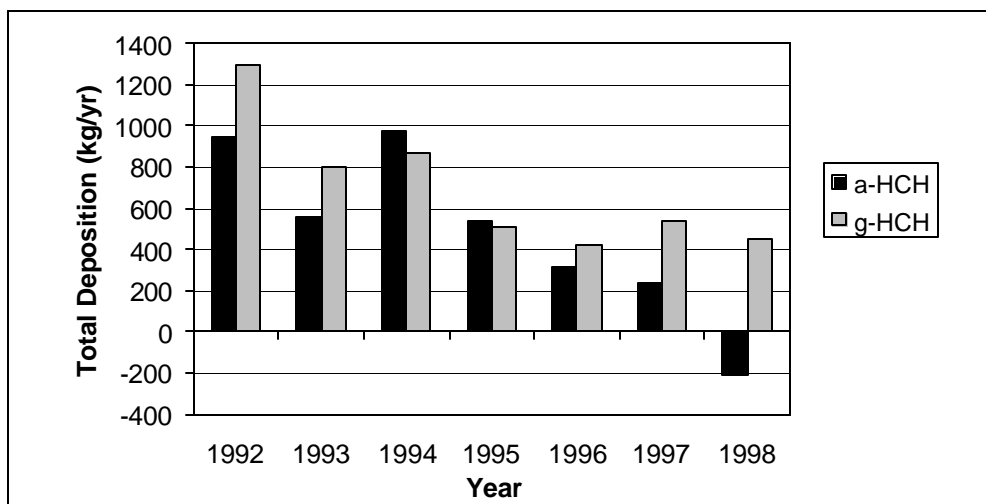
**Table 4. Annual total deposition flows (kg/yr) to all of the Great Lakes. Each annual regional sum includes data from at least three lakes. R-squared and x-intercept values are from the regression of total deposition flows against year for each species. Xintercept values represent the year when equilibrium (flow = 0 kg/yr) will be achieved. All r-squared values shown are significant at the 95% confidence level or greater. Those that are not significant are represented by NS.**

Species	Year							r-squared	x-intercept
	1992	1993	1994	1995	1996	1997	1998		
$\alpha$ -HCH	950	560	980	540	310	240	-210	0.785	1998
$\gamma$ -HCH	1300	800	870	510	420	540	450	0.736	2001
dieldrin	-960	-1400	-1300	-410	-420	-290	-370	0.628	1999
<i>p,p'</i> -DDD	17	28	30	2.7	16	44	25	NS	NS
<i>p,p'</i> -DDE	-70	-75	-65	21	4	-13	-55	NS	NS
<i>p,p'</i> -DDT	190	320	160	56	47	90	26	0.620	1998
HCB	-130	-94	-160	-26	-27	-1.5	15	0.741	1997
PCB 18	-160	-200	-200	-82	-61	-30	-33	0.772	1999
PCB 44	-59	-64	-72	-65	-16	-24	-28	0.579	2001
PCB 52	-80	-77	-71	-19	16	24	22	0.893	1996
PCB 101	-55	-49	-46	-16	0.6	-1.5	-2.6	0.861	1997
sum-PCB	-2900	-3100	-3100	-940	-1300	-730	-690	0.779	1999
phenanthrene	-4500	-4400	-4800	9500	3100	7100	5900	0.605	1994
pyrene	2200	2100	1600	4900	1700	2600	1800	NS	NS
B[ <i>b+k</i> ]F	820	700	620	3700	2400	3000	2700	NS	NS
B[ <i>a</i> ]P	840	840	610	1200	680	1000	830	NS	NS
Pb	160000	100000	79000	34000	41000	99000	78000	NS	NS
As	17000	13000	10000	4100	6800	2200	2200	0.882	1998
Se	27000	21000	18000	4300	5600	1200	1000	0.894	1997
Cd	9800	46000	3400	2100	2800	4700	6200	NS	NS

The difference between pollutants that have banned from use and those that are currently used stands out quite clearly. Throughout the years  $\gamma$ -HCH, a currently-used pesticide, almost always has the highest regional loading values other than the currently-emitted PAHs and metals. In 1994 and 1995, however,  $\gamma$ -HCH flows were close to  $\alpha$ -HCH values. The total deposition values for the banned chemicals dieldrin, *p,p'*-DDE, and HCB, however, are negative, indicating that the lakes themselves are acting as sources of these compounds to the atmosphere. Dieldrin regional flows have been negative in all years and have decreased by approximately a third from 1992 to 1998. This decreasing trend is moving towards equilibrium, which is estimated to have been achieved around 1999. Interestingly, *p,p'*-DDD and DDT total regional flows have been consistently positive across time. This indicates that the Great Lakes basin is still acting as a sink for these chemicals that have been banned from use for over 20 years. In fact, no significant trend

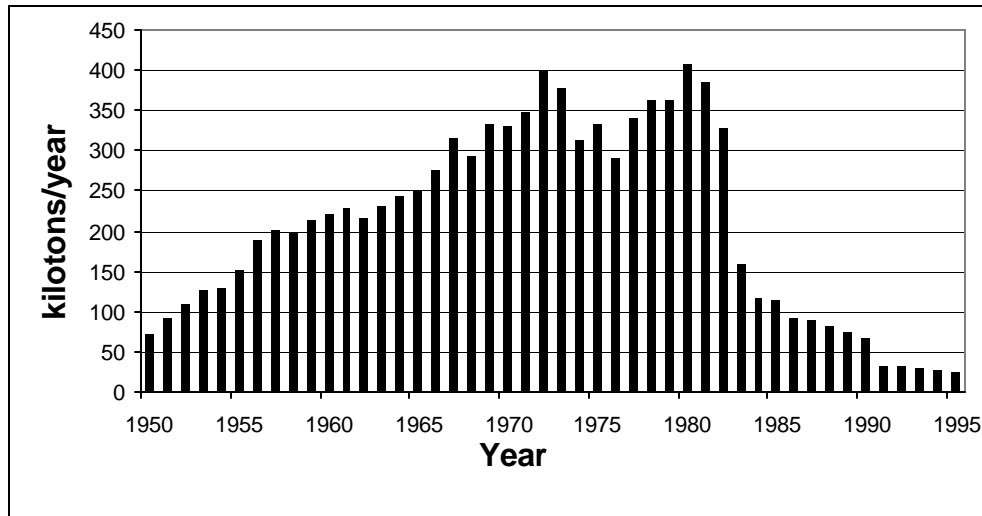
towards equilibrium could be found for  $p,p'$ -DDD. Based on the previously mentioned regression, however,  $p,p'$ -DDT should be very near equilibrium already.

The regional flows over time for both  $\alpha$ - and  $\gamma$ -HCH are shown in Figure 3.  $\alpha$ -HCH shows a statistically significant decrease across the Great Lakes basin and has a net volatilization for the first time in 1998. Flows for  $\gamma$ -HCH, on the other hand, have remained relatively stable since their decrease in 1995. Again, this is probably because its use is restricted rather than banned. However, r-squared values indicate that  $\gamma$ -HCH is showing a real trend towards equilibrium and will achieve it around the year 2001.



**Figure 3. Total flows of a- and g-HCH (kg/yr) over all Great Lakes**

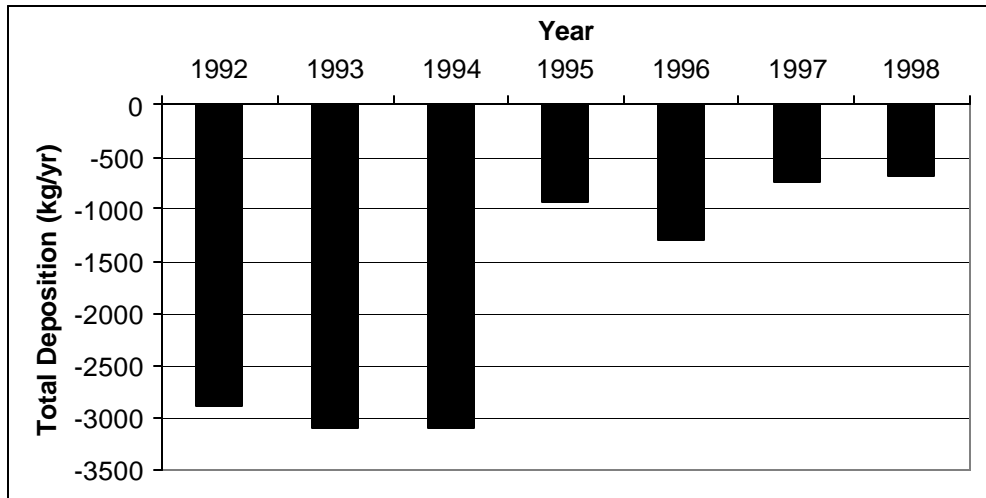
The observed decline in  $\alpha$ -HCH has more meaning when viewed against larger trends in HCH usage. Li (1999) surveyed usage of technical HCH on a global scale from 1948 through 1997. Technical HCH contains five isomers, of which 65% is the  $\alpha$ -isomer and 14% is the  $\gamma$ -isomer. From Li's work, a plot of annual global usage was created from 1950 to 1995 (see Figure 4). It is estimated that during this period, approximately 10 million tonnes of technical HCH was used. It is apparent from the graph that three main declines in global usage occurred. The first began in the early seventies when countries such as Canada (1971), Japan (1973) and the United States (1976) banned usage. Another major drop occurred in 1983 when China banned the use of this pesticide, and a third decrease began in 1990 when the former Soviet Union completely banned its use, and India banned its use for agricultural purposes.



**Figure 4. Global technical HCH usage in kilotons per year. This figure is taken from Li, 1999.**

Data collected by IADN and reported as flows correspond to the tail end of Figure 4. Given that technical HCH usage has been banned by some of the countries that used it most, it is not surprising that we are seeing decreasing atmospheric deposition to the Great Lakes. The downward trend observed in Figure 3 is probably due to the depletion of HCH stores in the soil and vegetation. Judging from Figure 4, we can expect to see continuing decreases in HCH flows to the Great Lakes region.

As HCH flows to the Great Lakes have decreased, so have PCB total flows out of the basin (see Figure 5). Between 1992 and 1998, PCB volatilization has decreased dramatically, with the largest change occurring between 1994 and 1995 when flows dropped from  $-3100$  kg/yr to  $-940$  kg/yr. With an  $r$ -squared of 0.779, sum-PCB flows are showing a real trend towards equilibrium, and should have achieved it around 1999. The largest contributors to this trend were Lakes Superior and Michigan, where total deposition flows for total PCBs dropped by a factor of five and six, respectively.



**Figure 5. Total flow (kg/yr) of PCBs over all the Great Lakes from 1992-1998. 1997 and 1998 data represent suite-PCB while previous years represent total PCBs.**

In general, the regional total deposition of PAHs has remained stable over the years. This is further supported by the lack of a significant trend in Table 4 for most PAHs. Benzo[*b+k*]fluoranthene appears to have increased since 1995, but this is most likely due to the fact that lake water concentrations for this compound from 1992-1994 were for benzo[*k*]fluoranthene only. Phenanthrene, however, stands out from the other PAHs. Total regional flows for this currently emitted pollutant were net volatilization for 1992 through 1994, though one would expect the region to experience net inputs of this compound since it is still released into the atmosphere. It is important to keep in mind, however, that water concentration data for PAHs are lacking in most lakes for most years (see Table A4), making true regional trends hard to assess.

Metals generally have the highest total flows into the lakes. While flows for all metals have decreased over time, they are still quite large. For example, lead inputs to the Great Lakes basin in 1992 totaled 160000 kg/yr. By 1998, these flows had dropped to 78000 kg/yr, but they were still 13 times higher than phenanthrene total deposition and 170 times higher than  $\gamma$ -HCH flows to the entire region, also currently-used or produced chemicals. As with the PAHs, lead and cadmium show no significant trend, but arsenic and selenium do show significant declining trends. These results may be somewhat misleading, however. No wet deposition estimates are available in 1997 and 1998 for arsenic and selenium, but all previous years total deposition estimates from Table 4 include wet deposition. Given that wet deposition is on average four times higher than dry deposition for these two metals, and assuming that wet deposition patterns would follow those of lead and cadmium, that is an increase in 1997 and 1998, we can assume that a significant portion of the regional total deposition estimate for arsenic and selenium is unaccounted for in 1997 and 1998 in Table 4. Thus, while the estimates represent all available data for arsenic and selenium in 1997 and 1998, they most likely do not represent true deposition to the region. If precipitation-phase data had been collected, it is highly probable that declining trends would not be significant for arsenic and selenium just as they are for lead

and cadmium.

## 5. Conclusions

Downward fluxes for pesticides in 1997 and 1998 ranged from 0.01 ng/m<sup>2</sup>/day to 40 ng/m<sup>2</sup>/day, with in-use pesticides such as  $\gamma$ -HCH accounting for the highest fluxes. Volatilization fluxes for those pesticides banned from use were almost 10 times greater than those for currently used pesticides, reaching -37 ng/m<sup>2</sup>/day at their highest. PCB and HCB downward fluxes ranged from 0.02 ng/m<sup>2</sup>/day to 11 ng/m<sup>2</sup>/day across the basin. Volatilization fluxes for these banned commercial chemicals were on the same order as those for banned pesticides. Suite-PCB volatilization fluxes increased from west to east across the basin. Downward fluxes for PAHs ranged from 0.3 ng/m<sup>2</sup>/day to 530 ng/m<sup>2</sup>/day with volatilization fluxes ranging from -0.00001 to -240 ng/m<sup>2</sup>/day. Where water concentration data are available, volatilization of PAHs was almost always less than net inputs. Fluxes for metals ranged from 13 to 840 ng/m<sup>2</sup>/day for dry deposition and from 130 to 5400 ng/m<sup>2</sup>/day for wet deposition. Since the metals analyzed by IADN are nonvolatile, they are not measured in the gas phase. The PAHs and metals measured by IADN are currently emitted through anthropogenic means into the atmosphere and thus have downward (air to water) fluxes much greater than those of the pesticides and PCBs that have been banned from use.

Current (1997-1998) fluxes (ng/m<sup>2</sup>/day) were compared across time and space to better understand loadings trends in the Great Lakes. Pesticide wet deposition fluxes seem to be generally decreasing over time except for  $\gamma$ -HCH at Lakes Huron and Ontario. Since  $\gamma$ -HCH is still in use, this trend is expected. Volatilization of dieldrin from Lake Ontario is the largest pesticide flux observed. The magnitude of PCB wet deposition fluxes is similar for Lakes Superior, Erie, and Michigan. Lake Erie, however, seems unique in that all PCBs measured there reached peak fluxes around 1994 and 1995 and then decreased in the following three years. Gas exchange of PCBs has been, for the most part, in the direction of net volatilization consistently over time with only Lake Michigan showing signs of nearing air-water equilibrium. Wet and dry deposition of PAHs shows no real temporal trend, but spatial analysis indicates that fluxes have increased from west to east across the basin. Gas exchange fluxes for Lakes Superior and Erie for all PAHs show net absorption over time. Metal fluxes for Lakes Huron and Ontario are similar over the years with dry deposition showing no real trend and wet deposition decreasing from 1992-1996 for Cd and Pb, then increasing in 1997 and 1998.

All of the flows and fluxes mentioned above are based on IADN master station data. These stations are remote sites, one on each lake, which measure what are considered to be Great Lakes background contaminant levels. However, spatial differences exist across each lake for many of the compounds we monitor, particularly near urban areas, where atmospheric deposition from cities can be much greater than that from remote sites. In an attempt to assess the impact of urban areas on lakewide loadings, and in accordance with the 1995-1996 loadings report, depositional data from IADN's Chicago site were ex-



trapolated onto Lake Michigan loadings. The impact of Chicago pollution on a small sub-area of Lake Michigan was then compared to loadings calculated at the remote master station. Results demonstrate that urban inputs have a minor lakewide effect for most pesticides. There does, however, seem to be a large effect on *cis*- and *trans*-chlordane, drastically changing lakewide volatilization and markedly increasing total mass loadings. Urban inputs also have a strong effect on the net gas exchange of PCBs. PAHs, currently emitted urban pollutants, show consistently large urban effects in all deposition categories.

In an attempt to explore a more tangible means of examining the loadings results, estimates were investigated on a Great Lakes basin-wide basis by summing the total deposition flow (kg/yr) of each substance over all five lakes for each year. These sums give a good approximation of the larger, regional atmospheric deposition to the Great Lakes. Total deposition for  $\alpha$ -HCH showed a decreasing trend, going from 950 kg/yr in 1992 to -210 kg/yr in 1998. Dieldrin and *p,p'*-DDE, two organochlorine pesticides banned from use, had negative total deposition across time, indicating that the lakes are acting as a source of these chemicals to the atmosphere. Sum-PCB total deposition across the basin also showed net volatilization for all years. Even so, PCB flows out of the Great Lakes have decreased dramatically over time, with the largest drop occurring between 1994 and 1995 when total PCB flows went from -3100 kg/yr to -940 kg/yr. PAHs and metals had the largest regional deposition. PAH flows have, for the most part, remained stable across time. While total loads of metals to the Great Lakes basin have decreased over time, the region was still receiving 78000 kg of lead in 1998.

With seven years of data, IADN has been succeeding at the task of determining the atmospheric loadings of toxic substances to the Great Lakes. This biennial report helps to further elucidate trends in the region and provide a more robust picture. Despite the vast array of compounds measured by IADN, some general trends have emerged among them over time. Loads of banned pesticides and commercial chemicals have decreased over the years, with the lakes themselves actually becoming the main source of these substances to the atmosphere. These trends also suggest that most of the restricted pesticides and PCBs measured by IADN are approaching air-water equilibrium. For those chemicals that are currently emitted, like PAHs, metals, and some pesticides, their loads have remained relatively constant through time. Furthermore, the temporal urban effects analysis in this report continues to raise concerns about the impact of urban pollution on the loadings to the Great Lakes.

## Acknowledgements

Special thanks to the many people whose work supported the production of the IADN loadings results.

- Site operators on Lake Superior – Donald B. Keith and Patricia Keith (Eagle Harbor), Ron Perala (Brule River), Carl Nielsen (Sibley), Larry Barnett (Turkey Lakes)
- Site operator and coordinator on Lake Michigan – Tom van Zoeren (Sleeping Bear Dunes), Nasrim Khalili (IIT- Chicago)
- Site operators on Lake Huron – Floyd Orford (Burnt Island), Terry Romphf (Grand Bend)
- Site coordinator on Lake Erie – Dr. Kim Irvine (Sturgeon Point), Gary Moulard (Pt Pelee), Tony Bucsis (Rock Point)
- Site operator on Lake Ontario – Darrell Smith (Point Petre)
  
- Karen Arnold and Matt O'Dell at Indiana University
- Brian Martin, Jim Woods, Frank MacLean, Chris Green, Andrew Elford, and Helena Dryfhout-Clark at the Centre for Atmospheric Research Experiments of Environment Canada's Meteorological Service of Canada
- Nick Alexandrou, Ky Su, Richard Park, Kulbir Banwait, Art Tham, Cecilia Shin, and Murray Smith at the Organics Analysis Laboratory of Environment Canada's Meteorological Service of Canada
- Michael Comba , Ed Sverko, and the staff of the Organic Analysis Laboratory at Environment Canada's National Laboratory for Environmental Testing
- Bruce Harrison and MaryLou Archer at the Ecosystem Health Division of Environment Canada's Ontario Region
- The staff at Philips Analytical
- Peter Fowlie of Cornerstone Science
  
- The U.S. EPA's Great Lakes National Program Office, Serge L'Italien at Environment Canada's Ontario Region Ecosystem Health Division, Derek Muir and Bill Strachan at Environment Canada's National Water Research Institute, and Environment Canada's Meteorological Service of Canada for generous provision of lake water concentration data used in this report

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## **Appendix A. Selected Data Used in Calculating IADN Loadings**

**Table A1. Summary of meteorological data at IADN master stations, 1992-1998**

<b>Lake</b>	<b>Parameter</b>	<b>1992</b>	<b>1993</b>	<b>1994</b>	<b>1995</b>	<b>1996</b>	<b>1997</b>	<b>1998</b>
Superior	Annual Precipitation (mm)	665	990	665	904	1148	696	1282
	Average Water Surface Temperature (K)	278.4	278.4	278.4	279.2	277.7	276.3	279.1
	Average Wind Speed (m/s)	3.02	3.09	2.99	3.20	2.84	2.75	2.77
Michigan	Annual Precipitation (mm)	866	1214	866	1182	1200	777	1793
	Average Water Surface Temperature (K)	280.0	280.0	280.0	282.5	280.8	279.4	281.9
	Average Wind Speed (m/s)	2.88	2.94	2.90	3.04	2.89	2.84	2.83
Huron	Annual Precipitation (mm)	1110	1110	896	931	908	917	810
	Average Water Surface Temperature (K)	280.8	280.8	281.0	281.8	280.4	278.8	280.8
	Average Wind Speed (m/s)	2.85	3.50	3.38	3.69	3.36	3.35	3.20
Erie	Annual Precipitation (mm)	1073	1389	1073	933	823	933	1242
	Average Water Surface Temperature (K)	283.6	283.6	283.6	284.3	283.4	281.8	283.2
	Average Wind Speed (m/s)	2.85	2.77	2.63	2.99	2.78	3.07	2.71
Ontario	Annual Precipitation (mm)	1021	1021	837	816	1000	958	965
	Average Water Surface Temperature (K)	283.6	283.6	283.6	284.3	283.4	281.8	283.2
	Average Wind Speed (m/s)	4.78	4.98	4.66	4.71	4.51	4.77	4.26



**Table A2. Lake water concentrations for IADN 1997-1998 loadings calculations**

Substance	Lake Superior				Lake Michigan				Lake Huron				Lake Erie				Lake Ontario			
	Conc. (ng/L)	n	COV (%)	Source	Conc. (ng/L)	n	COV (%)	Source	Conc. (ng/L)	n	COV (%)	Source	Conc. (ng/L)	n	COV (%)	Source	Conc. (ng/L)	n	COV (%)	Source
$\alpha$ -HCH	2.04	94	1	1, 2, 3, 4, 5	0.398	5	8	3	0.465	5	11	3	0.322	46	3	3, 6	0.449	50	6	3, 6, 7
$\gamma$ -HCH	0.439	111	1	1, 2, 3, 4, 5	0.119	5	3	3	0.0923	5	55	3	0.252	46	3	3, 6	0.232	50	6	3, 6, 7
Dieldrin	0.145	89	1	1, 2, 4, 5	-	-	-	-	-	-	-	-	0.143	35	8	6	0.141	27	6	6, 7
<i>cis</i> -chlordane	0.0113	94	0	1, 2, 3, 4, 5	0.00713	5	27	3	0.00345	5	15	3	0.00934	46	6	3, 6	0.00493	32	8	3, 6, 7
<i>trans</i> -chlordane	0.0200	94	0	1, 2, 3, 4, 5	0.00466	5	30	3	0.00144	5	14	3	0.0124	46	4	3, 6	0.00262	32	6	3, 6, 7
$\alpha$ -endosulfan	0.00575	64	7	1, 4, 5	-	-	-	-	-	-	-	-	0.0187	40	4	6	0.0272	20	64	6
<i>p, p'</i> -DDE	0.00285	69	4	1, 3, 4, 5	0.0102	5	56	3	0.00324	5	25	3	0.0338	46	2	3, 6	0.0222	25	14	3, 6
<i>p, p'</i> -DDT	0.00866	65	10	1, 4, 5	0.00359	5	37	3	0.00209	5	21	3	0.0667	46	0	3, 6	0.00509	25	13	3, 6
<i>p, p'</i> -DDD	0.00021	47	37	3, 4, 5	0.00034	5	0	3	0.000340	5	0	3	-	-	-	-	-	-	-	-
HCB	0.00909	70	2	1, 3, 4, 5	0.0101	5	17	3	0.00816	5	11	3	0.0120	46	6	3, 6	0.0115	25	4	3, 6
PCB18	0.00150	5	21	3	0.00308	5	27	3	0.00220	5	16	3	0.00816	6	48	3	0.00498	5	40	3
PCB44	0.00156	5	86	3	0.00427	5	20	3	0.00167	5	62	3	0.00816	6	70	3	0.00466	5	55	3
PCB52	0.00138	5	56	3	0.000175	5	0	3	0.000838	5	177	3	0.000175	6	0	3	0.00268	5	209	3
PCB101	0.00026	5	23	3	0.00264	5	21	3	0.00118	5	11	3	0.00315	6	87	3	0.00203	5	22	3
suite-PCB	0.0471	5	21	3	0.0469	5	9	3	0.0530	5	4	3	0.132	6	14	3	0.0781	5	11	3
Phenanthrene	0.568	27	5	1	-	-	-	-	-	-	-	-	1.37	40	6	6	1.41	20	20	6
Pyrene	0.175	27	5	1	-	-	-	-	-	-	-	-	0.385	40	16	6	0.557	20	20	6
B[b+k]F	0.0310	13	38	1	-	-	-	-	-	-	-	-	0.164	40	11	6	0.303	20	20	6
B[a]P	0.0331	27	1	1	-	-	-	-	-	-	-	-	0.147	40	5	6	0.169	20	20	6
<i>trans</i> -nonachlor	0.00182	66	3	2, 3, 4, 5	0.00562	5	55	3	0.00242	5	18	3	0.00348	6	37	3	0.00302	12	22	3, 7
[1,2,3- <i>cd</i> ]P	0.0494	28	5	1	-	-	-	-	-	-	-	-	0.144	40	15	6	-	-	-	-
endosulfan sulfate	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-
sum-PAH (UN ECE)	0.113	-	11	1	-	-	-	-	-	-	-	-	0.456	-	6	6	-	-	-	-

Data Sources:







1. EHD cruise (CCGV Limnos) of spring and summer 1997 analyzed by NLET, under contract to Maxxam Analytics; 2. CCGV Limnos cruise of spring and summer 1997 analyzed by MSC; 3. U.S. R/V Lake Guardian cruise of spring 1997 analyzed by U.S. EPA GLNPO; 4. CCGV Limnos cruise of spring and summer 1997 analyzed by NWRI; 5. CCGV Limnos cruise of spring 1998 analyzed by NWRI; 6. EHD cruises (CCGV Limnos) of spring 1998 on Lake Ontario and spring and summer 1998 on Lake Erie analyzed by NLET, under contract to Maxxam Analytics; 7. CCGV Limnos cruise of spring 1998 analyzed by MSC

**Table A3. IADN concentration data availability from 1992 to 1998 for all master stations**

Agency Media		Year						
		1992	1993	1994	1995	1996	1997	1998
<b>Substance</b>								
<b>EPA: Eagle Harbor, Sleeping Bear Dunes, Sturgeon Pt., IIT-Chicago</b>								
Gas								
PCBs	Eagle Harbor							
	Sleeping Bear Dunes							
	Sturgeon Point							
	IIT-Chicago							
	Pesticides							
	PAHs							
Particle								
	PCBs							
	Pesticides							
	PAHs							
	metals							
Precipitation								
	PCBs							
	Pesticides							
	PAHs							
	metals							
<b>MSC: Burnt Island, Pt. Petre</b>								
Gas								
	PCBs							
	Pesticides							
	PAHs							
Particle								
	PCBs							
	Pesticides							
	PAHs							
	metals							
<b>EHD: Burnt Island, Pt. Petre</b>								
Precipitation								
	PCBs							
	Pesticides							
	PAHs							
	metals							
<b>NWRI: Burnt Island, Pt. Petre</b>								
Precipitation								
	PCBs							
	Pesticides							
	metals							



**Table A5. Lake water concentration availability for additional substances added to loadings calculations in 1995**

Additional Species	Year			
	1995	1996	1997	1998
<b>Water Concentration: Non-IADN Sources</b>				
<b><i>α</i>-endosulfan</b>				
Superior				
Michigan				
Huron				
Erie				
Ontario				
<b><i>cis</i>-chlordane, <i>trans</i>-chlordane</b>				
Superior				
Michigan				
Huron				
Erie				
Ontario				
<b><i>trans</i>-nonachlor</b>				
Superior				
Michigan				
Huron				
Erie				
Ontario				
<b>Endosulfan Sulfate</b>				
Superior				
Michigan				
Huron				
Erie				
Ontario				
<b>Indeno[1,2,3-<i>cd</i>]pyrene</b>				
Superior				
Michigan				
Huron				
Erie				
Ontario				
				

## **Appendix B. Annual IADN Fluxes (ng/m<sup>2</sup>/d) for 1997-1998**

**Table B1. Annual atmospheric fluxes to Lake Superior for 1997**

Banned Organochlorine Pesticides	Lake Superior 1997											
	Wet Deposition		Dry Deposition		Gas Exchange						Total Mass Flux	
					Gas Absorption		Volatilization		Net Gas Exchange			
	Mean ng/m <sup>2</sup> /d	COV %	Mean ng/m <sup>2</sup> /d	COV %	Mean ng/m <sup>2</sup> /d	COV %	Mean ng/m <sup>2</sup> /d	COV %	Mean ng/m <sup>2</sup> /d	COV %	Mean ng/m <sup>2</sup> /d	COV %
$\alpha$ -HCH	1.2	78	0.097	260	19	79	-28	50	-9.7	150	-8.4	170
dieldrin	0.59	69	0.51	140	1.4	140	-8.7	50	-7.2	59	-6.1	71
cis-chlordane	0.13	190	0.083	170	0.48	110	-1	50	-0.56	120	-0.35	210
trans-chlordane	0.038	180	0.054	140	0.51	180	-2.2	50	-1.7	77	-1.6	82
trans-nonachlor	0.042	180	0.049	130	0.11	110	-0.29	50	-0.18	100	-0.089	230
p,p'-DDD	0.011	240	0.037	250	0.2	140	-0.0019	62	0.2	140	0.25	120
p,p'-DDE	0.085	82	-	-	0.35	220	-0.29	50	0.062	980	0.15	410
p,p'-DDT	0.13	130	0.047	100	0.29	140	-0.2	51	0.088	340	0.27	130
In-Use Pesticides	Lake Superior 1997											
	Wet Deposition		Dry Deposition		Gas Exchange						Total Mass Flux	
					Gas Absorption		Volatilization		Net Gas Exchange			
	Mean ng/m <sup>2</sup> /d	COV %	Mean ng/m <sup>2</sup> /d	COV %	Mean ng/m <sup>2</sup> /d	COV %	Mean ng/m <sup>2</sup> /d	COV %	Mean ng/m <sup>2</sup> /d	COV %	Mean ng/m <sup>2</sup> /d	COV %
$\gamma$ -HCH (lindane)	0.89	140	0.14	150	5.7	140	-3.4	50	2.3	280	3.3	200
$\alpha$ -endosulfan	0.52	87	0.97	130	1.9	210	-0.89	50	1	320	2.5	140
endosulfan sulfate	0.24	100	0.11	170	-	-	-	-	-	-	-	-
Banned Organochlorine Commercial Chemicals	Lake Superior 1997											
	Wet Deposition		Dry Deposition		Gas Exchange						Total Mass Flux	
					Gas Absorption		Volatilization		Net Gas Exchange			
	Mean ng/m <sup>2</sup> /d	COV %	Mean ng/m <sup>2</sup> /d	COV %	Mean ng/m <sup>2</sup> /d	COV %	Mean ng/m <sup>2</sup> /d	COV %	Mean ng/m <sup>2</sup> /d	COV %	Mean ng/m <sup>2</sup> /d	COV %
HCB	0.034	57	-	-	2.6	66	-2	50	0.59	160	0.62	150
PCB 18	0.078	130	-	-	0.13	100	-0.3	54	-0.17	120	-0.092	250
PCB 44	0.089	150	-	-	0.25	130	-0.27	99	-0.019	810	0.07	290
PCB 52	0.1	200	-	-	0.27	110	-0.22	75	0.051	350	0.15	180
PCB 101	0.1	110	-	-	0.17	120	-0.038	55	0.13	150	0.23	97
suite-PCB	2.5	110	-	-	4.2	110	-11	54	-7.1	100	-4.6	170
PAHs and Metals	Lake Superior 1997											
	Wet Deposition		Dry Deposition		Gas Exchange						Total Mass Flux	
					Gas Absorption		Volatilization		Net Gas Exchange			
	Mean ng/m <sup>2</sup> /d	COV %	Mean ng/m <sup>2</sup> /d	COV %	Mean ng/m <sup>2</sup> /d	COV %	Mean ng/m <sup>2</sup> /d	COV %	Mean ng/m <sup>2</sup> /d	COV %	Mean ng/m <sup>2</sup> /d	COV %
PHEN	9.1	180	3.2	130	100	96	-47	50	55	150	67	130
PYR	5.7	140	2.5	120	8.2	120	-6.2	50	2.1	370	10	120
B[b+k]F	7.7	140	5.6	100	3.3	67	-0.045	63	3.3	67	17	73
B[a]P	2.4	87	1.3	120	1.3	59	-0.13	50	1.2	60	4.9	55
I[1,2,3-cd]P	4.6	160	2.5	130	0.83	60	-2.4E-05	50	0.83	60	7.9	100
sum-PAH (UN ECE)	15	91	9.4	110	5.4	44	-0.17	40	5.3	45	30	58
Pb	-	-	-	-	-	-	-	-	-	-	-	-
As	-	-	-	-	-	-	-	-	-	-	-	-
Se	-	-	-	-	-	-	-	-	-	-	-	-
Cd	-	-	-	-	-	-	-	-	-	-	-	-

**Table B2. Annual atmospheric fluxes to Lake Superior for 1998**

Banned Or- ganochlorine Pesti- cides	Lake Superior 1998											
	Wet Deposition		Dry Deposition		Gas Exchange						Total Mass Flux	
					Gas Absorption		Volatilization		Net Gas Exchange			
	Mean ng/m <sup>2</sup> /d	COV %	Mean ng/m <sup>2</sup> /d	COV %	Mean ng/m <sup>2</sup> /d	COV %	Mean ng/m <sup>2</sup> /d	COV %	Mean ng/m <sup>2</sup> /d	COV %	Mean ng/m <sup>2</sup> /d	COV %
α-HCH	0.22	78	0.063	250	21	98	-37	50	-16	170	-16	170
dieldrin	0.38	69	0.6	120	1	140	-10	50	-9.5	54	-8.5	61
cis-chlordane	0.092	190	0.069	120	0.32	91	-1.2	50	-0.88	60	-0.72	78
trans-chlordane	0.024	180	0.053	120	0.21	110	-2.5	50	-2.3	51	-2.2	53
trans-nonachlor	0.019	180	0.033	130	0.12	95	-0.31	50	-0.19	75	-0.14	110
p,p'-DDD	0.025	240	0.021	110	0.17	120	-0.0026	62	0.17	120	0.22	97
p,p'-DDE	0.077	82	-	-	0.19	91	-0.33	50	-0.13	140	-0.053	360
p,p'-DDT	0.13	130	0.059	140	0.31	140	-0.26	51	0.049	440	0.24	120
In-Use Pesticides												
	Wet Deposition		Dry Deposition		Gas Exchange						Total Mass Flux	
					Gas Absorption		Volatilization		Net Gas Exchange			
	Mean ng/m <sup>2</sup> /d	COV %	Mean ng/m <sup>2</sup> /d	COV %	Mean ng/m <sup>2</sup> /d	COV %	Mean ng/m <sup>2</sup> /d	COV %	Mean ng/m <sup>2</sup> /d	COV %	Mean ng/m <sup>2</sup> /d	COV %
γ-HCH (lindane)	0.34	140	0.075	130	4.2	99	-4.3	50	-0.15	1400	0.27	800
α-endosulfan	0.16	87	0.95	110	1.5	220	-0.91	50	0.62	370	1.7	150
endosulfan sulfate	0.12	100	0.061	150	-	-	-	-	-	-	-	-
Banned Or- ganochlorine Commercial Chemicals												
	Wet Deposition		Dry Deposition		Gas Exchange						Total Mass Flux	
					Gas Absorption		Volatilization		Net Gas Exchange			
	Mean ng/m <sup>2</sup> /d	COV %	Mean ng/m <sup>2</sup> /d	COV %	Mean ng/m <sup>2</sup> /d	COV %	Mean ng/m <sup>2</sup> /d	COV %	Mean ng/m <sup>2</sup> /d	COV %	Mean ng/m <sup>2</sup> /d	COV %
HCB	0.02	57	-	-	2.9	63	-2.1	50	0.85	160	0.87	160
PCB 18	0.038	130	-	-	0.13	93	-0.31	54	-0.18	89	-0.14	120
PCB 44	0.18	150	-	-	0.21	110	-0.28	99	-0.076	500	0.1	470
PCB 52	0.055	200	-	-	0.25	98	-0.23	75	0.018	480	0.073	190
PCB 101	0.12	110	-	-	0.15	97	-0.041	55	0.11	120	0.23	81
suite-PCB	2.3	110	-	-	3.5	96	-11	54	-7.8	74	-5.5	110
PAHs and Metals												
	Wet Deposition		Dry Deposition		Gas Exchange						Total Mass Flux	
					Gas Absorption		Volatilization		Net Gas Exchange			
	Mean ng/m <sup>2</sup> /d	COV %	Mean ng/m <sup>2</sup> /d	COV %	Mean ng/m <sup>2</sup> /d	COV %	Mean ng/m <sup>2</sup> /d	COV %	Mean ng/m <sup>2</sup> /d	COV %	Mean ng/m <sup>2</sup> /d	COV %
PHEN	3.7	180	2.5	120	81	93	-54	50	28	200	34	170
PYR	2.8	140	2.4	140	8.2	130	-7.2	50	0.94	560	6.1	120
B[b+k]F	4.9	140	4.8	110	3.4	87	-0.063	63	3.4	88	13	70
B[a]P	1.6	87	1.2	150	1.3	60	-0.17	50	1.1	63	3.9	61
I[1,2,3-cd]P	2.4	160	2.1	150	0.8	60	-3.3E-05	50	0.8	60	5.3	94
sum-PAH (UN ECE)	8.9	91	8.2	110	5.5	58	-0.24	40	5.3	59	22	57
Pb	-	-	-	-	-	-	-	-	-	-	-	-
As	-	-	-	-	-	-	-	-	-	-	-	-
Se	-	-	-	-	-	-	-	-	-	-	-	-
Cd	-	-	-	-	-	-	-	-	-	-	-	-

**Table B3. Annual atmospheric fluxes to Lake Michigan for 1997**

Banned Or- ganochlorine Pes- ticides	Lake Michigan 1997											
	Wet Deposition		Dry Deposition		Gas Exchange						Total Mass Flux	
					Gas Absorption		Volatilization		Net Gas Exchange			
	Mean ng/m <sup>2</sup> /d	COV %	Mean ng/m <sup>2</sup> /d	COV %	Mean ng/m <sup>2</sup> /d	COV %	Mean ng/m <sup>2</sup> /d	COV %	Mean ng/m <sup>2</sup> /d	COV %	Mean ng/m <sup>2</sup> /d	COV %
$\alpha$ -HCH	1.9	260	0.16	410	22	110	-7.3	51	14	150	16	130
dieldrin	1.2	120	0.73	140	3.4	100	-	-	-	-	-	-
<i>cis</i> -chlordane	0.47	190	0.12	180	0.85	90	-0.75	57	0.1	170	0.69	140
<i>trans</i> -chlordane	0.052	130	0.069	140	0.56	94	-0.58	59	-0.017	200	0.1	120
<i>trans</i> -nonachlor	0.037	130	0.054	130	0.19	110	-0.97	74	-0.77	110	-0.68	130
<i>p,p'</i> -DDD	0.017	110	0.036	130	0.17	150	-0.0044	50	0.17	150	0.22	120
<i>p,p'</i> -DDE	0.12	79	-	-	1.1	99	-1.2	75	-0.073	270	0.047	470
<i>p,p'</i> -DDT	0.2	220	0.052	110	0.73	120	-0.11	62	0.62	130	0.87	110
In-Use Pesticides												
	Wet Deposition		Dry Deposition		Gas Exchange						Total Mass Flux	
					Gas Absorption		Volatilization		Net Gas Exchange			
	Mean ng/m <sup>2</sup> /d	COV %	Mean ng/m <sup>2</sup> /d	COV %	Mean ng/m <sup>2</sup> /d	COV %	Mean ng/m <sup>2</sup> /d	COV %	Mean ng/m <sup>2</sup> /d	COV %	Mean ng/m <sup>2</sup> /d	COV %
$\gamma$ -HCH (lindane)	1.5	290	0.11	150	8.4	110	-1.2	50	7.2	120	8.8	110
$\alpha$ -endosulfan	0.69	230	1.7	200	39	170	-	-	-	-	-	-
endosulfan sulfate	0.44	300	0.2	160	-	-	-	-	-	-	-	-
Banned Or- ganochlorine Commercial Chemicals												
	Wet Deposition		Dry Deposition		Gas Exchange						Total Mass Flux	
					Gas Absorption		Volatilization		Net Gas Exchange			
	Mean ng/m <sup>2</sup> /d	COV %	Mean ng/m <sup>2</sup> /d	COV %	Mean ng/m <sup>2</sup> /d	COV %	Mean ng/m <sup>2</sup> /d	COV %	Mean ng/m <sup>2</sup> /d	COV %	Mean ng/m <sup>2</sup> /d	COV %
HCB	0.035	48	-	-	3.1	64	-2.3	53	0.74	160	0.78	150
PCB 18	0.039	65	-	-	0.32	120	-0.65	57	-0.39	100	-0.35	110
PCB 44	0.062	75	-	-	0.31	110	-0.78	54	-0.52	68	-0.46	78
PCB 52	0.079	55	-	-	0.48	110	-0.03	50	0.45	120	0.53	100
PCB 101	0.051	93	-	-	0.20	100	-0.42	54	-0.24	79	-0.19	100
suite-PCB	1.2	44	-	-	6.2	120	-8.8	51	-3.2	170	-2	280
PAHs and Metals												
	Wet Deposition		Dry Deposition		Gas Exchange						Total Mass Flux	
					Gas Absorption		Volatilization		Net Gas Exchange			
	Mean ng/m <sup>2</sup> /d	COV %	Mean ng/m <sup>2</sup> /d	COV %	Mean ng/m <sup>2</sup> /d	COV %	Mean ng/m <sup>2</sup> /d	COV %	Mean ng/m <sup>2</sup> /d	COV %	Mean ng/m <sup>2</sup> /d	COV %
PHEN	15	340	4.6	130	160	81	-	-	-	-	-	-
PYR	11	340	4.7	140	15	84	-	-	-	-	-	-
B[ <i>b+k</i> ]F	16	140	8.7	120	4.4	66	-	-	-	-	-	-
B[ <i>a</i> ]P	5.7	160	2.3	140	1.4	62	-	-	-	-	-	-
I[1,2,3- <i>cd</i> ]P	9.4	160	4	150	1.3	75	-	-	-	-	-	-
sum-PAH (UN ECE)	31	91	15	120	7.2	45	-	-	-	-	-	-
Pb	-	-	-	-	-	-	-	-	-	-	-	-
As	-	-	-	-	-	-	-	-	-	-	-	-
Se	-	-	-	-	-	-	-	-	-	-	-	-
Cd	-	-	-	-	-	-	-	-	-	-	-	-



**Table B4. Annual atmospheric fluxes to Lake Michigan for 1998**

Banned Or- ganochlorine Pes- ticides	Lake Michigan 1998											
	Wet Deposition		Dry Deposition		Gas Exchange						Total Mass Flux	
					Gas Absorption		Volatilization		Net Gas Exchange			
	Mean ng/m <sup>2</sup> /d	COV %	Mean ng/m <sup>2</sup> /d	COV %	Mean ng/m <sup>2</sup> /d	COV %	Mean ng/m <sup>2</sup> /d	COV %	Mean ng/m <sup>2</sup> /d	COV %	Mean ng/m <sup>2</sup> /d	COV %
α-HCH	0.8	260	0.043	150	18	110	-8.8	51	9.7	170	11	150
dieldrin	1.5	120	0.87	120	2.1	140	-	-	-	-	-	-
cis-chlordane	0.52	190	0.15	160	0.61	99	-0.82	57	-0.21	500	0.46	320
trans-chlordane	0.058	130	0.085	120	0.4	110	-0.63	59	-0.23	300	-0.087	810
trans-nonachlor	0.042	130	0.039	130	0.2	100	-0.99	74	-0.79	90	-0.71	100
p,p'-DDD	0.14	110	0.047	130	0.26	140	-0.0055	50	0.25	150	0.44	93
p,p'-DDE	0.13	79	-	-	0.7	110	-1.3	75	-0.57	320	-0.44	420
p,p'-DDT	0.14	220	0.093	260	0.79	170	-0.13	62	0.66	190	0.89	150
In-Use Pesticides												
	Wet Deposition		Dry Deposition		Gas Exchange						Total Mass Flux	
					Gas Absorption		Volatilization		Net Gas Exchange			
	Mean ng/m <sup>2</sup> /d	COV %	Mean ng/m <sup>2</sup> /d	COV %	Mean ng/m <sup>2</sup> /d	COV %	Mean ng/m <sup>2</sup> /d	COV %	Mean ng/m <sup>2</sup> /d	COV %	Mean ng/m <sup>2</sup> /d	COV %
γ-HCH (lindane)	0.61	290	0.089	220	10	160	-1.4	50	8.9	180	9.6	170
α-endosulfan	0.49	230	6.6	310	40	170	-	-	-	-	-	-
endosulfan sulfate	0.42	300	0.17	160	-	-	-	-	-	-	-	-
Banned Or- ganochlorine Commercial Chemicals												
	Wet Deposition		Dry Deposition		Gas Exchange						Total Mass Flux	
					Gas Absorption		Volatilization		Net Gas Exchange			
	Mean ng/m <sup>2</sup> /d	COV %	Mean ng/m <sup>2</sup> /d	COV %	Mean ng/m <sup>2</sup> /d	COV %	Mean ng/m <sup>2</sup> /d	COV %	Mean ng/m <sup>2</sup> /d	COV %	Mean ng/m <sup>2</sup> /d	COV %
HCB	0.039	48	-	-	3.6	67	-2.3	53	1.2	180	1.2	180
PCB 18	0.065	65	-	-	0.40	96	-0.66	57	-0.26	240	-0.2	310
PCB 44	0.13	75	-	-	0.28	120	-0.8	54	-0.52	130	-0.39	180
PCB 52	0.091	55	-	-	0.55	98	-0.031	50	0.52	100	0.61	86
PCB 101	0.082	93	-	-	0.24	110	-0.44	54	-0.20	230	-0.12	390
suite-PCB	1.8	44	-	-	7.4	100	-9	51	-1.6	2600	0.2	21000
PAHs and Metals												
	Wet Deposition		Dry Deposition		Gas Exchange						Total Mass Flux	
					Gas Absorption		Volatilization		Net Gas Exchange			
	Mean ng/m <sup>2</sup> /d	COV %	Mean ng/m <sup>2</sup> /d	COV %	Mean ng/m <sup>2</sup> /d	COV %	Mean ng/m <sup>2</sup> /d	COV %	Mean ng/m <sup>2</sup> /d	COV %	Mean ng/m <sup>2</sup> /d	COV %
PHEN	8.5	340	5.8	130	130	77	-	-	-	-	-	-
PYR	6.4	340	7.1	170	19	110	-	-	-	-	-	-
B[b+k]F	13	150	14	140	5.3	93	-	-	-	-	-	-
B[a]P	4.2	160	4.1	210	1.5	100	-	-	-	-	-	-
I[1,2,3-cd]P	6.6	160	8.4	210	1.8	170	-	-	-	-	-	-
sum-PAH (UN ECE)	24	94	26	130	8.5	69	-	-	-	-	-	-
Pb	-	-	-	-	-	-	-	-	-	-	-	-
As	-	-	-	-	-	-	-	-	-	-	-	-
Se	-	-	-	-	-	-	-	-	-	-	-	-
Cd	-	-	-	-	-	-	-	-	-	-	-	-

**Table B5. Annual atmospheric fluxes to Lake Huron for 1997**

Banned Or- ganochlorine Pes- ticides	Lake Huron 1997											
	Wet Deposition		Dry Deposition		Gas Exchange						Total Mass Flux	
					Gas Absorption		Volatilization		Net Gas Exchange			
	Mean ng/m <sup>2</sup> /d	COV %	Mean ng/m <sup>2</sup> /d	COV %	Mean ng/m <sup>2</sup> /d	COV %	Mean ng/m <sup>2</sup> /d	COV %	Mean ng/m <sup>2</sup> /d	COV %	Mean ng/m <sup>2</sup> /d	COV %
α-HCH	3.3	240	-	-	8.3	68	-9	51	-0.67	3000	2.6	830
dieldrin	1.2	160	-	-	1.4	110	-	-	-	-	-	-
cis-chlordane	0.17	-	-	-	0.27	110	-0.43	52	-0.16	510	0.01	-
trans-chlordane	0.21	-	-	-	0.19	110	-0.22	52	-0.032	680	0.18	-
trans-nonachlor	-	-	-	-	0.12	110	-0.57	53	-0.45	110	-	-
p,p'-DDD	1.2	960	-	-	0.046	91	-0.0046	50	0.042	98	1.2	960
p,p'-DDE	0.97	1900	-	-	0.21	110	-0.44	56	-0.23	280	0.74	2500
p,p'-DDT	2.1	2500	-	-	0.33	100	-0.068	54	0.26	120	2.4	2200
In-Use Pesticides												
	Wet Deposition		Dry Deposition		Gas Exchange						Total Mass Flux	
					Gas Absorption		Volatilization		Net Gas Exchange			
	Mean ng/m <sup>2</sup> /d	COV %	Mean ng/m <sup>2</sup> /d	COV %	Mean ng/m <sup>2</sup> /d	COV %	Mean ng/m <sup>2</sup> /d	COV %	Mean ng/m <sup>2</sup> /d	COV %	Mean ng/m <sup>2</sup> /d	COV %
γ-HCH (lindane)	4.8	160	-	-	3.5	110	-0.98	74	2.6	150	7.4	120
α-endosulfan	1.3	230	-	-	5.1	150	-	-	-	-	-	-
endosulfan sulfate	-	-	-	-	-	-	-	-	-	-	-	-
Banned Or- ganochlorine Commercial Chemicals												
	Wet Deposition		Dry Deposition		Gas Exchange						Total Mass Flux	
					Gas Absorption		Volatilization		Net Gas Exchange			
	Mean ng/m <sup>2</sup> /d	COV %	Mean ng/m <sup>2</sup> /d	COV %	Mean ng/m <sup>2</sup> /d	COV %	Mean ng/m <sup>2</sup> /d	COV %	Mean ng/m <sup>2</sup> /d	COV %	Mean ng/m <sup>2</sup> /d	COV %
HCB	0.17	490	-	-	2	63	-2.7	51	-0.73	150	-0.56	250
PCB 18	-	-	-	-	0.64	82	-0.65	53	-0.0085	710	-0.0085	710
PCB 44	-	-	-	-	0.12	97	-0.41	79	-0.29	150	-0.29	150
PCB 52	-	-	-	-	0.28	90	-0.18	180	0.092	280	0.092	280
PCB 101	-	-	-	-	0.12	94	-0.24	51	-0.13	170	-0.13	170
suite-PCB	-	-	-	-	3.1	87	-13	50	-10	80	-10	80
PAHs and Metals												
	Wet Deposition		Dry Deposition		Gas Exchange						Total Mass Flux	
					Gas Absorption		Volatilization		Net Gas Exchange			
	Mean ng/m <sup>2</sup> /d	COV %	Mean ng/m <sup>2</sup> /d	COV %	Mean ng/m <sup>2</sup> /d	COV %	Mean ng/m <sup>2</sup> /d	COV %	Mean ng/m <sup>2</sup> /d	COV %	Mean ng/m <sup>2</sup> /d	COV %
PHEN	35	290	6.6	170	64	84	-	-	-	-	-	-
PYR	31	290	7.1	160	10	120	-	-	-	-	-	-
B[b+k]F	28	-	9.8	110	1.5	100	-	-	-	-	-	-
B[a]P	13	-	3.9	160	0.28	60	-	-	-	-	-	-
I[1,2,3-cd]P	20	1500	6.4	140	1.7	59	-	-	-	-	-	-
sum-PAH (UN ECE)	61	-	20	110	3.4	53	-	-	-	-	-	-
Pb	2800	1600	570	150	-	-	-	-	-	-	3400	1300
As	-	-	70	140	-	-	-	-	-	-	-	-
Se	-	-	27	230	-	-	-	-	-	-	-	-
Cd	130	1900	22	170	-	-	-	-	-	-	150	1600

**Table B6. Annual atmospheric fluxes to Lake Huron for 1998**

Banned Or- ganochlorine Pes- ticides	Lake Huron 1998											
	Wet Deposition		Dry Deposition		Gas Exchange						Total Mass Flux	
					Gas Absorption		Volatilization		Net Gas Exchange			
	Mean ng/m <sup>2</sup> /d	COV %	Mean ng/m <sup>2</sup> /d	COV %	Mean ng/m <sup>2</sup> /d	COV %	Mean ng/m <sup>2</sup> /d	COV %	Mean ng/m <sup>2</sup> /d	COV %	Mean ng/m <sup>2</sup> /d	COV %
α-HCH	1.8	240	-	-	6.8	67	-11	51	-3.8	220	-2	470
dieldrin	0.78	160	-	-	1.4	110	-	-	-	-	-	-
cis-chlordane	0.077	-	-	-	0.28	100	-0.48	52	-0.19	260	-0.11	-
trans-chlordane	0.16	-	-	-	0.19	97	-0.24	52	-0.051	460	0.11	-
trans-nonachlor	0.018	-	-	-	0.11	100	-0.55	53	-0.45	99	-0.43	-
p,p'-DDD	0.28	960	-	-	0.042	120	-0.0056	50	0.036	130	0.32	840
p,p'-DDE	0.27	1900	-	-	0.26	98	-0.49	56	-0.23	210	0.04	13000
p,p'-DDT	0.13	2500	-	-	0.27	160	-0.081	54	0.19	210	0.32	1000
In-Use Pesticides												
	Wet Deposition		Dry Deposition		Gas Exchange						Total Mass Flux	
					Gas Absorption		Volatilization		Net Gas Exchange			
	Mean ng/m <sup>2</sup> /d	COV %	Mean ng/m <sup>2</sup> /d	COV %	Mean ng/m <sup>2</sup> /d	COV %	Mean ng/m <sup>2</sup> /d	COV %	Mean ng/m <sup>2</sup> /d	COV %	Mean ng/m <sup>2</sup> /d	COV %
γ-HCH (lindane)	5.8	160	-	-	2.8	120	-1.1	74	1.7	180	7.5	130
α-endosulfan	1.5	230	-	-	4.8	140	-	-	-	-	-	-
endosulfan sulfate	-	-	-	-	-	-	-	-	-	-	-	-
Banned Or- ganochlorine Commercial Chemicals												
	Wet Deposition		Dry Deposition		Gas Exchange						Total Mass Flux	
					Gas Absorption		Volatilization		Net Gas Exchange			
	Mean ng/m <sup>2</sup> /d	COV %	Mean ng/m <sup>2</sup> /d	COV %	Mean ng/m <sup>2</sup> /d	COV %	Mean ng/m <sup>2</sup> /d	COV %	Mean ng/m <sup>2</sup> /d	COV %	Mean ng/m <sup>2</sup> /d	COV %
HCB	0.043	490	-	-	1.7	67	-2.5	51	-0.8	150	-0.76	160
PCB 18	-	-	-	-	0.38	71	-0.6	53	-0.22	180	-0.22	180
PCB 44	-	-	-	-	0.095	76	-0.4	79	-0.3	130	-0.3	130
PCB 52	-	-	-	-	0.22	78	-0.18	180	0.033	910	0.033	910
PCB 101	-	-	-	-	0.09	85	-0.25	51	-0.16	110	-0.16	110
suite-PCB	-	-	-	-	1.9	76	-13	50	-11	73	-11	73
PAHs and Metals												
	Wet Deposition		Dry Deposition		Gas Exchange						Total Mass Flux	
					Gas Absorption		Volatilization		Net Gas Exchange			
	Mean ng/m <sup>2</sup> /d	COV %	Mean ng/m <sup>2</sup> /d	COV %	Mean ng/m <sup>2</sup> /d	COV %	Mean ng/m <sup>2</sup> /d	COV %	Mean ng/m <sup>2</sup> /d	COV %	Mean ng/m <sup>2</sup> /d	COV %
PHEN	33	290	2.7	150	46	70	-	-	-	-	-	-
PYR	17	290	4.3	150	10	80	-	-	-	-	-	-
B[b+k]F	19	-	7.2	130	1.3	63	-	-	-	-	-	-
B[a]P	9.6	-	2.6	150	0.4	59	-	-	-	-	-	-
I[1,2,3-cd]P	16	1500	4.7	170	2.1	59	-	-	-	-	-	-
sum-PAH (UN ECE)	45	-	14	120	3.8	40	-	-	-	-	-	-
Pb	1300	1600	330	150	-	-	-	-	-	-	1600	1300
As	-	-	55	170	-	-	-	-	-	-	-	-
Se	-	-	26	210	-	-	-	-	-	-	-	-
Cd	190	1900	13	160	-	-	-	-	-	-	200	1800

**Table B7. Annual atmospheric fluxes to Lake Erie for 1997**

Banned Organochlorine Pesticides	Lake Erie 1997											
	Wet Deposition		Dry Deposition		Gas Exchange						Total Mass Flux	
					Gas Absorption		Volatilization		Net Gas Exchange			
	Mean ng/m <sup>2</sup> /d	COV %	Mean ng/m <sup>2</sup> /d	COV %	Mean ng/m <sup>2</sup> /d	COV %	Mean ng/m <sup>2</sup> /d	COV %	Mean ng/m <sup>2</sup> /d	COV %	Mean ng/m <sup>2</sup> /d	COV %
$\alpha$ -HCH	0.58	130	0.026	130	16	79	-7.1	50	8.7	120	9.3	110
dieldrin	0.98	120	0.9	120	3.2	140	-12	51	-8.7	130	-6.8	170
cis-chlordane	1.3	130	0.28	120	1.1	98	-1.1	50	0.035	490	1.6	110
trans-chlordane	0.08	120	0.12	130	0.86	100	-1.8	50	-0.89	180	-0.69	230
trans-nonachlor	0.053	93	0.083	130	0.25	110	-0.68	62	-0.43	170	-0.29	260
p,p'-DDD	0.079	330	0.077	190	0.37	170	-	-	-	-	-	-
p,p'-DDE	0.2	160	-	-	1.6	100	-4.3	50	-2.7	110	-2.5	120
p,p'-DDT	0.24	150	0.13	160	1.4	110	-2.4	50	-1.1	180	-0.73	280
In-Use Pesticides												
	Wet Deposition		Dry Deposition		Gas Exchange						Total Mass Flux	
					Gas Absorption		Volatilization		Net Gas Exchange			
	Mean ng/m <sup>2</sup> /d	COV %	Mean ng/m <sup>2</sup> /d	COV %	Mean ng/m <sup>2</sup> /d	COV %	Mean ng/m <sup>2</sup> /d	COV %	Mean ng/m <sup>2</sup> /d	COV %	Mean ng/m <sup>2</sup> /d	COV %
$\gamma$ -HCH (lindane)	0.56	140	0.14	170	7.5	130	-3	50	4.4	200	5.1	170
$\alpha$ -endosulfan	0.48	180	1.3	120	31	160	-0.0047	50	31	160	33	150
endosulfan sulfate	-	-	0.23	160	-	-	-	-	-	-	-	-
Banned Organochlorine Commercial Chemicals												
	Wet Deposition		Dry Deposition		Gas Exchange						Total Mass Flux	
					Gas Absorption		Volatilization		Net Gas Exchange			
	Mean ng/m <sup>2</sup> /d	COV %	Mean ng/m <sup>2</sup> /d	COV %	Mean ng/m <sup>2</sup> /d	COV %	Mean ng/m <sup>2</sup> /d	COV %	Mean ng/m <sup>2</sup> /d	COV %	Mean ng/m <sup>2</sup> /d	COV %
HCB	0.032	150	-	-	2.8	66	-3.1	50	-0.29	460	-0.26	510
PCB 18	0.037	130	-	-	0.51	100	-1.9	70	-1.4	130	-1.4	130
PCB 44	0.045	190	-	-	1.3	150	-1.7	86	-0.37	600	-0.33	670
PCB 52	0.064	150	-	-	1	110	-0.033	50	0.96	120	1	120
PCB 101	0.057	140	-	-	0.45	120	-0.56	100	-0.12	3200	-0.063	6100
suite-PCB	1.4	140	-	-	11	110	-28	52	-17	140	-16	150
PAHs and Metals												
	Wet Deposition		Dry Deposition		Gas Exchange						Total Mass Flux	
					Gas Absorption		Volatilization		Net Gas Exchange			
	Mean ng/m <sup>2</sup> /d	COV %	Mean ng/m <sup>2</sup> /d	COV %	Mean ng/m <sup>2</sup> /d	COV %	Mean ng/m <sup>2</sup> /d	COV %	Mean ng/m <sup>2</sup> /d	COV %	Mean ng/m <sup>2</sup> /d	COV %
PHEN	31	210	17	120	530	99	-140	50	390	130	440	120
PYR	23	210	19	130	57	110	-18	52	39	180	81	110
B[b+k]F	33	120	40	97	4.4	67	-0.44	51	3.9	70	77	72
B[a]P	13	110	11	130	1.6	68	-1	50	0.54	140	25	81
I[1,2,3-cd]P	16	120	19	130	1	67	-0.00013	52	1	68	36	87
sum-PAH (UN ECE)	62	78	70	110	7	46	-1.5	38	5.5	53	140	65
Pb	-	-	-	-	-	-	-	-	-	-	-	-
As	-	-	-	-	-	-	-	-	-	-	-	-
Se	-	-	-	-	-	-	-	-	-	-	-	-
Cd	-	-	-	-	-	-	-	-	-	-	-	-

**Table B8. Annual atmospheric fluxes to Lake Erie for 1998**

Banned Or- ganochlorine Pesti- cides	Lake Erie 1998											
	Wet Deposition		Dry Deposition		Gas Exchange						Total Mass Flux	
					Gas Absorption		Volatilization		Net Gas Exchange			
	Mean ng/m <sup>2</sup> /d	COV %	Mean ng/m <sup>2</sup> /d	COV %	Mean ng/m <sup>2</sup> /d	COV %	Mean ng/m <sup>2</sup> /d	COV %	Mean ng/m <sup>2</sup> /d	COV %	Mean ng/m <sup>2</sup> /d	COV %
$\alpha$ -HCH	0.16	130	0.021	250	17	110	-7.4	50	9.9	160	10	160
dieldrin	0.26	120	0.82	130	2.7	170	-11	51	-8.6	130	-7.5	150
<i>cis</i> -chlordane	0.15	130	0.39	150	0.9	120	-0.99	50	-0.089	1600	0.45	340
<i>trans</i> -chlordane	0.014	120	0.11	130	0.74	110	-1.5	50	-0.77	130	-0.65	160
<i>trans</i> -nonachlor	0.011	93	0.035	130	0.19	120	-0.54	62	-0.35	160	-0.3	190
<i>p,p'</i> -DDD	0.077	330	0.084	140	0.47	160	-	-	-	-	-	-
<i>p,p'</i> -DDE	0.04	160	-	-	1.2	120	-3.9	50	-2.7	100	-2.7	100
<i>p,p'</i> -DDT	0.04	150	0.068	180	1.2	160	-2.5	50	-1.3	260	-1.2	280
In-Use Pesticides												
	Wet Deposition		Dry Deposition		Gas Exchange						Total Mass Flux	
					Gas Absorption		Volatilization		Net Gas Exchange			
	Mean ng/m <sup>2</sup> /d	COV %	Mean ng/m <sup>2</sup> /d	COV %	Mean ng/m <sup>2</sup> /d	COV %	Mean ng/m <sup>2</sup> /d	COV %	Mean ng/m <sup>2</sup> /d	COV %	Mean ng/m <sup>2</sup> /d	COV %
$\gamma$ -HCH (lindane)	0.054	140	0.054	160	7.2	120	-3.1	50	4.1	190	4.2	190
$\alpha$ -endosulfan	0.065	180	2.1	200	27	180	-0.0045	50	27	180	29	170
endosulfan sulfate	-	-	0.14	170	-	-	-	-	-	-	-	-
Banned Or- ganochlorine Commercial Chemicals												
	Wet Deposition		Dry Deposition		Gas Exchange						Total Mass Flux	
					Gas Absorption		Volatilization		Net Gas Exchange			
	Mean ng/m <sup>2</sup> /d	COV %	Mean ng/m <sup>2</sup> /d	COV %	Mean ng/m <sup>2</sup> /d	COV %	Mean ng/m <sup>2</sup> /d	COV %	Mean ng/m <sup>2</sup> /d	COV %	Mean ng/m <sup>2</sup> /d	COV %
HCB	0.024	150	-	-	2.3	68	-2.4	50	-0.1	310	-0.076	410
PCB 18	0.011	130	-	-	0.43	110	-1.5	70	-1.1	150	-1.1	150
PCB 44	0.039	190	-	-	0.68	150	-1.4	86	-0.67	370	-0.63	390
PCB 52	0.023	150	-	-	0.84	110	-0.027	50	0.81	110	0.83	110
PCB 101	0.026	140	-	-	0.39	110	-0.47	100	-0.079	2300	-0.053	3400
suite-PCB	0.49	140	-	-	9.5	110	-22	52	-13	180	-13	180
PAHs and Metals												
	Wet Deposition		Dry Deposition		Gas Exchange						Total Mass Flux	
					Gas Absorption		Volatilization		Net Gas Exchange			
	Mean ng/m <sup>2</sup> /d	COV %	Mean ng/m <sup>2</sup> /d	COV %	Mean ng/m <sup>2</sup> /d	COV %	Mean ng/m <sup>2</sup> /d	COV %	Mean ng/m <sup>2</sup> /d	COV %	Mean ng/m <sup>2</sup> /d	COV %
PHEN	9.5	210	14	130	520	90	-130	50	390	120	410	110
PYR	5.3	210	18	130	54	120	-18	52	37	180	60	120
B[ <i>b+k</i> ]F	10	130	47	120	4.6	74	-0.48	51	4.1	78	61	95
B[ <i>a</i> ]P	2.9	110	10	130	1.4	75	-1.1	50	0.36	200	13	100
I[1,2,3- <i>cd</i> ]P	3.1	120	21	140	0.94	81	-0.00014	52	0.94	81	25	120
sum-PAH (UN ECE)	16	82	79	120	7	53	-1.6	38	5.4	63	100	96
Pb	-	-	-	-	-	-	-	-	-	-	-	-
As	-	-	-	-	-	-	-	-	-	-	-	-
Se	-	-	-	-	-	-	-	-	-	-	-	-
Cd	-	-	-	-	-	-	-	-	-	-	-	-

**Table B9. Annual atmospheric fluxes to Lake Ontario for 1997**

Banned Organochlorine Pesticides	Lake Ontario 1997											
	Wet Deposition		Dry Deposition		Gas Exchange						Total Mass Flux	
					Gas Absorption		Volatilization		Net Gas Exchange			
	Mean ng/m <sup>2</sup> /d	COV %	Mean ng/m <sup>2</sup> /d	COV %	Mean ng/m <sup>2</sup> /d	COV %	Mean ng/m <sup>2</sup> /d	COV %	Mean ng/m <sup>2</sup> /d	COV %	Mean ng/m <sup>2</sup> /d	COV %
α-HCH	3.6	55	-	-	10	68	-13	50	-2.6	430	1	1100
dieldrin	0.87	74	-	-	3.1	130	-19	50	-15	88	-14	94
cis-chlordane	0.12	64	-	-	0.71	110	-1	51	-0.31	740	-0.19	1200
trans-chlordane	0.16	72	-	-	0.6	110	-0.68	50	-0.074	2800	0.086	2400
trans-nonachlor	-	-	-	-	0.32	120	-1.3	55	-0.93	130	-	-
p,p'-DDD	0.33	53	-	-	0.13	110	-	-	-	-	-	-
p,p'-DDE	1.6	67	-	-	2	140	-5	52	-3	220	-1.4	480
p,p'-DDT	1.9	91	-	-	1.1	140	-0.25	52	0.83	170	2.7	83
<b>In-Use Pesticides</b>												
	Wet Deposition		Dry Deposition		Gas Exchange						Total Mass Flux	
					Gas Absorption		Volatilization		Net Gas Exchange			
	Mean ng/m <sup>2</sup> /d	COV %	Mean ng/m <sup>2</sup> /d	COV %	Mean ng/m <sup>2</sup> /d	COV %	Mean ng/m <sup>2</sup> /d	COV %	Mean ng/m <sup>2</sup> /d	COV %	Mean ng/m <sup>2</sup> /d	COV %
γ-HCH (lindane)	7.2	63	-	-	4.6	120	-3.6	50	0.99	360	8.2	70
α-endosulfan	2	220	-	-	26	200	-0.0095	81	26	200	28	190
endosulfan sulfate	-	-	-	-	-	-	-	-	-	-	-	-
<b>Banned Organochlorine Commercial Chemicals</b>												
	Wet Deposition		Dry Deposition		Gas Exchange						Total Mass Flux	
					Gas Absorption		Volatilization		Net Gas Exchange			
	Mean ng/m <sup>2</sup> /d	COV %	Mean ng/m <sup>2</sup> /d	COV %	Mean ng/m <sup>2</sup> /d	COV %	Mean ng/m <sup>2</sup> /d	COV %	Mean ng/m <sup>2</sup> /d	COV %	Mean ng/m <sup>2</sup> /d	COV %
HCB	0.13	130	-	-	3.3	64	-6.7	50	-3.4	93	-3.3	96
PCB 18	-	-	-	-	1.3	95	-2.6	64	-1.3	170	-1.3	170
PCB 44	-	-	-	-	0.49	100	-2	74	-1.5	130	-1.5	130
PCB 52	-	-	-	-	0.97	96	-1	210	-0.029	21000	-0.029	21000
PCB 101	-	-	-	-	0.37	100	-0.72	55	-0.34	240	-0.34	240
suite-PCB	-	-	-	-	7.2	95	-34	51	-27	91	-27	91
<b>PAHs and Metals</b>												
	Wet Deposition		Dry Deposition		Gas Exchange						Total Mass Flux	
					Gas Absorption		Volatilization		Net Gas Exchange			
	Mean ng/m <sup>2</sup> /d	COV %	Mean ng/m <sup>2</sup> /d	COV %	Mean ng/m <sup>2</sup> /d	COV %	Mean ng/m <sup>2</sup> /d	COV %	Mean ng/m <sup>2</sup> /d	COV %	Mean ng/m <sup>2</sup> /d	COV %
PHEN	57	140	7.3	120	150	82	-240	80	-91	260	-27	930
PYR	64	170	10	130	21	80	-38	88	-16	270	58	200
B[b+k]F	31	180	32	100	1.5	59	-0.98	100	0.51	190	64	100
B[a]P	12	230	8.6	130	0.32	59	-1.4	65	-1.1	120	20	150
I[1,2,3-cd]P	21	260	17	130	2	59	-	-	-	-	-	-
sum-PAH (UN ECE)	63	130	57	110	3.8	39	-	-	-	-	-	-
Pb	3000	210	730	140	-	-	-	-	-	-	3700	170
As	-	-	94	200	-	-	-	-	-	-	-	-
Se	-	-	87	140	-	-	-	-	-	-	-	-
Cd	150	110	24	110	-	-	-	-	-	-	170	98

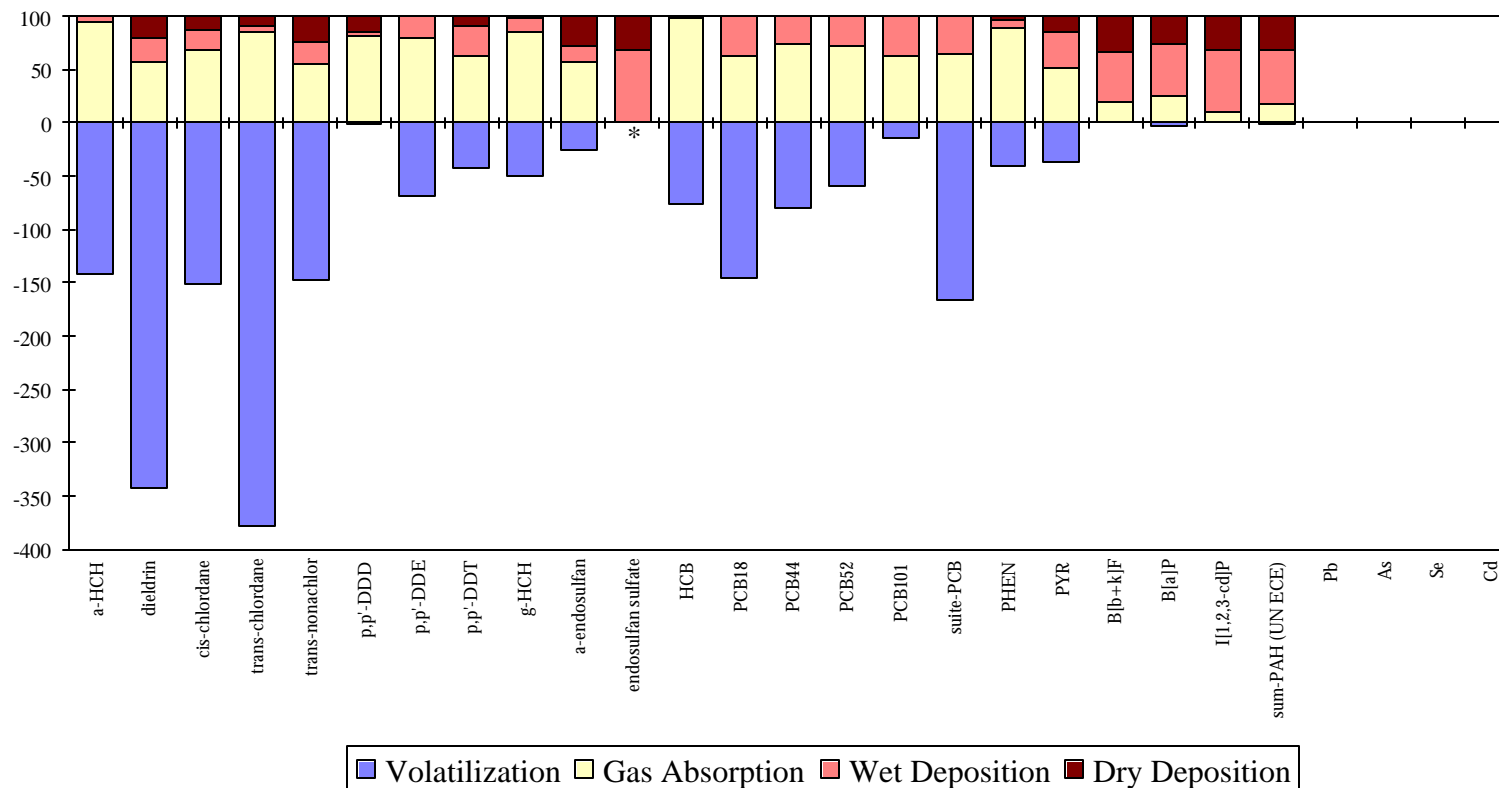
**Table B10. Annual atmospheric fluxes to Lake Ontario for 1998**

Banned Or- ganochlorine Pesti- cides	Lake Ontario 1998											
	Wet Deposition		Dry Deposition		Gas Exchange						Total Mass Flux	
					Gas Absorption		Volatilization		Net Gas Exchange			
	Mean ng/m <sup>2</sup> /d	COV %	Mean ng/m <sup>2</sup> /d	COV %	Mean ng/m <sup>2</sup> /d	COV %	Mean ng/m <sup>2</sup> /d	COV %	Mean ng/m <sup>2</sup> /d	COV %	Mean ng/m <sup>2</sup> /d	COV %
$\alpha$ -HCH	3.1	55	-	-	7.6	70	-14	50	-6.4	150	-3.3	300
dieldrin	0.65	74	-	-	2.3	120	-20	50	-17	80	-16	85
<i>cis</i> -chlordane	0.13	64	-	-	0.54	120	-1	51	-0.49	240	-0.36	330
<i>trans</i> -chlordane	0.12	72	-	-	0.38	110	-0.67	50	-0.29	240	-0.17	410
<i>trans</i> -nonachlor	0.045	-	-	-	0.21	110	-1.1	55	-0.93	100	-0.89	-
<i>p,p'</i> -DDD	0.29	53	-	-	0.095	110	-	-	-	-	-	-
<i>p,p'</i> -DDE	0.56	67	-	-	1.5	120	-5.1	52	-3.5	130	-2.9	160
<i>p,p'</i> -DDT	0.18	91	-	-	0.84	130	-0.28	52	0.56	170	0.74	130
In-Use Pesticides												
	Wet Deposition		Dry Deposition		Gas Exchange						Total Mass Flux	
					Gas Absorption		Volatilization		Net Gas Exchange			
	Mean ng/m <sup>2</sup> /d	COV %	Mean ng/m <sup>2</sup> /d	COV %	Mean ng/m <sup>2</sup> /d	COV %	Mean ng/m <sup>2</sup> /d	COV %	Mean ng/m <sup>2</sup> /d	COV %	Mean ng/m <sup>2</sup> /d	COV %
$\gamma$ -HCH (lindane)	4.8	63	-	-	4.3	130	-3.8	50	0.49	620	5.3	81
$\alpha$ -endosulfan	4	220	-	-	30	220	-0.0091	81	30	220	34	200
endosulfan sulfate	-	-	-	-	-	-	-	-	-	-	-	-
Banned Or- ganochlorine Commercial Chemicals												
	Wet Deposition		Dry Deposition		Gas Exchange						Total Mass Flux	
					Gas Absorption		Volatilization		Net Gas Exchange			
	Mean ng/m <sup>2</sup> /d	COV %	Mean ng/m <sup>2</sup> /d	COV %	Mean ng/m <sup>2</sup> /d	COV %	Mean ng/m <sup>2</sup> /d	COV %	Mean ng/m <sup>2</sup> /d	COV %	Mean ng/m <sup>2</sup> /d	COV %
HCB	0.08	130	-	-	2.6	69	-5.8	50	-3.2	90	-3.1	93
PCB 18	-	-	-	-	0.75	72	-2.2	64	-1.5	130	-1.5	130
PCB 44	-	-	-	-	0.3	91	-1.8	74	-1.5	120	-1.5	120
PCB 52	-	-	-	-	0.62	84	-0.92	210	-0.3	470	-0.3	470
PCB 101	-	-	-	-	0.26	92	-0.68	55	-0.41	140	-0.41	140
suite-PCB	-	-	-	-	4.6	81	-31	51	-26	86	-26	86
PAHs and Metals												
	Wet Deposition		Dry Deposition		Gas Exchange						Total Mass Flux	
					Gas Absorption		Volatilization		Net Gas Exchange			
	Mean ng/m <sup>2</sup> /d	COV %	Mean ng/m <sup>2</sup> /d	COV %	Mean ng/m <sup>2</sup> /d	COV %	Mean ng/m <sup>2</sup> /d	COV %	Mean ng/m <sup>2</sup> /d	COV %	Mean ng/m <sup>2</sup> /d	COV %
PHEN	49	140	7.5	160	180	91	-240	80	-59	330	-2.5	8300
PYR	31	170	15	180	39	130	-39	88	0.52	2500	47	130
B[ <i>b+k</i> ]F	37	190	46	220	7.1	230	-1.1	100	6	270	89	140
B[ <i>a</i> ]P	11	230	9.9	190	0.43	80	-1.6	65	-1.2	130	20	160
I[1,2,3- <i>cd</i> ]P	14	260	21	190	1.8	59	-	-	-	-	-	-
sum-PAH (UN ECE)	62	130	76	170	9.3	170	-	-	-	-	-	-
Pb	5400	210	840	110	-	-	-	-	-	-	6200	180
As	-	-	150	130	-	-	-	-	-	-	-	-
Se	-	-	67	160	-	-	-	-	-	-	-	-
Cd	230	110	24	140	-	-	-	-	-	-	250	100

## **Appendix C. Relative Loadings of IADN Substances, 1997-1998**

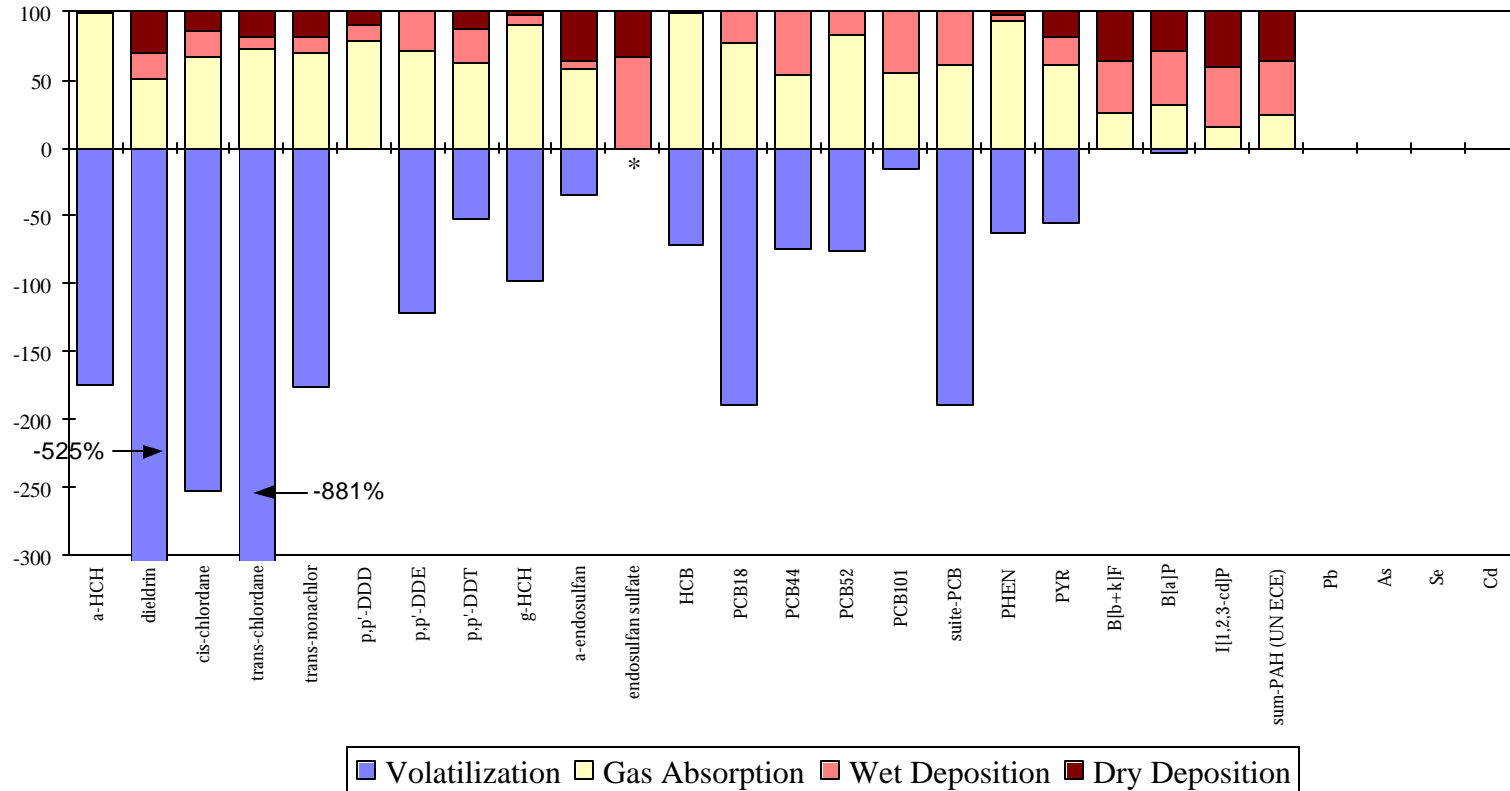


### Lake Superior 1997



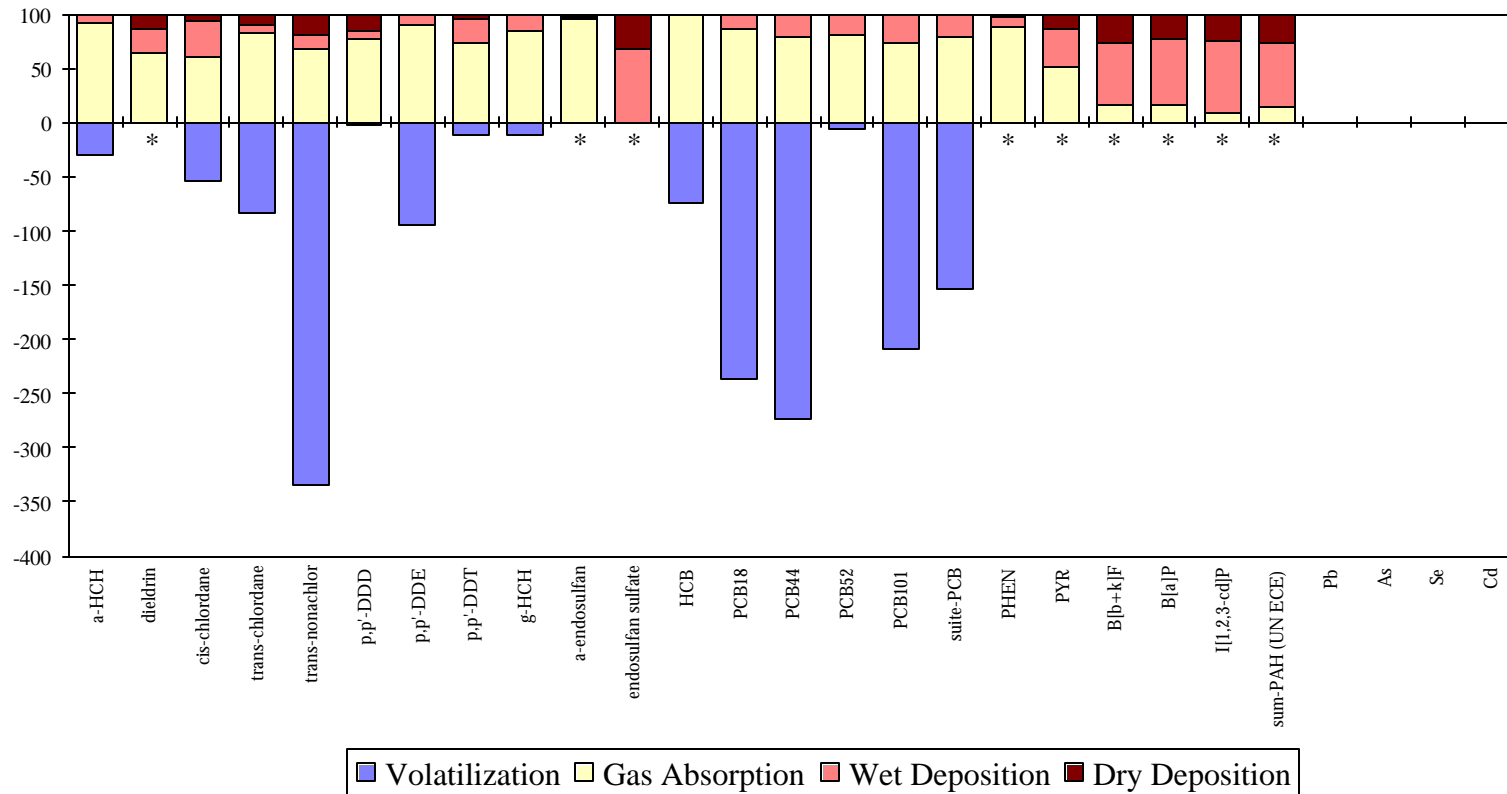
**Figure C1. Loadings as a proportion of total deposition to Lake Superior for 1997. \* indicates substances for which no volatilization estimates could be made because of lack of water concentration data. Refer to Table A3 for missing air concentration data.**

### Lake Superior 1998



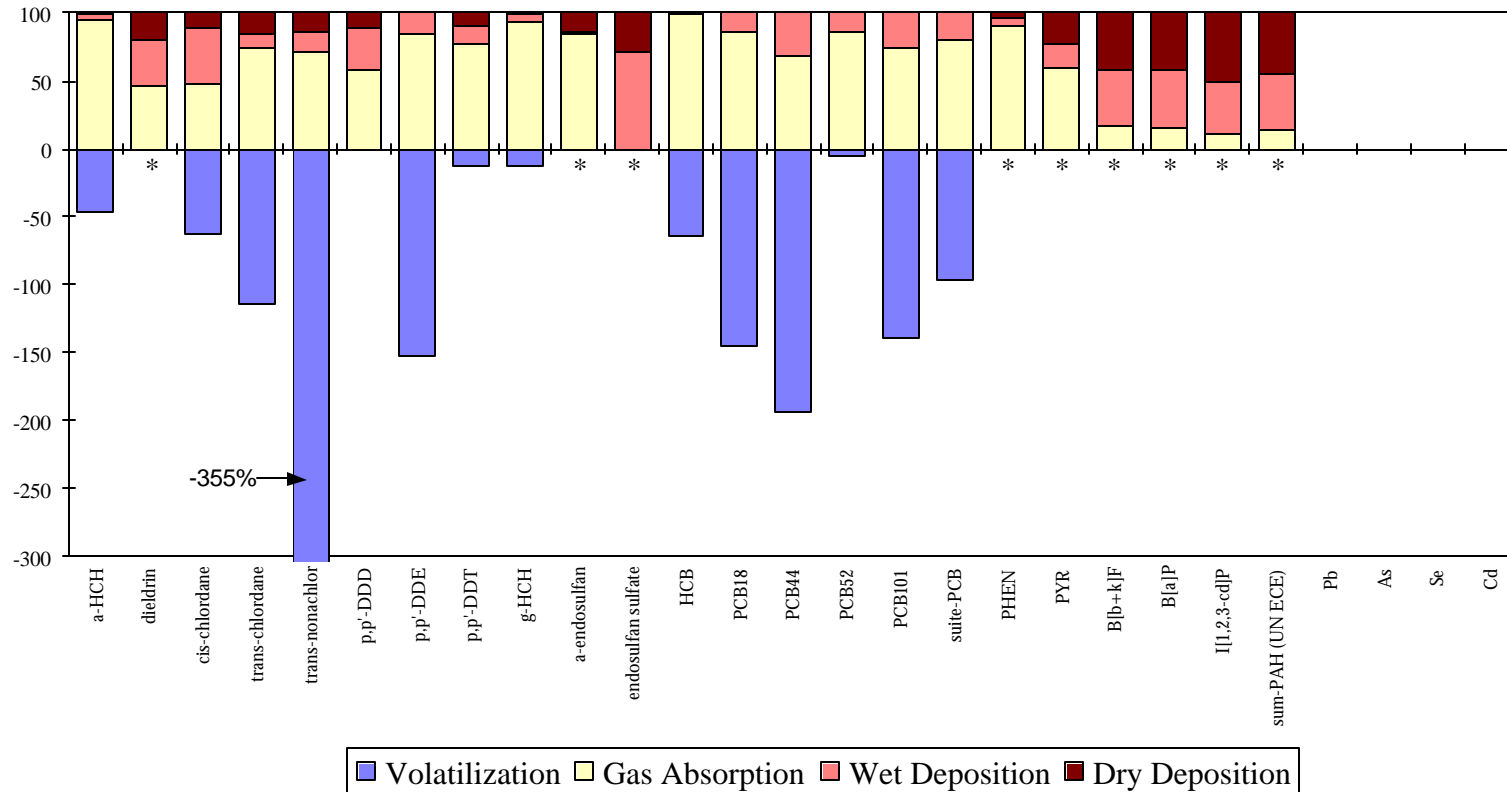
**Figure C2. Loadings as a proportion of total deposition to Lake Superior for 1998. \* indicates substances for which no volatilization estimates could be made because of lack of water concentration data. Refer to Table A3 for missing air concentration data.**

### Lake Michigan 1997



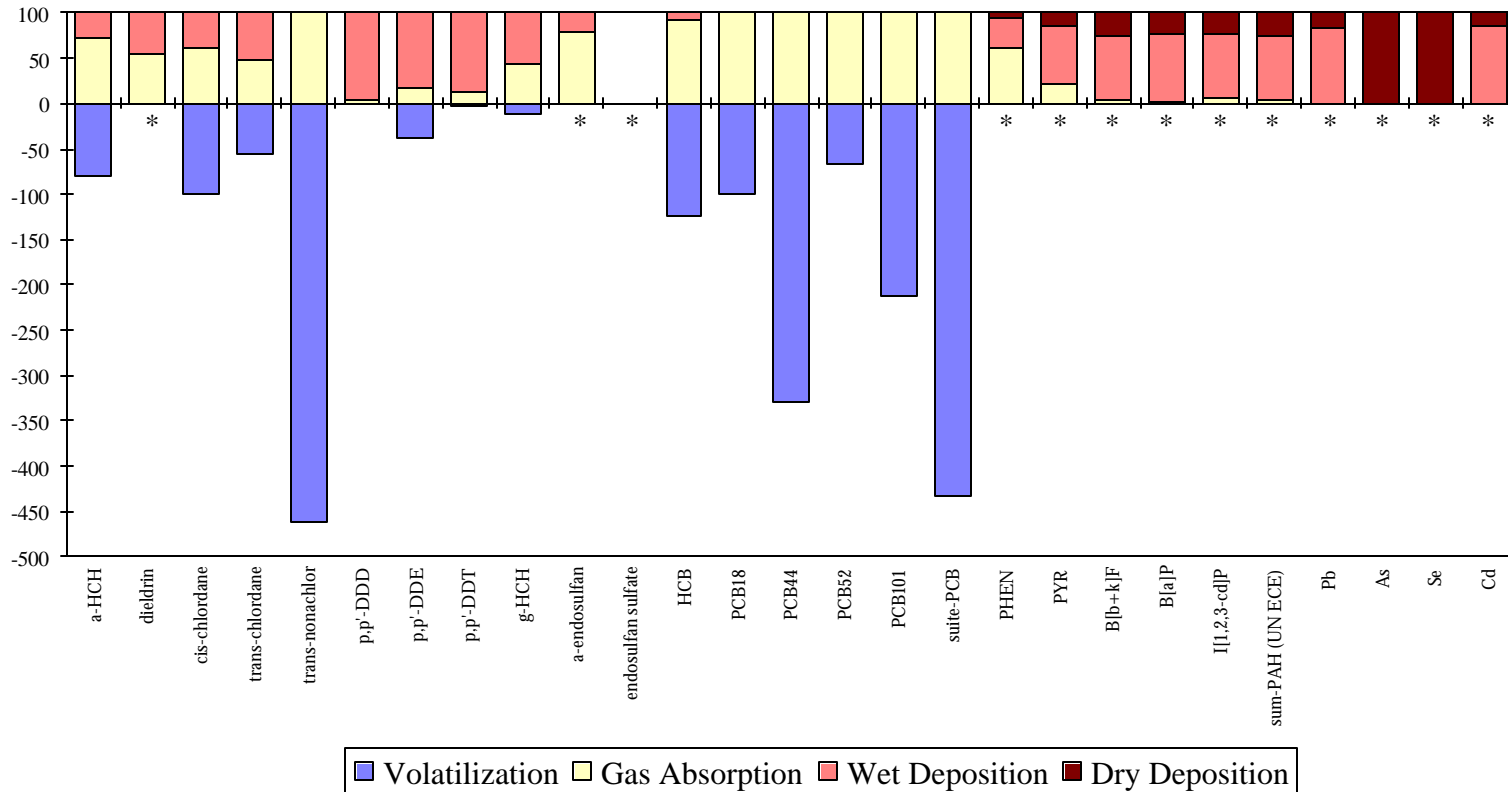
**Figure C3. Loadings as a proportion of total deposition to Lake Michigan for 1997. \* indicates substances for which no volatilization estimates could be made because of lack of water concentration data. Refer to Table A3 for missing air concentration data.**

### Lake Michigan 1998



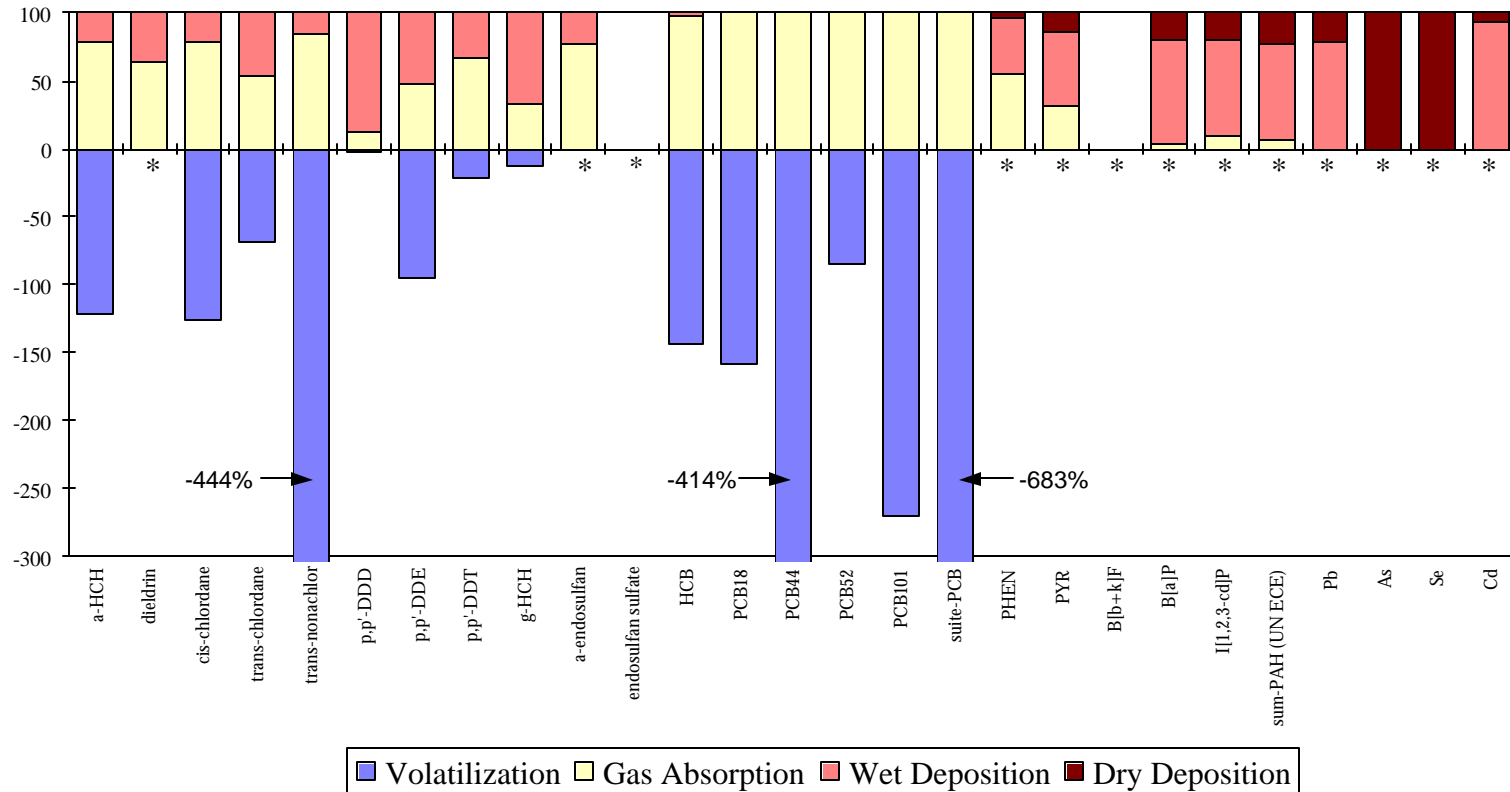
**Figure C4. Loadings as a proportion of total deposition to Lake Michigan for 1998. \* indicates substances for which no volatilization estimates could be made because of lack of water concentration data. Refer to Table A3 for missing air concentration data.**

### Lake Huron 1997



**Figure C5. Loadings as a proportion of total deposition to Lake Huron for 1997. \* indicates substances for which no volatilization estimates could be made because of lack of water concentration data. Refer to Table A3 for missing air concentration data.**

### Lake Huron 1998



**Figure C6. Loadings as a proportion of total deposition to Lake Huron for 1998. \* indicates substances for which no volatilization estimates could be made because of lack of water concentration data. Refer to Table A3 for missing air concentration data.**

Lake Erie 1997

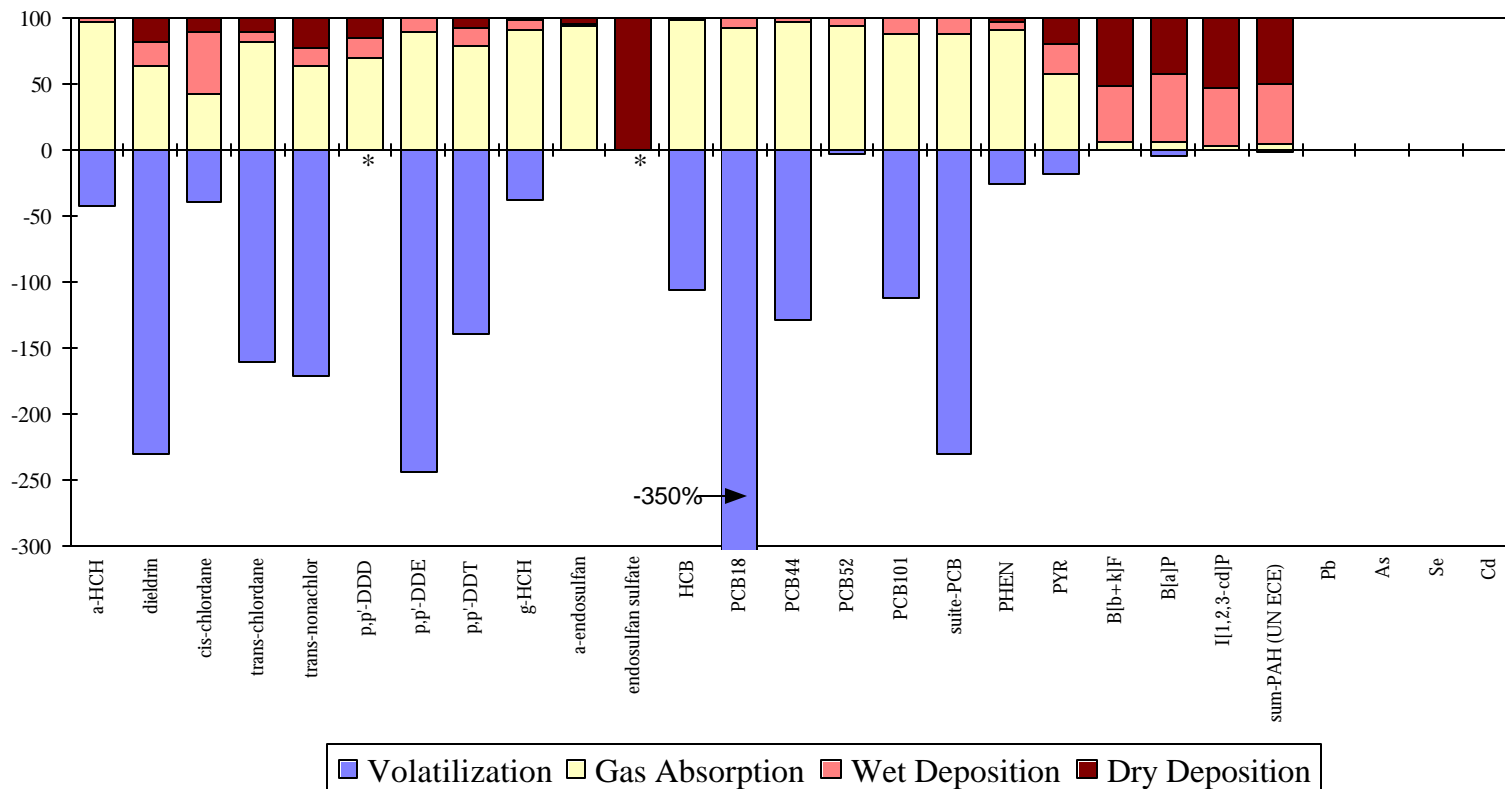
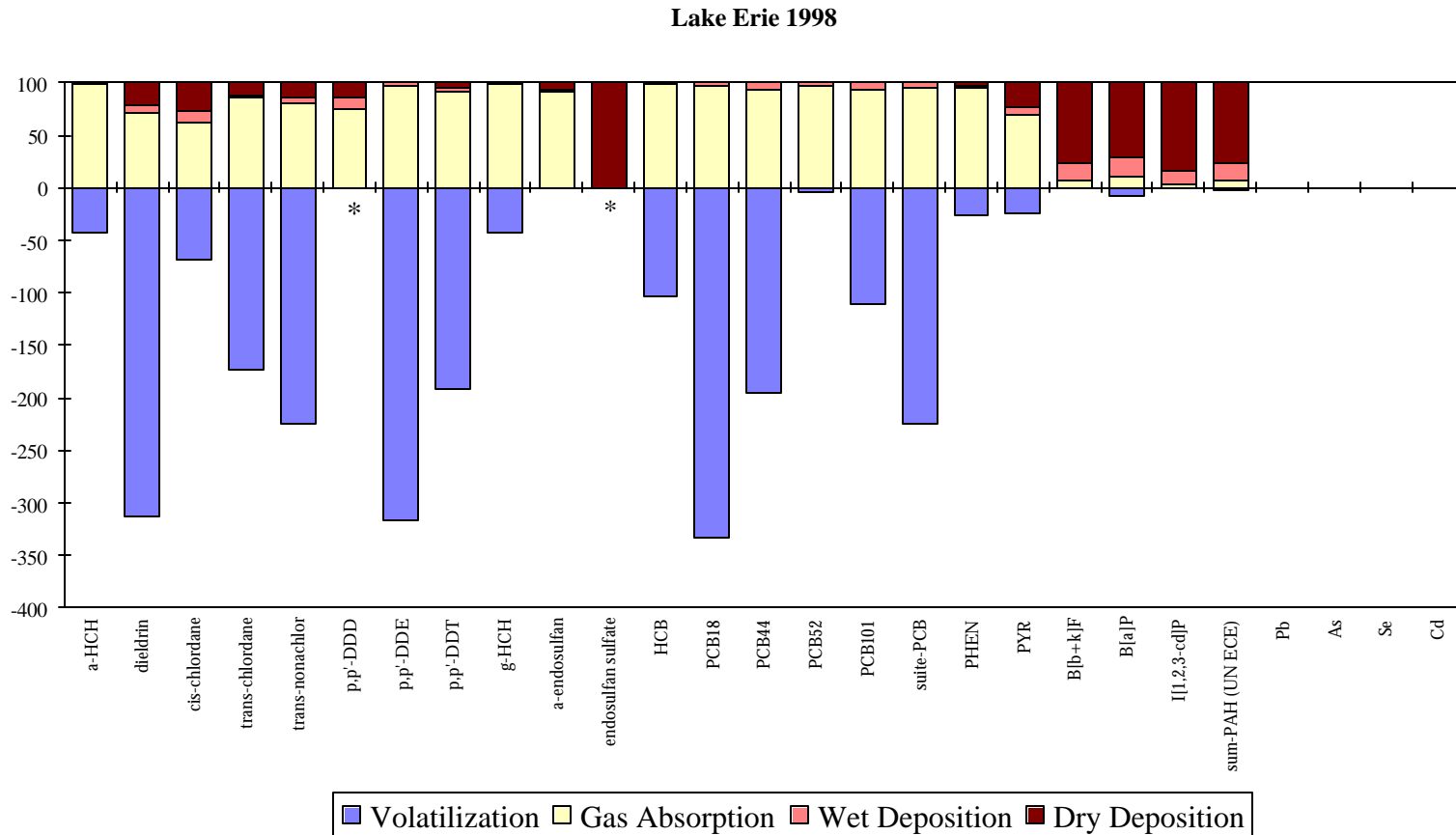


Figure C7. Loadings as a proportion of total deposition to Lake Erie for 1997. \* indicates substances for which no volatilization estimates could be made because of lack of water concentration data. Refer to Table A3 for missing air concentration data.



**Figure C8. Loadings as a proportion of total deposition to Lake Erie for 1998. \* indicates substances for which no volatilization estimates could be made because of lack of water concentration data. Refer to Table A3 for missing air concentration data.**



Lake Ontario 1997

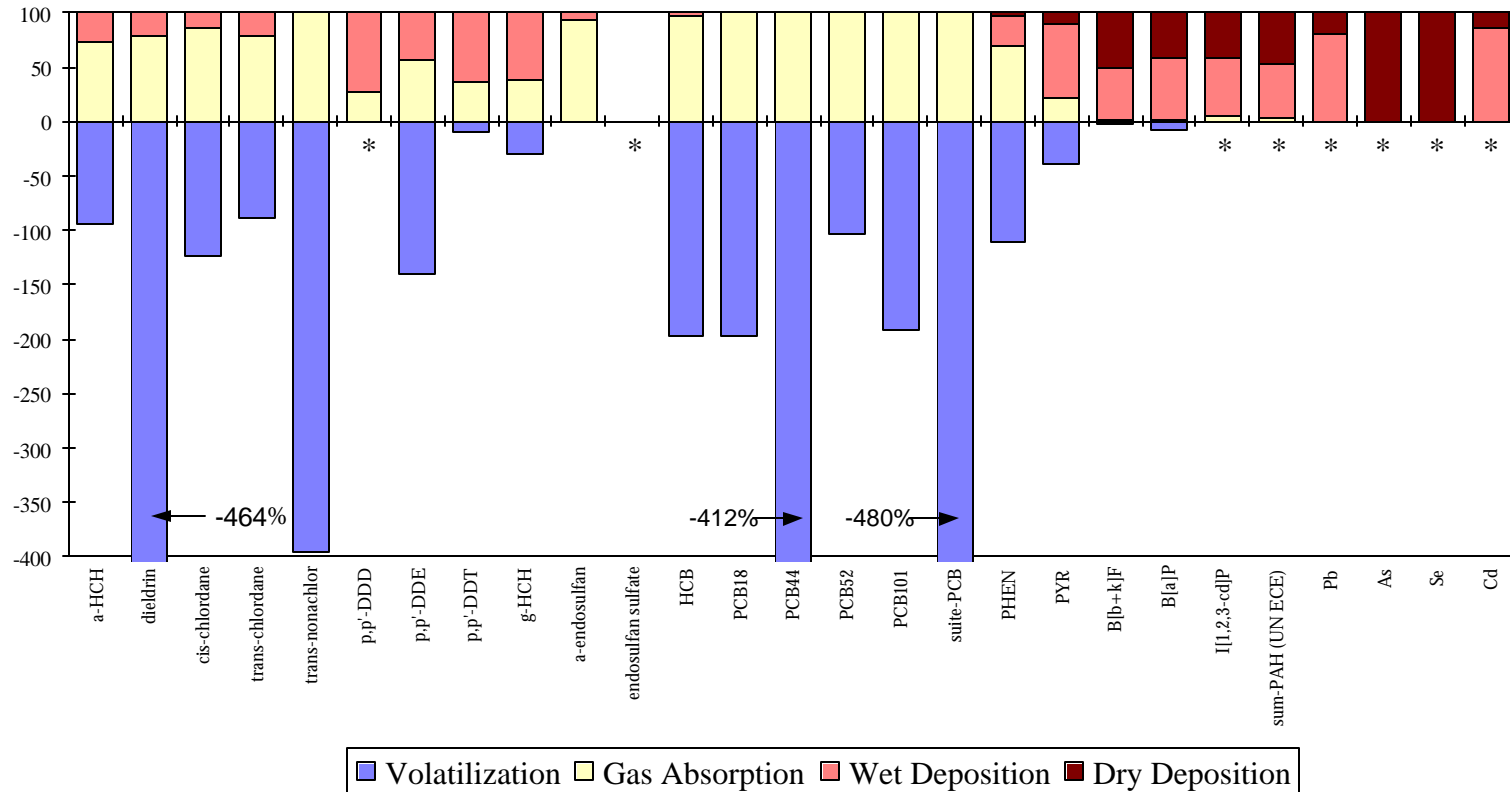
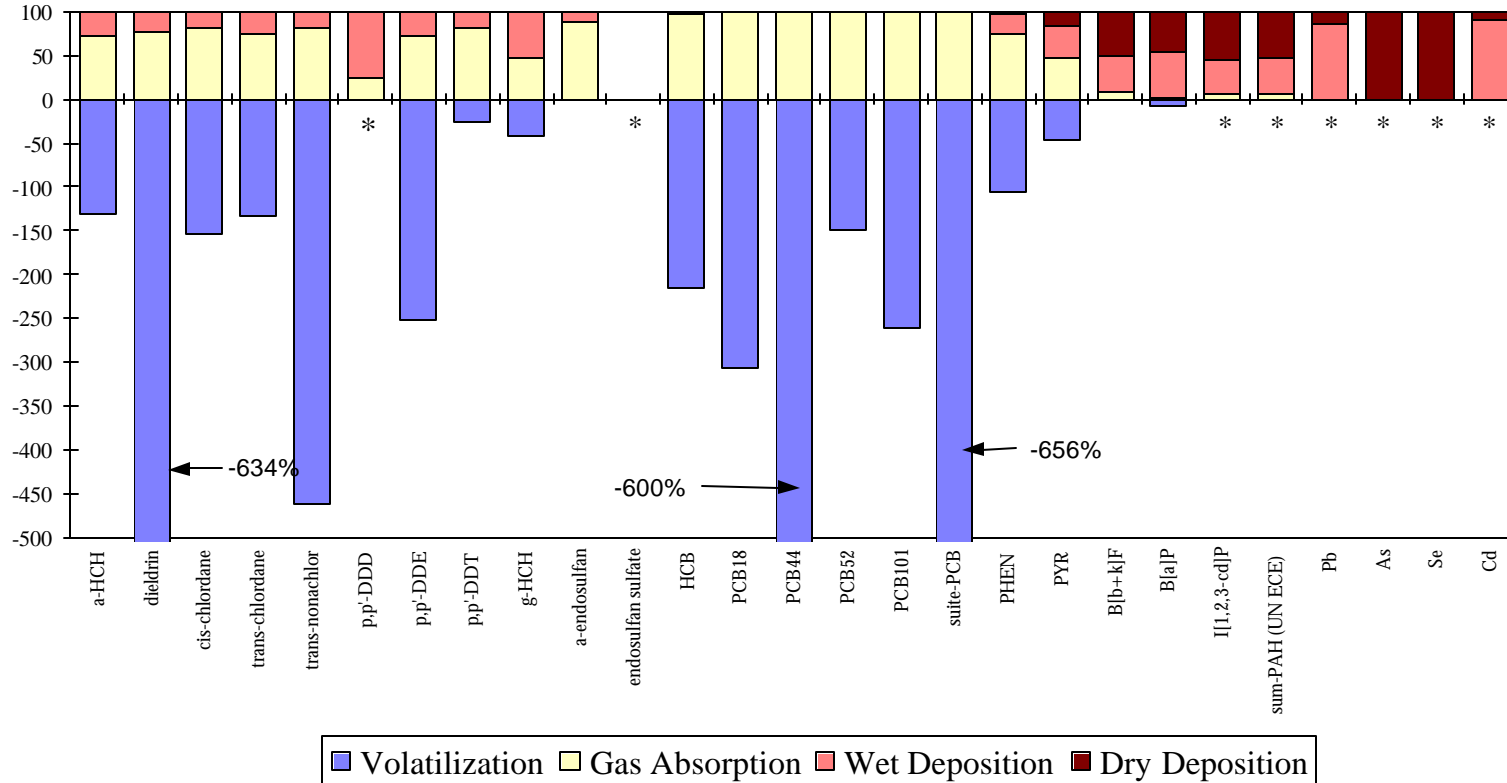


Figure C9. Loadings as a proportion of total deposition to Lake Ontario for 1997. \* indicates substances for which no volatilization estimates could be made because of lack of water concentration data. Refer to Table A3 for missing air concentration data.

### Lake Ontario 1998



**Figure C10. Loadings as a proportion of total deposition to Lake Ontario for 1998. \* indicates substances for which no volatilization estimates could be made because of lack of water concentration data. Refer to Table A3 for missing air concentration data.**

## **Appendix D. Annual IADN Flows (kg/yr), 1992-1998**

**Table D1. IADN annual flows (kg/yr) from 1992 to 1998 for Lake Superior**

		Wet Deposition	Dry Deposition	Net Gas Exchange	Total Deposition			Wet Deposition	Dry Deposition	Net Gas Exchange	Total Deposition	
α-HCH	1992	78	1.1	390	470	PCB101	1992	1.7	0.45	-42	-40	
	1993	33	5.6	450	490		1993	1.6	0.74	-39	-37	
	1994	38	12	710	760		1994	1.6	2.3	-25	-21	
	1995	28	4.7	-230	-200		1995	3.1	1.4	-0.94	3.6	
	1996	71	-	-240	-170		1996	2.6	-	22	25	
	1997	35	2.9	-290	-250		1997	3	-	3.9	6.9	
	1998	6.5	1.9	-470	-460		1998	3.5	-	3.2	6.7	
γ-HCH	1992	62	0.65	140	200	ΣPCB	1992	58	27	-1300	-1200	
	1993	14	2.7	47	64		1993	110	25	-1200	-1100	
	1994	19	2.4	95	120		1994	63	86	-1000	-850	
	1995	9.5	1.9	65	76		1995	70	47	-300	-180	
	1996	38	-	43	81		1996	90	-	-380	-290	
	1997	27	4.2	69	100		1997	75	-	-210	-140	
	1998	10	2.2	-4.6	7.6		1998	70	-	-230	-160	
dieldrin	1992	21	7.4	-500	-470	suite-PCB	1992	260	100	-5500	-5100	
	1993	62	6.3	-540	-470		phenan-threne	1992	180	60	-6800	-6600
	1994	11	25	-500	-460			1993	130	310	-6800	-6400
	1995	34	15	-240	-190			1994	500	59	5700	6300
	1996	20	-	-200	-180			1995	190	100	2200	2500
	1997	18	15	-220	-190			1996	270	96	1600	2000
	1998	11	18	-280	-250			1997	110	75	830	1000
p,p'-DDD	1992	-	6.1	-	-	pyrene		1992	160	120	980	1300
	1993	17	0.1	-	-		1993	140	74	67	280	
	1994	10	-	-	-		1994	210	220	-58	370	
	1995	0.89	0.5	-9.3	-7.9		1995	460	54	2600	3100	
	1996	0.8	-	4.3	5.1		1996	120	91	210	420	
	1997	0.32	1.1	6	7.4		1997	170	76	61	310	
	1998	0.74	0.64	5	6.4		1998	83	71	28	180	
p,p'-DDE	1992	2.6	0.39	-	-	B(k)F	1992	120	52	140	310	
	1993	3.8	1.2	-	-		1993	130	13	20	160	
	1994	4	-	-	-		1994	92	58	70	220	
	1995	4.6	0.96	-18	-12		1995	640	159	190	990	
	1996	2.1	-	-14	-12		1996	219	152	98	470	
	1997	2.5	-	1.9	4.4		1997	229	170	99	500	
	1998	2.3	-	-4	-1.7		1998	149	143	100	390	
p,p'-DDT	1992	6.2	1.6	21	29	B(b+k)F	1992	140	58	22	220	
	1993	59	1.9	12	73		B(a)P	1992	160	14	21	200
	1994	48	-	-	-			1993	92	39	35	170
	1995	1.7	2.4	2	6.1			1994	170	17	76	260
	1996	3.2	-	7.8	11			1995	49	35	34	120
	1997	3.9	1.4	2.6	7.9			1996	72	39	35	150
	1998	3.8	1.8	1.5	7.1			1997	49	36	33	120
HCB	1992	5.3	0.67	47	53	Pb		1992	-	-	-	-
	1993	25	19	15	59		1993	-	16000	-	-	
	1994	1.2	0.37	16	18		1994	-	26000	-	-	
	1995	1.5	0.42	24	26		1995	-	-	-	-	
	1996	1.2	-	22	23		1996	-	-	-	-	
	1997	1	-	18	19		1997	-	-	-	-	
	1998	0.6	-	25	26		1998	-	-	-	-	
PCB18	1992	0.92	0.26	-71	-70	As	1992	-	-	-	-	
	1993	3.1	0.39	-74	-71		1993	-	5600	-	-	
	1994	1.6	2.9	-71	-67		1994	-	2900	-67	-	
	1995	2	1.9	-14	-10		1995	-	-	-	-	
	1996	3.4	-	14	17		1996	-	-	-	-	
	1997	2.3	-	-5	-2.7		1997	-	-	-	-	
	1998	1.1	-	-5.5	-4.4		1998	-	-	-	-	
PCB44	1992	1.4	0.45	-19	-17	Se	1992	-	-	-	-	
	1993	9.1	0.92	-14	-4		1993	-	1800	-	-	
	1994	1.7	2.8	-7.3	-2.8		1994	-	3100	-	-	
	1995	2.2	1.7	-8.3	-4.4		1995	-	-	-	-	
	1996	1.9	-	51	53		1996	-	-	-	-	
	1997	2.7	-	-0.57	2.1		1997	-	-	-	-	
	1998	5.3	-	-2.3	3		1998	-	-	-	-	
PCB52	1992	1.1	0.31	-13	-12	Cd	1992	-	-	-	-	
	1993	1.8	0.56	-8	-5.6		1993	-	2100	-	-	
	1994	2.1	3.1	5	10		1994	-	4400	-	-	
	1995	2.6	2.4	-9.9	-4.9		1995	-	-	-	-	
	1996	2.6	-	34	37		1996	-	-	-	-	
	1997	3.1	-	1.5	4.6		1997	-	-	-	-	
	1998	1.6	-	0.54	2.1		1998	-	-	-	-	

**Table D2. IADN annual flows (kg/yr) from 1992 to 1998 for Lake Michigan**

		Wet Deposition	Dry Deposition	Net Gas Exchange	Total Deposition			Wet Deposition	Dry Deposition	Net Gas Exchange	Total Deposition	
α-HCH	1992	62	1.5	52	120	PCB101	1992	1.4	0.33	-26	-24	
	1993	44	4	81	130		1993	0.81	0.66	-23	-22	
	1994	98	6.3	120	220		1994	1.7	1.1	-29	-26	
	1995	56	4.5	350	410		1995	2.1	1.6	-9.3	-5.6	
	1996	15	-	300	320		1996	1.4	-	-9.1	-7.7	
	1997	40	3.4	300	340		1997	1.1	-	-3.79	-2.7	
	1998	17	0.91	200	220		1998	1.7	-	-4.3	-2.6	
γ-HCH	1992	65	1.1	870	940	ΣPCB	1992	52	16	-1300	-1200	
	1993	120	2	250	370		1993	86	24	-1200	-1100	
	1994	47	1.4	490	540		1994	71	39	-1400	-1300	
	1995	26	2.2	190	220		1995	78	41	-330	-210	
	1996	6.6	-	110	120		1996	48	-	-320	-270	
	1997	31	2.3	150	180		suite -PCB	1997	25	-	-51.4	-26
	1998	13	1.9	190	200		suite-PCB	1998	38	-	-34	4
dieldrin	1992	58	8	-	-	phenan- threne	1992	350	110	-	-	
	1993	55	7.2	-	-		1993	230	100	-	-	
	1994	62	23	-	-		1994	160	160	-	-	
	1995	47	20	-	-		1995	360	82	-	-	
	1996	30	-	-	-		1996	220	100	-	-	
	1997	24	15	-	-		1997	320	97	-	-	
	1998	31	18	-	-		1998	180	120	-	-	
p,p'-DDD	1992	-	3.8	-	-	pyrene	1992	220	140	-	-	
	1993	6.4	0.98	-	-		1993	220	140	-	-	
	1994	16	-	-	-		1994	130	170	-	-	
	1995	1.6	1.9	-	-		1995	340	95	-	-	
	1996	1.8	-	-	-		1996	140	110	-	-	
	1997	0.36	0.76	3.5	4.6		1997	220	99	-	-	
	1998	2.9	0.99	5.3	9.2		1998	130	150	-	-	
p,p'-DDE	1992	3.8	0.48	-	-	B(k)F B(k)F B(k)F B(b+k)F B(b+k)F B(b+k)F B(b+k)F	1992	130	56	-	-	
	1993	11	1.4	-	-		1993	110	43	-	-	
	1994	3.5	-	-	-		1994	73	63	-	-	
	1995	7.4	1.4	-	-		1995	480	198	-	-	
	1996	3.9	-	-	-		1996	258	197	-	-	
	1997	2.5	-	-1.5	1		1997	338	183	-	-	
	1998	2.7	-	-12	-9.3		1998	278	288	-	-	
p,p'-DDT	1992	22	2.3	44	68	B(a)P	1992	170	77	-	-	
	1993	56	6.2	35	97		1993	170	42	-	-	
	1994	58	-	-	-		1994	77	63	-	-	
	1995	9.7	0.67	-	-		1995	160	37	-	-	
	1996	9	-	-	-		1996	84	41	-	-	
	1997	4.3	1.1	13	18		1997	120	48	-	-	
	1998	3	2	14	19		1998	89	86	-	-	
HCB	1992	2.6	0.38	24	27	Pb	1992	-	-	-	-	
	1993	13	13	1.2	27		1993	-	16000	-	-	
	1994	1.1	0.19	-10	-8.7		1994	-	-	-	-	
	1995	1.4	0.35	8.3	10		1995	-	-	-	-	
	1996	0.91	-	8.3	9.2		1996	-	-	-	-	
	1997	0.75	-	16	17		1997	-	-	-	-	
	1998	0.83	-	26	27		1998	-	-	-	-	
PCB18	1992	0.91	0.18	-69	-68	As	1992	-	-	-	-	
	1993	1.3	0.46	-74	-72		1993	-	820	-	-	
	1994	1.6	1.2	-75	-72		1994	-	1300	-	-	
	1995	2.6	1.1	-24	-20		1995	-	-	-	-	
	1996	1.7	-	-24	-22		1996	-	-	-	-	
	1997	0.82	-	-6.2	-5.4		1997	-	-	-	-	
	1998	1.4	-	-5.5	-4.1		1998	-	-	-	-	
PCB44	1992	1.4	0.32	-44	-42	Se	1992	-	-	-	-	
	1993	1.2	0.67	-42	-40		1993	-	910	-	-	
	1994	9.1	1.4	-52	-42		1994	-	-	-	-	
	1995	1.8	1.1	-20	-17		1995	-	-	-	-	
	1996	0.97	-	-22	-21		1996	-	-	-	-	
	1997	1.3	-	-8.2	-6.9		1997	-	-	-	-	
	1998	2.8	-	-11	-8.2		1998	-	-	-	-	
PCB52	1992	1.2	0.24	-56	-55	Cd	1992	-	-	-	-	
	1993	1.1	0.55	-55	-53		1993	-	4500	-	-	
	1994	2.3	1.4	-60	-56		1994	-	-	-	-	
	1995	3	2	3	8		1995	-	-	-	-	
	1996	2.2	-	2.2	4.4		1996	-	-	-	-	
	1997	1.7	-	7.1	8.8		1997	-	-	-	-	
	1998	1.9	-	11	13		1998	-	-	-	-	

**Table D3. IADN annual flows (kg/yr) from 1992 to 1998 for Lake Huron**

		Wet Deposition	Dry Deposition	Net Gas Exchange	Total Deposition			Wet Deposition	Dry Deposition	Net Gas Exchange	Total Deposition	
α-HCH	1992	170	-	-	-	PCB101	1992	11	-	-	-	
	1993	140	-	-500	-360		1993	6	-	0.77	-	
	1994	150	-	-490	-340		1994	1.9	-	0.74	-	
	1995	220	-	-50	170		1995	-	-	-7.7	-	
	1996	160	-	-63	97		1996	-	-	-6.1	-	
	1997	72	-	-15	57		1997	-	-	-2.7	-	
	1998	40	-	-82	-42		1998	-	-	-3.4	-	
γ-HCH	1992	-	-	-	-	ΣPCB	1992	180	-	-	-	
	1993	260	-	-34	230		1993	130	-	-490	-360	
	1994	120	-	-19	100		1994	110	-	-460	-350	
	1995	110	-	32	140		1995	-	-	-240	-	
	1996	93	-	29	120		1996	-	-	-230	-	
	1997	100	-	56	160		suite-PCB	1997	-	-	-23	-
	1998	130	-	37	170		suite-PCB	1998	-	-	-24	-
dieldrin	1992	-	-	-	-	phenanthrene	1992	-	-	-	-	
	1993	13	-	-760	-750		1993	640	90	-	-	
	1994	15	-	-720	-710		1994	320	71	-	-	
	1995	19	-	-	-		1995	250	63	-	-	
	1996	41	-	-	-		1996	390	110	-	-	
	1997	26	-	-	-		1997	760	140	-	-	
	1998	17	-	-	-		1998	730	59	-	-	
p,p'-DDD	1992	-	-	-	-	pyrene	1992	-	-	-	-	
	1993	-	-	-	-		1993	350	130	-	-	
	1994	1.8	-	-	-		1994	190	77	-	-	
	1995	3.8	-	-	-		1995	220	61	-	-	
	1996	6.7	-	-	-		1996	350	130	-	-	
	1997	27	-	0.91	28		1997	680	160	-	-	
	1998	6.1	-	0.79	6.9		1998	380	94	-	-	
p,p'-DDE	1992	10	-	-	-	B(k)F	1992	-	-	-	-	
	1993	-	-	-	-		B(k)F	1993	-	83	-	-
	1994	3.4	-	-	-		B(k)F	1994	-	48	-	-
	1995	9.6	-	-	-		B(b+k)F	1995	610	-	-	-
	1996	7.8	-	-	-		B(b+k)F	1996	-	257	-	-
	1997	21	-	-5	16		B(b+k)F	1997	600	215	-	-
	1998	6	-	-5	1		B(b+k)F	1998	420	158	-	-
p,p'-DDT	1992	22	-	-	-	B(a)P	1992	-	-	-	-	
	1993	-	-	2.5	-		1993	-	110	-	-	
	1994	4.1	-	2.5	6.6		1994	-	56	-	-	
	1995	10	-	-	-		1995	350	-	-	-	
	1996	18	-	-	-		1996	-	100	-	-	
	1997	45	-	5.7	51		1997	280	86	-	-	
	1998	2.8	-	4	6.8		1998	210	56	-	-	
HCB	1992	5.8	-	-35	-29	Pb	1992	100000	11000	-	110000	
	1993	11	-	-14	-3		1993	64000	8000	-	72000	
	1994	3.3	-	-18	-15		1994	47000	11000	-	58000	
	1995	3.6	-	-35	-31		1995	15000	7600	-	23000	
	1996	1.3	-	-28	-27		1996	18000	13000	-	31000	
	1997	3.6	-	-16	-12		1997	61000	12000	-12	73000	
	1998	0.92	-	-17	-16		1998	28000	7300	-	35000	
PCB18	1992	17	-	-	-	As	1992	11000	2200	-	13000	
	1993	4	-	-28	-24		1993	7500	1700	-24	9200	
	1994	0.94	-	-29	-28		1994	6500	1200	-	7700	
	1995	-	-	-14	-		1995	2200	710	-	2900	
	1996	-	-	-15	-		1996	2700	2900	-	5600	
	1997	-	-	-0.18	-		1997	-	1500	-	-	
	1998	-	-	-4.9	-		1998	-	1200	-	-	
PCB44	1992	20	-	-	-	Se	1992	17000	2700	-	20000	
	1993	5.2	-	-10	-4.8		1993	12000	2400	-	14000	
	1994	2.5	-	-10	-7.5		1994	10000	2600	-	13000	
	1995	-	-	-17	-		1995	2700	110	-	2800	
	1996	-	-	-15	-		1996	3100	1100	-	4200	
	1997	-	-	-6.3	-		1997	-	590	-	-	
	1998	-	-	-6.6	-		1998	-	570	-	-	
PCB52	1992	7.6	-	-	-	Cd	1992	6600	470	-	7100	
	1993	11	-	-6.8	4.2		1993	2900	310	-	3200	
	1994	2.6	-	-6.7	-4.1		1994	2300	410	-	2700	
	1995	-	-	-7.3	-		1995	1400	170	-	1600	
	1996	-	-	-5.5	-		1996	2000	310	-	2300	
	1997	-	-	2	-		1997	2900	490	-	3400	
	1998	-	-	0.72	-		1998	4100	280	-	4400	

**Table D4. IADN annual flows (kg/yr) from 1992 to 1998 for Lake Erie**

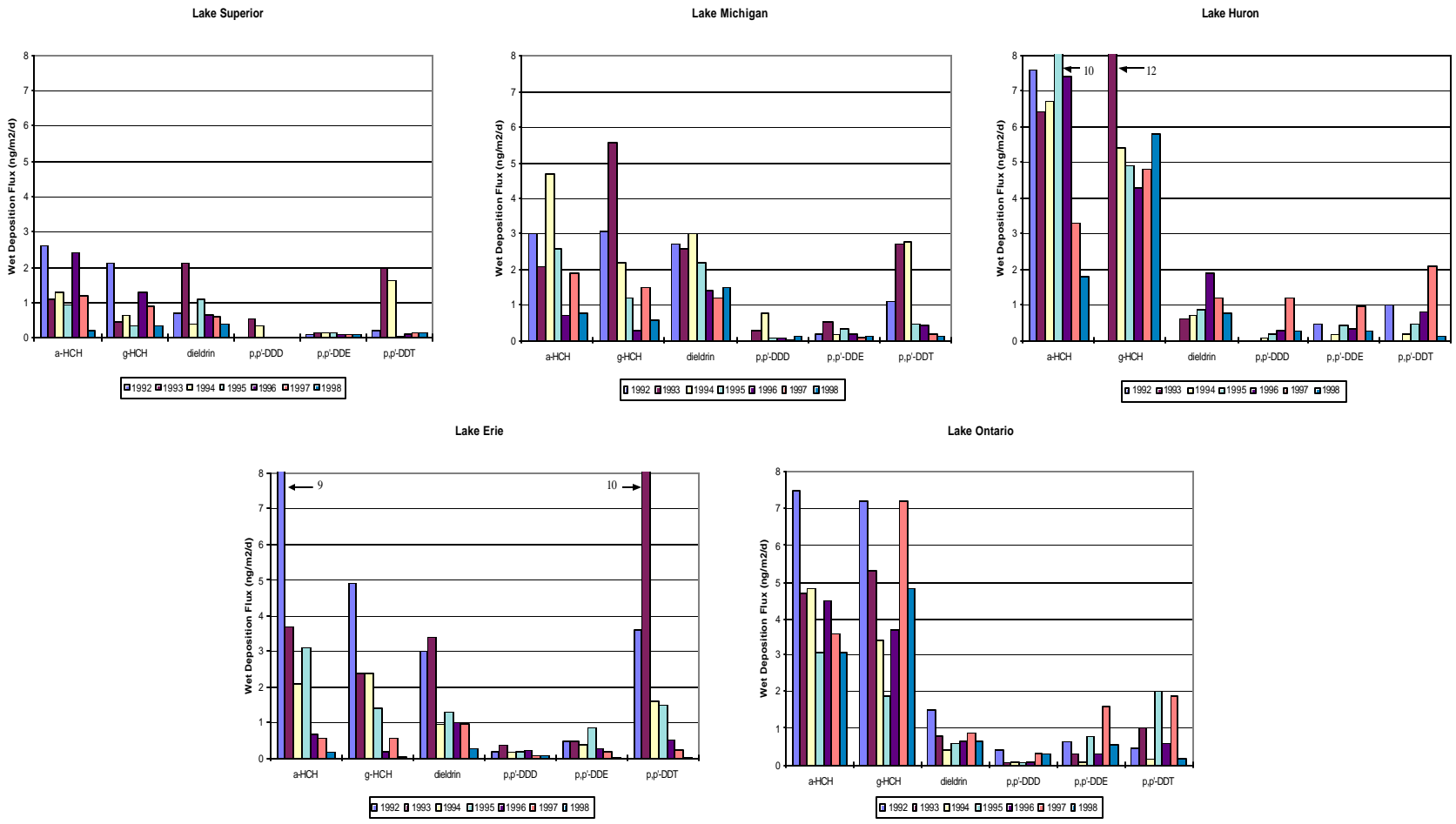
		Wet Deposition	Dry Deposition	Net Gas Exchange	Total Deposition			Wet Deposition	Dry Deposition	Net Gas Exchange	Total Deposition	
α-HCH	1992	84	1.2	140	230	PCB101	1992	0.48	0.28	-2.2	-1.4	
	1993	35	2	290	330		1993	0.81	0.37	1	2.2	
	1994	19	3.4	300	320		1994	1.1	0.73	-2.6	-0.77	
	1995	29	0.9	120	150		1995	1.4	1.4	-5.2	-2.4	
	1996	6.3	-	44	50		1996	0.51	-	-6.6	-6.1	
	1997	5.4	0.25	81	87		1997	0.54	-	-1.1	-0.56	
γ-HCH	1992	46	0.45	60	110	1998	0.25	-	-0.74	-0.49		
	1993	23	1.3	83	110	ΣPCB	1992	21	16	-200	-160	
	1994	22	0.83	68	91		1993	26	14	-100	-60	
	1995	13	0.6	45	59		1994	41	29	-200	-130	
	1996	1.7	-	47	49		1995	58	32	-220	-130	
	1997	5.3	1.3	41	48		1996	18	-	-310	-290	
1998	0.51	0.51	38	39	suite-PCB		1997	13	-	-160	-150	
dieldrin	1992	28	5.6	-300	-270	suite-PCB	1998	4.6	-	-120	-120	
	1993	32	3.7	-120	-84	phenanthrene	1992	500	86	-	-	
	1994	8.9	18	-110	-83		1993	360	100	-	-	
	1995	12	11	-110	-87		1994	210	190	-	-	
	1996	9.4	-	-110	-100		1995	530	160	1600	2300	
	1997	9.2	8.4	-82	-64		1996	91	190	-770	-490	
1998	2.4	7.7	-80	-70	1997		290	160	3600	4100		
p,p'-DDD	1992	1.9	2	-	-	1998	89	140	3600	3800		
	1993	3.4	0.21	-	-	pyrene	1992	330	110	-	-	
	1994	1.4	-	-	-		1993	310	130	-	-	
	1995	1.7	1.2	-	-		1994	160	250	-	-	
	1996	2	-	-	-		1995	360	210	290	860	
	1997	0.75	0.72	-	-		1996	58	260	-80	240	
1998	0.72	0.79	-	-	1997		210	180	370	760		
p,p'-DDE	1992	4.6	0.53	-	-	1998	50	170	350	570		
	1993	4.6	0.65	-	-	B(k)F	1992	150	60	-	-	
	1994	3.6	-	-	-		1993	140	84	-	-	
	1995	7.8	1.2	-	-		1994	81	100	-	-	
	1996	2.6	-	-	-		B(b+k)F	1995	560	580	52	1200
	1997	1.8	-	-26	-24		B(b+k)F	1996	158	430	15	600
1998	0.37	-	-25	-25	B(b+k)F		1997	305	376	37	720	
p,p'-DDT	1992	34	4.3	20	58	B(b+k)F	1998	93	441	39	570	
	1993	98	2.1	30	130	B(a)P	1992	180	63	-	-	
	1994	15	-	14	-		1993	190	57	-	-	
	1995	14	1.5	-	-		1994	97	98	-	-	
	1996	4.9	-	-	-		1995	190	100	-0.75	290	
	1997	2.3	1.2	-9.9	-6.4		1996	50	120	-8	160	
1998	0.38	0.64	-12	-11	1997		120	100	5.1	230		
HCB	1992	0.88	0.2	-17	-16	1998	27	98	3.3	130		
	1993	5.4	6.4	0.76	13	Pb	1992	-	-	-	-	
	1994	0.4	0.21	-7	-6.4		1993	-	13000	-	-	
	1995	0.73	0.22	-1	-0.05		1994	-	13000	-	-	
	1996	0.34	-	-5.7	-5.4		1995	-	-	-	-	
	1997	0.3	-	-2.8	-2.5		1996	-	-	-	-	
1998	0.23	-	-0.97	-0.74	1997		-	-	-	-		
PCB18	1992	0.34	0.12	-17	-17	1998	-	-	-	-		
	1993	0.57	0.21	-15	-14	As	1992	-	-	-	-	
	1994	0.56	0.74	-15	-14		1993	-	1500	-14	-	
	1995	1.4	0.6	-22	-20		1994	-	1400	-	-	
	1996	0.43	-	-26	-26		1995	-	-	-	-	
	1997	0.35	-	-13	-13		1996	-	-	-	-	
1998	0.1	-	-10	-9.9	1997		-	-	-	-		
PCB44	1992	0.55	0.23	-5.4	-4.6	1998	-	-	-	-		
	1993	0.81	0.43	-1.1	0.14	Se	1992	-	-	-	-	
	1994	1.6	0.83	-5.2	-2.8		1993	-	2800	-	-	
	1995	0.96	1.2	-11	-8.8		1994	-	2400	-	-	
	1996	0.32	-	-16	-16		1995	-	-	-	-	
	1997	0.43	-	-3.4	-3		1996	-	-	-	-	
1998	0.37	-	-6.3	-5.9	1997		-	-	-	-		
PCB52	1992	0.42	0.28	-5.8	-5.1	1998	-	-	-	-		
	1993	0.729	0.33	-2.2	-1.1	Cd	1992	-	-	-	-	
	1994	1	0.85	-5.2	-3.4		1993	-	1100	-	-	
	1995	1.7	1.3	-8.6	-5.6		1994	-	1500	-3.4	-	
	1996	0.65	-	-13	-12		1995	-	-	-	-	
	1997	0.6	-	9	9.6		1996	-	-	-	-	
1998	0.21	-	7.6	7.8	1997		-	-	-	-		
					1998	-	-	-	-			

**Table D5. IADN annual flows (kg/yr) from 1992 to 1998 for Lake Ontario**

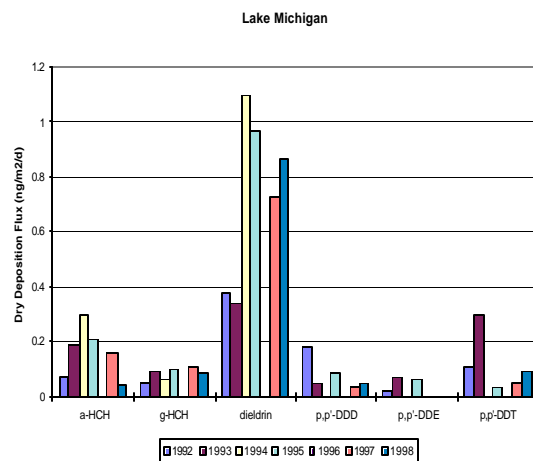
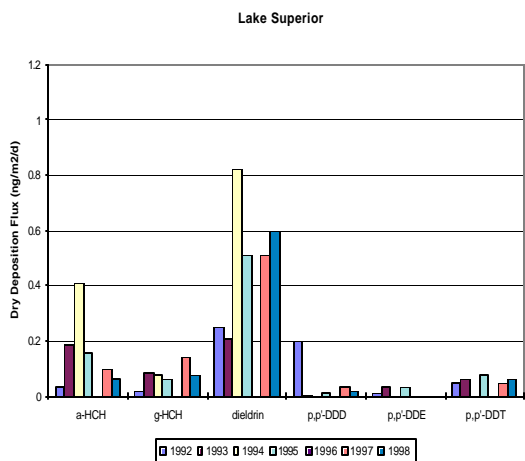
		Wet Deposition	Dry Deposition	Net Gas Exchange	Total Deposition			Wet Deposition	Dry Deposition	Net Gas Exchange	Total Deposition	
α-HCH	1992	52	-	-80	-28	PCB101	1992	1.7	-	-1.6	0.1	
	1993	32	-	-54	-22		1993	2.3	-	-2.4	-0.1	
	1994	33	-	-23	10		1994	0.62	-	-0.84	-0.22	
	1995	21	-	-5.7	15		1995	1.8	-	-6.2	-4.4	
	1996	31	-	-11	20		1996	1.3	-	-5.4	-4.1	
	1997	25	-	-18	7		1997	-	-	-2.4	-	
	1998	21	-	-44	-23		1998	-	-	-2.9	-	
γ-HCH	1992	50	-	-8.2	42	ΣPCB	1992	56	-	-450	-390	
	1993	37	-	-1.5	36		1993	89	-	-570	-480	
	1994	24	-	7.9	32		1994	15	-	-450	-440	
	1995	13	-	12	25		1995	38	-	-230	-190	
	1996	26	-	17	43		1996	26	-	-230	-200	
	1997	50	-	6.8	57		suite-PCB	1997	-	-	-19	-
	1998	33	-	3.4	36		suite-PCB	1998	-	-	-18	-
dieldrin	1992	11	-	-330	-320	phenanthrene	1992	70	41	-510	-400	
	1993	5.4	-	-200	-190		1993	540	25	-	-	
	1994	3	-	-180	-180		1994	380	44	-	-	
	1995	3.9	-	-230	-230		1995	110	28	-	-	
	1996	4.5	-	-210	-210		1996	250	63	-	-	
	1997	6	-	-110	-100		1997	390	51	-630	-190	
	1998	4.5	-	-120	-120		1998	340	52	-410	-18	
p,p'-DDD	1992	2.9	-	-	-	pyrene	1992	64	88	-46	110	
	1993	0.36	-	-	-		1993	470	55	-	-	
	1994	0.49	-	-	-		1994	220	48	-	-	
	1995	0.42	-	-	-		1995	130	46	-	-	
	1996	0.59	-	-	-		1996	260	83	-	-	
	1997	2.3	-	-	-		1997	450	72	-110	410	
	1998	2	-	-	-		1998	210	100	3.6	310	
p,p'-DDE	1992	4.4	-	-96	-92	B(k)F	1992	33	92	-9.7	120	
	1993	2	-	-99	-97		B(k)F	1993	-	79	-	-
	1994	0.61	-	-80	-79		B(k)F	1994	-	29	-	-
	1995	5.3	-	-	-		B(b+k)F	1995	173	105	-	-
	1996	2	-	-	-		B(b+k)F	1996	311	249	-	-
	1997	11	-	-21	-10		B(b+k)F	1997	210	225	3.5	440
	1998	3.9	-	-24	-20		B(b+k)F	1998	259	315	41	620
p,p'-DDT	1992	3.3	-	4.4	7.7	B(a)P	1992	54	86	-	-	
	1993	7.2	-	5.5	13		1993	-	79	-	-	
	1994	1.1	-	8.2	9.3		1994	-	43	-	-	
	1995	14	-	-	-		1995	63	29	-	-	
	1996	4.1	-	-	-		1996	110	60	-	-	
	1997	13	-	5.7	19		1997	81	60	-7.6	130	
	1998	1.3	-	3.9	5.2		1998	76	69	-8	140	
HCB	1992	6.1	-	-170	-160	Pb	1992	40000	4500	-	45000	
	1993	3	-	-190	-190		1993	27000	5300	-	32000	
	1994	0.62	-	-150	-150		1994	15000	6100	-	21000	
	1995	0.73	-	-32	-31		1995	7600	3300	-	11000	
	1996	0.87	-	-28	-27		1996	5000	5100	-	10000	
	1997	0.87	-	-24	-23		1997	21000	5100	-	26000	
	1998	0.56	-	-22	-21		1998	37000	5800	-	43000	
PCB18	1992	2.5	-	-19	-17	As	1992	2900	570	-	3500	
	1993	0.28	-	-22	-22		1993	3100	720	-	3800	
	1994	0.35	-	-19	-19		1994	2100	630	-	2700	
	1995	0.81	-	-18	-17		1995	970	210	-	1200	
	1996	0.71	-	-16	-15		1996	580	610	-	1200	
	1997	-	-	-8.7	-		1997	-	650	-	-	
	1998	-	-	-10	-		1998	-	1000	-	-	
PCB44	1992	3.6	-	-19	-15	Se	1992	5500	1000	-	6500	
	1993	5.7	-	-21	-15		1993	5000	1600	-	6600	
	1994	0.66	-	-18	-17		1994	3900	1800	-	5700	
	1995	2	-	-20	-18		1995	1300	150	-	1500	
	1996	1.4	-	-19	-18		1996	1100	340	-	1400	
	1997	-	-	-10	-		1997	-	600	-	-	
	1998	-	-	-10	-		1998	-	460	-	-	
PCB52	1992	1.6	-	-18	-16	Cd	1992	2600	96	-	2700	
	1993	1.4	-	-22	-21		1993	1300	99	-	1400	
	1994	0.93	-	-18	-17		1994	550	130	-	680	
	1995	2.5	-	-11	-8.5		1995	530	49	-	580	
	1996	2.5	-	-9.5	-7		1996	390	100	-	490	
	1997	-	-	-0.2	-		1997	1100	170	-	1300	
	1998	-	-	-2.1	-		1998	1600	170	-	1800	



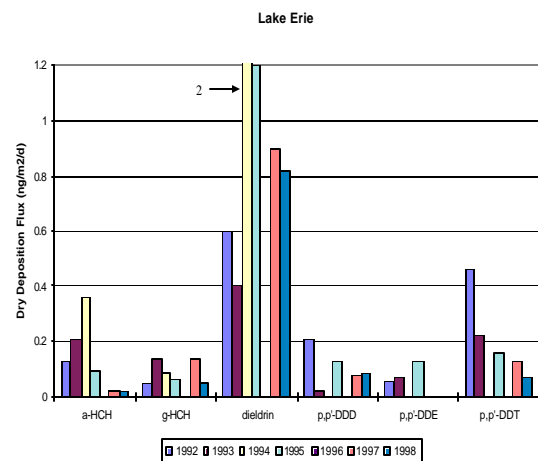
## **Appendix E. Annual Fluxes to the Great Lakes, 1992-1998**



**Figure E1. Annual average wet deposition flux (ng/m<sup>2</sup>/day) of organochlorine pesticides**

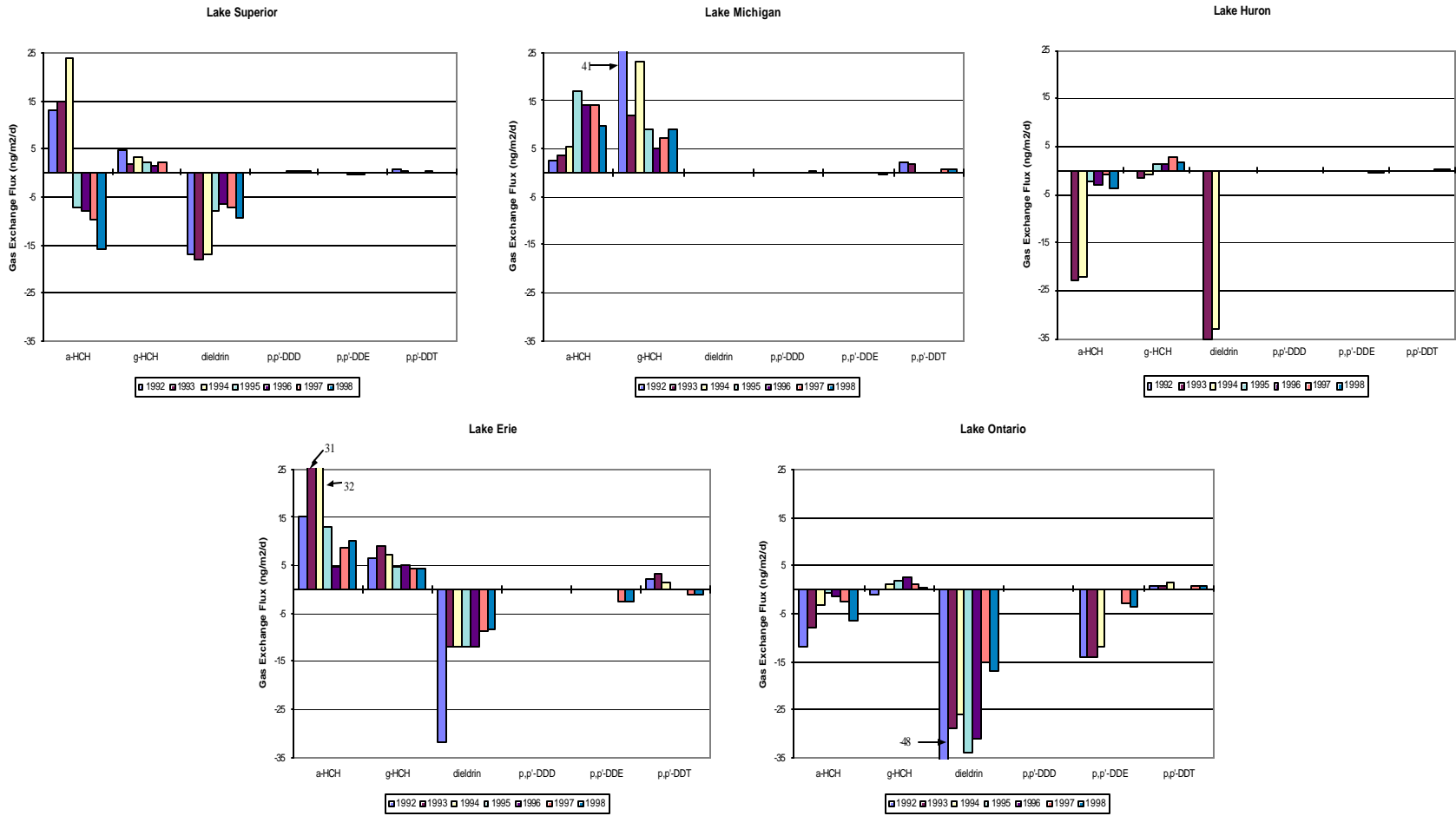


Data not available for Lake Huron

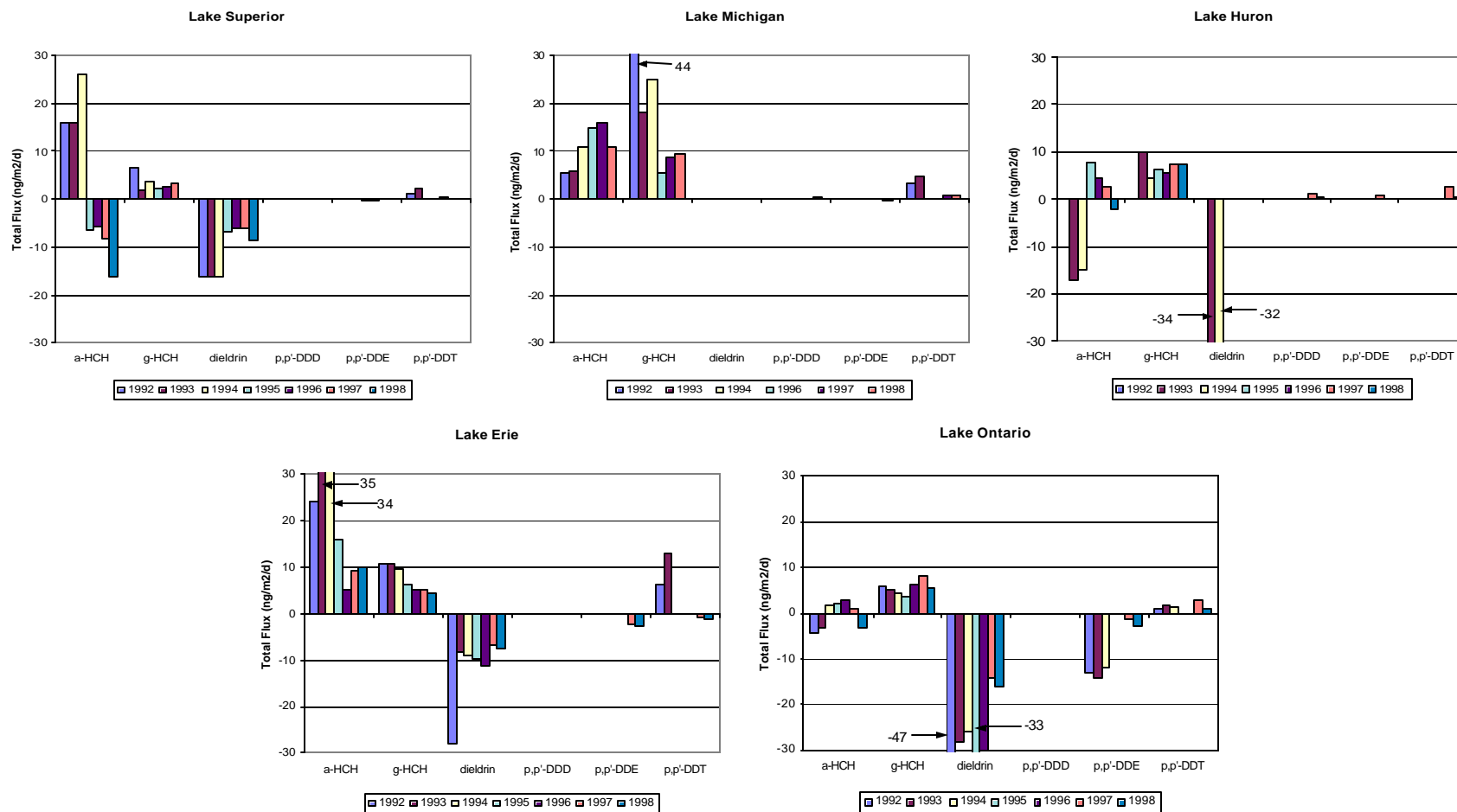


Data not available for Lake Ontario

**Figure E2. Annual average dry deposition flux (ng/m<sup>2</sup>/day) of organochlorine pesticides**



**Figure E3. Annual average net gas exchange flux (ng/m<sup>2</sup>/day) of organochlorine pesticides. Positive values denote net gas absorption, negative values denote net volatilization.**



**Figure E4. Annual average total flux (ng/m<sup>2</sup>/day) of organochlorine pesticides**

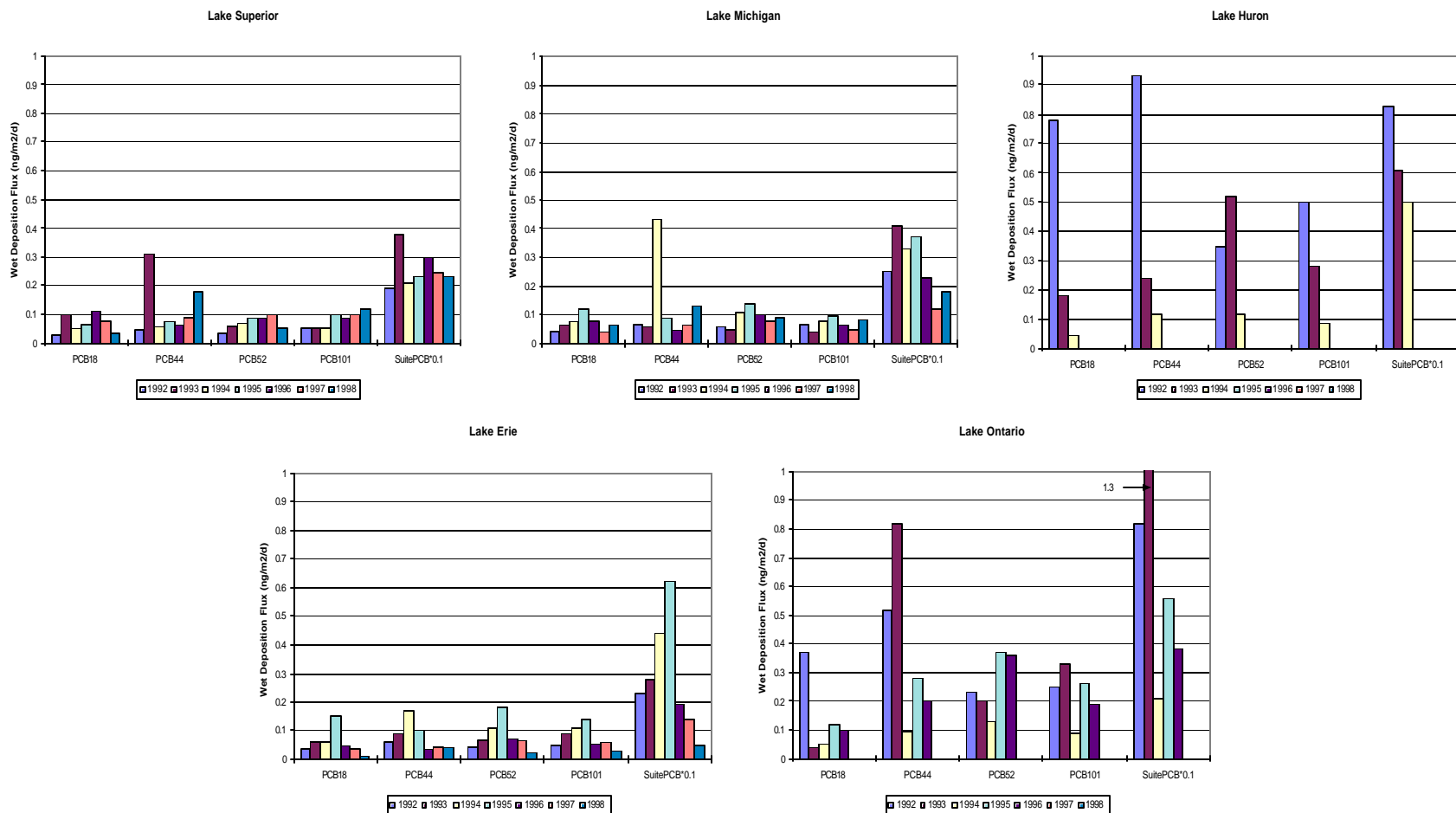
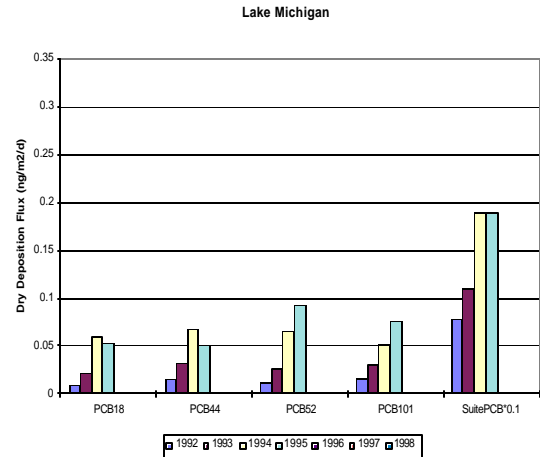
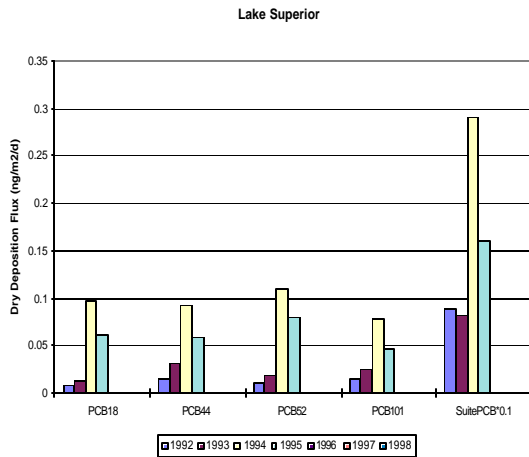
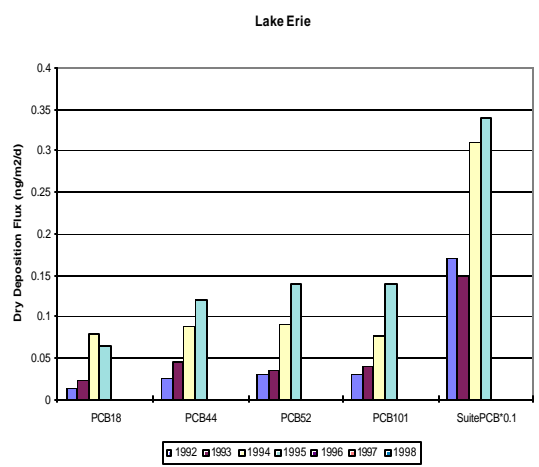


Figure E5. Annual average wet deposition flux (ng/m<sup>2</sup>/day) of PCBs. Note that sum-PCB is multiplied by 0.10.

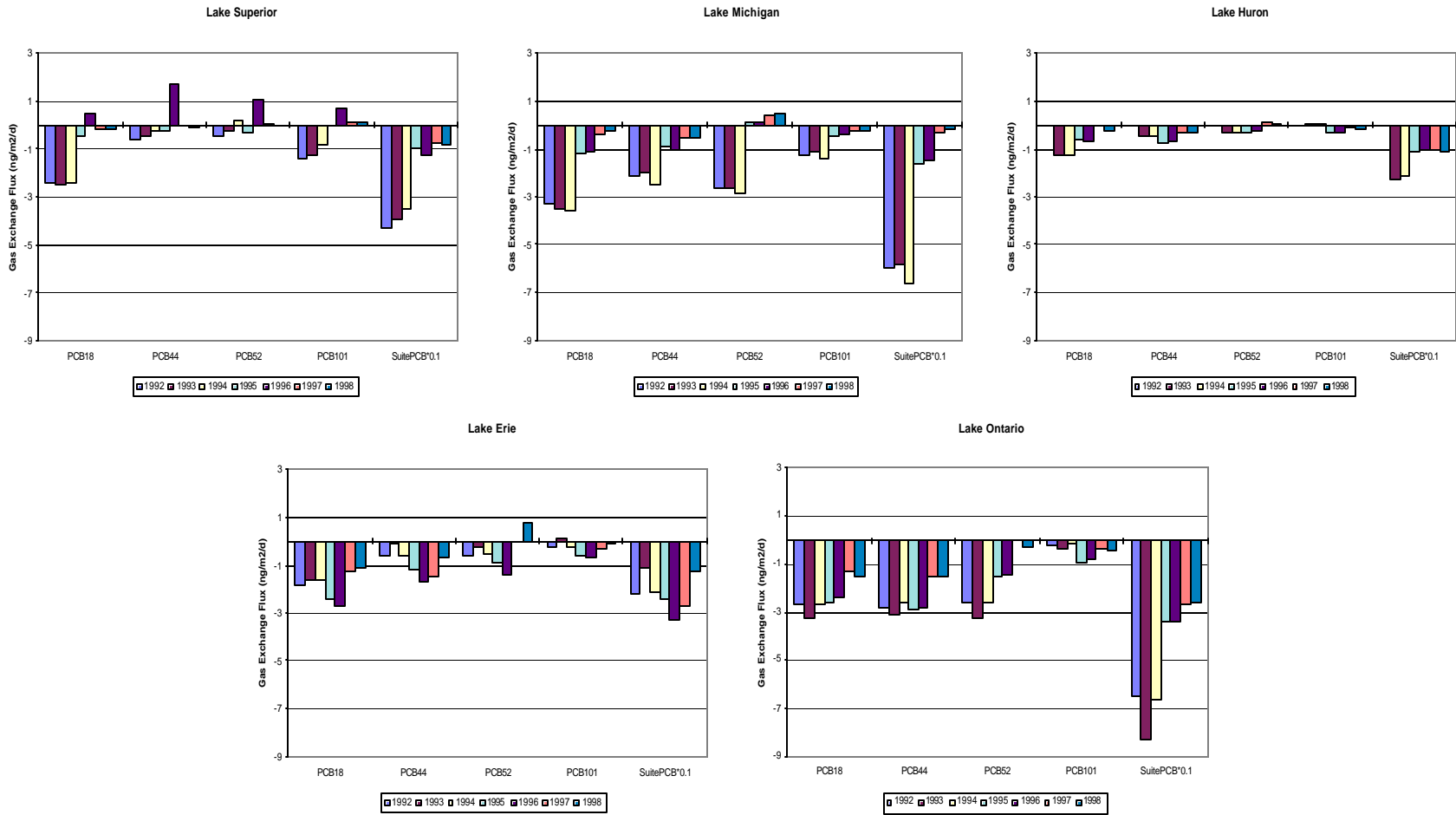


Data not available for Lake Huron



Data not available for Lake Ontario

**Figure E6. Annual average dry deposition flux (ng/m<sup>2</sup>/day) of PCBs. Note that sum-PCB is multiplied by 0.10.**



**Figure E7. Annual average net gas exchange flux (ng/m<sup>2</sup>/day) of PCBs. Note that sum-PCB is multiplied by 0.10. Positive values denote net gas absorption, negative values denote net volatilization.**



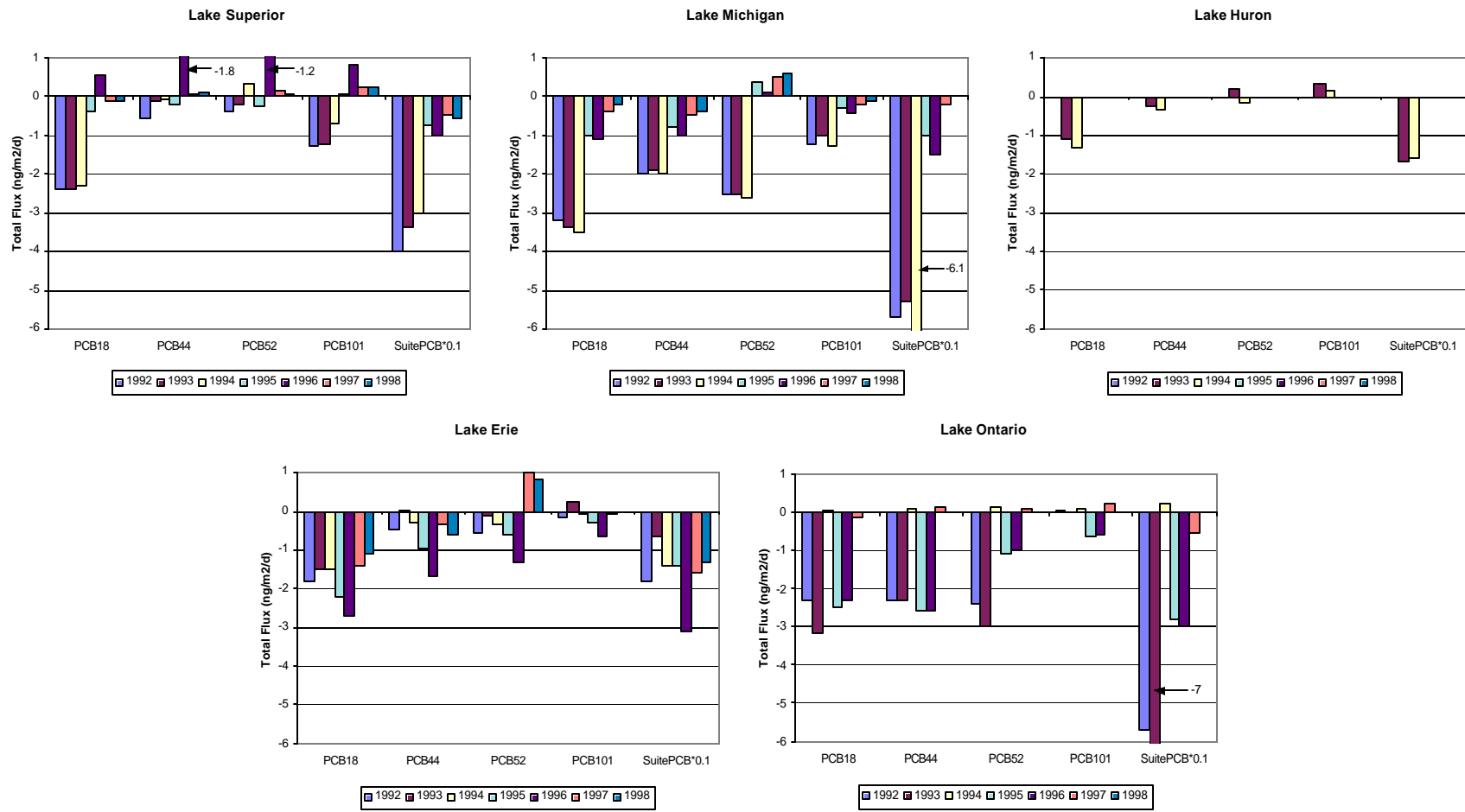


Figure E8. Annual average total flux (ng/m<sup>2</sup>/day) of PCBs. Note that sum-PCB is multiplied by 0.10.

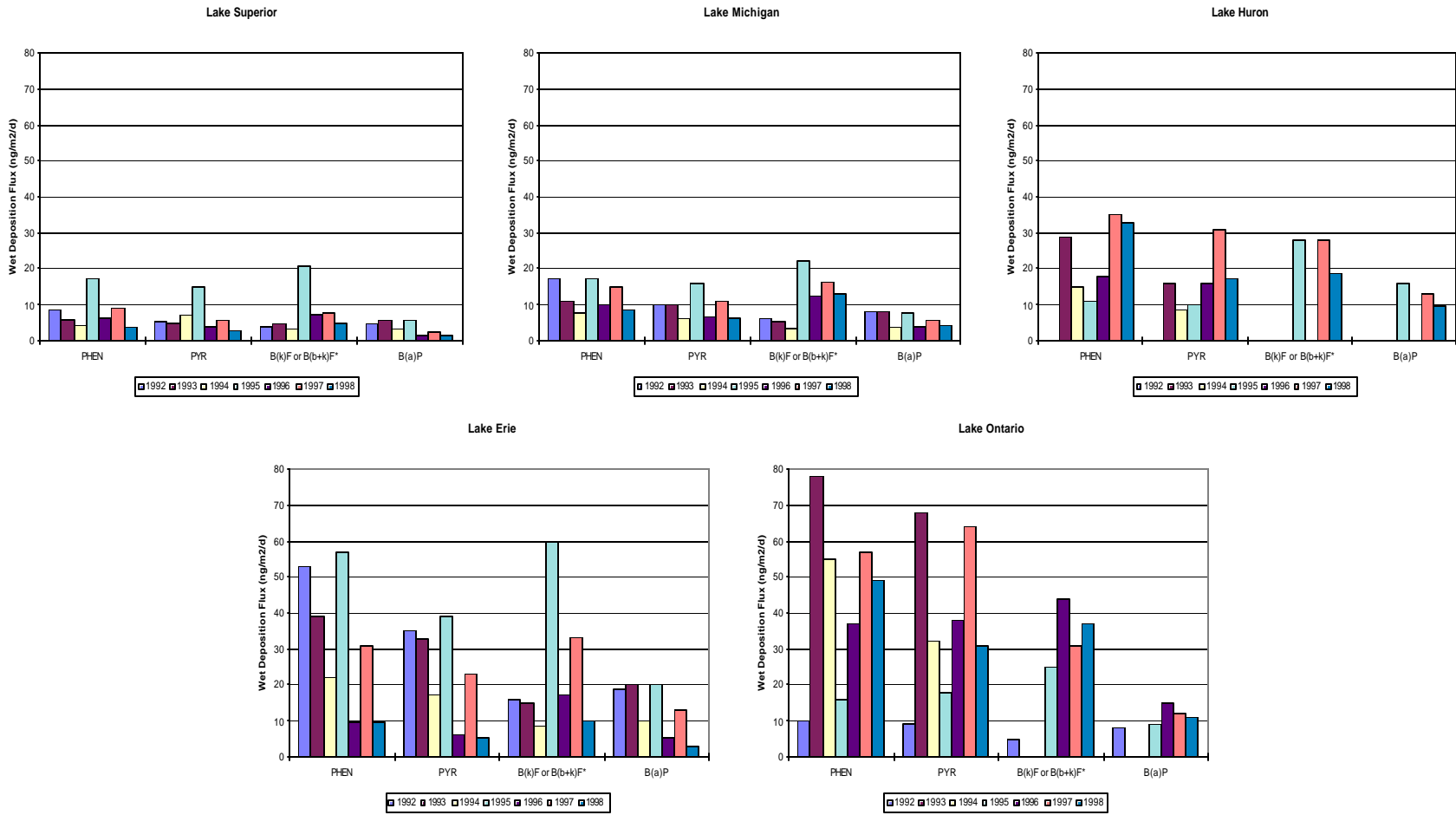


Figure E9. Annual average wet deposition flux (ng/m<sup>2</sup>/day) of PAHs. \* 1992-1994 : B(b)F; 1995-1998 : B(b+k)F

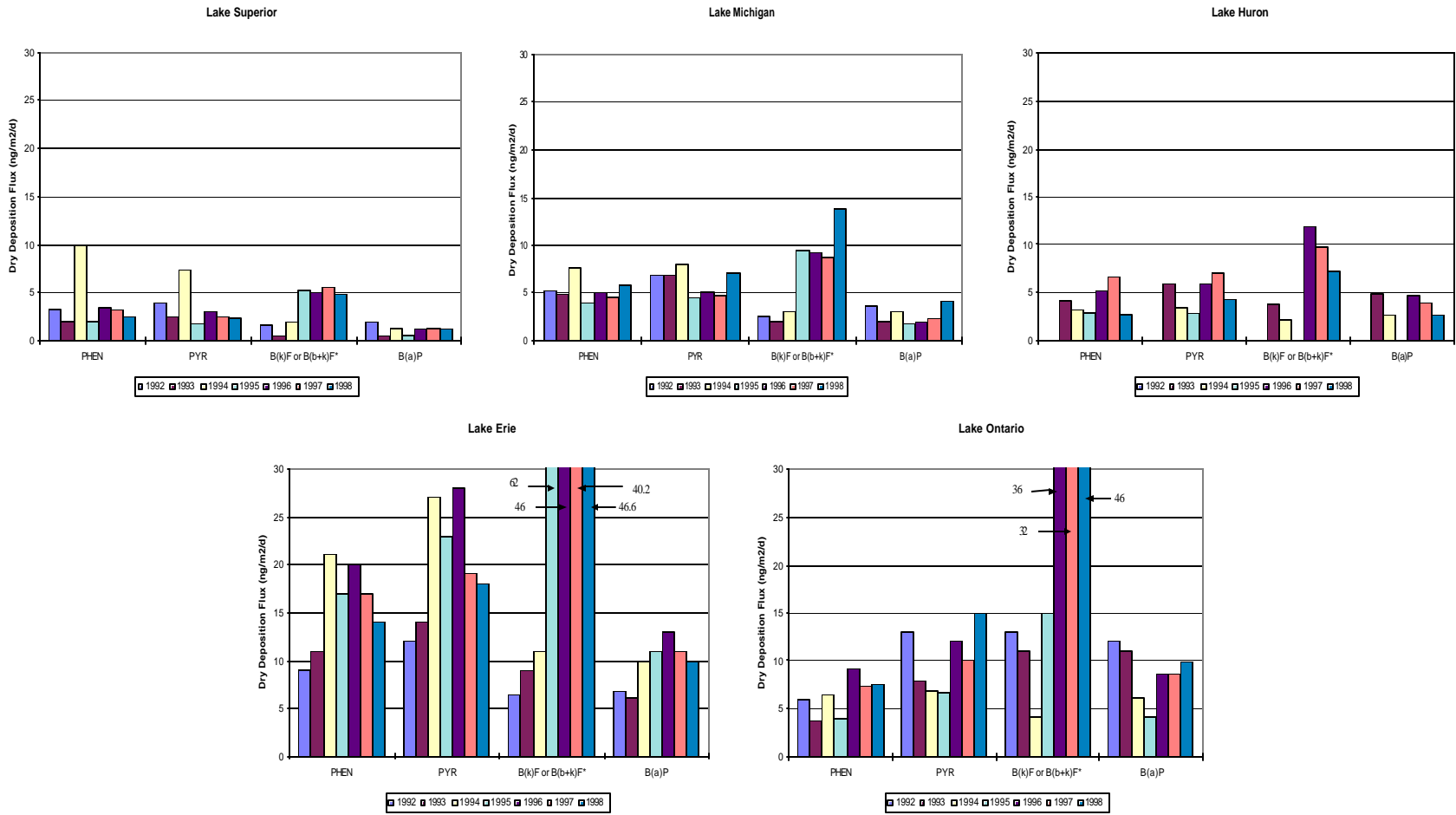
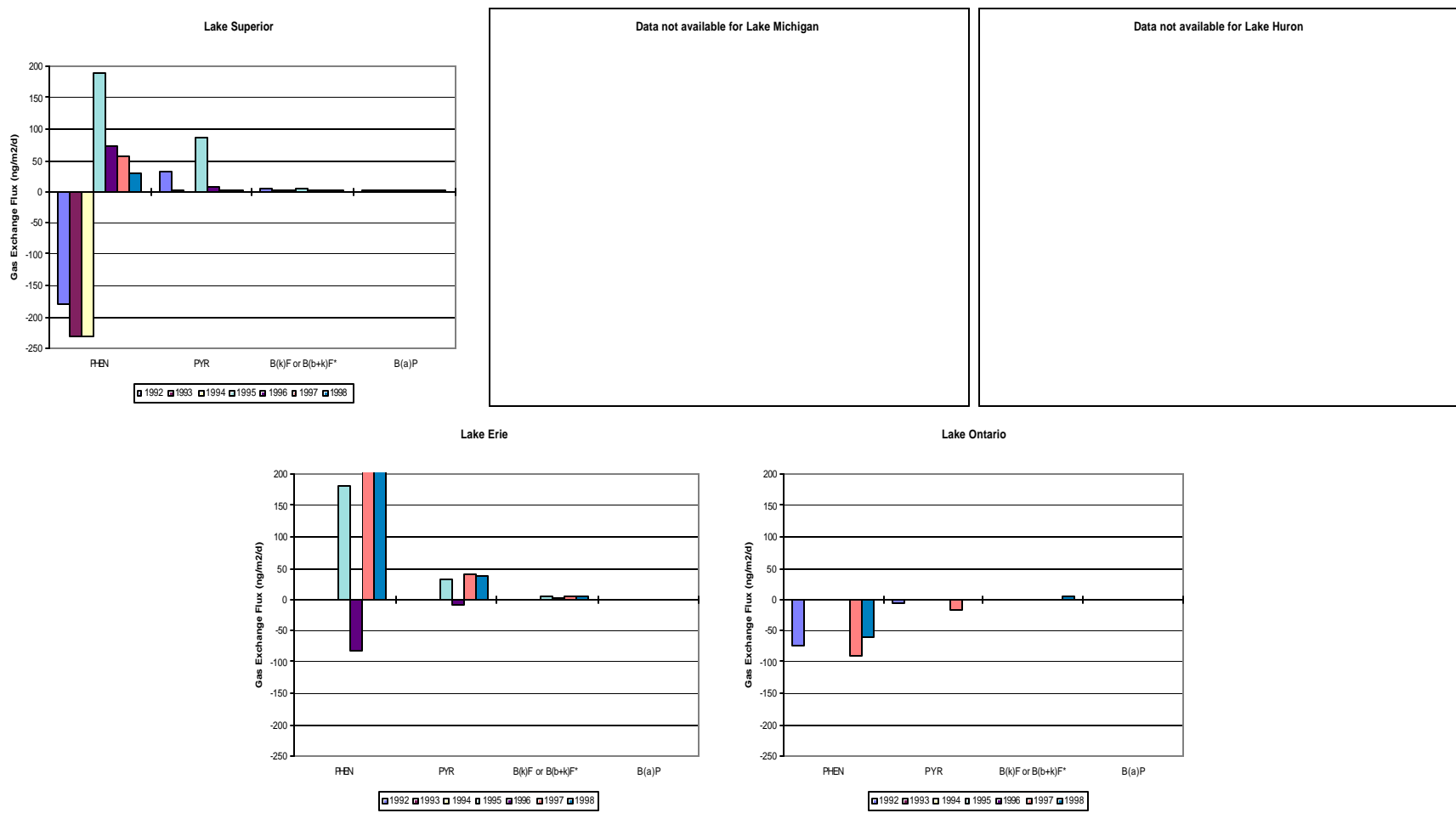


Figure E10. Annual average dry deposition flux (ng/m<sup>2</sup>/day) of PAHs. \* 1992-1994 : B(b)F; 1995-1998 : B(b+k)F



**Figure E11. Annual average net gas exchange flux (ng/m<sup>2</sup>/day) of PAHs. Positive values denote net gas absorption, negative values denote net volatilization. \* 1992-1994 : B(b)F; 1995-1998 : B(b+k)F**

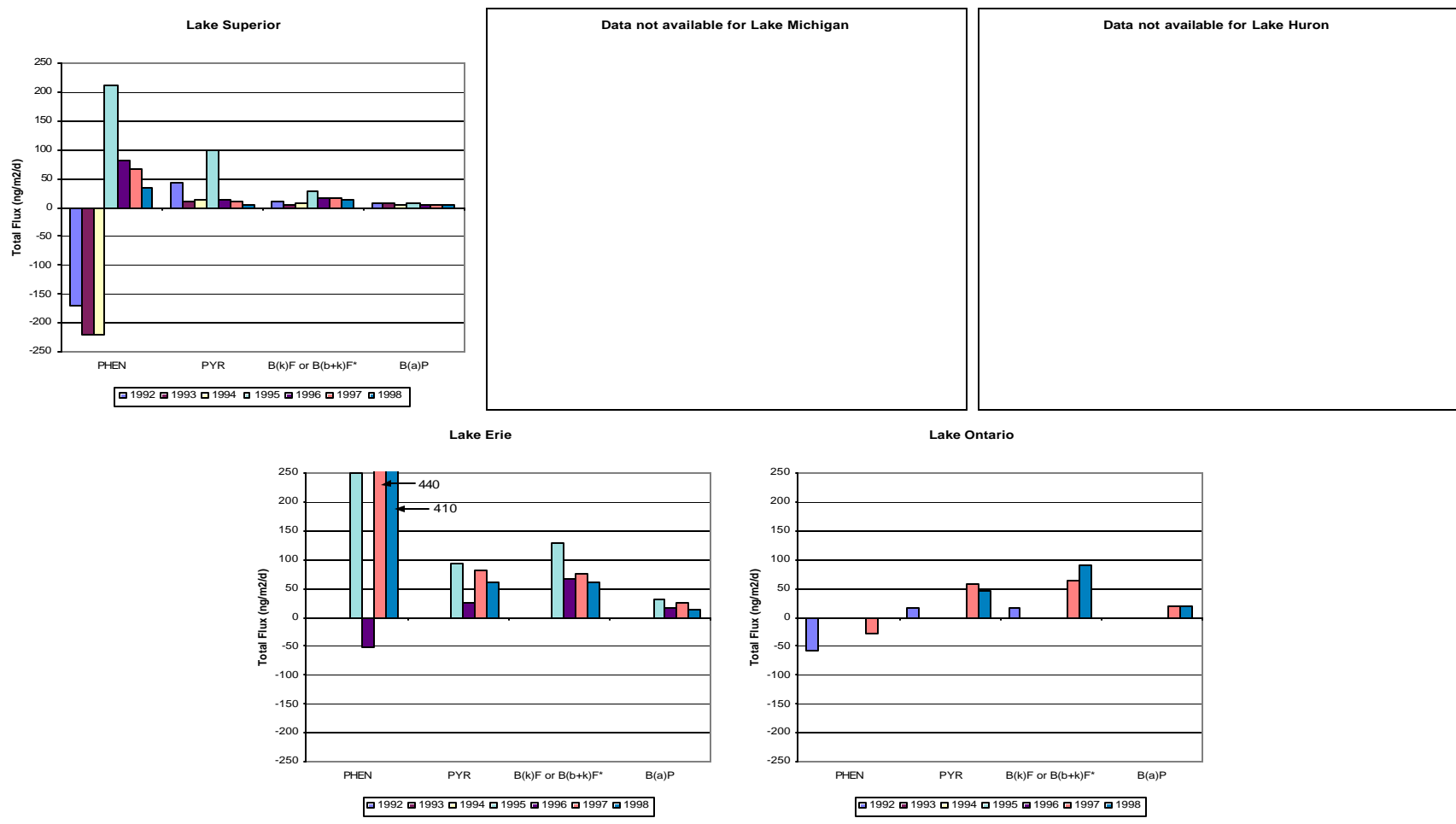


Figure E12. Annual average total flux (ng/m<sup>2</sup>/day) of PAHs. \* 1992-1994 : B(b)F; 1995-1998 : B(b+k)F

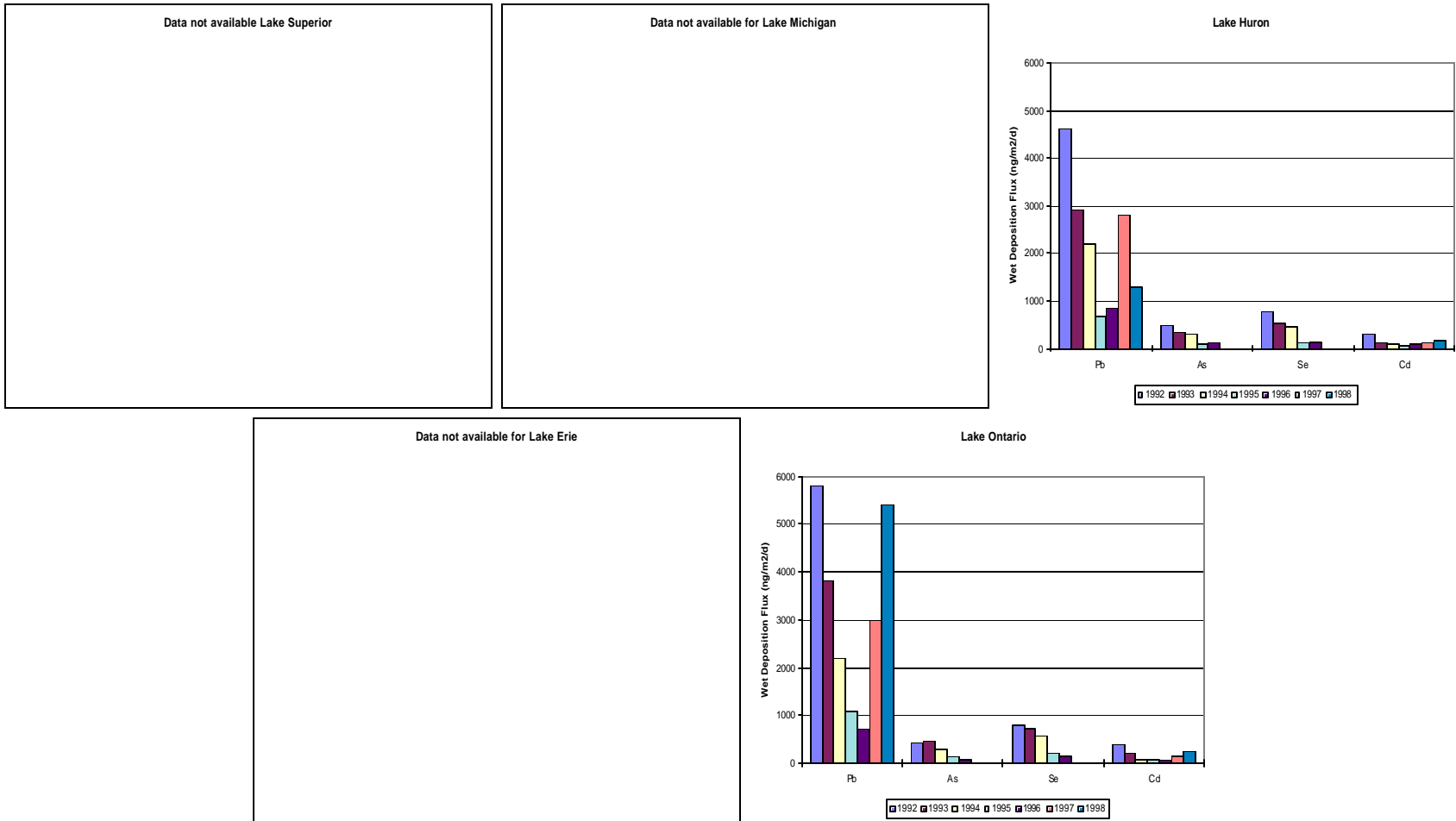


Figure E13. Annual average wet deposition flux (ng/m<sup>2</sup>/day) of metals

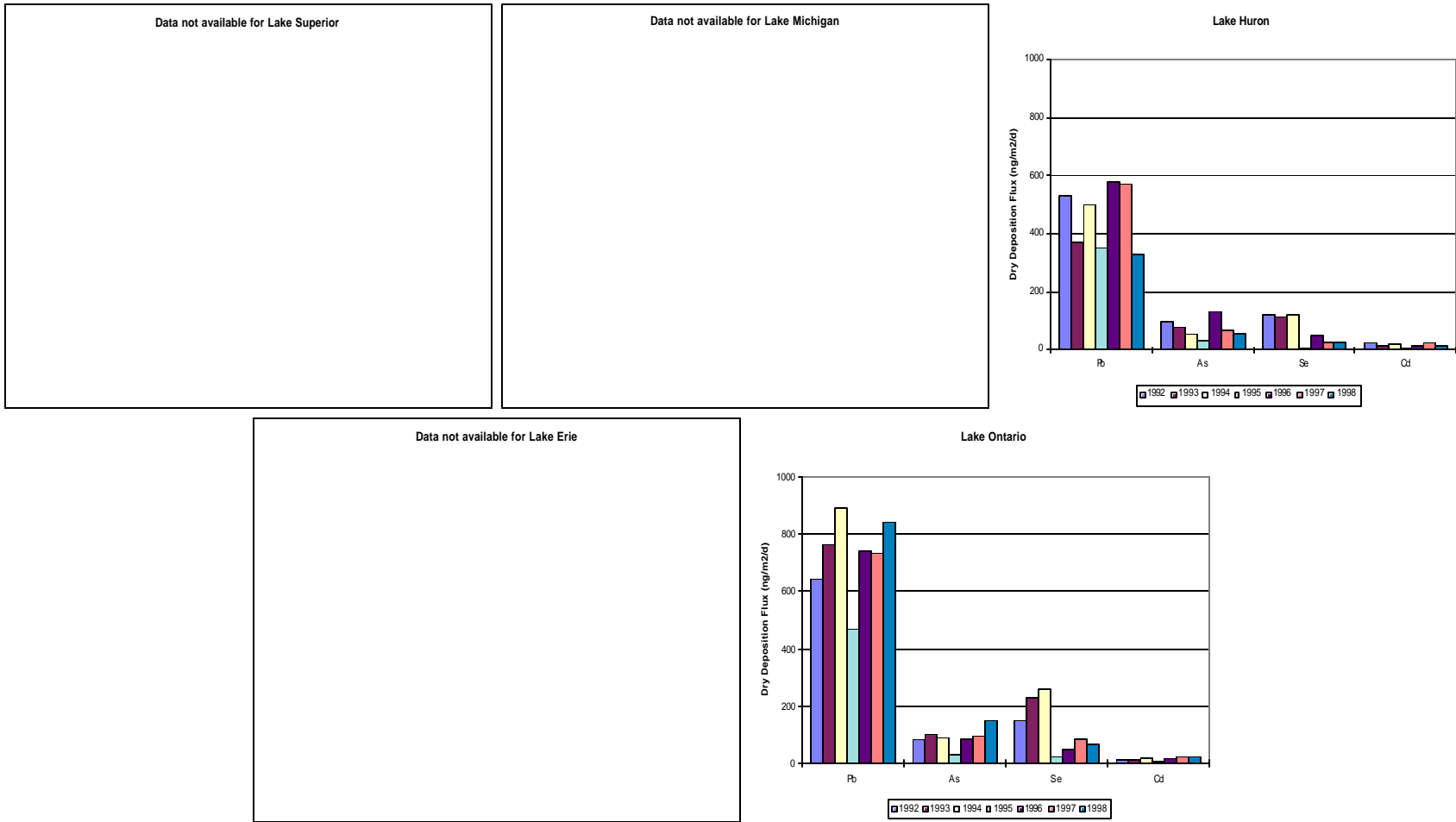
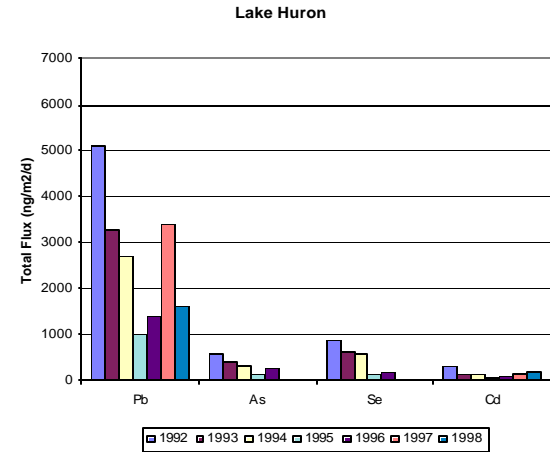


Figure E14. Annual average dry deposition flux (ng/m<sup>2</sup>/day) of metals

Data not available Lake Superior

Data not available for Lake Michigan



Data not available for Lake Erie

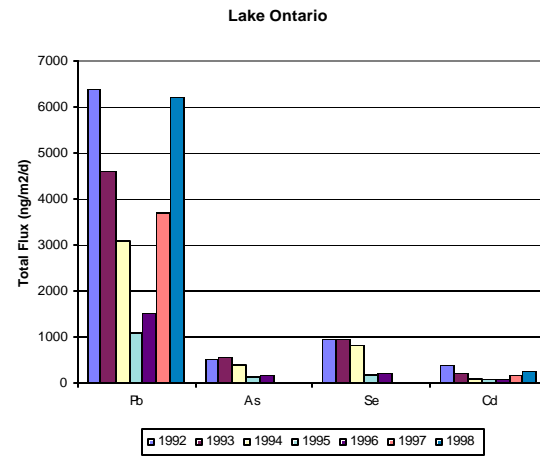


Figure E15. Annual average total flux (ng/m<sup>2</sup>/day) of metals



## **Appendix F. Annual Fugacity Ratios for IADN Substances, 1992-1998**

**Table F1. Fugacity ratios (absorption over volatilization) for gas exchange loadings over the Great Lakes**

		Lake Superior	Lake Michigan	Lake Huron	Lake Erie	Lake Ontario			Lake Superior	Lake Michigan	Lake Huron	Lake Erie	Lake Ontario
$\alpha$ -HCH	1992	1.7	1.1	-	1.5	0.62	$p,p'$ -DDD	1992	-	-	-	-	-
	1993	1.7	1.1	0.38	3.4	0.67		1993	-	-	-	-	-
	1994	2.3	1.2	0.38	3.5	0.88		1994	-	-	-	-	-
	1995	0.8	3.9	0.84	1.9	1		1995	1.3	-	-	-	-
	1996	0.73	4	0.78	1.4	0.9		1996	2.7	-	-	-	-
	1997	0.66	3.1	0.9	2.2	0.79		1997	100	39	10	-	-
	1998	0.56	2.1	0.65	2.3	0.55		1998	65	45	7.6	-	-
	$\gamma$ -HCH	1992	2.1	12	-	2		0.82	$p,p'$ -DDE	1992	-	-	-
1993		1.4	4.1	0.69	2.8	0.95	1993	-		-	-	-	0.17
1994		1.8	7.1	0.79	2.4	1.2	1994	-		-	-	-	0.23
1995		1.7	11	1.9	2.9	1.6	1995	0.34		-	-	-	-
1996		1.5	7.5	1.9	3	1.9	1996	0.29		-	-	-	-
1997		1.7	7.2	3.7	2.4	1.3	1997	1.2		0.96	0.48	0.37	0.4
1998		0.92	7.3	2.5	2.3	1.1	1998	0.59		0.56	0.51	0.31	0.29
dieldrin		1992	0.14	-	-	0.1	0.06	$p,p'$ -DDT		1992	4.4	13	-
	1993	0.11	-	0.062	0.31	0.15	1993		2.8	11	2.3	39	4
	1994	0.14	-	0.062	0.26	0.16	1994		-	-	2.4	19	6.1
	1995	0.19	-	-	0.21	0.08	1995		1.2	-	-	-	-
	1996	0.18	-	-	0.16	0.07	1996		2	-	-	-	-
	1997	0.17	-	-	0.27	0.17	1997		1.4	6.5	4.8	0.57	4.2
	1998	0.097	-	-	0.23	0.12	1998		1.2	6.1	3.2	0.48	3.1
	$\alpha$ -endosulfan	1992	-	-	-	-	-		HCB	1992	1.7	1.3	0.0011
1993		-	-	-	-	-	1993	1.2		1	0.74	1	0.12
1994		-	-	-	-	-	1994	1.2		0.86	0.67	0.72	0.17
1995		0.95	-	-	910	2300	1995	1.4		1.2	0.56	0.96	0.41
1996		0.65	-	-	530	2000	1996	1.4		1.2	0.57	0.78	0.46
1997		2.1	-	-	6600	2800	1997	1.3		1.3	0.74	0.93	0.49
1998		1.7	-	-	6000	3300	1998	1.4		1.5	0.68	0.96	0.45
<i>cis</i> -chlordane		1992	-	-	-	-	-	PCB18		1992	0.084	0.11	-
	1993	-	-	-	-	-	1993		0.083	0.093	0.22	0.29	0.25
	1994	-	-	-	-	-	1994		0.073	0.082	0.17	0.19	0.26
	1995	0.73	1	0.54	2.1	0.59	1995		0.26	0.14	0.52	0.14	0.28
	1996	0.55	0.79	0.56	1.2	0.58	1996		1.9	0.13	0.4	0.067	0.31
	1997	0.45	1.1	0.62	1.1	0.69	1997		0.43	0.36	1	0.27	0.51
	1998	0.26	0.76	0.62	0.91	0.52	1998		0.41	0.59	0.63	0.29	0.33
	<i>trans</i> -chlordane	1992	-	-	-	-	-		PCB44	1992	0.26	0.21	-
1993		-	-	-	-	-	1993	0.46		0.29	0.18	0.91	0.14
1994		-	-	-	-	-	1994	0.68		0.12	0.17	0.49	0.15
1995		1.4	0.83	0.45	1.7	0.44	1995	0.64		0.29	0.14	0.45	0.11
1996		0.51	0.7	0.51	0.96	0.39	1996	3.4		0.17	0.12	0.2	0.095
1997		0.22	1	0.85	0.51	0.89	1997	0.93		0.29	0.3	0.75	0.24
1998		0.083	0.65	0.79	0.5	0.57	1998	0.73		0.35	0.24	0.48	0.17
<i>trans</i> -nonachlor		1992	-	-	-	-	-	PCB52		1992	0.43	0.16	-
	1993	-	-	-	-	-	1993		0.65	0.21	0.42	0.86	0.2
	1994	-	-	-	-	-	1994		1.2	0.14	0.37	0.58	0.26
	1995	0.35	0.18	0.17	0.53	0.26	1995		0.66	1.7	0.35	0.48	0.3
	1996	0.31	0.14	0.18	0.4	0.2	1996		2.4	1.5	0.43	0.22	0.34
	1997	0.38	0.21	0.22	0.38	0.25	1997		1.2	12	1.5	30	0.97
	1998	0.4	0.2	0.19	0.36	0.18	1998		1.1	18	1.2	32	0.67

**Table F1 (continued)**

		Lake Superior	Lake Michigan	Lake Huron	Lake Erie	Lake Ontario
PCB101	1992	0.11	0.2	-	0.69	0.71
	1993	0.22	0.34	1.4	1.2	0.62
	1994	0.47	0.17	1.4	0.58	0.85
	1995	0.93	0.31	0.18	0.42	0.23
	1996	2.6	0.28	0.22	0.28	0.28
	1997	4.5	0.35	0.47	0.79	0.52
	1998	3.7	0.54	0.37	0.84	0.38
ΣPCB	1992	0.15	0.13	-	0.4	0.24
	1993	0.26	0.21	0.15	0.69	0.11
	1994	0.3	0.1	0.16	0.32	0.19
	1995	0.53	0.23	0.28	0.31	0.21
	1996	0.28	0.22	0.21	0.091	0.17
	1997	0.38	0.52	0.23	0.38	0.21
	1998	0.32	0.84	0.15	0.42	0.15
suite-PCB suite-PCB	1992	0.5	-	-	-	0.69
	1993	0.36	-	-	-	-
	1994	0.35	-	-	-	-
	1995	4.1	-	-	1.3	-
	1996	2.5	-	-	0.84	-
	1997	2.1	-	-	3.6	0.63
	1998	1.5	-	-	3.8	0.71
pyrene	1992	3.6	-	-	-	0.79
	1993	1.2	-	-	-	-
	1994	0.86	-	-	-	-
	1995	15	-	-	1.4	-
	1996	2.4	-	-	0.87	-
	1997	1.4	-	-	3.2	0.58
	1998	1.1	-	-	3	1
B(k)F B(k)F B(k)F B(b+k)F B(b+k)F B(b+k)F B(b+k)F	1992	70	-	-	-	0.28
	1993	10	-	-	-	-
	1994	36	-	-	-	-
	1995	36	-	-	3.4	-
	1996	23	-	-	1.8	-
	1997	71	-	-	9.8	1.5
	1998	53	-	-	9.6	6.4
B(a)P	1992	1.4	-	-	-	-
	1993	1.3	-	-	-	-
	1994	1.6	-	-	-	-
	1995	14	-	-	0.96	-
	1996	8.3	-	-	0.62	-
	1997	10	-	-	1.6	0.22
	1998	7.3	-	-	1.4	0.27
I(1,2,3-cd)P	1992	-	-	-	-	-
	1993	-	-	-	-	-
	1994	-	-	-	-	-
	1995	28000	-	-	2500	-
	1996	30000	-	-	1700	-
	1997	35000	-	-	8000	-
	1998	24000	-	-	6800	-
sum-PAH (UNECE)	1992	-	-	-	-	-
	1993	-	-	-	-	-
	1994	-	-	-	-	-
	1995	28	-	-	2.4	-
	1996	19	-	-	1.4	-
	1997	20	-	-	4.6	-
	1998	23	-	-	4.4	-