

# AMBIENT LEVELS AND DRY DEPOSITION FLUXES OF MERCURY TO LAKES HURON, ERIE AND St. CLAIR

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**Abstract.** Ambient concentrations and dry deposition fluxes of Hg in the gas and particle phase to Lakes St. Clair, Erie and Huron were estimated with a hybrid receptor-deposition model (HRD). The ambient gas and particulate phase Hg concentrations were predicted to vary by a factor of 12 to 18 during the transport of air masses traversing the lakes. The ensemble average deposition fluxes of fine particle Hg ranged from 7  $\mu\text{g}/\text{m}^2\text{-h}$  to 15.3  $\mu\text{g}/\text{m}^2\text{-h}$  over Lake St. Clair, 0.5 to 4.2  $\mu\text{g}/\text{m}^2\text{-h}$  over Lake Huron and 5.1 to 20.6  $\mu\text{g}/\text{m}^2\text{-h}$  over Lake Erie. The deposition flux of coarse particle Hg was in the range of 50 to 84  $\mu\text{g}/\text{m}^2\text{-h}$  over Lake St. Clair, 4.7 to 24.2  $\mu\text{g}/\text{m}^2\text{-h}$  over Lake Huron and 5.1 to 20.6  $\mu\text{g}/\text{m}^2\text{-h}$  over Lake Erie. Gaseous Hg volatilized at a rate of 0.21 to 0.52  $\text{ng}/\text{m}^2\text{-h}$  from Lake Huron and 0.13 to 0.36 from Lake Erie. Gas phase Hg was deposited at a rate of 5.9  $\text{ng}/\text{m}^2\text{-h}$  and/or volatilized at a rate of 0.5  $\text{ng}/\text{m}^2\text{-h}$  from Lake St. Clair depending upon the location of the sampling site used in the HRD model. The effect of meteorological conditions, particle size distributions and type and location of the sampling sites played an important role in the transfer of atmospheric Hg to and/or from the lakes.

## 1. Introduction

Mercury is emitted into the atmosphere from a multitude of natural sources and from previously contaminated natural waters. Emissions from anthropogenic sources including coal and oil combustion, municipal solid waste and sewage sludge incineration, and smelting operations are more significant (EPA, 1993). As a consequence of the increase of anthropogenic emissions in industrialized countries some investigations indicate that ambient Hg concentrations have increased by 1.5% per year in the Northern Hemisphere and by 1.2% per year in the Southern Hemisphere during the 1977-1990 period (Slemer and Langer, 1992). In addition, increases in Hg deposition rates have been reported for the north temperate lake systems in Wisconsin, Minnesota and Sweden (Slemer and Langer, 1992; Swain *et al.*, 1992; Meger, 1986; Lindqvist *et al.*, 1991; Nater and Grigal, 1992). Swain *et al.* (1992) reported a 2% increase in Hg deposition rates in remote lakes in Wisconsin and Minnesota. Moreover, analysis of lake sediments and dated soils (Rada *et al.*, 1989; Norton *et al.*, 1990) as well as peat bogs (Norton *et al.*, 1990) suggest that Hg deposition may have doubled during the last century. It is now widely accepted that atmospheric deposition is an important pathway in the transfer of Hg to natural waters.

This paper presents estimates of deposition fluxes and exchange rates of gas phase Hg to Lakes St. Clair, Huron and Erie. Ambient concentrations and particle size distributions of atmospheric Hg measured in the Detroit urban/industrial area in April 1992 (Keeler *et al.*, 1994a) and March 1994 are used as input to a hybrid receptor-deposition model (HRD) to estimate the ambient concentrations, deposition fluxes, and exchange rates of the gas phase Hg at the air-water interface. The HRD model accounts for temporal and spatial variations in the ambient concentration, deposition flux and gas exchange rate associated with changing meteorological conditions, particle size distributions, and hydrodynamics (i.e., wave frequency and spray formation).

## 2. Methods

### 2.1 SAMPLING SITES

During a 10-day intensive study in April 1992 (Keeler *et al.* 1994a), ambient Hg concentrations for both gas and particle phases were measured at two urban sites: Site-5 was located in downtown Detroit close to Zug Island, which is one of the most industrialized areas in Southeastern Michigan. Anthropogenic Hg is emitted (in this area) from a large number of emission sources including coke ovens, iron and steel plants, incinerators, power generation facilities, and lime and cement operations (Pirrone *et al.*, 1994). The second site (Site-1) was located in a mixed residential/industrial land use area which is northwest of the Detroit Incinerator (GDRRA), one of the largest municipal solid waste incinerator facilities in the US which has been operating in Detroit since 1989.

In March 1994, an 18-day intensive study was performed in Detroit at the urban site (Site-5) to determine ambient Hg concentrations in the gas phase as well as the Hg associated with fine fraction ( $<2.5 \mu\text{m}$ ) and total suspended particulate matter. In addition, a six stage-microorifice cascade impactor (MOI) was used to determine the particle size distribution of particulate Hg in the atmosphere. The dry deposition velocity of particulate Hg in each particle size over Lakes Erie, St. Clair and Huron was then calculated.

### 2.2 ANALYTICAL PROCEDURES

Total particulate Hg was collected using an open-faced Teflon filter pack (Savillex) with a 47 mm diameter glass fiber filter (Gelman Science Type A/E) for 24 hours at a nominal flow rate of 30 L/min. Particulate Hg in the fine fraction ( $<2.5 \mu\text{m}$ ) was collected onto a 47 mm diameter glass fiber filter (Gelman Science Type A/E) after removal of particles in the coarse fraction using a Teflon-coated aluminum cyclone (University Research Glassware, N. C.). A six stage-MOI was used to measure the size distribution of the particulate Hg (Keeler *et al.*, 1994b). Samples were collected onto 37 mm glass fiber filters at a nominal flow rate of 30 L/min for 24 hours. After sampling, the filters were individually placed in acid-cleaned petri dishes and stored at  $-40 \text{ }^\circ\text{C}$  until analysis. Gas phase Hg was collected using gold-coated glass beads at a nominal flow rate of 300 mL/min for 12 and 24 hours.

Each glass fiber filter was extracted in 30 mL of 10%  $\text{HNO}_3$ , placed in a CEM MDS-2000 microwave oven for 20 minutes at  $160 \text{ }^\circ\text{C}$ , and then allowed to react for 12 hours at room temperature. After digestion, 10 mL of extract were removed with an Eppendorf pipet and placed into 30 mL acid cleaned polyethylene bottle for trace metals analysis using an ICP-MS. The remaining extract was used for Hg analysis after addition of 0.25 mL of  $\text{BrCl}$  to oxidize all the mercury to  $\text{Hg}^{++}$ . The glass fiber filters from the MOI were extracted with 10 mL of 10%  $\text{HNO}_3$  at  $160 \text{ }^\circ\text{C}$  for 20 minutes, allowed to react for 12 hours at room temperature, and oxidized to  $\text{Hg}^{++}$  with 0.25 mL of  $\text{BrCl}$ .

The Hg analysis was performed using dual amalgamation with Cold Vapor Atomic Fluorescence Spectrometry (Fitzgerald and Gill, 1979). Excess  $\text{BrCl}$  was removed by adding 100  $\mu\text{L}$  of hydroxylamine hydrochloride to 5 mL aliquots of filter extract and allowed to react for 5 min. Subsequently, 0.5 mL of stannous chloride were added to reduce the  $\text{Hg}^{++}$  to  $\text{Hg}^0$ , which was subsequently purged from the solution with an Hg-free nitrogen stream and concentrated onto a gold-coated sand trap. Calibration curves covering the expected concentration levels were analyzed on a daily basis and the check of standards was performed every six samples.

### 2.3 MODELING

The atmospheric transport of contaminants and their transfer to natural waters is affected by their chemical transformations and vapor-particle partitioning during transport, meteorological parameters, and wave dynamics and spray formation at the air-water interface. In order to estimate the deposition fluxes of particulate and gas phase Hg to Lakes St. Clair, Huron and Erie, the Hybrid Receptor-Deposition Model (Pirrone *et al.* 1994a; 1994b) developed to estimate the deposition flux of trace metals and semivolatile organic compounds (SOCs) to Lake Michigan was modified for Hg.

### 3. Results and Discussion

This study was aimed at assessing the levels of atmospheric Hg in the urban area of Detroit and estimating the ambient concentration, dry deposition flux and the air-water gas exchange rate of Hg over Lakes St. Clair, Huron and Erie. To accomplish these objectives, 24-hour integrated samples were used in the HRD model to evaluate the distribution of ambient concentrations and deposition fluxes of Hg in the particle and gas phase over the lakes. Dry deposition fluxes were determined for Hg in the gas phase and in the fine and coarse fraction measured at the industrial/urban sites in April 1992 and March 1994, and for particulate Hg with a mass median diameter (MMD) corresponding to the 50% collection efficiency of each MOI stage (see Table I). Major details on the particle size distribution of Hg measured with MOI can be found in Keeler *et al.* (1994b).

Table I shows ambient concentrations of Hg in the particle and gas phase measured in Detroit during April 1992 and March 1994. In March 1994 the particulate Hg concentration in the fine fraction ranged from 12 pg/m<sup>3</sup> to 190 pg/m<sup>3</sup>, which represents 86% of the total suspended particulate Hg. This finding agrees very well with the MOI data (obtained as sum of the last four MOI stages) which show that 86% of the atmospheric Hg collected on glass fiber filters of the last four MOI stages was in the fine fraction. Particulate Hg concentrations measured in March 1994 were found to be lower than those measured at the same location in Detroit in April 1992. The particulate Hg concentration measured in April 1992 ranged from 57 pg/m<sup>3</sup> to 1230 pg/m<sup>3</sup> at Site-5 and from 69 pg/m<sup>3</sup> to 1086 pg/m<sup>3</sup> at Site-1 with arithmetic means of 297 pg/m<sup>3</sup> at Site-5 and 342 pg/m<sup>3</sup> at Site-1. Ambient concentrations of Hg in the gas phase during April 1992 ranged from 0.41 ng/m<sup>3</sup> to 3.5 ng/m<sup>3</sup> at Site-5 and from 9.5 ng/m<sup>3</sup> to over 70 ng/m<sup>3</sup> at Site-1. The high concentrations found at Site-1 suggest the impact of a significant local source. However, the lower particulate Hg concentrations measured in Detroit during March 1994 were likely due to the prevailing west-northwesterly winds during this period.

In April 1992 the prevailing winds were from the south-southeast suggesting transport of atmospheric Hg emitted from the industrial area of Detroit which includes coal combustion plants (e.g., coke ovens, iron and steel plants), municipal solid waste plants and sewage sludge incineration plants (Pirrone *et al.*, 1994). These source types account for a large fraction (>45%) of anthropogenic emissions of Hg in the United States (EPA, 1993).

Changes in the ambient concentrations of particulate and gas phase Hg with distance from the sampling sites are shown in Figure 2 for the forward trajectories which started in Detroit and traversed Lake St. Clair and Lake Erie on 9 April 1992 and on 7 March 1994 (Figure 1). The calculated Hg concentrations along the over-water trajectories were obtained using the Hg concentrations and particle size distributions measured at the sampling sites (Site-1 and Site-5) as input values in the HRD model.

TABLE I

Ambient concentrations of Hg measured at Site-1 and Site-5 in the fine and coarse fraction ( $\text{pg}/\text{m}^3$ ), in the gas phase ( $\text{ng}/\text{m}^3$ ), and on particles collected on each Micro-Orifice Impactor (MOI) stage ( $\text{pg}/\text{m}^3$ ). The mass median diameters ( $\mu\text{m}$ ) corresponding to a 50% collection efficiency of each MOI stage is given.

| Particle Size                   | Sampling Period  | Mass Median Diameter | Site - 1 | Site - 5 |
|---------------------------------|------------------|----------------------|----------|----------|
| Fine (<2.5 $\mu\text{m}$ )      | April 5-17, 1992 | 0.6                  | 290      | 252      |
| Coarse (>2.5 $\mu\text{m}$ )    | ibid.            | 5.0                  | 51.3     | 44.5     |
| Gas phase                       | ibid.            |                      | 48.8     | 3.4      |
| Fine (<2.5 $\mu\text{m}$ )      | March 1-18, 1994 | 0.6                  |          | 86       |
| Coarse (>2.5 $\mu\text{m}$ )    | ibid.            | 5.0                  |          | 12       |
| <b>Micro-orifice Impactor :</b> | ibid.            |                      |          |          |
| Stage-1                         |                  | 5.0                  |          | 6        |
| Stage-2                         |                  | 2.5                  |          | 6        |
| Stage-3                         |                  | 1.0                  |          | 14       |
| Stage-4                         |                  | 0.6                  |          | 33       |
| Stage-5                         |                  | 0.18                 |          | 30       |

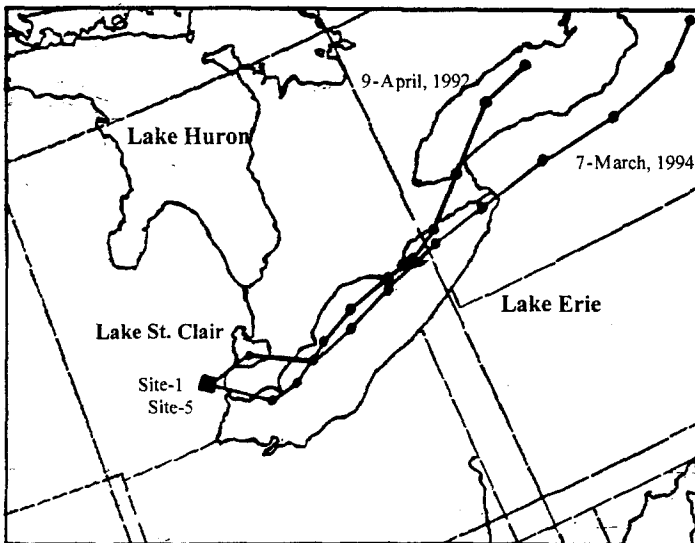


Fig. 1. Forward trajectories which started in Detroit and traversed Lake St. Clair and Lake Erie on 9 April 1992 and 7 March 1994.

The concentrations of particulate Hg in the fine and coarse fraction (Figure 2b) decreased by a factor of 12 and 18, respectively, when the air mass moved from the sampling site to the lakes. The decrease in ambient concentration was mainly due to atmospheric dispersion during transport with deposition accounting for only 10 and 22% of the overall decrease in the Hg concentrations in the fine and coarse fractions, respectively. The gas phase Hg concentrations decreased along the over-water trajectory by a factor of 15 compared to that measured at the sampling sites on 9 April 1992. Figure 1-c shows the decrease in the ambient concentration along the over-water trajectories that traversed Lakes St. Clair and Erie on 7 March 1994. The overall decrease in the ambient Hg concentrations are similar to those obtained for the fine and coarse fraction along the trajectory that traversed the lakes on 9 April 1992 (Figure 2b). However, the increase in the ambient concentration observed when the air mass was at a distance of 10-20 km from the sampling site (Figure 2c) was due to a decrease in the mixing layer depth (from 1080 m to 660 m) occurring over the lake during transport. The Hg levels over the lakes reached the minimum value of 10-20 pg/m<sup>3</sup> in the particle phase and 1.5-2 ng/m<sup>3</sup> in the gas phase at a distance > 40 km from the sampling sites. These Hg levels are similar to the regional background levels observed in the Great Lakes Region (Keeler *et al.*, 1994b; Burke *et al.*, 1994).

Figure 3 shows the change in dry deposition flux of Hg in the particle and gas phase with distance from the sampling sites when the air masses moved from Detroit over Lake Erie on 9 April 1992 and 7 March 1994. The exchange rates of gas phase Hg at the air-water interface, F-1 and F-5 (Figure 3a), were calculated using the measured concentrations at sampling sites Site-1 and Site-5, respectively, and a total Hg concentration in the dissolved aqueous phase of 0.5 ng/L (Gill and Bruland, 1990). Since measurements were not available to estimate the spatial and temporal variations of the total Hg in the dissolved aqueous phase, a Hg<sup>0</sup> concentration of 0.05 ng/L was assumed constant throughout the lakes area. This value was used to evaluate the exchange rate of gas phase Hg at the air-water interface. The exchange rate of gas phase Hg increased up to a factor of 2 during transport. Similar changes were found in other critical parameters effecting the gas exchange at the air-water interface such as the overall air-water transfer coefficient, K<sub>OL</sub>, which ranged from 1.1 to 4 cm/h, Henry's law constant, H, which ranged from 120 to 175 Pa/m<sup>3</sup>-mole, the temperature at the air-water interface, T<sub>a-w</sub>, which varied by 50% on average, the wind speed, W<sub>s</sub>, which ranged from 2.3 to 0.8 m/h and the mixing layer depth, Z<sub>mix</sub>, which ranged from 1080 to 620 m. The increase found in the K<sub>OL</sub> was associated with a decrease in the Z<sub>mix</sub> which led to an increase in the ambient concentrations along the over-water trajectory with higher exchange rates of the gas phase Hg. However, it is likely that the air-water exchange rate is actually more variable than that found in this study since changes in the Hg<sup>0</sup> concentration in the lakes were not measured. The dry deposition flux of fine and coarse fraction Hg decreased 10-fold along the over-water trajectory (Figure 3b). Similar variations were found in the deposition flux of particulate Hg obtained using the MOI data (Figure 3c). These changes ranged from 20-fold for Hg associated with 0.18 μm diameter particles (Stage-5 of the MOI) to 10-fold for Hg associated with 5 μm diameter particles (Stage-1 of the MOI). Variations in the dry deposition flux of particulate Hg are mainly due to changes in deposition velocity and ambient concentration along the trajectories traversing the lakes (Pirrone *et al.*, 1994a). Figure 4 shows the volatilization rates from the lakes and/or deposition to the lakes for each forward trajectory that traversed each lake during April 1992. The exchange rates of the gas phase Hg ranged from 0.8 ng/m<sup>2</sup>-h (deposition to the water surface) to -0.5 ng/m<sup>2</sup>-h (volatilization from the water surface) when Hg concentrations measured at Site-5 were used. When Hg concentrations measured at Site-1 are utilized, the HRD model estimates that only volatilization occurs from the

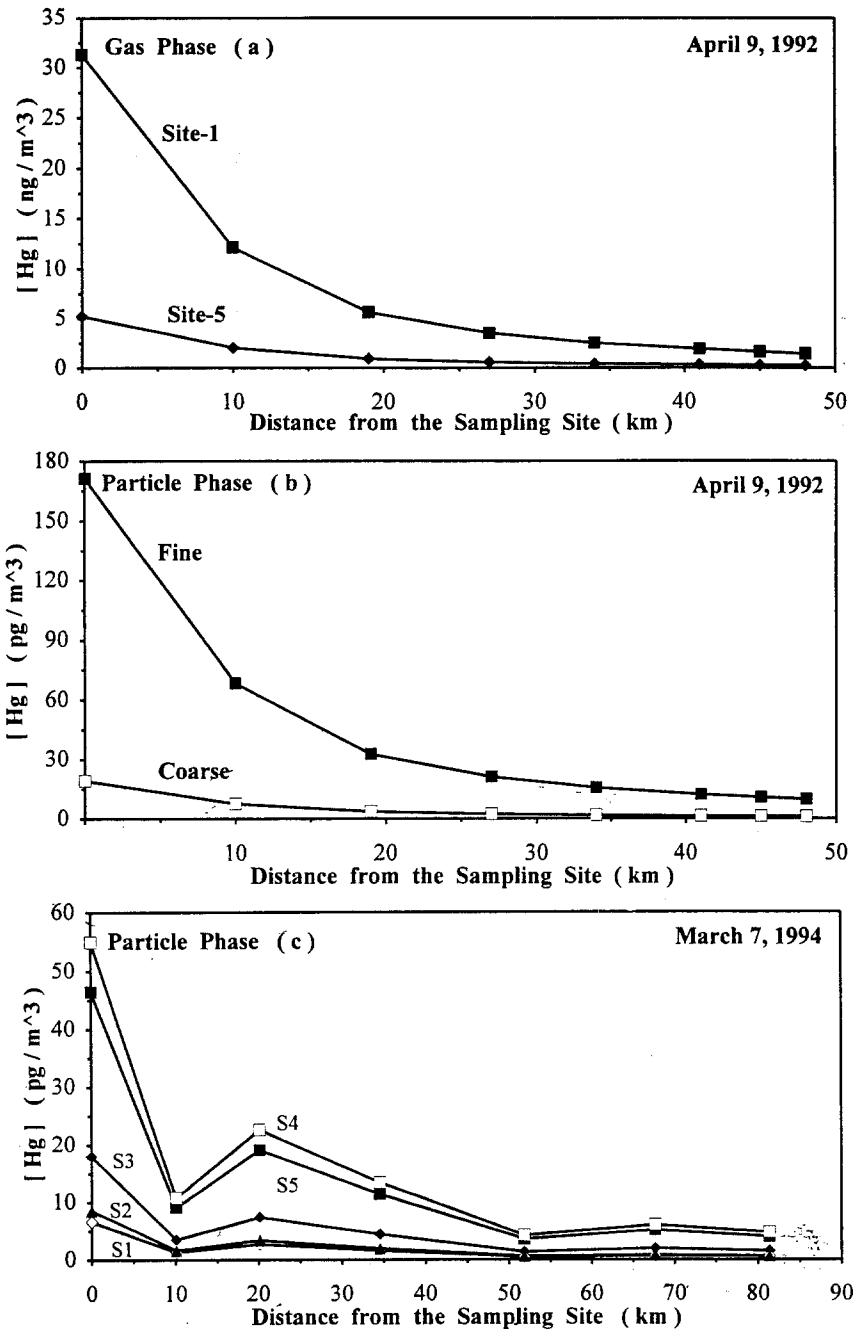


Fig. 2. Calculated Hg concentrations in the gas and particle phase along 24-hour forward trajectories traversing Lake St. Clair and Lake Erie on 9 April 1992 and on 7 March 1994 using the Hg concentrations measured at Site-1 and Site-5.

lakes. These findings suggest that based on the assumption of a constant Hg<sup>0</sup> concentration equal to 0.05 ng/L in the dissolved aqueous phase, the lakes are degassing Hg into the atmosphere.

The respective changes in dry deposition flux of particulate Hg in the fine and coarse fraction were 8-fold and 15-fold, respectively, when the concentrations measured at Site-1 were used, and 25-fold and 30-fold when the concentrations measured at Site-5 were used. The higher variations in the dry deposition flux of Hg over the lakes obtained using Site-5 data were likely due to larger variations in the activity of local emission sources located upwind of Site-5, since the effect of variations in the parameters controlling the transport of the air mass and particle deposition velocity over the water were similar for both sites.

The deposition flux of particulate Hg, obtained as averages of those obtained along each forward trajectory using the MOI data, shows 6-fold variations for the MOI-Stage1, 7-fold for the MOI-Stage2, 16-fold for the MOI-Stage3, 15-fold for the MOI-Stage4 and 7-fold for the MOI-Stage5 (Figure 5). The higher variations in the dry deposition flux of Hg associated with particles with a MMD=1 μm are in good agreement with results obtained by Pirrone *et al.* (1994a) for trace metals associated with 1.3 μm diameter particles.

Table II shows the estimated deposition fluxes of atmospheric Hg in the gas and particle phase using the measurements made at sites Site-1 and Site-5 in April 1992 and March 1994 as input values to the HRD model. Dry deposition fluxes of total particulate Hg (fine+coarse) obtained using the Hg concentrations measured in April 1992 at Site-1 were higher than those estimated using the Hg concentrations measured at Site-5. However, Lake Erie was more impacted by the urban plume generated in Detroit with dry deposition fluxes in the range of 112 to 125 pg/m<sup>2</sup>-h, followed by Lake St. Clair 67 to 109 pg/m<sup>2</sup>-h and Lake Huron 19 to 31 pg/m<sup>2</sup>-h. In March 1994 the dry deposition flux of particulate Hg (fine+coarse) was lower than that obtained during April 1992. However, Lake St. Clair is more impacted by the transport of air masses of urban origin with an average deposition flux of 57 pg/m<sup>2</sup>-h, followed by Lake Erie 24 pg/m<sup>2</sup>-h and Lake Huron 5.2 pg/m<sup>2</sup>-h. Hg volatilization rates from the lakes were predicted to range from -0.13 to -0.36 ng/m<sup>2</sup>-h for Lake Erie and -0.21 to -0.52 ng/m<sup>2</sup>-h for Lake Huron. Predicted Hg volatilization from Lake St. Clair was at a rate of -0.5 ng/m<sup>2</sup>-h when Hg concentrations measured at Site-5 were used, while deposition to the lake at a rate of 5.9 ng/m<sup>2</sup>-h was predicted when Hg concentrations measured at Site-1 were used. The volatilization rates obtained in this study were similar to those estimated by Vandal *et al.* (1991).

TABLE II

Dry deposition flux of Hg in the gas phase (ng/m<sup>2</sup>-h), in the particle phase (pg/m<sup>2</sup>-h) for each Micro-Orifice Impactor (MOI) stage, and in the fine and coarse fraction (ng/m<sup>2</sup>-h) calculated for Lakes St. Clair, Huron and Erie during April 1992 and March 1994. Negative values imply volatilization of the gas phase Hg from the lake.

|           | Lake St. Clair  |                 |                 | Lake Huron      |                 |                 | Lake Erie       |                 |                 |
|-----------|-----------------|-----------------|-----------------|-----------------|-----------------|-----------------|-----------------|-----------------|-----------------|
|           | Apr-1992 Site-1 | Apr-1992 Site-5 | Mar-1994 Site-5 | Apr-1992 Site-1 | Apr-1992 Site-5 | Mar-1994 Site-5 | Apr-1992 Site-1 | Apr-1992 Site-5 | Mar-1994 Site-5 |
| Gas phase | 5.9             | -0.5            |                 | -0.21           | -0.5            |                 | -0.36           | -0.13           |                 |
| MOI:      |                 |                 |                 |                 |                 |                 |                 |                 |                 |
| Stage-1   |                 |                 | 23.5            |                 |                 | 3.6             |                 |                 | 15.2            |
| Stage-2   |                 |                 | 5.8             |                 |                 | 1               |                 |                 | 5.1             |
| Stage-3   |                 |                 | 4.15            |                 |                 | 0.41            |                 |                 | 3.7             |
| Stage-4   |                 |                 | 2.3             |                 |                 | 0.17            |                 |                 | 1.5             |
| Stage-5   |                 |                 | 0.63            |                 |                 | 0.02            |                 |                 | 0.4             |
| Fine      | 25              | 15.3            | 7.1             | 6.8             | 4.2             | 0.52            | 20.6            | 18.5            | 5.1             |
| Coarse    | 84              | 51.4            | 50              | 24.2            | 15              | 4.7             | 104             | 93              | 19              |

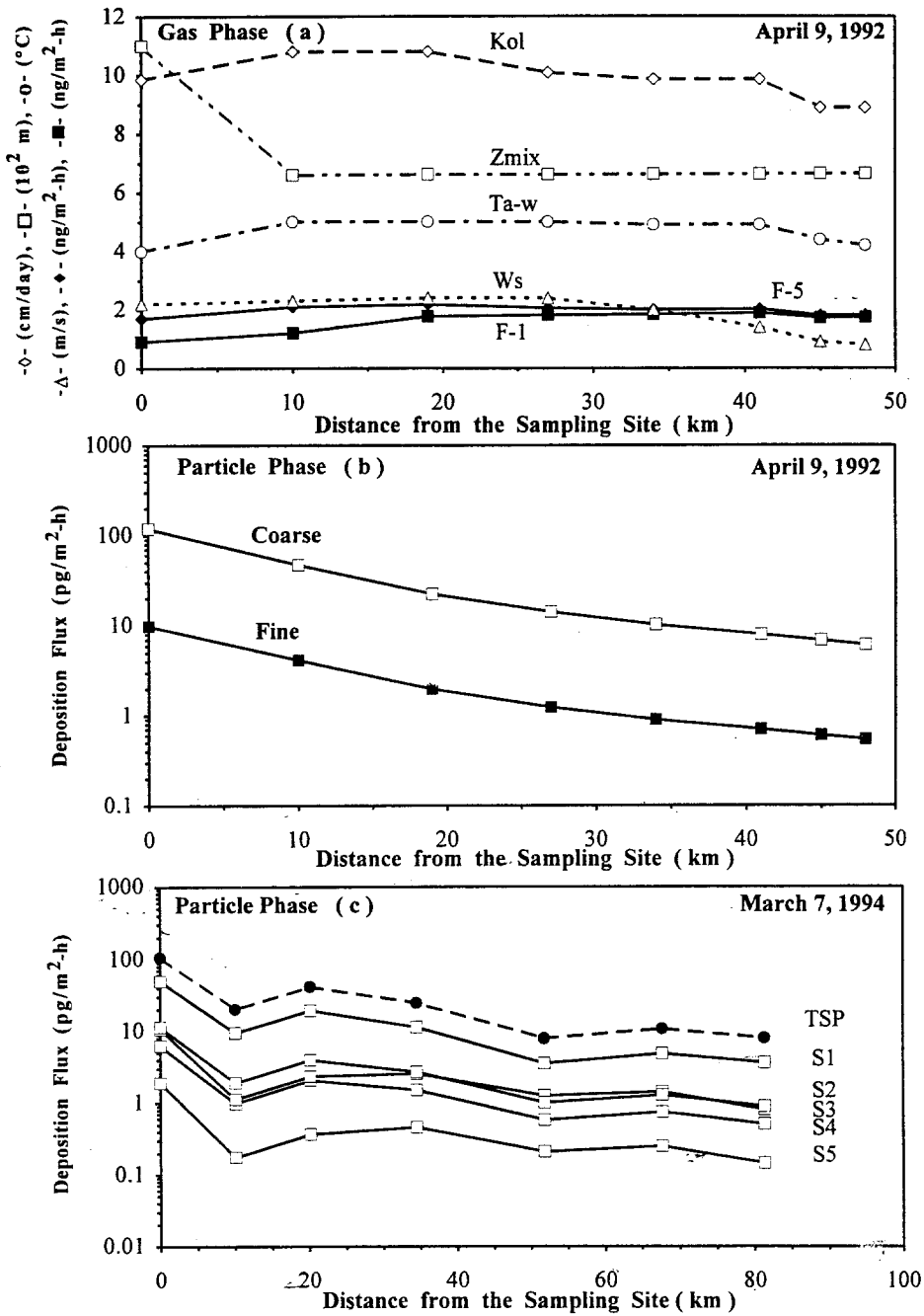


Fig. 3. Volatilization rates of the gas phase Hg and dry deposition fluxes of the particulate Hg along the forward trajectories that traversed Lake St. Clair and Lake Erie on 9 April 1992 and on 7 March 1994.



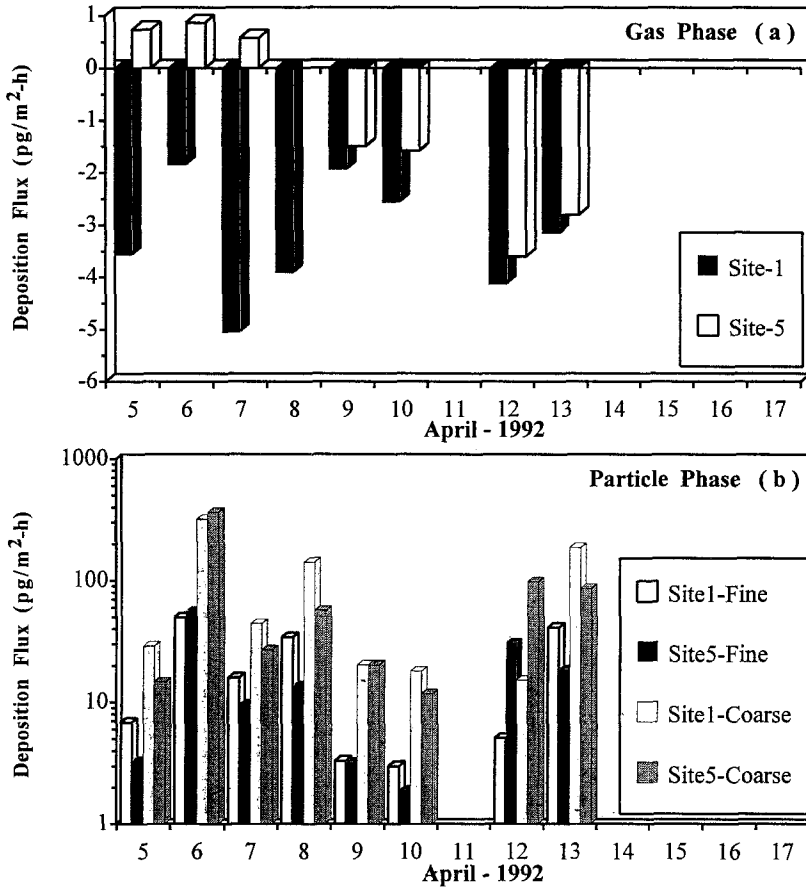


Fig. 4. Dry deposition fluxes of Hg in the gas and particle phase obtained as average of those calculated along each over-water trajectory traversing Lake St.Clair, Lake Huron and Lake Erie. Negative values imply volatilization of gas phase Hg from the lake.

**4. Summary and Conclusions**

Temporal and spatial variations of atmospheric concentrations and deposition fluxes of Hg associated with both the particle and gas phase over Lakes St. Clair, Erie and Huron were estimated using a hybrid receptor-deposition model (HRD). An evaluation of the variations in the particle deposition velocities and gas exchange rates of Hg during transport was performed. The overall variations in the deposition fluxes over the lakes ranged from 20-fold for 0.18 μm diameter particles to 10-fold for 5 μm diameter particles. The estimated exchange rates of the gas phase Hg were affected by the sampling site location and the fraction of Hg<sup>0</sup> in the total Hg dissolved in the aqueous phase. Therefore, gas phase Hg was predicted to volatilize from Lake Huron and Lake Erie at a rate of -0.21 to -0.5 ng/m<sup>2</sup>-h and -0.13 to -0.36 ng/m<sup>2</sup>-h, respectively. The model predicted that Lake St. Clair was degassing Hg at a rate of 0.5 ng/m<sup>2</sup>-h when Site-5 data were used, and when the higher ambient concentrations measured at Site-1 were utilized gas phase Hg was deposited to Lake St. Clair at a rate of 5.9 ng/m<sup>2</sup>-h. These results suggest that gas phase Hg can be deposited to the water surface (e.g., Lake St. Clair) when the air mass is near the emission sources, while it is re-emitted to the atmosphere at locations more distant.

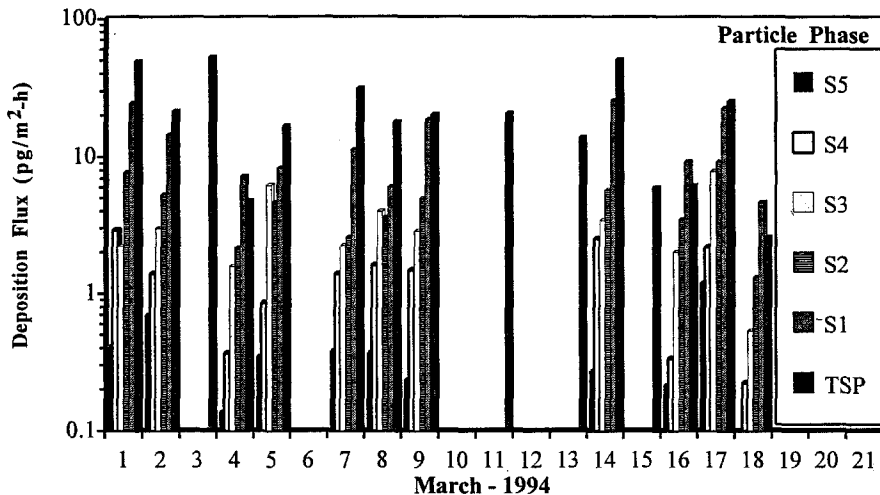


Fig. 5. Dry deposition fluxes of particulate Hg for each particle size range of the MOI stages, obtained as average of those calculated along each over-water trajectory that traversed Lakes St. Clair, Huron and Erie during March 1994.

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