

# Metals Associated with Suspended Sediments in Lakes Erie and Ontario, 2000–2002

Chris Marvin · Murray Charlton · Jacqui Milne · Lina Thiessen ·  
Joanne Schachtschneider · Gino Sardella · Ed Sverko

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**Abstract** Sediment traps were deployed in the three major basins of Lake Erie, and the central (Mississauga) basin of Lake Ontario, and refurbished seasonally over the period 2000–2002. In Lake Ontario, sediment down-flux rates and corresponding contaminant down-flux rates were highest in winter during periods of unstratified thermal conditions, and generally increased with depth due to the influence of resuspended bottom sediments during all sampling periods. Lake Ontario suspended sediments exhibited the highest concentrations of metals; concentrations of mercury and lead frequently exceeded guideline values for bottom sediments. Contaminant levels in Lake Ontario suspended sediments were similar to concentrations in bottom sediments in the same area. There was a spatial trend toward higher suspended sediment metals concentrations from the eastern basin to the western basin of Lake Erie, which is similar to the trend in bottom sediment contamination. In the eastern basin of Lake Erie, which is the deepest area of the lake, there was no trend in down-flux rate with depth in 2001; however, down-flux rates increased with depth in 2002. Suspended sediments in the western basin of Lake Erie were determined to be

largely resuspended bottom sediments; all western basin samples collected in the study exceeded the guideline value for mercury (0.486 µg/g).

**Keywords** Lake Ontario · Lake Erie · Suspended sediments · Metals · Mercury · Lead

## 1 Introduction

Suspended sediments are an important vector for contaminant transport through the water column in the Great Lakes. Sediment traps are simple yet effective apparatus for time-integrated sampling of suspended particulate. As a result, sediment traps can provide estimates of contaminant down-fluxes that are useful in investigating contaminant cycling, and for estimation of loadings and inventories. Previous studies have used sediment traps for studies of contaminants, nutrients, and sedimentation rates in the Great Lakes (Baker, Eisenreich, & Eadie, 1991; Chambers & Eadie, 1981; Charlton, 1983; Eadie, Chambers, Gardner, & Bell, 1984; Marvin, Sverko, Charlton, Thiessen, & Painter, 2004; Oliver & Charlton, 1984; Oliver, Charlton, & Durham, 1989).

The findings reported in this paper are complementary to a recent report on persistent organic pollutants (POPs) associated with suspended sediments in Lakes Erie and Ontario over the period 1997–2000 (Marvin et al., 2004). In this previous study, relatively higher concentrations of PCBs were

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C. Marvin (✉) · M. Charlton · J. Milne · L. Thiessen ·  
J. Schachtschneider · G. Sardella · E. Sverko  
Environment Canada, 867 Lakeshore Road,  
Burlington, Ontario, Canada L7R 4A6  
e-mail: Chris.Marvin@ec.gc.ca

**Table 1** Suspended sediment collection intervals, depths of sediment trap deployment, suspended sediment down-flux rates ( $\text{g m}^{-2} \text{day}^{-1}$ ) and available data for percent loss on ignition (bracketed values) for Lakes Erie and Ontario over the period 2001–2002

Station	Time interval	Depth	Down-flux ( $\text{g m}^{-2} \text{day}^{-1}$ )
Lake Erie 23	2 May–6 Jun 2001 (U/S)	20 m	0.66 (32)
Lake Erie 23	2 May–6 Jun 2001	40 m	0.64 (30)
Lake Erie 23	2 May–6 Jun 2001	50 m	0.69 (29)
Lake Erie 23	2 May–6 Jun 2001	59 m	0.73 (28)
Lake Erie 23	6 Jun–5 Jul 2001 (S)	20 m	1.04 (12)
Lake Erie 23	6 Jun–5 Jul 2001	40 m	0.88 (13)
Lake Erie 23	6 Jun–5 Jul 2001	50 m	0.88 (13)
Lake Erie 23	6 Jun–5 Jul 2001	59 m	0.87 (12)
Lake Erie 23	5 Jul–16 Aug 2001 (S)	20 m	0.66 (28)
Lake Erie 23	5 Jul–16 Aug 2001	40 m	0.34 (27)
Lake Erie 23	5 Jul–16 Aug 2001	50 m	0.41 (26)
Lake Erie 23	5 Jul–16 Aug 2001	59 m	0.41 (25)
Lake Erie 23	27 Sept–3 Nov 2001 (U)	21 m	40.0 (7.5)
Lake Erie 23	27 Sept–3 Nov 2001	41 m	27.7 (7.2)
Lake Erie 23	27 Sept–3 Nov 2001	51 m	35.3 (7.3)
Lake Erie 23	27 Sept–3 Nov 2001	60 m	45.0 (7.5)
Lake Erie 84	18 Apr–25 May 2001 (U/S)	18 m	2.44 (25)
Lake Erie 84	18 Apr–25 May 2001	21 m	2.83 (22)
Lake Erie 84	25 May–20 Jun 2001 (S)	18 m	0.44 (30)
Lake Erie 84	25 May–20 Jun 2001	21 m	1.62 (21)
Lake Erie 84	25 Jul–30 Aug 2001 (S)	19 m	1.34 (32)
Lake Erie 84	25 Jul–30 Aug 2001	22 m	4.14 (19)
Lake Erie 84	27 Sept–Nov 3 2001 (U)	18 m	150 (9.7)
Lake Erie 84	27 Sept–Nov 3 2001	21 m	210 (9.3)
Lake Erie 357	18 Apr–24 May 2001 (U)	9 m	72 (10)
Lake Erie 357	24 May–20 Jun 2001 (U)	9 m	140 (8.8)
Lake Erie 357	20 Jun–25 Jul 2001 (U)	9 m	31 (11)
Lake Erie 357	25 Jul–29 Aug 2001 (U)	9 m	110 (9.3)
Lake Erie 357	29 Aug–26 Sept 2001 (U)	9 m	230 (9.2)
Lake Ontario 403	1 Nov 2000–31 May 2001 (U)	20 m	0.78 (23)
Lake Ontario 403	1 Nov 2000–31 May 2001	60 m	0.87 (22)
Lake Ontario 403	1 Nov 2000–31 May 2001	100 m	0.97 (20)
Lake Ontario 403	1 Nov 2000–31 May 2001	140 m	1.13 (20)
Lake Ontario 403	1 Nov 2000–31 May 2001	165 m	1.92 (19)
Lake Ontario 403	1 Nov 2000–31 May 2001	175 m	2.78 (18)
Lake Ontario 403	31 May–24 Oct 2001 (U/S)	20 m	0.23 (41)
Lake Ontario 403	31 May–24 Oct 2001	60 m	0.24 (39)
Lake Ontario 403	31 May–24 Oct 2001	100 m	0.32 (34)
Lake Ontario 403	31 May–24 Oct 2001	140 m	0.30 (31)
Lake Ontario 403	31 May–24 Oct 2001	166 m	0.57 (26)
Lake Ontario 403	31 May–24 Oct 2001	174 m	0.97 (22)
Lake Erie 23	3 Nov 2001–10 Apr 2002 (U)	21 m	2.04 (7.1)
Lake Erie 23	3 Nov 2001–10 Apr 2002	41 m	2.73 (7.8)
Lake Erie 23	3 Nov 2001–10 Apr 2002	51 m	3.17 (8.1)
Lake Erie 23	3 Nov 2001–10 Apr 2002	60 m	3.37 (7.6)
Lake Erie 23	10 Apr–12 Jun 2002 (U/S)	20 m	2.41 (10)
Lake Erie 23	10 Apr–12 Jun 2002	40 m	2.69 (10)
Lake Erie 23	10 Apr–12 Jun 2002	50 m	2.90 (10)
Lake Erie 23	10 Apr–12 Jun 2002	60 m	3.35 (10)
Lake Erie 23	12 Jun–11 Jul 2002 (S)	20 m	1.86 (11)
Lake Erie 23	12 Jun–11 Jul 2002	40 m	1.46 (24)

**Table I** (continued)

Station	Time interval	Depth	Down-flux (g m <sup>-2</sup> day <sup>-1</sup> )
Lake Erie 23	12 Jun–11 Jul 2002	50 m	0.98 (22)
Lake Erie 23	12 Jun–11 Jul 2002	59 m	1.27 (14)
Lake Erie 23	11 Jul–11 Sept 2002 (S)	20 m	0.87 (16)
Lake Erie 23	11 Jul–11 Sept 2002	40 m	1.09 (15)
Lake Erie 23	11 Jul–11 Sept 2002	50 m	1.17 (14)
Lake Erie 23	11 Jul–11 Sept 2002	59 m	1.24 (14)
Lake Erie 23	11 Sept–31 Oct 2002 (S/U)	20 m	3.56 (11)
Lake Erie 23	11 Sept–31 Oct 2002	40 m	4.99 (6.8)
Lake Erie 23	11 Sept–31 Oct 2002	50 m	6.42 (7.1)
Lake Erie 23	11 Sept–31 Oct 2002	59 m	6.39 (7.1)
Lake Erie 84	24 Apr–5 Jun 2002 (U/S)	19 m	11.5 (14)
Lake Erie 84	24 Apr–5 Jun 2002	22 m	14.2 (14)
Lake Erie 84	11 Jul–21 Aug 2002 (S)	20 m	1.42 (21)
Lake Erie 84	11 Jul–21 Aug 2002	23 m	2.94 (16)
Lake Erie 84	21 Aug–17 Sept 2002 (S)	20 m	NA (47)
Lake Erie 84	21 Aug–17 Sept 2002	23 m	6.28 (18)
Lake Erie 84	17 Sept–23 Oct 2002 (S/U)	19 m	26.2 (11)
Lake Erie 84	17 Sept–23 Oct 2002	22 m	NA (10)
Lake Erie 357	23 Apr–5 Jun 2002 (U)	9 m	165 (12)
Lake Erie 357	5 Jun–17 Jul 2002 (U)	9 m	53 (13)
Lake Erie 357	17 Jul–12 Aug 2002 (U)	10 m	
Lake Erie 357	12 Aug–17 Sept 2002 (U)	9 m	
Lake Ontario 403	24 Oct 2001–5 Apr 2002 (U)	20 m	1.25 (21)
Lake Ontario 403	24 Oct 2001–5 Apr 2002	60 m	1.62 (20)
Lake Ontario 403	24 Oct 2001–5 Apr 2002	100 m	1.86 (20)
Lake Ontario 403	24 Oct 2001–5 Apr 2002	140 m	2.43 (20)
Lake Ontario 403	24 Oct 2001–5 Apr 2002	166 m	3.71 (20)
Lake Ontario 403	24 Oct 2001–5 Apr 2002	174 m	4.37 (19)
Lake Ontario 403	5 Apr–5 Nov 2002 (S/U)	20 m	0.76 (21)
Lake Ontario 403	5 Apr–5 Nov 2002	60 m	0.36 (29)
Lake Ontario 403	5 Apr–5 Nov 2002	100 m	0.37 (29)
Lake Ontario 403	5 Apr–5 Nov 2002	140 m	0.49 (25)
Lake Ontario 403	5 Apr–5 Nov 2002	166 m	2.37 (16)
Lake Ontario 403	5 Apr–5 Nov 2002	174 m	1.03 (22)

NA Denotes data not available. Thermal conditions over the periods of sediment trap deployment were classified as: *S* Stratified, *U* unstratified, *U/S* unstratified at deployment–stratified at retrieval, *S/U* stratified at deployment–unstratified at retrieval.

detected in suspended sediments, compared to bottom sediments, but also showed that current levels of contaminants in Lake Ontario suspended sediments represented 70–90% reductions over concentrations reported for similar samples collected in the mid-1980s.

In this paper, we report the results of an investigation of metals associated with suspended sediments collected at a range of sites encompassing all three major basins of Lake Erie, and in the Mississauga (central) basin of Lake Ontario over the period 2000–

2002. The previous study on POPs included data from only the western basin of Lake Erie; the current study provides greater spatial scope through addition of data from the central and eastern basins; physical properties of suspended sediments from the central and eastern basins of Lake Erie provide a more valid comparison with Lake Ontario suspended sediments, compared to suspended sediments from the western basin of Lake Erie which are heavily influenced by resuspended bottom sediments. Concentrations and down-fluxes were calculated for a suite of metals

including lead, mercury, cadmium, copper, nickel and zinc. Metal concentrations in suspended sediments were also assessed using the Canadian Sediment Quality Guideline values.

## 2 Materials and Methods

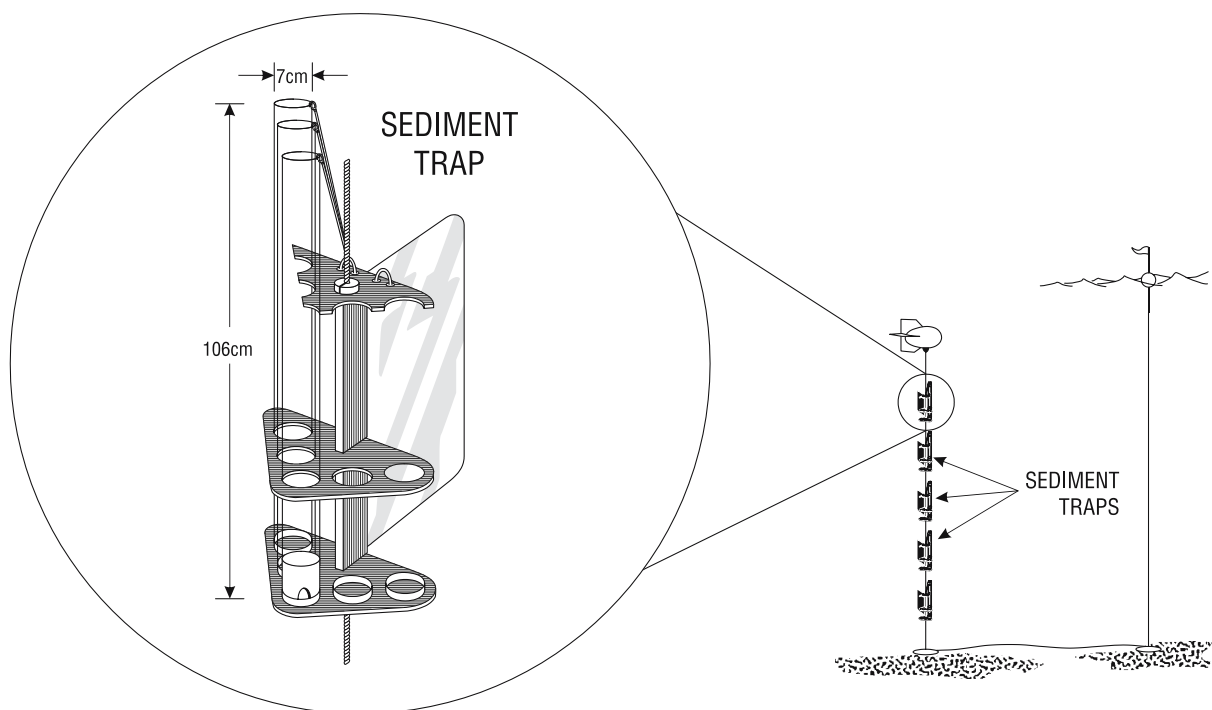
### 2.1 Sediment traps

Suspended sediments were collected over time and depth intervals (Table 1) at individual stations in the western basin (Station 357, 41°49'13" N 82°59'59"), central basin (Station 84, 41°56'30" N 81°39'33"), and eastern basin (Station 23, 42°30'13" N 79°52'29") of Lake Erie, and the Mississauga basin (Station 403, central basin, 43°35'50" N 78°13'48" W) of Lake Ontario over the period 2000–2002. Sediment trap assemblies were identical to those described in detail in Marvin et al. (2004), and consisted of five lengths of core tubing positioned in racks (Figure 1). Contents of cups representing suspended sediments at discrete depths were combined, refrigerated, and allowed to settle overnight prior to a second decanting; no preservatives were added. Samples were frozen,

freeze-dried and weighed. Loss on ignition (LOI) was determined by combustion in a muffle furnace at 500°C for 2 h.

### 2.2 Metals analyses

One-half gram of homogeneous freeze-dried sediment was weighed into a Teflon (TFM) vessel and 10 ml of nitric acid, 2 ml of hydrochloric acid, and 1 ml of hydrogen peroxide were added. The vessel was assembled, sealed, and fitted onto a microwave rotor, which was placed in a high-pressure microwave oven (MLS 1200 Mega, Milestone, Germany). The sample was digested in the microwave field with the temperature maintained at 200°C for 15 min. The vessel was removed from the microwave oven and the cooled digest was brought to 100 ml volume with deionized water. The high temperature and the strong oxidizing conditions of the digestion ensured destruction of the organic and inorganic components of the substrate. With the exception of siliceous bound elements, the strong acid microwave-assisted digestion provided effective extraction of analytes of interest. The resultant solution was analyzed for Al, Ba, Ca, Cr, Fe, Mn, P, K, Na, Sr and V by ICP-OES



**Figure 1** Sediment trap design and mooring assembly for depth integrated sampling of suspended sediments in Lake Ontario.

(IRIS, Thermo Jarrel Ash, Franklin, MA). As, Be, Bi, Cd, Cu, Co, Ga, La, Pb, Li, Mo, Ni, Rb, Sb, Tl, U and Zn were analyzed by ICP-MS (PQ2, VG Elemental, UK).

Total mercury was analyzed from the digest by Cold Vapour Atomic Absorption Spectrometry (CVAAS). Mercury (II) was reduced to elemental mercury in an automated continuous flow system by the action of stannous chloride. The mercury was sparged from the solution with a stream of purified air and passed through an absorption cell of a mercury analyzer (LDC/Milton-Roy, Riviera Beach, FL).

### 3 Results and Discussion

#### 3.1 Sediment trap deployments and suspended sediment fluxes

Suspended sediment samples were subjected to physical measurements including loss on ignition (LOI), and analyzed for metals. We recently published the results of a study that investigated the association of persistent organic pollutants (POPs), including PCBs and organochlorine pesticides, with suspended sediments from the western basin of Lake Erie and the central basin of Lake Ontario over the period 1997–2000 (Marvin et al., 2004). As with the previous study, we were unable to correlate percent LOI with TOC in suspended and bottom sediments from Lakes Erie and Ontario. Some studies have shown good correlation between LOI and TOC for some soils and sediments (Ball, 1964; Dean, 1974); however, clay content and carbonate carbon can greatly influence LOI, resulting in a significant overestimation of TOC (Dean, 1974).

Sediment trap moorings in the central and eastern basins of Lake Erie, and in Lake Ontario, consisted of vertical arrays to enable sampling of suspended sediments at depth intervals; the Lake Erie western basin (station 357) mooring was deployed at only one depth (roughly 9 m), due to the shallowness of water in this area. A listing of deployment dates for the Lake Ontario and Lake Erie sediment trap moorings is shown in Table I. For each period of deployment, dry suspended sediment down-flux rates and corresponding percent LOI are also shown. Table I also lists limnological characteristics at each site in terms of stratified or unstratified conditions; these

designations were based on temperature and oxygen depth profiles. A transition in conditions from stratified–unstratified or from unstratified–stratified occurred during some periods of sediment trap deployment; these conditions are designated as ‘S/U’ or ‘U/S’, respectively, in Table I.

#### 3.2 Characterization of suspended sediments in Lake Ontario

Station 403 is located in the Mississauga (central) basin of Lake Ontario. Bottom sediments within the three major Lake Ontario basins are comprised of fine-grained material classified as mixtures of glacio-lacustrine clay, sand, silt and mud (Thomas, Kemp, & Lewis, 1972). Water circulation in Lake Ontario primarily occurs in a counter-clockwise direction, with a small clockwise gyre in the northwestern part of the lake (Beletsky, Saylor, & Schwab, 1999). This circulation pattern results in distribution of suspended sediment across the entire lake area; therefore, physical and chemical characteristics of bottom sediments at station 403 are assumed to be generally representative of sediments lake-wide (Marvin et al., 2002c; Mudroch, 1993).

Estimated dry suspended sediment down-fluxes in Lake Ontario over the period 2000–2002 ranged from 0.23 to 4.4 g m<sup>-2</sup> day<sup>-1</sup> (Table I). These values were quite similar to those reported at this site for the period 1997–2000 (Marvin et al., 2004), and those for stations offshore of the mouth of the Niagara River in 1980 (0.8–4.2 g m<sup>-2</sup> day<sup>-1</sup>, Charlton, 1983). The range of LOI values (range of percent LOI of 16–48%) was also similar to that of the 1997–2000 (range of percent LOI from 18 to 41%) and 1980 studies (16–33%). As with the previous studies, generally greater amounts of sediment were collected with increasing depth (Table I). Greater accumulation of suspended sediments with increased depth in Lake Ontario has been attributed to resuspended bottom sediments (Charlton, 1975; Charlton, 1983; Gasith, 1975; Marvin et al., 2004; Oliver et al., 1989). In addition, Lake Ontario exhibits a turbid layer in the water column directly above the lake bottom called the nepheloid layer. This turbid water mass feature is characterized by high suspended solids and light attenuation, and is present at depths greater than 60 m (Sandilands & Mudroch, 1983). Therefore, the combined contributions of the nepheloid layer and

resuspended bottom sediments result in higher suspended sediment fluxes closer to the lake bottom. This general trend of increased particle mass flux *versus* depth can exhibit seasonal variations, with highest fluxes occurring during periods of thermal stratification (Chambers & Eadie, 1981; Eadie et al., 1984).

The contribution of resuspended bottom sediments with low organic content to material accumulated in the sediment traps at greater depths was also evidenced by a trend toward decreased percent LOI with increased sampling depth (Table I). Percent LOI throughout the entire study period (2000–2002) for Lake Ontario suspended sediments ranged from 18 to 41%; typical total organic carbon (TOC) values in Lake Ontario surficial bottom sediments are on the order of 3%. Overall, the influence of resuspended bottom sediments is expected to result in higher down-flux rates and correspondingly lower percent LOI values. This relationship between suspended sediment down-flux rate and percent LOI for all of the 2000–2002 Lake Erie and Lake Ontario samples is shown in Figure 2. As expected, there is an apparent trend toward higher percent LOI with decreasing down-flux.

Lake Ontario moorings were deployed during the winter months in both years of the study (November 2000–May 2001 and October 2001–April 2002, Table I), which enabled a comparison with data from station 403 for the period 1982–1986 reported by Oliver et al. (1989) for suspended sediments collected over similar winter time intervals. As expected, accumulation rates during the 2000–2001 and 2001–2002 winter intervals increased over with depth over the range of 20–165 m (Table I). The range of suspended sediment down-flux rates at station 403 during the winter intervals ( $0.78\text{--}4.4\text{ g m}^{-2}\text{ day}^{-1}$ ) was similar to that reported by Oliver et al. for the same depth range during winter ( $1.2\text{--}3.6\text{ g m}^{-2}\text{ d}^{-1}$ ). Oliver et al. also observed higher down-flux rates in winter samples, compared to summer samples, which was attributed to increased lake mixing and resuspension of bottom sediments during unstratified conditions. In addition, storms can influence physical processes at greater depths during periods of isothermal conditions, resulting in an increased contribution of resuspended bottom sediments to material accumulated in sediment traps over the entire range of depths sampled. Intense winter storms in southern

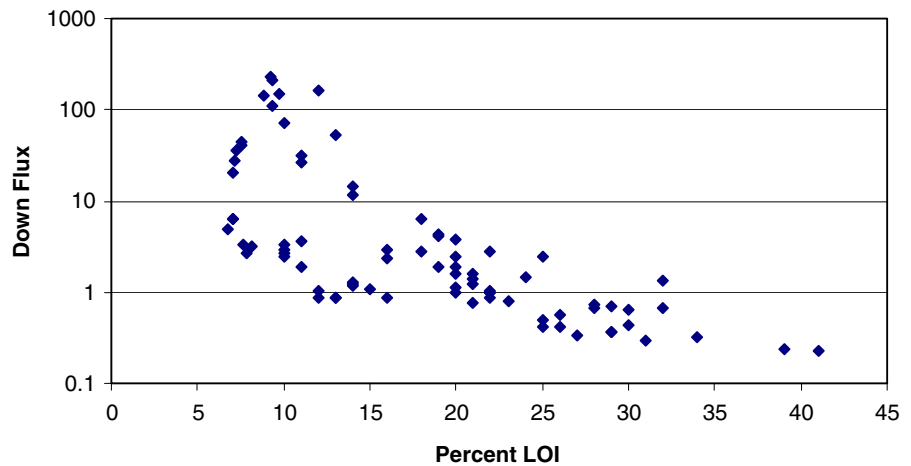
Lake Michigan in 1998 resulted in large-scale resuspension of bottom sediments and correspondingly large fluxes of contaminants (Bogdan, Budd, Eadie, & Hornbuckle, 2002); the March 1998 storm was estimated to have resulted in roughly 400 kg of PCBs being resuspended into the water column.

### 3.3 Characterization of suspended sediments in Lake Erie

Sediment traps were deployed in all three Lake Erie basins. Unlike Lake Ontario, where the three major basins exhibit relatively consistent patterns of bottom sediment contamination, there is a trend in the Lake Erie basins toward greater contamination from east-to-west (Painter et al., 2001). The western basin and south-central area of the central basin exhibit the highest degrees of sediment contamination as a result of tributary discharges and proximity to highly industrialized/urbanized areas.

Suspended sediment down-flux rates in the western basin of Lake Erie (station 357) in 2001–2002 ranged from 31 to  $230\text{ g m}^{-2}\text{ day}^{-1}$ , compared with a range of  $14\text{--}330\text{ g m}^{-2}\text{ day}^{-1}$  in the 1997–2000 study (Marvin et al., 2004). The down-flux rates in the western basin were generally much higher compared to down-flux rates at stations in the central and eastern basins of Lake Erie, and for the Lake Ontario station, during periods when the water columns were stratified (Table I). Primary inflow to Lake Erie is via the Detroit River, with a flow averaging roughly  $6,000\text{ m}^3\text{ s}^{-1}$ , which represents approximately 80% of the Lake Erie total. The western basin of Lake Erie is relatively shallow (mean depth of 9 m), which combined with tributary inputs results in high resuspension rates of bottom sediments. As a result, material accumulated in sediment traps in the western basin of Lake Erie have relatively low LOI values (Table I). Down-flux rates during the spring and fall collection periods in Lake Erie were generally greater than during the summer periods. Down-flux rates in the western basin during the April–June and August–September periods ranged from 72 to  $230\text{ g m}^{-2}\text{ day}^{-1}$ , which was substantially higher compared to the June–July collection periods (range of  $31\text{--}53\text{ g m}^{-2}\text{ day}^{-1}$ ). The higher spring/fall down-flux rates may be the result of increased tributary discharges as a result of greater rates of precipitation and runoff during these periods, compared to summer.

**Figure 2** Relationship between Lake Erie and Lake Ontario suspended sediment down-flux rate and percent loss on ignition (LOI).



Down-flux rates in the central and eastern basins of Lake Erie during periods when the water column was stratified were lower, compared to the western basin during the same time periods (Table I). At the central basin station (station 84), down-flux rates were consistently higher at the deeper interval (21–23 m), compared to the shallower interval (18–20 m). Correspondingly, percent LOI was typically lower at the deeper interval; both of these observations indicate the greater influence of resuspended bottom sediments at greater depth. Deployment periods characterized by a transition between stratified and unstratified conditions resulted in higher down-flux rates compared to periods of stratification. The September–November 2001 deployment corresponding to an unstratified period resulted in very high down-flux rates (150–210 g m<sup>-2</sup> day<sup>-1</sup>). Corresponding percent LOI values for these samples (9.3 and 9.7%) were relatively low.

Annual trends in sediment down-flux rates in the eastern basin of Lake Erie (station 23) were variable for the two years of the study. In 2001, there was no apparent trend with depth in down-flux rate or percent LOI. Dramatically higher down-flux rates, and correspondingly lower percent LOI values, were observed during the September–November 2001 period that corresponded to unstratified conditions (Table I). Relatively higher suspended sediment down-flux rates were also observed during the period of transition from stratified to unstratified conditions during the September–October 2002 deployment period. In contrast to the data from 2001, a trend toward higher down-flux rates with increasing depth was apparent in 2002 in the eastern basin of Lake Erie. Interestingly,

this trend in down-flux rate was not accompanied by a corresponding trend toward decreasing percent LOI values.

### 3.4 Suspended sediment metals down-fluxes

Suspended sediment annual mean contaminant concentrations and daily contaminant down-flux rates for Lakes Erie and Ontario over the period 2000–2002 are shown in Table II; daily contaminant down-fluxes were calculated using the mean daily sediment down-flux rates from Table I. Suspended sediment concentrations, represented by both annual means and monthly/seasonal values, were highest in the western basin of Lake Erie (station 357) and at the Lake Ontario station, compared to the central and eastern basins of Lake Erie. However, estimates of metals down-flux rates were orders of magnitude greater in the western basin of Lake Erie, compared to Lake Ontario (Table II). In the western basin of Lake Erie, the shallow depth (9 m), bathymetry, and prolonged periods of high-suspended sediment loadings contribute to a prolonged contaminant down flux of material primarily comprised of resuspended bottom sediments. The predominant influence of these resuspended bottom sediments resulted in artificially high down-flux rates, and an absence of any seasonal trends in down-flux rates in the western basin of Lake Erie.

Metals concentrations in suspended sediments in the central (station 84) and eastern (station 23) basins of Lake Erie showed little inter-annual variation, and were relatively low compared to the western basin (Table II). The better suspended sediment quality in

the eastern and central basins of Lake Erie was reflected in a similar trend in bottom sediment contamination (Painter et al., 2001). The only anomalies were unusually high concentrations of zinc in the eastern basin samples in summer 2002, and in the central basin samples in fall 2002; these concentrations exceeded the PEL (Table II).

There were apparent seasonal trends in metals down-flux rates in the central and eastern basins of Lake Erie. In general, metals down-flux rates were higher in the winter, spring, and fall sampling periods that are typical of periods of unstratified conditions, which is a direct result of the similar trend in the suspended sediment down-flux rates. This seasonal trend was more definitive in 2001 in Lake Erie, compared to 2002. In the eastern basin of Lake Erie in 2002, down-flux rates for the winter sampling period (November 2001–April 2002) were relatively low. There is increased lake-mixing and resuspension of bottom sediments during the period of unstratified conditions resulting in higher suspended sediment down-flux rates, and therefore higher metals down-flux rates. Additional factors resulting in increased metals down-flux rates during periods of unstratified conditions in Lake Erie could have included storms that influence physical processes at greater depths during periods of unstratified conditions, resulting in an increased contribution of resuspended bottom sediments to material accumulated in sediment traps over the entire range of depths sampled, and increased tributary discharges as a result of greater rates of precipitation and runoff during the spring and fall periods, compared to summer.

There was significant seasonal variation in suspended sediment (Table I) and metals concentrations and down-fluxes (Table II) at the Lake Ontario station. In both years of the study, the winter sampling periods were characterized by substantially higher metals concentrations and higher metals down-fluxes. As with the central and eastern basins of Lake Erie, this seasonal trend was attributed to unstratified conditions resulting in increased suspended sediment down-flux rates due to an increased contribution of resuspended bottom sediments. Oliver et al. (1989) also observed higher down flux rates in the winter samples at station 403 in Lake Ontario, compared to summer samples, which was also attributed to increased lake-mixing and resuspension during the period of unstratified conditions. In the winter 2001

samples, samples corresponding to depths greater than 60 m exceeded the PEL for mercury (0.486  $\mu\text{g/g}$ ). In the winter 2002 samples, all samples exceeded the PEL for mercury, lead (91.3  $\mu\text{g/g}$ ), and nickel (75  $\mu\text{g/g}$ ); all samples below a depth of 20 m exceeded the PEL for zinc (314.8  $\mu\text{g/g}$ ). Metals concentrations and corresponding down-flux rates during periods of primarily stratified conditions (spring/summer/fall) were generally substantially lower, particularly for mercury and lead, compared to the winter sampling periods.

There was also a general trend toward increasing metals down-flux rates with increasing depth for both sampling periods for both years of the study in Lake Ontario. These observations are directly related to the increased suspended sediment down-flux rates attributed to increased trapping of resuspended surficial bottom sediment during unstratified periods as explained previously. There was also a general trend toward increasing metal concentrations with increasing depth in the winter 2001 samples. Concentrations in the winter 2002 samples were more consistent with depth. As with the winter samples, there was a trend toward increasing concentrations with increasing depth for most metals in the spring/summer/fall samples in 2001, while concentrations in the spring/summer/fall samples were more consistent with depth. However, there was generally a far greater range of concentrations over the 20- to 175-m depth intervals, compared to the winter samples. In the case of mercury, concentrations in the spring/summer/fall samples at the 20-m depth interval were approximately three-fold lower than concentrations in the 175-m samples. Presumably, the relatively high concentrations at the greatest depth intervals reflect the influence of resuspended bottom sediments. Bottom sediments are not resuspended into the water column during periods of stratified conditions to the same degree as during unstratified conditions, but are still a significant influence on concentrations of suspended sediments.

The spatial distribution in suspended sediment metals contamination in Lake Erie, where concentrations generally increased from east to west (Table II), was also reflected in the relative degree of bottom sediment contamination observed during a lake-wide survey conducted in 1997 (Painter et al., 2001). Similarly, concentrations of a variety of contaminant classes in surficial bottom sediments in



**Table II** Suspended sediment metals concentrations ( $\mu\text{g/g}$ ) and daily down-flux rates (bracketed values expressed in  $\mu\text{g m}^{-2} \text{day}^{-1}$ ) for Lakes Erie and Ontario for 2001–2002

Station/date <sup>a</sup>	Depth <sup>b</sup> (m)	Mercury	Lead	Cadmium	Copper	Nickel	Zinc
<sup>c</sup> CSQG PEL		0.486	91.3	3.53	196.6	75	314.8
Erie 23–Jun 2001	20 + 40 + 50 + 59	0.118 (0.080)	21.1 (14.3)	1.4 (0.95)	57 (39)	21.7 (14.8)	141 (96)
Erie 23–Jul 2001	20 + 40 + 50 + 59	0.108 (0.099)	21.1 (19.4)	0.5 (0.46)	48 (44)	42.2 (38.8)	245 (225)
Erie 23–Aug 2001	20 + 40 + 50 + 59	0.125 (0.057)	53.7 (24.7)	1.5 (0.69)	88 (40)	69.1 (31.8)	351 (161)
Erie 23–Nov 2001	21	0.088 (3.52)	29.7 (1,190)	0.8 (32)	41 (1,640)	53.0 (2,120)	154 (6,160)
Erie 23–Nov 2001	41	0.086 (2.38)	28.2 (780)	0.7 (19.4)	32 (890)	46.0 (1,270)	149 (4,130)
Erie 23–Nov 2001	51	0.083 (2.93)	28.3 (1,000)	0.8 (28)	33 (1,160)	46.7 (1,650)	140 (4,940)
Erie 23–Nov 2001	60	0.086 (3.87)	29.4 (1,320)	0.8 (36)	34 (1,530)	48.3 (2,170)	142 (6,390)
Erie 84–May 2001	18 + 21	0.180 (0.474)	36.3 (95.8)	1.6 (4.2)	57 (150)	39.9 (105)	220 (581)
Erie 84–Jun 2001	18 + 21	0.230 (0.237)	50.6 (52.1)	2.0 (2.1)	69 (71)	45.8 (47.2)	239 (246)
Erie 84–Aug 2001	19 + 22	0.164 (0.449)	41.1 (113)	2.5 (6.9)	55 (151)	58.2 (159)	225 (617)
Erie 84–Nov 2001	18	0.207 (31.1)	58.0 (8,700)	1.7 (255)	48 (7,200)	62.8 (9,420)	228 (34,200)
Erie 84–Nov 2001	21	0.195 (41.0)	57.3 (12,000)	1.7 (357)	47 (9,870)	60.4 (12,700)	231 (48,500)
Erie 357–May 2001	9	<b>1.10</b> (79.2)	66.3 (4,770)	1.8 (130)	50 (3,600)	65.8 (4,740)	238 (17,100)
Erie 357–Jun 2001	9	<b>0.715</b> (100)	70.4 (9,890)	2.0 (280)	51 (7,140)	65.8 (9,200)	243 (34,000)
Erie 357–July 2001	9	<b>0.645</b> (20.0)	74.8 (2,320)	2.2 (68)	50 (1,550)	60.1 (1,860)	247 (7,660)
Erie 357–Aug 2001	9	<b>0.713</b> (78.4)	72.4 (7,960)	2.3 (253)	51 (5,610)	64.3 (7,070)	254 (27,900)
Erie 357–Sept 2001	9	<b>0.780</b> (179)	73.8 (17,000)	2.3 (529)	54 (12,400)	63.5 (14,600)	249 (54,300)
Ont 403–May 2001	20	0.427 (0.333)	61.6 (48.0)	1.6 (1.2)	92 (72)	58.0 (45.2)	198 (154)
Ont 403–May 2001	60	0.469 (0.408)	72.3 (62.9)	1.7 (1.5)	88 (77)	62.7 (54.5)	282 (245)
Ont 403–May 2001	100	<b>0.575</b> (0.558)	78.6 (76.2)	1.8 (1.7)	96 (93)	67.7 (65.7)	285 (276)
Ont 403–May 2001	140	<b>0.503</b> (0.568)	81.2 (90.9)	1.8 (2.0)	95 (107)	68.4 (77.3)	309 (349)
Ont 403–May 2001	165	<b>0.588</b> (1.13)	88.3 (170)	2.1 (4.0)	104 (200)	73.2 (141)	316 (607)
Ont 403–May 2001	175	<b>0.565</b> (1.57)	<b>91.8</b> (255)	2.2 (6.1)	103 (286)	73.9 (205)	308 (856)
Ont 403– Oct 2001	20	0.101 (0.023)	25.1 (5.7)	1.6 (0.37)	77 (18)	33.3 (7.7)	134 (30.8)
Ont 403–Oct 2001	60	0.122 (0.029)	20.2 (4.8)	1.9 (0.46)	118 (28)	25.9 (6.2)	331 (79)
Ont 403–Oct 2001	100	0.170 (0.054)	23.1 (7.4)	1.6 (0.51)	103 (33)	30.3 (9.7)	203 (65)
Ont 403–Oct 2001	140	0.255 (0.077)	35.3 (10.6)	1.7 (0.51)	111 (33)	34.7 (10.4)	210 (63)
Ont 403–Oct 2001	166	0.316 (0.180)	46.1 (26.2)	1.6 (0.91)	158 (90)	45.1 (25.7)	221 (126)
Ont 403–Oct 2001	175	<b>0.496</b> (0.481)	48.8 (47.3)	1.6 (1.6)	116 (113)	54.8 (53.2)	291 (282)
Erie 23–Apr 2002	21	0.079 (1.61)	29.8 (698)	0.7 (14.3)	32 (653)	46.5 (949)	269 (5,490)
Erie 23–Apr 2002	41	0.101 (0.276)	33.8 (92.3)	0.8 (2.2)	37 (101)	49.8 (136)	157 (429)
Erie 23–Apr 2002	51	0.094 (0.298)	32.4 (109)	0.8 (2.5)	32 (101)	45.5 (144)	126 (399)
Erie 23–Apr 2002	60	0.090 (0.303)	31.0 (104)	0.7 (2.4)	34 (115)	45.7 (154)	148 (499)
Erie 23–Jun 2002	20	0.054 (0.130)	19.1 (46.0)	0.5 (1.2)	126 (304)	33.9 (81.7)	138 (465)
Erie 23–Jun 2002	40	0.077 (0.207)	19.2 (51.6)	0.5 (1.3)	159 (428)	32.7 (87.9)	197 (530)
Erie 23–Jun 2002	50	0.049 (0.142)	17.5 (50.8)	0.5 (1.5)	81 (235)	32.3 (93.7)	114 (330)
Erie 23–Jun 2002	60	0.062 (0.208)	20.3 (60.8)	0.5 (1.7)	39 (131)	33.7 (113)	121 (405)
Erie 23–Jul 2002	20 + 40 + 50 + 59	0.144 (0.200)	36.8 (51.1)	2.5 (3.5)	52 (72)	44.3 (61.6)	<b>546</b> (759)
Erie 23–Sept 2002	20 + 40	0.064 (0.063)	19.3 (18.9)	0.9 (9.0)	54 (53)	51.0 (50.0)	150 (147)
Erie 23–Sept 2002	50 + 59	0.076 (1.21)	21.3 (25.8)	0.8 (1.0)	159 (192)	47.4 (57.3)	213 (258)
Erie 23–Oct 2002	20	0.044 (0.157)	16.1 (57.3)	1.1 (3.9)	60 (214)	33.8 (120)	190 (676)
Erie 23–Oct 2002	40	0.044 (0.220)	18.9 (94.3)	0.6 (3.0)	164 (818)	42.9 (214)	128 (639)
Erie 23–Oct 2002	50	0.048 (0.308)	20.6 (132)	0.5 (3.2)	64 (411)	44.0 (282)	128 (822)
Erie 23–Oct 2002	59	0.058 (0.371)	19.9 (127)	0.5 (3.2)	42 (268)	41.7 (266)	116 (741)
Erie 84–Jun 2002	19	0.133 (1.53)	38.2 (439)	1.2 (13.8)	39 (449)	49.4 (568)	168 (1,930)
Erie 84–Jun 2002	22	0.141 (2.00)	38.9 (552)	1.7 (24.1)	56 (795)	51.8 (736)	208 (2,950)
Erie 84–Jul 2002	20 + 23	0.180 (NA)	50.4 (NA)	1.6 (NA)	95 (NA)	58.8 (NA)	222 (NA)
Erie 84–Aug 2002	20 + 23	0.150 (0.441)	46.5 (101)	2.2 (4.8)	67 (146)	67.1 (146)	181 (395)
Erie 84–Sept 2002	20 + 23	0.190 (1.19)	55.5 (349)	2.2 (138)	144 (904)	60.1 (377)	<b>359</b> (2,250)
Erie 84–Oct 2002	19	0.186 (4.87)	54.1 (1,420)	1.7 (44.5)	72 (1,890)	65.8 (1,720)	220 (5,760)

**Table II** (continued)

Station/date <sup>a</sup>	Depth <sup>b</sup> (m)	Mercury	Lead	Cadmium	Copper	Nickel	Zinc
Erie 84–Oct 2002	22	0.169 (4.43)	50.1 (NA)	1.6 (NA)	87 (NA)	59.0 (NA)	<b>603</b> (NA)
Erie 357–Jun 2002	9	<b>0.604</b> (99.7)	66.7 (11,000)	1.7 (281)	51 (8,400)	64.4 (10,600)	231 (38,100)
Erie 357–Jul 2002	9	<b>0.677</b> (35.9)	68.1 (3,610)	2.1 (111)	54 (2,860)	64.8 (3,430)	244 (12,900)
Erie 357–Aug 2002	10	<b>0.671</b> (NA)	69.5 (NA)	2.2 (NA)	53 (NA)	63.2 (NA)	241 (NA)
Erie 357–Sept 2002	9	<b>0.641</b> (NA)	69.1 (NA)	2.1 (NA)	51 (NA)	64.2 (NA)	249 (NA)
Ont 403–Apr 2002	20	<b>0.572</b> (0.715)	<b>117</b> (146)	2.3 (2.9)	110 (138)	<b>85.0</b> (106)	311 (389)
Ont 403–Apr 2002	60	<b>0.556</b> (0.900)	<b>109</b> (177)	2.2 (3.6)	286 (463)	<b>85.1</b> (138)	<b>349</b> (565)
Ont 403–Apr 2002	100	<b>0.587</b> (1.09)	<b>95.9</b> (178)	2.1 (3.9)	121 (225)	<b>85.6</b> (159)	<b>314</b> (584)
Ont 403–Apr 2002	140	<b>0.561</b> (1.36)	<b>113</b> (275)	2.2 (5.3)	233 (566)	<b>82.9</b> (201)	<b>315</b> (765)
Ont 403–Apr 2002	166	<b>0.597</b> (2.21)	<b>104</b> (386)	2.4 (8.9)	130 (482)	<b>87.1</b> (323)	<b>327</b> (1,210)
Ont 403–Apr 2002	174	<b>0.632</b> (2.76)	<b>102</b> (446)	2.5 (10.9)	123 (538)	<b>96.2</b> (420)	<b>336</b> (1,470)
Ont 403–Nov 2002	20	0.109 (0.083)	21.2 (16.1)	1.3 (1.0)	100 (76)	27.7 (21.1)	145 (110)
Ont 403–Nov 2002	60	0.108 (0.039)	35.4 (12.7)	2.2 (0.8)	91 (32.8)	27.5 (9.9)	305 (110)
Ont 403–Nov 2002	100	0.157 (0.058)	28.2 (10.4)	1.5 (0.6)	134 (49.6)	24.2 (9.0)	<b>362</b> (134)
Ont 403–Nov 2002	140	0.183 (0.090)	68.4 (33.5)	2.1 (1.0)	131 (64.2)	39.9 (19.6)	<b>510</b> (250)
Ont 403–Nov 2002	166	0.266 (0.630)	57.9 (137)	1.8 (4.3)	108 (256)	42.9 (102)	<b>329</b> (780)
Ont 403–Nov 2002	174	0.382 (0.393)	82.5 (85.0)	2.3 (2.4)	116 (119)	55.8 (57.5)	<b>343</b> (353)

Metals down-flux rates were calculated using the mean daily sediment down-flux rates from the values shown in Table I. Values in bold type denote exceedances of the PEL guideline.

<sup>a</sup> Dates represent the end of the time intervals specified in Table I.

<sup>b</sup> Multiple depths correspond to composite samples. Metals down-flux rates for composite samples were calculated using mean sediment down-flux rates for depths shown.

<sup>c</sup> CSQG PEL corresponds to the Canadian Sediment Quality Guidelines Probable Effect Level (CCME, 1999).

NA Denotes data not available.

the western basin of Lake Erie in 1997 were similar to those across the major depositional basins of Lake Ontario in 1998 (Marvin et al., 2002b). Since material collected during the winter months at station 403 in Lake Ontario, and at station 357 in the western basin of Lake Erie throughout the year, represented a significant contribution from resuspended bottom sediments, comparisons were made with data from the most recent surficial sediment surveys. The comparison of concentrations in lead and mercury in bottom sediments of Lakes Erie (1997) and Ontario (1998), and the annual mean concentrations of mercury and lead in suspended sediments in 2001 and 2002 calculated using data in Table II, is shown in Table III.

For Lake Erie, annual mean suspended sediment concentrations of mercury and lead were statistically very similar to bottom sediment concentrations (Table III), although most suspended sediment concentrations were higher, compared to bottom sediments. In addition, there was very little inter-annual variation in metals concentrations. In our previous study of persistent organic pollutants associated with

suspended sediments in the western basin of Lake Erie (station 357, Marvin et al., 2004), PCB contamination in suspended sediments was greater, compared to bottom sediments. The relatively higher PCB concentrations in suspended sediments were attributed to potential contamination associated with material discharged from the Detroit River. The Detroit River continues to be a vector for active loadings of contaminants, including PCBs and dioxins and furans, which may originate from areas within the river, or from other sources including the upstream lakes and connecting channels (Marvin et al., 2002a). The similarities between bottom and suspended sediment concentrations across the entire breadth of Lake Erie implicate bottom sediments as a primary influence on suspended sediment quality. In the western basin, the relatively shallow depth, bathymetry, and prolonged periods of high-suspended sediment loadings contribute to a prolonged contaminant down flux of material primarily comprised of resuspended bottom sediments.

Due to the substantial seasonal variation in metals concentrations in suspended sediments in Lake Ontario, data presented in Table III are presented for

**Table III** Comparison of annual mean concentrations of mercury and lead in suspended sediments ( $\mu\text{g/g}$ ) in 2001 and 2002 vs. bottom sediments in Lakes Erie and Ontario

	Erie 23 (East)	Erie 84 (Central)	Erie 357 (West)	Ontario 403 (Central)
2001				
Suspended sediments				
Mercury	0.099 $\pm$ 0.017	0.195 $\pm$ 0.025	<b>0.790 <math>\pm</math> 0.179</b>	<b>0.521 <math>\pm</math> 0.065</b> (Winter) 0.243 $\pm$ 0.148 (Spring/Summer/Fall)
Lead	30.2 $\pm$ 11.0	48.7 $\pm$ 9.7	71.5 $\pm$ 3.4	79.0 $\pm$ 11.0 (Winter) 33.1 $\pm$ 12.3 (Spring/Summer/Fall)
Bottom sediments				
Mercury	0.068 $\pm$ 0.042	0.163 $\pm$ 0.104	0.410 $\pm$ 0.275	<b>0.600 <math>\pm</math> 0.450</b>
Lead	22.0 $\pm$ 13.4	45.4 $\pm$ 24.3	44.4 $\pm$ 18.4	74.0 $\pm$ 46.0
2002				
Suspended Sediments				
Mercury	0.072 $\pm$ 0.027	0.160 $\pm$ 0.023	<b>0.650 <math>\pm</math> 0.033</b>	<b>0.584 <math>\pm</math> 0.028</b> (Winter) 0.200 $\pm$ 0.106 (Spring/Summer/Fall)
Lead	23.7 $\pm$ 6.9	47.7 $\pm$ 6.9	68.4 $\pm$ 1.3	<b>107 <math>\pm</math> 7.7</b> (Winter) 48.9 $\pm$ 24.3 (Spring/Summer/Fall)
Bottom Sediments				
Mercury	0.068 $\pm$ 0.042	0.163 $\pm$ 0.104	0.410 $\pm$ 0.275	<b>0.600 <math>\pm</math> 0.450</b>
Lead	22.0 $\pm$ 13.4	45.4 $\pm$ 24.3	44.4 $\pm$ 18.4	74.0 $\pm$ 46.0

Bottom sediment data for Lake Erie from Painter et al. (2001). Bottom sediment data for Lake Ontario from Marvin et al. (2002b). Suspended sediment data for Lake Ontario represents mean values for winter and combined spring/summer/fall sampling periods. Concentrations in bold face indicate exceedances of the Canadian Sediment Quality Guideline Probable Effect Level (PEL).

winter and spring/summer/fall sampling periods. As with the Lake Erie data, little inter-annual variation was observed. The influence of resuspended bottom sediments to the suspended sediment pool over the winter period during unstratified conditions was clearly evident. Lead and mercury concentrations in winter suspended sediments in Lake Ontario over the period 2000–2002 were similar to bottom sediment concentrations. In contrast, suspended sediment concentrations of mercury and lead during the spring/summer/fall sampling timeframes, which comprised significant periods of stratified conditions, were substantially lower than bottom sediments. In contrast to Lake Erie, these results indicate that suspended sediments entering Lake Ontario via the Niagara River and other tributaries contains significantly lower contaminant burdens than bottom sediments. These results corroborate reports that many known sources of contaminants in the Niagara River watershed have been significantly reduced (Durham & Oliver, 1983; Marvin et al., 2002c), and that further declines in contaminant levels in bottom sediments may be expected. In our previous study of persistent

organic pollutants associated with suspended sediments in Lake Ontario (Marvin et al., 2004), significant declines in both down-flux rates and concentrations of contaminants including PCBs and organochlorine pesticides in suspended sediments over the period from the early 1980s to the late 1990s were observed.

#### 4 Conclusions

Characterization of material collected in sediment traps in both lakes Erie and Ontario indicated that resuspended bottom sediments can considerably influence contaminant concentrations in the water column during periods of thermal unstratification, e.g., winter. Assessment of down-flux rates and percent LOI values indicated that material collected in sediment traps in the western basin of Lake Erie during all periods of deployment were primarily resuspended bottom sediments. The influence of these bottom sediments was indicated by low percent LOI values and high down-flux rates.

Similarly, suspended sediment collected in Lake Ontario during winter exhibited lower percent LOI and higher down-flux rates, compared to periods of stratified conditions. In addition, concentrations of lead and mercury in suspended sediments during winter in Lake Ontario were roughly two-fold higher, compared to samples from spring/summer/fall. Mercury and lead concentrations in winter suspended sediments from Lake Ontario were very similar to concentrations in bottom sediments, which further indicated the influence of bottom sediments on these samples. In contrast, suspended sediment samples collected in Lake Ontario over the period May–October exhibited relatively higher percent LOI values and lower down-flux rates; metals concentrations in these samples were roughly two-fold lower than typical levels in bottom sediments in the same area. The variation in percent LOI values and down-flux rates with depth in lake Ontario showed a transition from a predominance of suspended sediments high in organic content in the top 100 m of the water column, to a predominance of resuspended bottom sediments with lower organic content in the bottom 75 m of the water column. These results also indicate that suspended material entering Lake Ontario from the watershed, or via atmospheric deposition, is less contaminated than the existing bottom sediments. These observations also corroborate our conclusions regarding temporal trends in bottom sediment contamination in Lake Ontario; significant declines have occurred over the past 30 years, and the improved quality of material entering the lake suggest that further reductions in metals contamination are likely.

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