The Effect of a Large Resuspension Event in Southern Lake Michigan on the Short-term Cycling of Organic Contaminants

Jerel J. Bogdan[1], Judith W. Budd[2], Brian J. Eadie[3], and Keri C. Hornbuckle[4],*

[1]*Malcolm Pirnie, Inc.*
Orchard Park, New York 14127

[2]*Geological Engineering and Sciences*
Michigan Technological University
Houghton, Michigan 49931

[3]*Great Lakes Environmental Research Laboratory (NOAA)*
Ann Arbor, Michigan 48105

[4]*Dept. Civil and Environmental Engineering*
University of Iowa
Iowa City, Iowa 52242

ABSTRACT. *In January and March, 1998, a series of intense, northerly wind-driven storms suspended sediment over the entire coastline of the southern basin of Lake Michigan. The effect of large scale resuspension on organic contaminant cycling was investigated using a two-pronged sample collection strategy that included analysis of settling sediment trap material and discrete air and water samples collected before and after a major resuspension event. It was found that major resuspension events result in a large flux of contaminants. For example, 6.2 ng/cm² ΣPCB (sum of 89 congener peaks) and 175 ng/cm² ΣPAHs (sum of 31 compounds) fell through the water column in the southern basin between November and May but almost half of that occurred in the month of March after a series of intense storms induced a large-scale resuspension event in that month. Assuming the concentration of contaminants in settling sediments is similar throughout the basin, the March event brought ~400 kg of ΣPCBs and ~13,000 kg ΣPAHs into the water column. Furthermore, the data indicate that concentrations of dissolved phase ΣPCB and ΣPAHs declined significantly (α = 0.05) after the event and after resuspended sediment had settled from the surface waters. As a result of the depressed dissolved concentrations at the surface, the potential for gas-phase input to the lake increases on the southwestern coastal region near Chicago, IL and Gary, IN. The potential input of gas-phase contaminants was 8 kg for ΣPCBs and 2,200 kg for ΣPAHs over the 40-day lifetime of the near-shore event.*

INDEX WORDS: *Resuspension, PCBs, PAHs, organic contaminants, gas-phase exchange, Lake Michigan.*

INTRODUCTION

Delivery of new materials and mobilization of sedimentary materials may have an important impact on the cycling of organic contaminants in coastal systems. For example, resuspension may cause release of buried contaminants in older sediments or may cause removal of contaminants through increased particulate fluxes. To examine the role of resuspension events on organic contaminant cycling, a field study was conducted in southern Lake Michigan during several large resuspension events in the winter/spring of 1998. One resuspension event occurred in mid to late January 1998. A larger resuspension event, generated by two strong late winter-early spring storms, was first observed in early March and subsided over a ~40 day period.

Intense winter/spring episodes of sediment resuspension occur frequently in southern Lake Michigan (Eadie *et al.* 1996), however, the turbidity

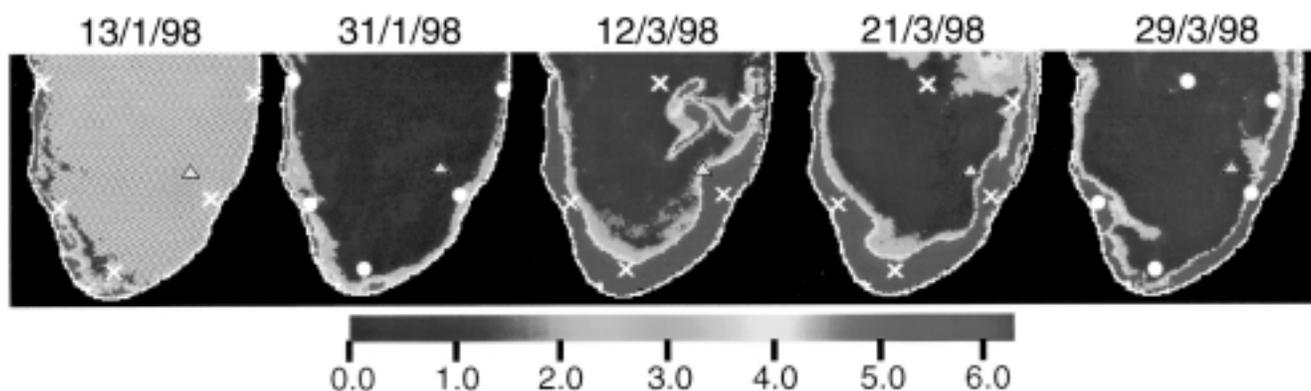*Corresponding author. E-mail: keri-hornbuckle@uiowa.edu

FIG. 1. *Satellite images of AVHRR reflectance. The scale is from 0% reflectance (dark blue) to 6% reflectance (red). The 31 January and 29 March images include the sites of discrete air and water sampling. Water sampling locations are noted as white circles, the yellow triangle notes where the sediment trap was deployed. The 13/1/98, 12/3/98, and 21/3/98 images show the spatial coverage of the 1998 March resuspension event. Although samples were not collected during these 3 days, each of the sampling locations are plotted as an X for spatial comparison with the resuspension region.*

plume event of March 1998 was one of the largest of recent record (Lou *et al.* 2000). Plume development corresponded with two large storm events that occurred on 9 and 21 March. On those 2 days strong winds reached 17m/s, primarily from the northeast, and wave heights reached 5 m in the southern basin. These storms produced a significant resuspension event as observed in satellite imagery. At full development, the resulting coastal plume covered a 300 km length of the coast of the southern basin and was visible by satellite for 6 weeks before calming in late April. A spectacular offshore-spiraling eddy was observed 3 days after the 9 March storm, due to meandering of the strong offshore currents driven by a two-gyre wind-driven circulation pattern, shown in Figure 1 (Lou *et al.* 2000). Schwab *et al.* (2000) estimated total mass of suspended sediment in the southern basin of the lake in March 1998 was 7.5×10^9 kg. For perspective, the United State Geological Survey's Southern Lake Michigan Coastal Erosion Study (1988 to 1992) (Folger *et al.* 1994) reported an average *annual* sediment budget for the southern basin of Lake Michigan from records of the last 100 years to balance at 3×10^9 kg (Folger *et al.* 1994). Thus, the 40-day 1998 event was twice the 100-year annual sediment budget for the entire southern basin in a single, large-magnitude event. The initiation of the plume in 1998 does not appear to coincide with the development of thermal stratification or springtime ice-out as suggested by Mortimer (1988).

This work was done in conjunction with a large,

multidisciplinary effort involving over forty principle investigators and sixteen institutions, as part of Episodic Events—Great Lakes Experiment (EEGLE). The effect of the March storms on contaminant cycling processes was evaluated. Polycyclic aromatic hydrocarbons (PAHs) and polychlorinated biphenyl congeners (PCBs) were used as representative of persistent organic pollutants (POPs). Using these 121 compounds, the effect was examined in two ways. First, the total mass of contaminant introduced into the water column was determined as a result of the storm events. Second, changes in the distribution and potential fluxes of these POPs in water, air, and suspended sediments was assessed for two sampling expeditions.

METHODS

This work addresses changes in the cycling of POPs using a two-pronged strategy of discrete and time-integrated sampling. Discrete samples of water and air were collected before and after the resuspension event. The first sampling expedition (prior to the event) was conducted from 28 January to 4 February 1998, and the second sampling expedition (after the 9 and 22 March storms) was from 28 to 30 March 1998. The locations of the water sample sites are indicated in Figures 1 and 2 and Table 1. The transects over which air sampling was conducted are shown in Figure 2. In addition, subsamples of settling sediment was analyzed from an integrated sequencing trap that was deployed from
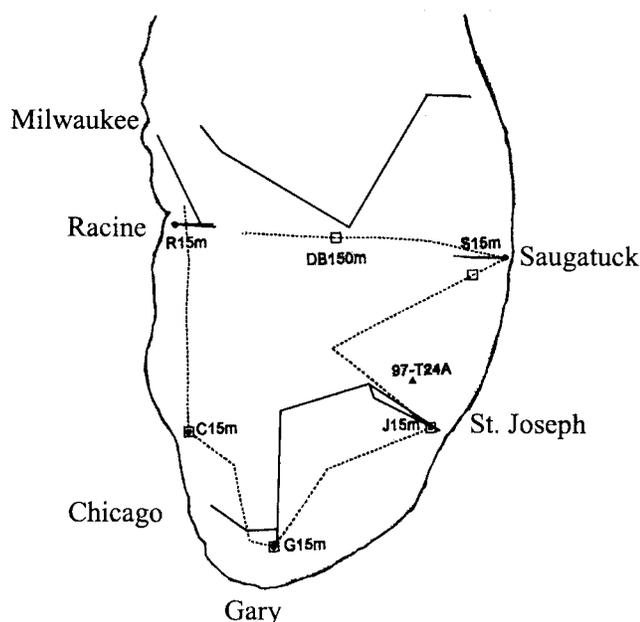
FIG. 2. Sampling locations for the spring 1998 sampling cruises. The solid line indicates the collection transect for the January air sampling, the dotted line is the transect for the March air sampling. January water sampling sites are indicated by solid circles. March water sampling sites are indicated by open squares. The sediment trap deployment site (97-T24A) is indicated by a triangle.

25 November 1997 to 24 May 1998 at 42.291 latitude, –86.638 longitude (Figs. 1 and 2). The trap was located in 56 m deep water at 30 m from the surface. The trap collected settling sediment generally every 9 days, yielding twenty sediment trap samples over the deployment period. Whereas the discrete sampling provides details of the gas, aerosol, dissolved, and particulate phases at one point in time, the sediment trap material provides an integrated view of chemical concentrations and downward fluxes that were changing through time.

Sampling Procedures

Discrete water and air sampling was conducted aboard the U.S. EPA's Research Vessel R/V *Lake Guardian*. These methods are detailed elsewhere and briefly reviewed here (Bogdan 1999, Hornbuckle *et al.* 1993, Hornbuckle *et al.* 1994, Pearson *et al.* 1996). Water samples consisted of glass fiber filters through which ~500 L of water was passed,

and XAD-2 resin through which ~200 L of filtered water was passed. Air samples consisted of quartz fiber filters and XAD-2 resin in series. Air samples were collected from a long pole (yard-arm) that swings out over the water, about 3 m from the ship's bow. Most of the water sampling sites were located at the 15 m isopleth although all were pumped from the near surface (~2 m) water. One site was located in deep waters but only sampled during the 28 to 30 March cruise.

The twenty sediment trap samples were collected from a sequencing sediment trap deployed at a location chosen because of its close proximity to an offshore transport feature of the plume event that has been observed in satellite imagery (Eadie *et al.* 1996). It is also a region of maximum sediment accumulation in Lake Michigan. A computer-controlled carousel containing twenty-three 60 mL polyethylene bottles was situated beneath the trap funnel. An electric motor rotated the carousel to the next collection bottle every 9 days. The collection bottles were poisoned with 6 mL of chloroform to prevent biological decay and filled with distilled water prior to deployment. Upon retrieval of the trap, the material was screened at NOAA/GLERL to remove large particles (500 μm) and then the finer particle fraction was split into two equal subsamples. One subsample was sent to the University of Iowa for POPs analysis, while the other fraction was analyzed for total and organic carbon as well as for total and organic nitrogen.

Analytical Procedures

The samples were analyzed for a total of 32 individual PAH compounds and 89 individual PCB congeners and/or congener groups. Methods used to extract, clean up, concentrate, and analyze air samples were consistent with those used during the Lake Michigan Mass Balance Project (United States Environmental Protection Agency 1997a, United States Environmental Protection Agency 1997b), with the exception of the water removal step and the order of PAH analysis. The following is a brief description of the procedures.

The sediment trap subsamples were decanted and mixed with Na_2SO_4 to remove water, injected with PAH and PCB surrogate standards, and extracted in a Soxhlet apparatus with dichloromethane. The overlying water/chloroform mixture was back-extracted with dichloromethane and water and recombined with the sediment extract. The extracts were concentrated by rotoevaporator to 2 mL and

TABLE 1. *Summary of water samples obtained during January and March 1998.*

Sampling Location	Date	Lat	Long	TSM [mg/L]	f_{oc}	Surface Water Temp (C°)	Average Wind speed (m/s)[a]	Wind direction (deg)[b]	Water Current (deg)[c]
Racine 15 m	28-Jan-98	42.750	–87.725	4.15	0.04	0.6	2.5	SW	—
Chicago 15 m	29-Jan-98	42.098	–87.622	—	—	1.6	3.0	SW	—
Gary Dup1 15 m	31-Jan-98	41.751	–87.225	0.82	0.21	1.6	2.4	W	—
Gary Dup2 15 m	31-Jan-98	41.751	–87.225	0.82	0.21	1.6	2.4	W	—
St. Joseph 15 m	4-Feb-98	42.142	–86.551	1.35	0.15	2.1	1.9	SW	—
Saugatuck 15 m	1-Feb-98	42.690	–86.238	1.97	0.10	1.0	2.2	S	—
Chicago 15 m	28-Mar-98	42.098	–87.621	1.02	0.15	1.2	8.7	SW	S
Gary 15 m	29-Mar-98	41.750	–87.226	1.54	0.10	1.8	7.8	SW	E
St. Joseph 15 m	29-Mar-98	42.142	–86.549	1.0*	0.10*	1.7	4.4	S	SW
Saugatuck 15 m	29-Mar-98	42.631	–86.384	1.0*	0.10*	2.3	4.8	SW	S
Basin 150 m	30-Mar-98	42.731	–86.999	0.66	0.16	—	4.0	S	S

[a] Wind speed is the average for 48 hours prior to water sampling using measurements from the nearest meteorological station.
[b] Wind direction is the average for 48 hours prior to water sampling from the nearest meteorological station (see text for algorithm).
[c] Water current direction is the average for 48 hours prior to water sampling using hourly modeled data (Schwab *et al.* 2000). Model results are not available for the January dates.
* Data are estimated from satellite data of the plume extent, and values measured 10 days earlier.

injected with PAH internal standards d10-acenaphthalene, d10-phenanthrene, d10-pyrene, and d12-benzo[e]pyrene. Next the extracts were transferred to autosampler vials and analyzed for PAH compounds by gas chromatography—selective ion mass spectrometry in electronic impact mode (HP5973 mass selective detector with HP6890 capillary GC equipped with electric pressure control and autoinjector). Subsequent to the PAH analysis, the extracts were transferred to a silica gel (3% deactivated) column and eluted with three solvent mixtures of increasing polarity. The first fraction was eluted with hexane and contained the PCBs. This fraction was concentrated by rotoevaporator and nitrogen stream and injected with PCB internal standards 2,4,6-trichlorobiphenyl (IUPAC #30), 2,2′,3,4,4′,5,6,6′-octachlorobiphenyl (IUPAC #204). The extract was then concentrated to approximately 200 uL and analyzed by gas chromatography with electron capture detection (HP 6890 GC with ^{63}Ni µECD).

The analytical quality of the data was determined through recoveries of surrogate compounds, duplicate sampling, detection limit studies, and various blanks. Of the 91 samples collected and analyzed for this study, 35% were quality control samples, consisting of field blanks, trip blanks, solvent blanks, and matrix spikes. Every sample was spiked with surrogate standards and recoveries of PAH sur-

rogates were 69% ± 25% for d10-fluorene, 80% ± 17% for d10-fluoranthene, and 88% ± 21% for d12-perylene (n=88). Recoveries of PCB surrogates were 47% ± 13% for Congener 14, 60% ± 16% for Congener 65, and 67% ± 19% for Congener 166 (n=88). The lower PCB recoveries are reflective of the loss of extract for PAH analysis prior to sample cleanup and is not due to poor sample handling. All PAH and PCB concentrations determined in the sample extracts were corrected for surrogate recovery.

Replicate (side-by-side and/or sequential) samples were collected during the January expedition to evaluate reproducibility of the methods. The relative percent difference (RPD) in replicate samples is due to real random error associated with sample collection, handling, extraction, and analysis, as well as to variability in each sample matrix and sampling device. Mean RPD values for dissolved ΣPAHs and ΣPCBs were 60% and 8%, respectively. Mean RPDs for the gaseous phase ΣPAHs and ΣPCBs were 20% and 33%; higher RPD values for water particulate and airborne particulate phases were observed (28.6% to 131.2%), indicative of less precision for sampling of these phases.

The percent blank mass (PBM) was calculated for each sample matrix as the mean mass of field blanks divided by the mean mass of samples. Dissolved, water particulate, and gas phase samples all

had PBM values of < 13% whereas higher PBM were found for airborne particulate (42 to 50%) and sediment trap samples (18 to 28%). Water and air samples were not corrected for blank values. The field blank values observed in the sediment trap samples may be attributable to absorption of dissolved contaminants by the chloroform in the sediment collection bottles. All sediment trap bottles, including three field blanks, were deployed filled with the chloroform (6 mL) and distilled water. When not open for sample collection, the remaining trap bottles were sealed with Teflon interface, pressed by an o-ring. As particles fell into the trap bottles, a small amount of lake water could diffuse in and out of bottles, introducing the possibility of extraction of dissolved phase compounds by the liquid organic solvent chloroform. Therefore, the mass of each individual PCB and PAH mass in sediment trap samples was corrected for this contamination by subtracting the average mass found in the field blanks.

RESULTS

Sediment and Organic Carbon

Satellite images and sediment trap mass fluxes (Figs. 1 and 3) illustrate evidence for two periods of winter/spring resuspension in southern Lake Michigan. The satellite images indicate that a resuspension event occurred in early January. It appears that this event was short-lived and limited to about 5 km off the coast, although sediment trap collection indicates broad coverage across the southern basin during the first week of January (unpublished data, B.J. Eadie 2001). The second event was initiated by the 9 March storm. High settling fluxes were measured near the southeast coast all through March, returning to background levels in the beginning of April (Fig. 3).

The origin of the settling material collected at T24 is not known exactly, but may include resuspension of previously deposited material, erosion of coastal material, riverine sediments, and atmospheric particles. Hydrodynamic modeling and drifter studies indicate that circulation of the lake during the January and March events was strongly counterclockwise (cyclonic) south and west of the site but often simultaneously clockwise (anticyclonic) north of the site. Satellite imagery of surface reflectance (Fig. 1) and sediment transport modeling (Schwab *et al.* 2000) suggest that the sediment delivered to T24 is primarily from the regions east and south of the site.
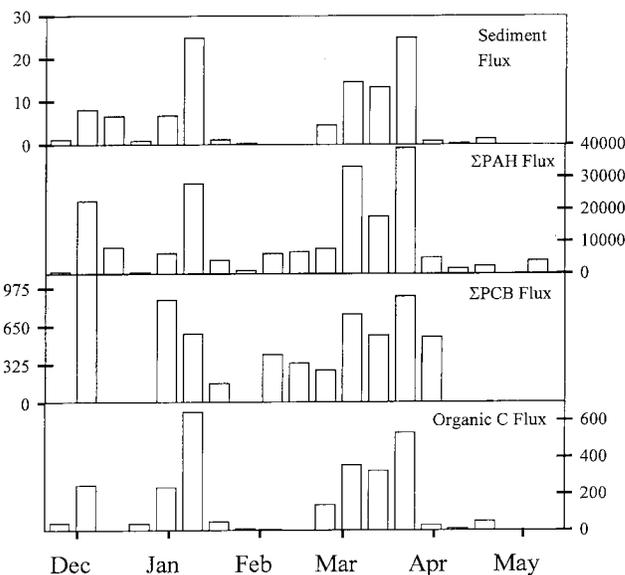

FIG. 3. *Blank-corrected results from the sequencing sediment trap, starting at top plot: Sediment flux (g/m²/d), ΣPAH settling flux (ng/m²/d), ΣPCB settling flux (ng/m²/d), Σ PAH concentration on settling particles (ng/g), ΣPCB concentration on settling particles (ng/g), and organic carbon flux (mg/m²/d). Each vertical bar represents a 9-day sequential collection period beginning on 25 November 1997. The hatched areas through each plot identifies the two plume events transpiring in January and March 1998 as observed in the temporal profile of sediment flux.*

A NOAA/GLERL expedition conducted on 16 to 19 March included measurements of total suspended matter concentration (TSM). They reported TSM values of 12 to 34 mg/L (Schwab *et al.* 2000) which is about ten times the annual average. No POPs sampling was conducted during the GLERL expedition: the POPs expedition described here was conducted independently about 1 week later. By this time, the surface concentrations of suspended sediment had returned to the values measured prior to the March event (1.38 ± 0.58 mg/L in 28 January to 4 February and 1.07 ± 0.44 mg/L in 28 to 30 March). The fraction of organic carbon (f_{oc}) on TSM varied in space (0.04 to 0.21) but this variability could not be attributed to the extent of the resuspension coverage. This may be due to significant settling of the resuspended particles at the surface

TABLE 2.   *Chemical concentrations in settling sediment.*

Sample Name	Date Open	Date Closed	organic carbon, foc, g/g	ΣPCB [ng/g]	ΣPAH [ng/g]
T2642	25/11/97	4/12/97	0.035	2.7	490
T2643	4/12/97	13/12/97	0.031	148	2,790
T2644	13/12/97	22/12/97	NA	8.0	1,230
T2645	22/12/97	31/12/97	0.042	1.3	448
T2646	31/12/97	9/1/98	0.035	130	940
T2647	9/1/98	18/1/98	0.026	23.6	1,120
T2648	18/1/98	27/1/98	0.041	147	2,560
T2649	27/1/98	5/2/98	0.033	102	3,040
T2650	5/2/98	14/2/98	0.033	3,530	53,200
T2651	14/2/98	23/2/98	0.036	3,680	73,300
T2652	23/2/98	4/3/98	0.031	61.9	1,740
T2653	4/3/98	13/3/98	0.025	51.9	2,280
T2654	13/3/98	22/3/98	0.024	43.0	1,330
T2655	22/3/98	31/3/98	0.021	36.5	1,570
T2656	31/3/98	9/4/98	0.034	673	5,910
T2657	9/4/98	18/4/98	0.031	376	5,120
T2658	18/4/98	27/4/98	0.037	100	1,700
T2659	27/4/98	6/5/98	NA	72.5	372
T2660	6/5/98	15/5/98	NA	2,710	120,000
T2661	15/5/98	17/5/98	0.045	1,810	7,570

or an inability to distinguish the borders of the plume during the expeditions.

Organic carbon on suspended particles may be used as a tracer of biogenic particles (Weyhenmeyer *et al.* 1995). Sediments collected in the settling traps were low in organic carbon (Table 2), indicative of resuspension of weathered bottom sediments and/or mineral particles. The lowest f_{oc} values were recorded during the high sediment flux periods in mid January and March. During the NOAA/GLERL expedition on 17 March, investigators found very low organic carbon contents (f_{oc} = 0.02 to 0.04) in shallow waters (10 m to 20 m depth) near St. Joseph and Gary (Winkelman *et al.* 1998). Larger organic carbon contents (f_{oc} > 0.12) were reported in deeper waters. A full spatial analysis of the plume coverage using satellite images and measurements taken from the research vessels continues, but the measurements of organic carbon content and TSM suggest that the low organic carbon material had settled from the surface waters by 28 March, when the second POPs expedition began. Except for the January Racine site, higher organic carbon content (f_{oc} up to 0.21, Table 1) was found during both cruises than measured in any of the trap samples.

PCBs and PAHs in Settling Sediment, Dissolved Water, and Suspended Particles

Settling sediment fluxes (g/m^2/d) are typically highest in the winter unstratified period and lowest in the summertime when the lake is stratified and biologically productive (Eadie 1997). Fluxes of contaminants were a strong function of the sediment flux. In southern Lake Michigan, the settling flux of ΣPCBs and ΣPAHs ranged from 19.42 to 1,052 ng/m^2/d and 0.8 to 35.8 μg/m^2/d respectively. ΣPCB settling fluxes measured in this study were larger than those measured in Lake Superior (Baker *et al.* 1991), in the Canadian Experimental Lakes Area (Jeremiason *et al.* 1999), and comparable to Lake Ontario (80 to 1,900 ng/m^2/d) (Oliver *et al.* 1989) during winter unstratified conditions. During periods of low settling fluxes, very high concentrations of POPs were measured on the settling particles (ΣPCB = 700 to 3,700 ng/g and ΣPAH = 53,000 to 110,000 ng/g). Although the these POP masses exceed the blank values and method detection limits and pass quality control criteria, these concentrations are unusually large. Since the mass of sediment was very small in these cases, it is possible that a small amount of contamination interfered with the analyses.

ΣPCB dissolved phase concentrations were sig-

nificantly lower in March (128.4 pg/L average ± 41.8 pg/L std.dev.) as compared to January levels (219.3 pg/L ± 54.4 pg/L). ΣPAH dissolved-phase concentrations showed the same trend, with 10.7 ng L^{-1} ± 16.7 ng/L in March as compared to January (17.7 ng/L ± 9.5 ng/L). However, paired t-test for the sites that were sampled during both results in significant difference (95 % CI) for the dissolved phase PCBs but is not statistically significant for PAHs.

Spatial and temporal trends in the POP associated with suspended sediments (TSM) were more complicated to interpret than the dissolved phase concentrations. Near urban areas in the southwest, the concentration of particle bound POPs decreased in March, whereas concentrations went up in the southeastern region. Particulate phase PCB and PAH concentrations decreased near Chicago and Gary (by 15% to 30%), but increased substantially at the St. Joseph and Saugatuck sites (by 240% to 680%).

When normalized to organic carbon, the contaminant concentrations on settling particles are comparable to those on suspended particles (Fig. 4) (α = 0.05). These findings are markedly different than those of similar studies in Lake Superior (Baker *et al.* 1991, Jeremiason *et al.* 1998) and two lakes in The Netherlands (Koelmans *et al.* 1997), where investigators found that concentrations of PCBs and PAHs on settling particles captured in summertime were enriched 2 to 100 times relative to suspended particles. These findings suggest that the winter/springtime settling and suspended sediments are both similarly contaminated with PCBs and PAHs as a function of organic carbon content during the unstratified winter and spring transition.

DISCUSSION

Surficial bottom sediments are contaminated in southern Lake Michigan. A survey completed by the Lake Michigan Mass Balance study (U.S.EPA) indicates that surficial sediment PCB concentrations are approximately 80 ng/g at the T24 deployment site and significantly higher about 20 km north (United States Environmental Protection Agency 2001). This region of highly contaminated sediments of concentrations exceeding 150 ng/g includes the central deep basin almost to the eastern coast from the St. Joseph region to the 43.5ϒ latitude north of Muskegon. Surficial bottom sediment ΣPCB concentrations in the southern coastal and western regions of the basin are much less contami-
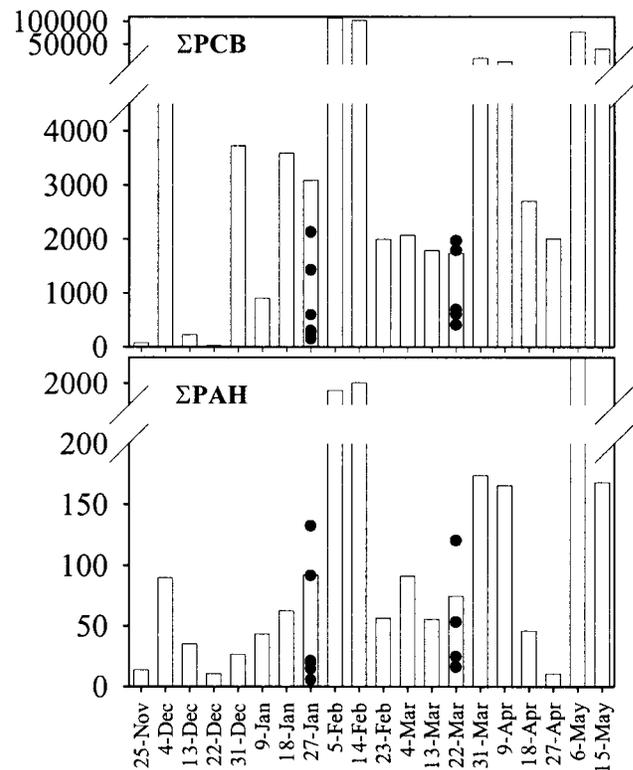

FIG. 4.   Concentrations of ΣPCB (top, in ng/g OC) and ΣPAH (bottom, in mg/g OC) on sediment normalized to organic carbon. The bars represent the concentrations on settling sediment. The solid points represent the concentrations measured in suspended sediment. The dates on the x-axis are the first day of the sediment trap collection periods. The discrete samples of suspended sediment were collected on 29 January to 1 February, and on 29 to 31 March.

nated, less than 40 ng/g (Warren and Horvatin 2001). (PAHs in surficial bottom sediments have not been reported by the LMMB). The average mass-weighted ΣPCB concentration in settling sediment from T24 was 63 ng/g over the entire sampling period and 53 ng/g during the March event, which is similar in magnitude to the surficial sediments in the mid-depth (30 to 80 m) region (Warren and Horvatin 2001). The ΣPCB concentrations in the T24 trap suggest that both local and regionally transported sediments contribute to the falling sediment at this site and are chemically representative of the total load of sediment introduced into the water column during the March 1998 period.

The March resuspension event introduced a large

TABLE 3. *Settling fluxes and predicted resuspension of contaminants in southern Lake Michigan.*

Compound	USEPA Priority Pollutants (16 PAHs) 40CFR Part 423.17	25 Nov. through 15 May Settling (ng/cm^2)	March Resuspension Event Settling (ng/cm^2)	Total Mass Reuspended During March Event (kg)
ΣPCBs	✓	6.2	2.77	398
2-Methyl naphthalene		4.4	1.6	240
1-Methyl naphthalene		2.4	0.9	126
Biphenyl		0.9	0.4	56
Acenapthylene	✓	0.9	0.4	53
Acenapthene	✓	1.7	0.8	110
Dibenzofuran		3.0	1.4	199
Fluorene	✓	4.3	1.8	265
1-Methyl fluorene		0.4	0.0	5
Dibenzothiophene		3.1	1.5	222
Phenanthrene	✓	30.8	14.6	2,137
Anthracene	✓	1.9	1.1	168
Benzo[h]quinoline		0.7	0.3	44
2 Methyl phenanthrene		8.3	3.9	564
Methylene phenanthrene		0.5	0.4	54
1-Methyl phenanthrene		17.2	7.0	1,025
3,6-Dimethyl phenanthrene		2.5	1.1	166
Fluoranthene	✓	16.8	9.8	1,432
Pyrene	✓	13.3	8.1	1,180
1,2-Benzo[a]fluoranthene		1.4	0.8	123
Retene		3.5	1.1	162
2,3-Benzofluorene		0.6	0.3	50
Benzo[a]anthracene	✓	5.8	3.5	518
Chrysene	✓	9.7	5.8	847
Benzo[b]fluoranthene	✓	7.1	3.8	559
Benzo[k]fluoranthene	✓	4.9	2.7	401
Benzo[e]pyrene		6.6	3.5	516
Benzo[a]pyrene	✓	6.4	3.3	476
Perylene		2.0	1.1	156
Indeno[123Cd]pyrene	✓	6.1	3.1	451
Dibenzo[ah]anthracene	✓	1.4	0.7	109
Benzo[ghi]perylene	✓	6.1	3.0	440
ΣPAH		174.8	87.9	12,854

amount of sediment and contaminants into the Lake Michigan water column. If it is assumed that the average ΣPCB concentration measured can be extrapolated to the whole lake, then 400 kg of ΣPCB was introduced as a result of this storm period. A similar calculation for ΣPAH compounds results in 13,000 kg ΣPAH resuspended, including 9,100 kg of PAH listed as U.S EPA priority pollutants (Table 3). While this represents a large input of toxic compounds into the lake, it is a small fraction of the total lake-wide inventory of Lake Michigan: Golden *et al* (1993) estimated the total sediment-associated burden of ΣPCBs in Lake Michigan at 75,000 kg. Although this is a small portion of the total sediment inventory, the resupended material may represent the portion of bottom sediments annually available for biological uptake. Recently, Stapleton *et al.* (2001) found that settling sediments in Grand Traverse Bay, Lake Michigan, were major sources of contaminants to the lower food chain, second only to atmospheric sources.

Partitioning of PCBs and PAHs in Water

Resuspended contaminated sediment may re-equilibrate with the water as it is transported. Several investigators have suggested that rapid re-equilibration of contaminants between suspended sediments, water, and air may drive contaminants to and from the various media as a function of particle concentration, biological productivity, and/or organic carbon loadings in the water (Dachs *et al.* 1999, Dachs *et al.* 2000, Jeremiason *et al.* 1999, Kidd *et al.* 2001, Soderstrom *et al.* 2000). Unfortunately, it is difficult to model or measure this phenonemon, because natural systems are often highly heterogeneous over time and space with respect to atmospheric contaminant concentrations, suspended particle concentrations and characteristics, and contaminant weathering on bottom sediments. This study provides a unique opportunity to examine the potential for indirect shunting of contaminants between air, water and sediment. This study is unique because of 1) the magnitude of the resuspension event; 2) the timing of the sampling directly before and after the large resuspension event; and 3) the spatial and temporal concurrence of air, water, and sediment sampling.

To examine redistribution of contaminants between these phases, the distribution of contaminants before and after the resuspension event was assessed, as was the change in absolute concentrations. The change in absolute concentrations was significant for PCBs in the dissolved phase. Since gas exchange of contaminants in southern Lake Michigan is dynamic and significant to the whole lake contaminant loadings, the potential for a change in atmospheric loading was also considered as a result of particle/water redistribution after resuspension.

The effect of the resuspension event on partitioning of POPs between dissolved and particulate phases was evaluated for all dissolved and particle pairs of water samples by calculating congener/compound-specific, organic carbon normalized, distribution coefficient, Koc (L/kg OC):

$$Koc = \frac{\dfrac{C_p}{(TSM)C_d}}{f_{oc}} \qquad (1)$$

where C_p is the particle-associated POP concentration (ng/L), C_d is the dissolved POP (ng/L), and TSM is the total suspended matter (kg/L). The partitioning of PCBs and PAHs to aquatic particles is

expected to increase as the hydrophobicity (K_{ow}) increases (Achman *et al.* 1996, Karickhoff *et al.* 1979, Schwarzenbach *et al.* 1993). This was true for the PAH compounds but less so for PCBs (Bogdan 1999). It is difficult to evaluate the equilibrium status for POPs in natural systems: kinetics (Borglin *et al.* 1996), colloids (Butcher *et al.* 1998, Tye *et al.* 1996), and other factors confound interpretation of field Koc as a measure of chemical equilibrium. Nevertheless, comparing the Koc between the two sampling periods can help determine changes in the system as caused by this large-scale resuspension event. Figure 5 illustrates the change in Koc for individual compounds before and after the event. In all cases, the plotted data for the correlation were biased *above* the 1:1 line.

This suggests that the plume event not only affected the particle/water distribution of POPs, but also that the particles suspended in the water column became enriched relative to the dissolved concentrations after the resuspension event. This is especially true for the samples collected near the eastern coast, at Saugatuck and St. Joseph. (Because of the large differences in partitioning, this finding is not particularly sensitive to f_{oc}, hence the difference not due to uncertainty in f_{oc} values.) Samples collected near the western coast have similar particle/dissolved distributions before and after the event.

It is unclear why the suspended particles collected at the eastern sites appear enriched in POPs in late March (Table 4). One possibility is that a shift in wind/wave direction on 26–27 March pushed the higher contaminated sediments from the region north of the site. Southerly winds during these 2 days created a circulation pattern with onshore flow at this site rather than the typical offshore flow. Another possibility is that suspended particles from the Gary/Chicago region were effectively moved eastward during the March storms. The Gary/Chicago region had consistently higher PCB and PAHs particulate-phase concentrations. Investigations are ongoing to evaluate the relative rates of particle/water partitioning and particle transport.

Potential for Atmopheric Deposition of PCBs and PAHs

The effect of lower dissolved phase concentrations was evaluated with respect to potential increases in net gas-phase inputs of POPs to Lake Michigan. Instantaneous net ΣPCB and ΣPAH
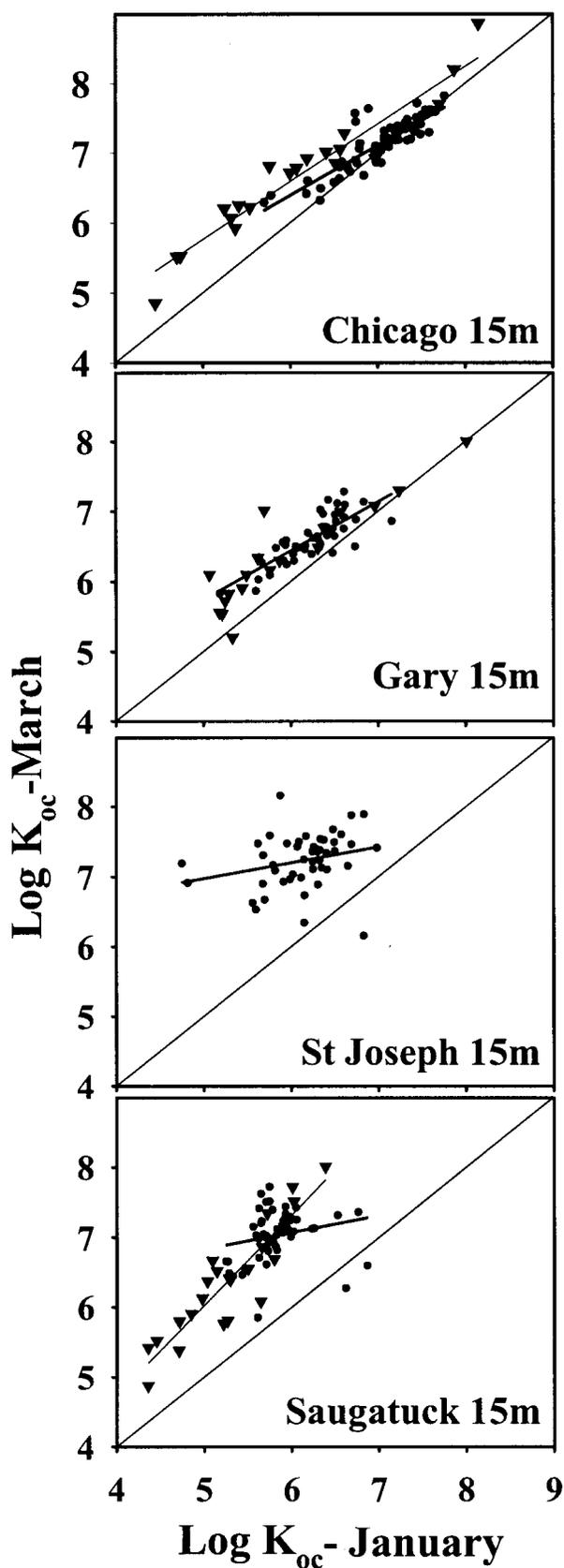
fluxes were calculated for the samples collected both before and after the resuspension event in March 1998. Net fluxes were determined on a congener/compound-specific basis. This model has been applied a number of times in the Great Lakes (Achman *et al.* 1993, Hornbuckle *et al.* 1993, Hornbuckle *et al.* 1994, Hornbuckle *et al.* 1995). Gas exchange flux determinations are based on dissolved and gaseous phase PCB and PAH concentrations (Table 5) as well as on Henry's Law constant, which were corrected for measured surface water temperatures. The effect of wind speed on the mass transfer coefficients was estimated using a two parameter Weibull distribution function. Net gas exchange of contaminants was calculated for individual PCB congeners and PAH compounds for all discrete sites sampled for this study (Livingstone and Imboden 1993). For all the sites measured, ΣPAHs varied from -2.3 to -66 μg/m^2/d, while that of ΣPCBs varied from -58 to -259 ng/m^2/d. By convention, negative values are used to describe losses from the atmosphere to the water so net *inputs* of gas phase contaminants to the water were calculated for every site. For this region, net gas-phase inputs are expected and have been reported by several researchers (Miller *et al.* 2001, Simcik *et al.* 1997, Zhang *et al.* 1999).

Potential gas-phase inputs of PCBs and PAHs increased after the resuspension event at most sites that were measured before and after the event. Wind speeds, wind direction, water temperatures, and gas-phase POPs concentrations were roughly the same during both the January and March expeditions, so this difference is not due to changes in the mass transfer coefficient or Henry's law constant, but to changes in the concentration gradient between air and water. The gas phase concentrations of PCBs and PAHs measured in this study were similar to those recently measured during other studies in southern Lake Michigan (Miller *et al.* 2001, Simcik *et al.* 1997, Zhang *et al.* 1999). Therefore, the increase in net gas-phase input is a result of the re-distribution of these compounds in the water column, and is not due to changes in air concentrations or meteorology.

Because calculated gas-phase inputs of the conta-

FIG. 5. *Log Koc—from samples collected in March versus Log Koc for samples collected in January. Triangles represent PAH compounds, solid circles represent PCB congeners.*

TABLE 4.   Chemical concentrations in water.

Sampling Location	Date	Σ_dPCB [pg/L]	Σ_pPCB [pg/L]	Σ_dPAH [ng/L]	Σ_pPAH [ng/L]
Racine 15 m	28-Jan-98	290	240	23	15
Chicago 15 m	29-Jan-98	250	350	21	22
Gary Dup1 15 m	31-Jan-98	170	44	16	3.6
Gary Dup2 15 m	31-Jan-98	160	100	20	3.3
St. Joseph 15 m	4-Feb-98	250	60	12	2.9
Saugatuck 15 m	1-Feb-98	280	28	14	1.0
Chicago 15 m	28-Mar-98	170	300	15	18
Gary 15 m	29-Mar-98	100	63	6.8	2.4
St. Joseph 15 m	29-Mar-98	160	410	17	LOST
Saugatuck 15 m	29-Mar-98	80	140	10	5.6
Basin 150 m	30-Mar-98	96	68	LOST	5.6

minants consistently increased after resuspension, this natural event may bring about a *short-term increased input of POPs* to the lake via gas exchange. This differential increase in net input is as much as 130 ng/m^2/d for ΣPCBs and 36,000 ng/m^2/d for ΣPAHs in the southwestern region (Fig. 6). The magnitude of the combined effect on increased contaminant gas-phase input to the lake was estimated by multiplying the surface area of the annual aver-

age urban air plume influence zone (1,575 km^2) (Green *et al.* 2000) by the approximate duration of the sediment resuspension event in the nearshore region (~40 days) and the average increase in gas depositional flux of PCBs (122 ng/m^2/d) and PAHs (35 μg/m^2/d) observed after the sediment resuspension event. The estimated mass of gas phase PCBs and PAHs absorbed by the lake over the 40 day resuspension period due to the air-water-sediment

TABLE 5.   Volume-based gaseous (subscript g) and airborne particulate (subscript p) phase ΣPCB and ΣPAH concentrations and ancillary measurements for air samples obtained in January and March 1998.

Sampling Transect Name	Σ_gPAH [ng/m^3]	Σ_pPAH [ng/m^3]	Σ_gPCB [ng/m^3]	Σ_pPCB [pg/m^3]	Air temp (C °)	Ave. Wind speed (m/s)[a]	Wind direction (deg)[b]	Ship Wind Speed [m/s][c]	Ship Wind Direction (deg) [c]
January Sampling Expedition									
Milwaukee/ Racine Transect	79	1.8	0.90	52	1.22	0.1	NW	7.8	E
Chicago/Gary Dup1 Transect	150	19	0.66	40	1.75	1.7	S	5.9	SW
Chicago/ Gary Dup2 Transect	120	3.9	0.65	32	1.75	1.7	S	5.9	SW
Saugatuck Transect	52	2.3	0.33	24	4.2	4.6	S	11.6	S
St. Joseph Transect	46	1.4	0.41	21	0.06	2.0	N	8.3	NE
Basin Transect [d]	53	0.8	0.38	53	–1.1	—	—	9.1	E
March Sampling Expedition									
Chicago Transect	94	2.0	0.66	14	11.0	8.6	SW	8.3	SW
Gary Transect	220	3.6	0.85	30	10.0	1.0	SW	7.2	S
St. Joseph Transect	24	5.5	0.38	14	10.0	3.5	SW	7.2	S
Basin Transect	73	4.2	0.68	15	8.5	7.1	S	9.2	S

[a] Wind speed is the average of hourly measurements reported by the nearest meteorological station during the air sampling period.
[b] Wind direction is the average of hourly measurements reported by the nearest meteorological station during the air sampling period.
[c] Meteorological data averaged from hourly measurements aboard the research vessel.
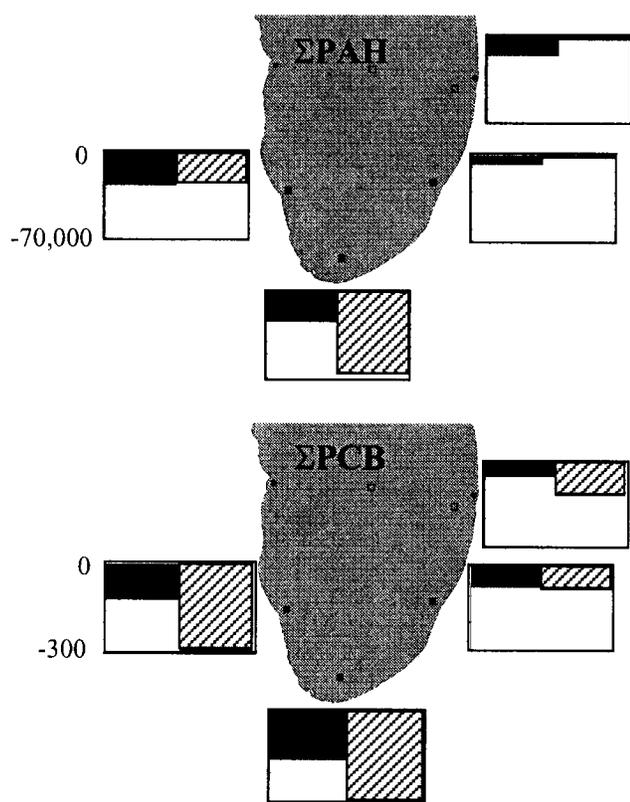[d] The buoy in south-central Lake Michigan was not deployed during the January expedition.

FIG 6. Potential gas exchange fluxes (ng/m²/d) of ΣPAHs (top plot) and ΣPCB (bottom plot). Only fluxes calculated for air and water pair collected at the same site in January and March are illustrated. The black bars are the net fluxes calculated from the January samples, the hatched bars are the net fluxes calculated from the March samples. The black point nearest each flux plot indicates the water sampling site. Air samples were collected at transects, see Figure 2. The open circles are water sampling sites that were not measured in both January and March and so are not compared in this figure. Note that in all cases, atmospheric inputs are calculated, and indicated as negative values to reflect loss from the atmosphere.

couple between the Chicago urban air plume and the sediment resuspension event is 8 kg and 2,200 kg, respectively.

The reason why the resuspension event causes such a large increase in potential net air deposition is because of the proximity of the coastal resuspension zone to the Chicago/Gary urban industrial region. In this region, concentrations of gas-phase PCBs and PAH are dramatically higher than those found in other regions of the Great Lakes (Green *et*

al. 2000, Holsen 1991, Miller *et al.* 2001, Odabasi *et al.* 1999, Offenberg and Baker 2000, Simcik *et al.* 1999, Zhang *et al.* 1999). The additional loading of PCBs and PAHs due to this coupled process represents a previously unknown load of POPs to Lake Michigan. When compared to reports of Zhang *et al.* (1999) and Miller *et al.* (2001), the predicted load of 8 kg of PCBs represents a significant portion (~57%) of the estimated entire *annual* gas phase input to proximate waters of the urban complex at the southwestern shore of the lake. This analysis of the change in gas exchange fluxes is a critical finding. If gas exchange is not considered, then one might conclude that resuspension in the Great Lakes has no effect (or a positive, cleansing effect) on contaminant cycling. While this may be true for resuspension of non-volatile contaminants (Brassard *et al.* 1997), one cannot exclude the atmospheric role when considering cycling of semi-volatile organic compounds.

CONCLUSIONS

Northerly winter/spring storms introduce a large amount of sediment into Lake Michigan's southern basin. This material is contaminated with PCBs and PAHs and are a large source of the contaminants to the lake. It is estimated that during the largest resuspension event studied, in March 1998, approximately of 400 kg of ΣPCBs and 13,000 kg ΣPAH entered the water column. However, resuspension *does not* result in an increase in (dissolved) water concentrations of contaminants from desorption from sediments. In fact, the opposite is found. The distribution of contaminants between the dissolved and particulate phases change as a result of large-scale resuspension, with a greater proportional of the total water concentration associated with the particle phase shortly after the resuspension event when the resuspended particles have settled from the near surface waters.

In the Gary/Chicago region, the redistribution from dissolved to particulate phase is primarily a result of a drop in dissolved phase concentrations, perhaps a result of transport of newly eroded and relatively clean sediments that scavenged dissolved chemical. In any case, the decrease in dissolved concentrations causes an increased potential for atmospheric deposition to the water in this region characterized by large sources of atmospheric PCBs and PAHs. This load is as much as half the annual atmospheric load to the region and would not be detected under normal monitoring conditions.

A determination of the long-term fate of resuspended contaminants is beyond the scope of this paper. It is not clear what fraction of the resuspended material is available for biological uptake and what fraction simply returns to the bottom sediments. The spatial distribution and origin of the resuspended sediments also needs further examination.

REFERENCES

Achman, D.R., Hornbuckle, K.C., and Eisenreich, S.J. 1993. Volatilization of polychlorinated biphenyls from Green Bay, Lake Michigan. *Environ. Sci. Technol.* 27:75–87.

———, Brownawell, B.J., and Zhang, L.C. 1996. Exchange of polychlorinated biphenyls between sediment and water in the Hudson River Estuary. *Estuaries* 19:950–965.

Baker, J.E., Eisenreich, S.J., and Eadie, B.J. 1991. Sediment trap fluxes and benthic recycling of organic-carbon, polycyclic aromatic hydrocarbons, and polychlorobiphenyl congeners in Lake Superior. *Environ. Sci. Technol.* 25:500–509.

Bogdan, J.J. 1999. A study of the cycling of organic contaminants in air and water: The pilot year of the Episodic Events—Great Lakes Experiment (EEGLE) Project. M.S. thesis, State University of New York at Buffalo, Buffalo, NY.

Borglin, S., Wilke, A., Jepsen, R., and Lick, W. 1996. Parameters affecting the desorption of hydrophobic organic chemicals from suspended sediments. *Environ. Toxicol. Chem.* 15:2254–2262.

Brassard, P., Kramer, J.R., and Collins, P.V. 1997. Dissolved metal concentrations and suspended sediment in Hamilton Harbour. *J. Great Lakes Res.* 23:86–96.

Butcher, J.B., Garvey, E.A., and Bierman, V.J. 1998. Equilibrium partitioning of PCB congeners in the water column: Field measurements from the Hudson River. *Chemosphere* 36:3149–3166.

Dachs, J., Eisenreich, S.J., Baker, J.E., Ko, F.C., and Jeremiason, J.D. 1999. Coupling of phytoplankton uptake and air-water exchange of persistent organic pollutants. *Environ. Sci. Technol.* 33:3653–3660.

———, Eisenreich, S.J., and Hoff, R.M. 2000. Influence of eutrophication on air-water exchange, vertical fluxes, and phytoplankton concentrations of persistent organic pollutants. *Environ. Sci. Technol.* 34:1095–1102.

Eadie, B.J. 1997. Probing particle processes in Lake Michigan using sediment traps. *Water Air Soil Poll.* 99:133–139.

———, Schwab, D.J., Assel, R.A., Hawley, N., Lansing, M.B., Miller, G.S., Morehead, N.R., Robbins, J.A., van Hoof, P.L., Leshkevich, G.L., Johengen, T.H., Lavrentyev, P., and Holland, R.E. 1996. Development of a recurrent coastal plume in Lake Michigan observed for the first time. EOS. Transactions of the American Geophysical Union. 77:337–338.

Folger, D.W., Colman, S.M., and Barnes, P.W. 1994. Overview of the southern Lake Michigan coastal erosion study. *J. Great Lakes Res.* 20:2–8.

Golden, K.A., Wong, C.S., Jeremiason, J.D., Eisenreich, S.J., Sanders, G., Hallgren, J., Swackhamer, D.L., Engstrom, D.R., and Long, D.T. 1993. Accumulation and preliminary inventory of organochlorines in Great Lakes sediments. *Water Science & Technology* 28:19–31.

Green, M.L., DePinto, J.V., Sweet, C., and Hornbuckle, K.C. 2000. Regional spatial and temporal interpolation of atmospheric PCBs: Interpretation of Lake Michigan mass balance data. *Environ. Sci. Technol.* 34:1833–1841.

Holsen, T.M., Noll, K.E., Liu, S.P., and Lee, W.J. 1991. Dry deposition of polychlorinated biphenyls in urban areas. *Environ. Sci. Technol.* 25:1075–1081.

Hornbuckle, K.C., Achman, D.R., and Eisenreich, S.J. 1993. Over-water and over-land polychlorinated biphenyls in Green Bay, Lake Michigan. *Environ. Sci. Technol.* 27:87–98.

———, Sweet, C.W., and Eisenreich, S.J. 1994. Seasonal variations in air-water exchange of polychlorinated biphenyls in Lake Superior. *Environ. Sci. Technol.* 28:1491–501.

———, Sweet, C.W., and Pearson, R.F. 1995. Assessing annual water-air fluxes of polychlorinated biphenyls in Lake Michigan. *Environ. Sci. Technol.* 29:869–877.

Jeremiason, J.D., Eisenreich, S.J., Baker, J.E., and Eadie, B.J. 1998. PCB decline in settling particles and benthic recycling of PCBs and PAHs in Lake Superior. *Environ. Sci. Technol.* 32:3249–3256.

———, Eisenreich, S.J., Paterson, M.J., Beaty, K.G., Hecky, R., and Elser, J.J. 1999. Biogeochemical cycling of PCBs in lakes of variable trophic status: A

paired-lake experiment. *Limnol. Oceanogr.* 44: 889–902.

Karickhoff, S., Brown, D., and Scott, T. 1979. Sorption of hydrophobic pollutants on natural sediment. *Water Res.* 13:241–248.

Kidd, K.A., Bootsma, H.A., Hesslein, R.H., Muir, D.C.G., and Hecky, R.E. 2001. Biomagnification of DDT through the benthic and pelagic food webs of Lake Malawi, East Africa: Importance of trophic level and carbon source. *Environ. Sci. Technol.* 35:14–20.

Koelmans, A.A., Gillissen, F., Makatita, W., and VandenBerg, M. 1997. Organic carbon normalisation of PCB, PAH and pesticide concentrations in suspended solids. *Water Res.* 31:461–470.

Livingstone, D.M., and Imboden, D.M. 1993. The nonlinear influence of wind-speed variablility on gas transfer in lakes. *Tellus* 45B:275–295.

Lou, J., Schwab, D.J., Beletsky, D., and Hawley, N. 2000. A model of sediment resuspension and transport dynamics in southern Lake Michigan. *J. Geophys. Res.* 105:6591–6610.

Miller, S.M., Green, M.L., DePinto, J.V., and Hornbuckle, K.C. 2001. Results from the Lake Michigan Mass Balance Study: Concentrations and fluxes of atmospheric polychlorinated biphenyls and *trans*-nonachlor. *Environ. Sci. Technol.* 35:278–285.

Mortimer, C.H. 1988. Discoveries and testable hypotheses arising from Coastal Zone Color Scanner imagery of southern Lake Michigan. *Limnol. Oceanogr.* 33:203–226.

Odabasi, M., Vardar, N., Sofuoglu, A., Tasdemir, Y., and Holsen, T.M. 1999. Polycyclic aromatic hydrocarbons (PAHs) in Chicago air. *Sci. Tot. Environ.* 227:57–67.

Offenberg, J.H., and Baker, J.E. 2000. PCBs and PAHs in southern Lake Michigan in 1994 and 1995: Urban atmospheric influences and long-term declines. *J. Great Lakes Res.* 26:196–208.

Oliver, B.G., Charlton, M.N., and Durham, R.W. 1989. Distribution, redistribution, and geochronology of polychlorinated biphenyl congeners and other chlorinated hydrocarbons in Lake Ontario sediments. *Environ. Sci. Technol.* 23:200–208.

Pearson, R.F., Hornbuckle, K.C., Eisenreich, S.J., and Swackhamer, D.L. 1996. PCBs in Lake Michigan water revisited. *Environ. Sci. Technol.* 30:1429–1436.

Schwab, D.J., Beletsky, D., and Lou, J. 2000. The 1998 coastal turbidity plume in Lake Michigan. *Estuarine, Coastal, and Shelf Science* 50:49–58.

Schwarzenbach, R.P., Gschwend, P.M., and Imboden, D.M. 1993. *Environmental Organic Chemistry*. New York, NY: Wiley Interscience.

Simcik, M.F., Zhang, H.X., Eisenreich, S.J., and Franz, T.P. 1997. Urban contamination of the Chicago/coastal Lake Michigan atmosphere by PCBs and PAHs during AEOLOS. *Environ. Sci. Technol.* 31:2141–2147.

_____, Eisenreich, S.J., and Lioy, P.J. 1999. Source apportionment and source/sink relationships of PAHs in the coastal atmosphere of Chicago and Lake Michigan. *Atmos. Environ.* 33:5071–5079.

Soderstrom, M., Nylund, K., Jarnberg, U., Lithner, G., Rosen, G., and Kylin, H. 2000. Seasonal variations of DDT compounds and PCB in a eutrophic and an oligotrophic lake in relation to algal biomass. *Ambio* 29:230–237.

Stapleton, H.M., Masterson, C., Skubinna, J., Ostrom, P., Ostrom, N.E., and Baker, J.E. 2001. Accumulation of atmospheric and sedimentary PCBs and toxaphene in a Lake Michigan food web. *Environ. Sci. Technol.* 35:3287–3293.

Tye, R., Jepsen, R., and Lick, W. 1996. Effects of colloids, flocculation, particle size, and organic matter on the adsorption of hexachlorobenzene to sediments. *Environ. Toxicol. Chem.* 15:643–651.

United States Environmental Protection Agency. 1997a. *Lake Michigan Mass Balance Study Methods Compendium. Volume 1: Sample Collection Techniques.* USEPA, Great Lakes National Program Office, Chicago, Illinois, June, 1997 EPA 905-R-97-012a.

_____. 1997b. *Lake Michigan Mass Balance Study Methods Compendium. Volume 2: Organic and Mercury Sample Analysis Techniques.* USEPA, Great Lakes National Program Office, Chicago, Illinois, June, 1997 EPA 905-R-97-012b.

_____. 2001. *Results of the Lake Michigan Mass Balance Study: Polychlorinated Biphenyls and trans-Nonachlor Data Report, Chapter 6: Sediments.* Great Lakes National Program Office, U.S. EPA, Chicago, IL, Dec. 2001 905R-01-011.

Warren, G., and Horvatin, P. Lake Michigan Mass Balance Study—2001. In *Abstracts 44th Annual Conference on Great Lakes Research*. Green Bay, WI, International Association for Great Lakes Research.

Weyhenmeyer, G.A., Meili, M., and Pierson, D.C. 1995. Simple Method to Quantify Sources of Settling Particles in Lakes—Resuspension Versus New Sedimentation of Material From Planktonic Production. *Mar. Freshw. Res.* 46:223–231.

Winkelman, A.G., Stabenau, E.R., and Eadie, B.J. 1998. *Particle Size Distribution and Concentration of Total Suspended Matter in Southern Lake Michigan: January 28–February 10, 1998.* Great Lakes Environmental Research Laboratory/National Oceanic and Atmospheric Administration, Ann Arbor, Michigan, ERL GLERL-105.

Zhang, H., Eisenreich, S.J., Franz, T.R., Baker, J.E., and Offenberg, J.H. 1999. Evidence of Increased Gaseous PCB Fluxes to Lake Michigan from Chicago. *Environ. Sci. Technol.* 33:2129–2137.