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The Magnitude and Origin of Contaminants Resuspended in Southern Lake Michigan

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Magnitude and origin of polychlorinated biphenyl (PCB) and dichlorodiphenyltrichloroethane (DDT) compounds resuspended in southern Lake Michigan

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Polychlorinated biphenyls (PCBs) and dichlorodiphenyltrichloroethane (DDT) compounds are introduced into the water as a result of large-scale storms and sediment resuspension in the southern basin of Lake Michigan. Settling and suspended sediments, as well as air and water samples, were collected in southern Lake Michigan over a 12 month period. Analysis of contaminant fluxes on settling particles shows that approximately 370 kg of PCBs and 110 kg of DDT compounds are resuspended in southern Lake Michigan during a single basin-wide event (January 1999). Examination of contaminant signals indicates strong regional and temporal source-receptor relationships between settling, suspended, and surficial sediments. The settling, suspended, and bottom surficial sediments in the shallow waters of the southern coastal region are enriched in lower molecular weight PCBs. The sediments in the water column and on the lake bottom in the deeper regions are enriched in higher molecular weight PCBs. Furthermore, falling sediments collected in the deeper regions of the lake are enriched in 4,4'-DDT. The unique contaminant signal in deep water regions is surprising and suggests a source/receptor relationship among the bottom sediments and the sediments suspended and settling above them.

INDEX TERMS: 4863 Oceanography: Biological and Chemical: Sedimentation; 4857 Oceanography: Biological and Chemical: Pollution; 1615 Global Change: Biogeochemical processes (4805); 9345 Information Related to Geographic Region: Large bodies of water (e.g., lakes and inland seas); KEYWORDS: persistent organic pollutants, sedimentation, principal component analysis, non-point source


1. Introduction

The bottom sediments of the Great Lakes are contaminated with persistent organic contaminants, including polychlorinated biphenyls (PCBs) and dichlorodiphenyltrichloroethane (DDT) compounds [Eisenreich et al., 1989; Golden et al., 1993; Oliver et al., 1989; Pearson et al., 1997; Schneider et al., 2001; Wong et al., 1995]. Resuspension of sediments may be an important continuing source of these compounds and could explain why concentrations of these compounds in Great Lakes fish continue to exceed the FDA action level and water quality objectives defined by Annex I of the Great Lakes Water Quality Agreement. PCBs and DDT both contribute to states’ and provinces’ decisions to set fish consumption advisories, and for good reason. PCBs are implicated as endocrine disrupters [Kester et al., 2000; Wolkowick et al., 2001], immuno-depressants [Weisiglusa-Kuperus et al., 2000], and known neurotoxins [Schantz et al., 2003]. Consumption of PCB-contaminated fish is correlated with learning disabilities and developmental delay in the children born to Great Lakes’ fish eaters [Buck et al., 2000; Jacobson and Jacobson, 1996]. DDT is no longer viewed as a probable cause of breast cancer [Safe, 2000; Snedeker, 2001], but studies have shown that the DDT and its degradation products are endocrine disrupters [National Research Council, 1999].

Lake Michigan has a long and intense history of exposure to these contaminants. Golden et al. [1993] estimated the total inventory of PCBs and DDT (parent and major degradation products) compounds in the lake to be 75,000 kg and 35,000 kg, respectively. The majority of these contaminant burdens are considered permanently buried, but some fraction of the surficial sediments may...
be reintroduced in the water column through resuspension. Of greatest concern is those regions of the lake where concentrations of PCBs and DDT in surficial sediment remain very high. A detailed survey of surficial sediment PCB concentrations was conducted as part of the U.S. Environmental Protection Agency’s Lake Michigan Mass Balance/Environmental Monitoring and Assessment (LMMB/EMAP) field study in 1994–1995. The results show that the highest concentrations of surficial PCBs in the southern basin are found in the regions of highest sediment accumulation, primarily at depths exceeding 60 m [U.S. Environmental Protection Agency (U.S. EPA), 2001] in the central and eastern region (Figure 1). However, this distribution does not follow the lake bathymetry. Currents and prevailing circulation trends also have major impacts on the contaminant accumulation. An earlier survey of surficial sediments conducted in 1975 report similar distributions of DDT [Frank et al., 1981].

[4] Bottom sediments may be resuspended during the wintertime unstratified period. This may be an important source of contaminants to the lake. However, it is not clear if the most highly contaminated sediments, found in the deepest waters, are accessible to mixing under any conditions. Bottom sediments in more than 60 m of water can occasionally be resuspended under intense mixing during winter storms when the lake is isothermal [Baker et al., 1991; Eadie et al., 1984; Hawley and Lee, 1999; Hawley et al., 1996; Schneider et al., 2002] but coastal surficial sediments are expected to be the most important source of the sediment transported to in the open and deep waters of southern Lake Michigan [Eadie et al., 1996, 2002].

Nearshore sediments are regularly resuspended, transported laterally and deposited into deeper regions. For example, in reporting a detailed field investigation of sediment transport in southern Lake Michigan, Hawley and Lee [1999] concluded that coastal sediments transported laterally is the major source of the settling sediment at the deeper sites in southern Lake Michigan. Also, Edgington and Robbins [1990], using radionuclide tracers, described how the southern basin of Lake Michigan is subjected to intense focusing processes that move sediment to deep regions.

[5] The first objective of this study was to determine the magnitude of organic contaminants resuspended in southern Lake Michigan, primarily during the winter period when the lake is not thermally stratified. The magnitude of resuspended PCB and DDT contaminants was examined using sequencing sediment traps deployed in the region. The second objective was to determine the source of the contaminated, resuspended sediment. To accomplish this, we exploit subtle but significant differences in PCB congener profiles. We examined the PCB congeners and DDT compounds in settling sediment, suspended sediment, and surficial bottom sediment.

[6] A series of field sampling efforts was conducted in southern Lake Michigan. The sampling strategy was designed to study large-scale resuspension events and organic contaminants. Sampling included: (1) settling trap material collected over one year in 1998–1999; (2) water, air, and suspended sediments collected during four sampling expeditions during the unstratified period of the same year. These samples are compared with data from a large survey of Lake Michigan surficial sediment. The locations of the
2. Methods

Large lake.

been used to examine the magnitude and source of sediments and contaminants resuspended in the open waters of a large lake.

2. Methods

This work was conducted as part of Episodic-Events-Great Lakes Experiment (EEGLE), a multi-investigator and multidisciplinary study of resuspension in southern Lake Michigan. The EEGLE project included deployment of sequencing sediment traps to capture settling sediment coordinated with a series of sampling expeditions timed to capture changes in the lake as a result of large-scale sediment resuspension. The sampling expeditions included sampling of air (vapor phase is reported here), and water (separated into suspended sediment and dissolved fractions).

Sequencing traps were successfully deployed at four locations in southern Lake Michigan, and collected samples between May 1998 and May 1999. The locations and deployment periods of the trap collection at these stations is listed in Table 1. From the two most northern stations (T12 and T28; Figure 1), the traps were successful in collecting the full carousel of 23 samples. Because of problems caused by zebra mussel contamination, the deployment at station T20 yielded only nine noncontinuous samples, while a battery failure at station T33 caused the trap to stop sampling after the twelfth bottle. Bottles from traps deployed at three other locations experienced problems with the batteries and the bottles from these sites were used as blanks. Details of the sediment trap collection procedures has been described elsewhere [Bogdan et al., 2002; Muzzi and Eadie, 2004; Schneider et al., 2002].

Water and air samples were collected during four sampling expeditions in the winter and spring of 1999. The water samples each consisted of 500 to 1200 L of lake water collected at less than 2 m depth. The particulate and dissolved phases in water and the particulate and gas phases in air are operationally defined by the separation on glass fiber filters and XAD-2 resin in series. These methods have been described elsewhere [Achman et al., 1993; Hornbuckle et al., 1993; U.S. EPA, 1997].

All sediment trap, suspended, dissolved-, and vapor-phase samples collected were analyzed for a total of 98 individual PCB congeners and/or congener groups and 25 pesticides at the University of Iowa. The extraction of the sediment trap bottles with a 1:1 acetone:hexane mixture used a repetitive sonication and centrifugation procedure to allow the analysis of the solids as well as the overlying water and chloroform preservative.

Table 1. Dates and Locations of Sequencing Sediment Trap Deployments

<table>
<thead>
<tr>
<th>Station</th>
<th>Deployment Dates</th>
<th>Trap Depth, m</th>
<th>Station Depth, m</th>
</tr>
</thead>
<tbody>
<tr>
<td>T12</td>
<td>June 1998 to May 1999</td>
<td>30/160</td>
<td></td>
</tr>
<tr>
<td>T20</td>
<td>October 1998 to March 1999</td>
<td>12/25</td>
<td></td>
</tr>
<tr>
<td>T28</td>
<td>June 1998 to May 1999</td>
<td>30/110</td>
<td></td>
</tr>
</tbody>
</table>

*Not a continuous chronology of samples.
surrogate recovery. \( \Sigma \text{PCB} \) and \( \Sigma \text{DDT} \) in the field blanks, at the 95% confidence level, were 1.8 ng ± 0.72 ng and 0.62 ng ± 0.66 ng, respectively.

[16] A quantitation limit (QL) for the settling sediment data was determined to distinguish sample compound masses that are significantly larger than the field blanks. The QL was calculated as three times the standard deviation of the compound mass in the field blanks. The QL was determined on a congener or compound specific basis and compared to the contaminant masses in the samples. As a result of high QL values, 10 PCB congeners were excluded from the entire data set. For the remaining congeners, the QL for \( \Sigma \text{PCB} \) was 5.0 ng/sample. For the DDT group, the QL ranged from 0.10 ng/sample for 4,4'-DDD to 4.4 ng/sample for 4,4'-DDE. Figures in the auxiliary materials illustrate the low level of potential contamination in the settling sediment.

2.2. Statistical Analysis (Normalization, SAS)

[17] PCB congener patterns in 164 samples were analyzed for correlations using principal component analysis (PCA) in SAS Analyst, version 8 (SAS Institute, Cary NC) with Varimax rotation. Prior to PCA analysis, each PCB congener mass was normalized to the total PCB concentration in the sample. PCA was performed with a normalized matrix of the PCBs in 16 suspended sediments, 18 dissolved phase, 12 vapor phase, 67 settling sediment traps, 47 bottom surficial sediments, and 4 Aroclor mixtures. Some deletion or summation of specific congeners was necessary to match the sets, resulting in a suite of 76 congener groups (see table in auxiliary material). Principal component analysis reduces the number of variables that describe the data set and identify a few variables or dimensions (eigenvectors) that adequately distinguish groups of samples from each other [Dunteman, 1989]. Without such a method it would be difficult, if not impossible, to compare the patterns of the 76 PCB congeners in the 164 samples to one another.

3. Results and Discussion

3.1. Resuspension of PCBs and DDT Compounds

[18] Sediment and contaminant settling fluxes exhibit highly episodic behavior in southern Lake Michigan, corresponding to regional storms during the unstratified period. In the 1998–1999 unstratified season, five resuspension events were recorded by the sequencing sediment trap systems (Figure 2). Two of the events were recorded at all the operating trap deployment sites: the first in late December through early January and the second in the first week of March. The December/January event was particularly strong. It was triggered by a storm in the eastern region on 30 December followed by another storm in the western region on 2 January (UCAR Nowcast archives). Recorded wave heights from tripods deployed near St. Joseph, Michigan City and Milwaukee reached a peak on 30 December 1998 (Hawley, NOAA/GLERL, personal communication), the largest between October and May of that year. On the same day, a resuspension event was recorded by Acoustic Doppler Current Profiler (ADCP) deployed near St. Joseph [Beletsky et al., 2000].
The mass of contaminants associated with the December/January event can be estimated from the settling fluxes, the resuspension event period, and relevant lake area coverage. The latter is unknown, but is illustrated by calculating a contaminant mass flux as if each trap were representative of the entire region. Table 2 includes the contaminant fluxes and length of the December/January event period for each trap. This method has major uncertainties that cannot be quantified, especially since the western region included no traps and we do not know if measured chemical fluxes are representative. However, if each trap were representative of the entire southern region, then $\Sigma$PCB resuspension ranges from 250 to 540 kg of material resuspended. $\Sigma$DDT resuspension ranges from 22 to 310 kg of material resuspended. Clearly PCBs have a more uniform impact, varying by about a factor of two, than does DDT - perhaps reflecting more years of input and more uniform impact, varying by about a factor of two, than DDT - perhaps reflecting more years of input and varying by about a factor of two, than does DDT - perhaps reflecting more years of input and potentially being a less well-mixed pool.

Table 2. $\Sigma$PCB and $\Sigma$DDT Resuspension During the December/January Event

<table>
<thead>
<tr>
<th>Site</th>
<th>Days$^a$</th>
<th>Start Day$^b$</th>
<th>End Day$^b$</th>
<th>$n^c$</th>
<th>$\Sigma$PCB</th>
<th>$\Sigma$DDT</th>
</tr>
</thead>
<tbody>
<tr>
<td>T12</td>
<td>48</td>
<td>1 Jan. 1999</td>
<td>18 Feb. 1999</td>
<td>4</td>
<td>15,947</td>
<td>10657</td>
</tr>
<tr>
<td>T28</td>
<td>36</td>
<td>1 Jan. 1999</td>
<td>6 Feb. 1999</td>
<td>3</td>
<td>18,727</td>
<td>4716</td>
</tr>
<tr>
<td>T20</td>
<td>36</td>
<td>20 Dec. 1998</td>
<td>25 Jan. 1999</td>
<td>3</td>
<td>8,547</td>
<td>1341</td>
</tr>
</tbody>
</table>

$^a$Number of days of elevated sedimentation during the December/January event period.
$^b$Inclusive dates of sediment trap bottle collection during the December/January event period.
$^c$Number of sediment trap bottles recording the December/January event period.

The resuspended sediment occurs. Even if desorption is thermodynamically feasible, there may be insufficient time before the sediment returns to the lake bottom. It is possible that there is no net change in contaminant burden, or in ecosystem exposure, especially since biological productivity is very low during the periods of the most active resuspension.

3.2. Contaminant Concentrations on Settling and Bottom Sediments

The variation in contaminant concentrations in settling sediments may be used to identify sources of the resuspended sediments. For some compounds the variation is related to major resuspension episodes. For example, $\Sigma$DDT concentrations in settling sediment increase during major resuspension events ($p = 0.001$ for the entire data set; Figure 3). This effect was not statistically significant for $\Sigma$PCBs (Table 3), consistent with other reports [Bogdan et al., 2002; Schneider et al., 2002]. Concentrations of $\Sigma$DDT in settling sediment ranged from $<2$ ng g$^{-1}$ to 44 ng g$^{-1}$, with the highest values observed in January to March at the central T12 trap site. These are similar to concentrations reported for bottom sediment in southern Lake Michigan. Sampling of bottom surficial sediments completed in 1975 [Frank et al., 1981] and in 1991 [Golden, 1994; Golden et al., 1993] report concentrations ranging from 20 to 70 ng g$^{-1}$ in the area below the T12 trap deployment site and exceeding 40 ng g$^{-1}$ in the eastern region near site T33.

A Principal Component Analysis (PCA) was conducted to examine similarities between sediment samples collected throughout the southern Lake Michigan basin. PCBs are very good analytes for the PCA method because each sample is described by many variables (congeners) with patterns that are difficult to identify or group by inspection. This method has been used to examine sources of PCBs in biota [Vuorinen et al., 1997], sediments [Ashley and Baker, 1999], water [Bremle and Larsson, 1997; Offenberg and Baker, 2000] and air [Helm and Bidleman,
2003]. In this study, PCA successfully identified PCB congener pattern differences and similarities between bottom sediments, suspended sediments, settling sediments, water and air.

[24] The first two principal components of the normalized matrix, which describe 72% of the total variability in the data, are plotted in Figure 4a. The first principal component (PC1) is an apparent measure of the molecular weight, hydrophobicity, and/or chlorination of the PCBs in a sample. Samples with congener distributions dominated by lower chlorinated congeners exhibit lower PC1 scores. Gas-phase and dissolved-phase samples are an example, in this study and others, of congener distributions enriched in the less chlorinated congeners (PCBs with two, three, and four chlorines). A correlation between chlorine number and PC1 was found and is illustrated in the auxiliary material. Chlorine number, \( \text{Cl}\# \), is defined in equation (1) and the results for various sample matrices are illustrated in Figure 5.

\[
\text{Cl}\# = \frac{\sum \text{congener}_i \cdot \text{Cl}_i}{\sum \text{PCB}}.
\]

[25] Congener, is the mass of congener \( i \) in the sample, \( \text{Cl}_i \) is the number of chlorines in congener \( i \), \( \sum \text{PCB} \) is the sum of the 76 congeners masses in the sample. For the total data set, the correlation between PC1 and \( \text{Cl}\# \) is significant but only describes about half of the variability (\( r^2 = 0.45 \)). For a subset of only the sediment samples plotted in Figures 4b–4d, the correlation is much stronger (\( R^2 = 0.86 \), see also auxiliary material). We conclude PC1 is primarily, but not exclusively, identifying the pattern differences due to PCB chlorination, molecular weight, or cocorrelated compound properties.

[26] The second principal component (PC2) best describes pattern differences for trap samples collected during periods of low resuspension and settling. This set of samples is labeled ‘low-sed settling’ in Figure 4a. These are trap samples that collected a small amount of settling sediment (<2 g m\(^{-2}\) d\(^{-1}\)). PC2 may be related to analytical

![Figure 3.](image)

Figure 3. Concentrations (ng/g) of the DDT compounds measured in settling sediment. The dotted line is the downward flux of sediment.

Table 3. Measured Concentrations of Contaminants in Sediment (Average ± Standard Deviation)

<table>
<thead>
<tr>
<th>Study Period</th>
<th>Sediment Matrix</th>
<th>( \sum \text{PCB} ), ng/g (All for the Same 76 Congeners)</th>
<th>( \sum \text{DDT} ), ng/g</th>
</tr>
</thead>
<tbody>
<tr>
<td>1998–1999</td>
<td>settling sediment</td>
<td>60 ± 34</td>
<td>16 ± 12</td>
</tr>
<tr>
<td>1998–1999</td>
<td>high sed(^a)</td>
<td>110 ± 150</td>
<td>8 ± 8</td>
</tr>
<tr>
<td>1999</td>
<td>suspended sediment</td>
<td>24 ± 20</td>
<td>6 ± 10</td>
</tr>
<tr>
<td>1994–1995</td>
<td>surficial bottom sediment(^b)</td>
<td>62 ± 58(^*)</td>
<td>NA(^d)</td>
</tr>
<tr>
<td>1991</td>
<td>surficial bottom sediment(^d)</td>
<td>126 ± 4</td>
<td>69 ± 7.1</td>
</tr>
</tbody>
</table>

\(^a\)During periods of high sediment settling fluxes (>2 g m\(^{-2}\) d\(^{-1}\)).
\(^b\)During periods of low sediment settling fluxes (<2 g m\(^{-2}\) d\(^{-1}\)).
\(^d\)DDT concentrations are not available for 1994–1995 surficial sediment.
\(^*\)Golden [1994] site 18, near site 34 of this study, four 1 cm increments.
sensitivity and detection limits. The samples with low PC2 scores have a large number of undetected congeners (see auxiliary material). However, PC2 is still a valuable component as it allows a statistical explanation for removing the ‘low-sed settling’ samples from additional analysis.

Figures 4b–4d are plots of the same results as in Figure 4a, with sample groups plotted separately for clarity. Three major findings result. First, the PCA consistently groups together those sediment samples collected in the deep waters of the southern basin, regardless of the sediment’s ‘state’ of suspension, settling or accumulation in bottom sediments. This grouping includes settling sediment collected at site T12 during resuspension events, suspended sediment collected at the DWS site, and surficial bottom sediment collected in deep water (see samples with PC1 scores near 5 in Figures 4b–4d). In all these cases, the congener distributions are enriched in highly chlorinated congeners (high PC1 scores). This type of signal would be expected from older sediment. Jeremiason et al. [1994], suggested that weathering of PCB patterns in the Great Lakes is primarily a function of volatilization of congeners rather than microbial degradation or sediment burial and results in enrichment of highly chlorinated PCBs and loss of the more volatile congeners. This is a subtle effect, however, and would probably not be detected without the PCA. (Congener distributions for representative samples and homolog distributions for all samples are provided in the auxiliary material. Concentrations of surficial sediment for each grouping in Figure 4d are also provided in the auxiliary material.)

Second, the PCA groups together those sediment samples collected in shallower waters of the southern region of the lake, regardless of the sediment sample matrix (suspended, surficial, or settling). Of these six sites, site 7 has a very high \( \sum PCB \) concentration (107 ng/g). The remaining sites of this ‘shallow’ group have much lower \( \sum PCB \) concentrations (7.7 ± 9.1 ng/g). In all these cases, the congener distributions are enriched in the less chlorinated congeners (low PC1 scores).

Thirdly, the bottom surficial sediment samples collected on the eastern and western coastal regions are indistinguishable with respect to PCB patterns, unlike the deep water sites and the shallow coastal region (Figure 4d). The \( \sum PCB \) concentrations are statistically different, however, even if the patterns are not. PCB concentrations are significantly higher (p < 0.001) among surficial sediments collected in the eastern region (87 ± 60 ng/g) as compared to those in the western region (13 ± 7.6 ng/g). Our trap
deployment and sampling expeditions did not include the western coastal region so settling sediment cannot be directly compared with the bottom sediment in this region.

3.4. PCB Congener Signals in Sediments

Vary Over Time

[30] PCB congener signals in the settling sediments change over the course of the year. Figure 6 describes this change in PCB signal for all four traps. The PCB signal is represented as the first principal component (PC1) so that it can be plotted two dimensionally. At sites T12 and T28, the sites with the complete annual record, this change is most evident. Since PC1 is primarily a function of a shift in molecular weight, or chlorination, the resuspension events are associated with heavier PCBs. PC1 is not completely a function of molecular weight, however. As discussed above, during the stratified period of low rates of sediment settling, the first principal component is not well correlated with molecular weight. So the PC1 scores during the spring, summer and early fall are not necessarily representative of molecular weight. In general, however, large storms resuspend high molecular weight PCBs, especially in the deeper regions of the lake. At other times of the year, the background PCBs in the suspended and settling sediments consist of more of the lower chlorinated congeners.

[31] The PCB signal starts to change prior to the major settling periods. At both the T12 and T28 sites, a series of steadily increasing PC1 scores are observed during the four to six weeks prior to the major event. No corresponding increase in settling fluxes is observed. At this time, the lake is still stratified at T12, but coastal regions of the lake are beginning to turn over. Particles collected in traps during this pre-event time may be reflective of coastal resuspension processes that move sediment from the coastal margins into the deep lake.

4. Conclusions

[32] During intense resuspension, the sediments on the bottom of southern Lake Michigan share the same chemical signal as the suspended and settling sediments in the water column above or nearby. This indicates a common source of sediments in each region, which we have classified as shallow, deep and a group of ‘other’ sites located in both the eastern and western coastal region.

[33] The sediments in the shallow regions of the lake have a distinct chemical signal. The settling, suspended and bottom surficial sediments in the shallow waters of the southern coastal region are more enriched in lower molecular weight PCB congeners than elsewhere. There are several known sources of contaminated sediment in the extreme southern region of the lake, including the Grand Calumet River and Indiana Harbor Ship Canal [U.S. EPA, 2003]. While Offenberg and Baker [2000] found that the Gary region was a source of higher PCB content particles, we found that the bottom surficial and settling sediments are not unusually contaminated. We did not find evidence of major sediment release from the canal during resuspension events, probably because flows from the canal are well controlled. Sediment in this region does not accumulate. It is rapidly transported to deeper regions. Therefore the chemical signal is reflective to background inputs, including atmospheric deposition [Green et al., 2000; Offenberg and Baker, 2000; Paode et al., 1998; Simcik et al., 1999].

[34] The sediments in the deep waters of the open lake have a distinct chemical signal. The sediments in settling, suspended, and surficial bottom sediments are enriched in higher molecular weight PCB congeners and the settling sediment is more enriched in 4,4'-DDT. This chemical signal is consistent with older sediments that have retained the less volatile contaminants and lost the lighter and more volatile compounds. The presence of 4,4'-DDT is notable. Detection of 4,4'DDT only in the central T12 trap suggests that deeper sediments were accessible only during the most intense storm related turbulence.

[35] The unique contaminant signal in deep water regions is surprising and suggests a source/receptor relationship among the bottom sediments and the sediments suspended and settling above them. Two explanations are proposed.
First, the sediments in the water column may have been resuspended from the surficial bottom sediments directly below them. While this is a reasonable explanation for the signals observed in the shallower regions, it is less plausible for those deeper regions of the lake. Sediments settling in deep regions are expected to originate from surficial sediments in coastal regions. The average depth of the sediments with similar chemical signals in this group is 120 m, a depth considered inaccessible to resuspension. In fact, there is clear satellite evidence that transport of some sediment from coastal regions to the open lake is ongoing during intense resuspension events [Eadie et al., 2002].

A second explanation is that the spatial differences between sediments in deep and shallow waters are a result of sorting by particle size or transport. Particle sorting by size is likely to occur during resuspension events: Larger particles fall more rapidly and close to their sources while finer particles fall slower and further from their source [Digiano et al., 1993; Roberts et al., 1998]. Higher molecular weight organic contaminants tend to associate with finer particles of higher organic carbon content. The combined effect of these factors may result in spatial and temporal differences in PCB signatures in the lake. We cannot address this fully as the particle size distributions and organic carbon contents by size in all the settling, suspended, and surficial sediments of our study are not available. However, we do not think that this effect can completely explain our observations. A PCB pattern difference as a result of particle size would have resulted in significant differences between the PCB patterns in settling, suspended and surficial sediments. In fact, we observe strong similarities in these sediments, when paired in time and space. We therefore conclude that deep and highly contaminated sediments are resuspended during high-intensity large-scale storms in unstratified waters.

[37] Acknowledgments. For assistance in sample collection, the authors thank the captain and crew of the U.S. EPA R/V Lake Guardian. The authors acknowledge Brian Conolly at the University of Iowa for his analytical assistance with the settling and suspended sediments, air and water sample analysis. Although the surficial sediment data were supplied by the Great Lakes National Program Office of the United States Environmental Agency (GLNPO/U.S. EPA), the authors acknowledge Patricia Van Hoof for PCB analysis of the surficial sediment samples and discussions of this manuscript. Financial support for this work was provided by GLNPO/U.S. EPA (GL995156-01-0, GL985762-01-0). The trap and water sampling work was conducted as part of the EEGLE (Episodic Events - Great Lakes Experiment) study, which is supported by the National Science Foundation (OCE-9726680) and the National Oceanic and Atmospheric Administration Coastal Ocean Program and GLERL.

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