# Analyzing the Spatial Distribution of Sediment Contamination in the Lower Great Lakes

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Despite significant reductions in contaminant concentrations over the past 30 years, large areas within Lake Erie and Lake Ontario still exceed Canadian sediment quality guidelines. Hexachlorobenzene (HCB), polychlorinated biphenyls (PCBs), lead (Pb) and mercury (Hg) can persist for long periods of time in the environment and cause significant ecosystem damage. Analyses of the spatial distribution of these contaminants were carried out using a GIS-based kriging technique. Initially, statistically valid results were obtained for three of four contaminants in Lake Erie (HCB, Pb, Hg) and two of four (HCB, Hg) in Lake Ontario. Acceptable concentration estimates were subsequently achieved for all contaminants following log-normal transformation kriging analyses. In general, the concentration of contaminants was lower in sediment collected in Lake Erie than in Lake Ontario. In many areas of Lake Erie, the concentrations were under both the probable effect level (PEL) and the threshold effect level (TEL), which relate to the severity of adverse biological effects that may be expected. Greater concentrations of these contaminants were observed in Lake Ontario sediments, which can be partly explained by the bathymetry and current circulation patterns in the lake.

Key words: spatial distribution, interpolation, kriging, log-normal, sediment contamination

## Introduction

The Great Lakes of North America extend 1200 km from west to east and their surrounding area is home to approximately one-quarter of Canada's population and more than one-tenth of the United States' population (U.S. EPA 1995). The lakes together with their connecting rivers/channels provide water for industrial production and a wide range of other consumptive uses including drinking water. They also function as transportation routes for natural resources and manufactured goods and provide a source for hydropower generation. Furthermore, the lakes are utilized for recreational activities including swimming, fishing and boating.

Some of the world's largest concentrations of industrial activity take place in the Great Lakes basin. Production is particularly concentrated in shoreline areas and amounts to 7% of the total U.S. production. The area is equally important for farming, supporting approximately 25% of the Canadian agricultural output (U.S. EPA 1995). The Canadian and American governments have recognized the need to respond to pollution concerns within the Great Lakes basin. In 1987, they signed the amended Great Lakes Water Quality Agreement. Among the recommendations in the agreement was the creation of a Lakewide Management Plan for each lower lake, adopting an ecosystem approach to address environmental issues (Lake Erie Lakewide Management Plan 2000; Lake Ontario Lakewide Management Plan 1998).

Epstein (2002) states that with the increase of industry and the boom in the production of synthetic organic chemicals and metals since the 1940s, a slow contamination throughout the Great Lakes basin began and indications of dangerous and substantial levels of toxicity became apparent. Runoff containing these substances as well as many other toxic pollutants, such as trace metals, has had many negative effects on the ecosystem (Forsythe 2004). Small particles in the water are very efficient at absorbing certain types of pollutants. These chemicals are associated with particles that sink to the bottom of the lakes and rivers. The combination of many of these polluted particles makes up what is known as contaminated sediment (Ashworth 1986). Sediment can be subsequently resuspended through the processes of harbour dredging, shipping and navigating, wind and wave action caused by storms, and biotic disturbances (U.S. EPA 1995). The negative consequences of using specific chemicals have led to banning their further use, while others have been voluntarily phased out.

Ouyang et al. (2003a) state that sediment contamination may pose a significant hazard to aquatic life. As a result of sediment contamination, water quality is reduced and subsequently, the population of fish and wildlife is jeopardized. Additionally, human health and development opportunities suffer (Currie 1994). For example, it has

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been shown that the concentration of contaminants in top predators is magnified due to bioaccumulation. This contamination has been linked to genetic deformities in cormorants (e.g., crossed bills), thinning of osprey and herring gull eggshells causing a decline in birth rates, and tumours in top predatory fish, such as the sauger (U.S. EPA 1995). An additional loading of pollution enters the water system from the atmosphere directly through precipitation. This is especially important due to the large surface area of the Great Lakes and their drainage basin and the prevailing annual wind patterns which bring emissions from distant industrial/manufacturing and coal-fired power generation into the area.

The study of contaminated sediments is one method to obtain information on water quality. The results can lead to assessment, management, remediation and restoration efforts in areas of concern. More specifically, determination of sediment chemistry enables the creation of guidelines and threshold levels within ecosystems and can lead to proper regulation of the waterways to ensure the health of the ecosystem (Crane and MacDonald 2003). Guidelines have been developed with respect to contaminant concentrations by the Canadian federal government. They specify the threshold effect level (TEL) and probable effect level (PEL) for sediment contamination. The TEL refers to the concentration below which adverse biological effects are expected to occur rarely, while the PEL defines the level above which adverse effects are expected to occur frequently (Canadian Council of Ministers of the Environment 1999). The guidelines for the contaminants examined in this study are outlined in Table 1.

## **Materials and Methods**

## Study Area

Knowledge of lake bathymetry and circulation processes is important for understanding the pattern of sediment distribution within the Great Lakes. Studies relating to sediment characterization, lake bathymetry, and current circulation were carried out by Thomas et al. (1976) for Lake Erie and Thomas et al. (1972) for Lake Ontario. The time-averaged circulation pattern in Lake Erie is characterized by eastward flowing currents along the

**TABLE 1.** Selected contaminants and federal guidelines(source: after Canadian Council of Ministers of theEnvironment 1999)

| Contaminant                      | TEL       | PEL        |
|----------------------------------|-----------|------------|
| Hexachlorobenzene (HCB)          | 20 ng/g   | 480 ng/g   |
| Polychlorinated biphenyls (PCBs) | 34.1 ng/g | 277 ng/g   |
| Lead (Pb)                        | 35 μg/g   | 91.3 μg/g  |
| Mercury (Hg)                     | 0.17 μg/g | 0.486 μg/g |

northern and southern shorelines with a westward flowing current in the middle of the lake. The majority of water circulation in Lake Ontario occurs in a counterclockwise direction with a small clockwise gyre in the northwestern part of the lake (Beletsky et al. 1999).

Lake Erie is the smallest of the Great Lakes (by volume) at 484 km<sup>3</sup>, which can be attributed to a shallow average depth of 19 m, and a relatively small surface area of approximately 25,700 km<sup>2</sup>. The main source of inflow to Lake Erie is from Lake Huron via the St. Clair River, Lake St. Clair and the Detroit River. The drainage basin encompasses parts of the American states of Michigan, Ohio, Pennsylvania, New York and Indiana, and the Canadian province of Ontario (Forsythe 2004). The main outflow is to Lake Ontario via the Niagara River and the Welland Canal. The average water retention in Lake Erie time is 2.6 years, the shortest of all the Great Lakes (U.S. EPA 1995). Lake Erie is exposed to the greatest effects of agriculture and industrial processes due to the fertile soils and industrial activity in its drainage basin. It is also the most densely populated of the five lake basins (Great Lakes Information Network 2004). Lake Erie is therefore a major sink for contaminants. The western basin exhibits the greatest degree of sediment contamination as a result of loadings via main tributaries including the Detroit and Maumee rivers (Painter et al. 2001).

Lake Ontario is more than three times larger than Lake Erie (by volume) at 1640 km<sup>3</sup>, although by surface area it is the smallest of the Great Lakes at 19,010 km<sup>2</sup> (Lake Ontario Lakewide Management Plan 1998). It is fed primarily by the waters of Lake Erie through the Niagara River and Welland Canal. Its drainage basin covers portions of the Canadian province of Ontario and the American state of New York. Approximately 93% of the water in Lake Ontario is drained to the northeast by the St. Lawrence River (Lake Ontario Lakewide Management Plan 1998). Lake Ontario has a water retention time of 6 years due to its higher volume of water when compared to Lake Erie. The western and northern Canadian shores are intensely developed with major urban industrial centres such as Hamilton and Toronto. Alternatively, the southern and eastern U.S. shores are much less urbanized (U.S. EPA 1995). In contrast to Lake Erie, contaminant concentrations including PCBs, lead and mercury are more evenly distributed across the three major depositional basins (Marvin et al. 2004, 2003a).

## Data Collection

Field research was conducted in 1997 to 1998 under the Environment Canada Great Lakes Sediment Assessment Program that provided sediment contamination data for 80 sites in Lake Erie and 71 sites in Lake Ontario using a mini-box core sampling procedure. Some specific sites were sampled in order to assess certain Lake Ontario Areas of Concern (AOCs) including Hamilton Harbour and the mouth of the Niagara River (Jakubek and Forsythe 2004). Details concerning the sampling and analysis procedures can be found in Marvin et al. (2003a) and Painter et al. (2001).

#### Statistical Methods – Kriging

The methods for interpolating spatial data can be divided into deterministic and probabilistic classes. Deterministic methods (e.g., inverse distance weighting) have a mathematical development based on assumptions about the functional form of the interpolator (Krivoruchko and Gotway 2004). Probabilistic methods have a foundation in statistical theory and assume a statistical model for the data. When probabilistic methods are used for interpolation, they are referred to as methods for spatial prediction. These predictors have standard errors that quantify the uncertainty associated with the predicted or interpolated values (Krivoruchko and Gotway 2004).

If the data follow a Gaussian distribution, the best predictor, the one that minimizes the prediction meansquared error, is a linear predictor (i.e., a linear combination of data values). Thus, ordinary kriging is optimal for Gaussian data only (Krivoruchko and Gotway 2004). Cross-validation is a general procedure that checks the compatibility between a set of data and its structural model (ASTM 1996 as found in Ouyang et al. 2003b). It is a simple way to compare various assumptions either about the model (e.g., the type of variogram and its parameters, the size of the kriging neighbourhood) or about the data (e.g., values that do not fit their neighbourhood, such as outliers or point-wise anomalies). In the cross-validation procedure, each sample value, C, at a location, x, is removed in turn from the data set and a value, C\*, at that location is estimated using the n - 1 samples (Wackernagel 2003). The difference between the measured value (C) and the crossvalidation estimated value  $(C^*)$  is the estimation error  $(C - C^*)$ , which gives an indication on how well the data value fit into the neighbourhood of the surrounding data values. If the average of the cross-validation errors is close to zero, one can say that there is no apparent bias, while a significant negative or positive average error can represent, respectively, systematic overestimation or underestimation (Osburn 2000).

The most common kriging predictor is a linear predictor, meaning that prediction at any location is obtained as a weighted average of the neighbouring data. Ordinary kriging assumes a constant, but unknown mean, and estimates the mean value as a constant in the searching neighbourhood (Krivoruchko and Gotway 2004). Thus, this approach models a spatial surface in deviations from a constant mean, where the deviations are spatially correlated. Even though the assumption of a constant mean is rather simple, the modelled surfaces can be quite complex (Krivoruchko and Gotway 2004), and if the pattern of spatial continuity of the data can be described visually using a variogram model, it is difficult to improve on the estimates that can be derived in the kriging process (Isaaks and Srivastava 1989).

Kriging analysis has been used for analysis by numerous scientists and engineers in mining and petroleum explorations, environmental studies, and even agricultural practices (Ouyang et al. 2003b). Few, however, have applied this tool to the estimation of large lake sediment contamination distribution. In this research, the Geostatistical Analyst extension of the ArcGIS software was used to interpolate the prediction surfaces.

## **Results and Discussion**

Data characteristics are important when trying to analyze spatial patterns. Tables 2 and 3 display the important characteristics of the data sets used in this research. Information is provided that relates to the data distributions and the levels of contamination in the sampling location data. It is interesting to note that no sampling sites exceeded the PEL for HCB or PCBs in either of the lakes.

The cross-validation procedure provides measures of accuracy for the predictions made using the ordinary kriging method (Johnston et al. 2001; Krivoruchko and Gotway 2004). The measures produced include the mean prediction error (MPE), average standard error (ASE) and standardized root-mean-squared prediction error (SRM-SPE). Values calculated for these measures are documented for Lake Erie in Table 4 and Lake Ontario in Table 5. Statistically valid results should have MPE results near 0, ASE values that are <20 (otherwise predictions are straying quite far from the measured locations), and SRMSPE values approaching 1 (Jakubek and Forsythe 2004; Forsythe et al. 2004). If the SRMSPE is greater than 1, there is an underestimation of the variability of the predictions and if the SRMSPE is less than 1, overestimation of the variability is the result (Johnston et al. 2001; Forsythe et al. 2004). Analysis and results for the mercury data set were published previously in Forsythe et al. (2004) and are provided here for comparison purposes.

### Hexachlorobenzene

Hexachlorobenzene (HCB) occurs as a by-product of several chlorination processes, in particular chlor-alkali production, during the manufacturing of solvents, and in the production of pesticides (Jakubek and Forsythe 2004). It can also be produced in the combustion of chlorinated organic chemicals, chlorine manufacturing, metal manufacturing and the incineration of municipal waste. Until 1984, HCB was used as a pesticide. Other uses include the manufacture of fireworks, ammunition and synthetic rubber. HCB is resistant to degradation, and adsorbs strongly on soil and sediment particles (Agency for Toxic Substances and Disease Registry 2002).

| Contaminant | No. of<br>sites | No. <tel< th=""><th>No. ≥TEL<br/>and <pel< th=""><th>No. ≥PEL</th><th>Minimum</th><th>Maximum</th><th>Average</th><th>Standard<br/>deviation</th></pel<></th></tel<> | No. ≥TEL<br>and <pel< th=""><th>No. ≥PEL</th><th>Minimum</th><th>Maximum</th><th>Average</th><th>Standard<br/>deviation</th></pel<> | No. ≥PEL | Minimum | Maximum | Average | Standard<br>deviation |
|-------------|-----------------|--|---|----------|---------|---------|---------|-----------------------|
| НСВ         | 57              | 57   | 0   | 0        | 0.00    | 11.826  | 1.594   | 2.237                 |
| PCBs        | 67              | 10   | 57  | 0        | 1.850   | 244.476 | 95.304  | 61.015                |
| Pb          | 55              | 22   | 32  | 1        | 4.979   | 104.273 | 43.084  | 23.425                |
| Hg          | 55              | 28   | 23  | 4        | 0.006   | 0.940   | 0.202   | 0.185                 |

TABLE 2. Data characteristics for Lake Erie (minimum, maximum, average and standard deviation in ng/g for HCB and PCBs; µg/g for Pb and Hg)

TABLE 3. Data characteristics for Lake Ontario (minimum, maximum, average and standard deviation in ng/g for HCB and PCBs; µg/g for Pb and Hg)

| Contaminant | No. of<br>sites | No. <tel< th=""><th>No. ≥TEL<br/>and <pel< th=""><th>No. ≥PEL</th><th>Minimum</th><th>Maximum</th><th>Average</th><th>Standard<br/>deviation</th></pel<></th></tel<> | No. ≥TEL<br>and <pel< th=""><th>No. ≥PEL</th><th>Minimum</th><th>Maximum</th><th>Average</th><th>Standard<br/>deviation</th></pel<> | No. ≥PEL | Minimum | Maximum | Average | Standard<br>deviation |
|-------------|-----------------|--|---|----------|---------|---------|---------|-----------------------|
| НСВ         | 71              | 32   | 39  | 0        | 0.312   | 57.969  | 23.161  | 18.717                |
| PCBs        | 71              | 17   | 54  | 0        | 2.601   | 254.763 | 100.226 | 71.123                |
| Pb          | 68              | 16   | 26  | 26       | 5.169   | 196.617 | 71.812  | 41.908                |
| Hg          | 71              | 12   | 17  | 42       | 0.015   | 1.138   | 0.586   | 0.353                 |

The cross-validation results for Lake Erie were very good and the predicted surface (Fig. 1) indicates that concentrations of HCB are low throughout the lake. Slightly higher concentrations (although well below the TEL) were found in the western basin which can be related to transport via the Detroit River in the direction of prevailing lake currents.

The predicted surface for Lake Ontario HCB (Fig. 2) contrasts sharply with that of Lake Erie. Higher concentrations are again found in the deep lake basins. The prediction surface almost perfectly estimated the variability and featured a SRMSPE value of 0.9748. The Niagara River watershed has been identified as a source of HCB to Lake Ontario (Williams et al. 2000).

## Polychlorinated Biphenyls (PCBs)

The manufacturing of PCBs was banned in Canada and the U.S. in 1977 after scientific evidence revealed that they were the cause of environmental and human health problems (U.S. EPA 1995). In the 1960s, PCBs were found to be the cause of the death of thousands of birds in the Irish Sea and in Sweden, and 1200 people in Japan were poisoned by rice oil containing PCBs (Canadian Council of Resource and Environment Ministers 1986; Hodgson and Levi 1997). PCBs are highly persistent; they degrade very slowly by weathering and microbial processes, and have the ability to bioaccumulate in the food chain (Canadian Council of Resource and Environment Ministers 1986; Ontario Ministry of the Environment 1999). In 1929, the Monsanto Company began to produce PCBs commercially for use as a cooling and insulating fluid for electrical equipment. They have also been used for a variety of other industrial purposes, including heat exchangers, plasticizers, hydraulic fluids and flame retardants. PCBs are still used in closed electrical systems, and can be found in landfills. Oliver and Bourbonniere (1985) found PCB concentrations in the western basin of Lake Erie to be much higher than in Lake St. Clair and Lake Huron, indicating the presence of primary sources along the Huron-Erie corridor. Drouillard et al. (2003) estimated that an area downstream of the Trenton Channel contains 62% of the total PCB mass balance for the Detroit River system. This area is highly industrialized, with coal power generation, and steel and chemical production predominating (Marvin et al. 2003b). The sediments in this area are fine-grained, vulnerable to resuspension and provide loadings to the western basin of Lake Erie during major storm events (Reitsma et al. 2003). Nettesheim (2003)

| TABLE 4. Kriging cross-validation results for Lake Erie |        |       |        |  |  |
|---|--------|-------|--------|--|--|
| Contaminant   | MPE    | ASE   | SRMSPE |  |  |
| НСВ   | 0.0122 | 2.115 | 0.917  |  |  |
| PCBs  | 0.1522 | 42.30 | 1.056  |  |  |
| Pb  | 0.4266 | 18.26 | 1.165  |  |  |
| Hg  | 0.0006 | 0.144 | 1.102  |  |  |

| TABLE 5. Kriging cross-validation results for Lake Ontario |        |       |        |  |  |
|--|--------|-------|--------|--|--|
| Contaminant  | MPE    | ASE   | SRMSPE |  |  |
| НСВ  | 0.4687 | 14.64 | 0.9748 |  |  |
| PCBs   | 2.5720 | 72.86 | 0.9336 |  |  |
| Pb   | 1.3490 | 39.77 | 0.9331 |  |  |
| Hg   | 0.0107 | 0.36  | 0.9474 |  |  |



Fig. 1. HCB kriging results for Lake Erie (inset: time-averaged circulation in Lake Erie—isobaths at 20 and 50 m). Source: modified after Beletsky et al. (1999).



Fig. 2. HCB kriging results for Lake Ontario (inset: time-averaged circulation in Lake Ontario—isobaths every 50 m). Source: modified after Beletsky et al. (1999).

states that PCBs exhibit a "strong urban effect," with atmospheric PCB concentrations higher in urban areas. The highest concentrations of PCBs in the air over the Great Lakes were found by McConnell et al. (1998) at the eastern and western extremes of Lake Erie, near Detroit and Buffalo, respectively. This indicates that local point source emissions to the air from industrial and urban areas are a significant source of PCBs through atmospheric deposition.

The results for Lake Erie (Fig. 3) have a higher than desired ASE value (42.30), and the MPE value of 0.1522 is also high; however, the SRMSPE result of 1.056 is very close to normal. The observed patterns can be related to lake bathymetry, currents and the proximity to urban areas. The plume along the southern shoreline eastward from Cleveland is in sharp contrast to that along the sparsely populated Canadian shoreline to the north. This area is representative of ambient background concentrations as it is out of the main circulation pattern emanating from the Detroit River. There are, however, some impacts as indicated by the higher than TEL concentrations found there.

PCBs were estimated to have high concentrations in the deep lake regions of Lake Ontario (Fig. 4). When the actual measured values are compared to the isobaths throughout the lake, the predictions seem reasonable, but high ASE results are the reason to suspect inconsistent outcomes from the kriging analysis. A possible explanation for these cross-validation results is a biased prediction supported by a MPE value of 2.572. The majority of the lake is estimated as having values higher than the TEL but lower than the PEL which corresponds well with the original data points. The large standard deviation of 71.123 does, however, adversely influence the results as it is indicative of a high amount of variation in the data set (Forsythe et al. 2004).

## Lead

Lead is routinely detected in sediments of the Great Lakes as a result of its historically heavy use, primarily as an additive in gasoline. The proximity of Lake Erie and Lake Ontario to industrial and agricultural areas makes it likely that point sources and runoff are also significant sources (Forsythe et al. 2004). Painter et al. (2001) observed that the spatial distribution of lead in Lake Erie showed the highest concentrations in the western basin and in the southern area of the central basin, while Marvin et al. (2003a) stated that the distribution in Lake Ontario was more consistent across the depositional basins.

The lead prediction result for Lake Erie (Fig. 5) had acceptable cross-validation results although the ASE value



Fig. 3. PCB kriging results for Lake Erie (inset: time-averaged circulation in Lake Erie—isobaths at 20 and 50 m). Source: modified after Beletsky et al. (1999).



Fig. 4. PCB kriging results for Lake Ontario (inset: time-averaged circulation in Lake Ontario—isobaths every 50 m). Source: modified after Beletsky et al. (1999).



Fig. 5. Lead kriging results for Lake Erie (inset: time-averaged circulation in Lake Erie—isobaths at 20 and 50 m). Source: modified after Beletsky et al. (1999).

of 18.26 is slightly higher than desirable. Areas of higher concentrations were found in the south-central part of the lake near Cleveland. The high concentration pocket (>PEL) corresponds well to the single sampling location with a value greater than the PEL value of 91.3 µg/g.

The prediction surface representing lead for Lake Ontario (Fig. 6) estimated the variability well with a SRMSPE value of 0.9331; however, it features an ASE value of 39.77. Therefore the results should be interpreted with some caution as they are not statistically valid. Again a possible reason for these results is the location of two contaminated sediment sampling sites in Hamilton Harbour which may skew the results in the western part of the lake. The higher concentrations in the middle of the Mississauga Basin are related to lake currents and bathymetry which combine for increased deposition of sediments in this area.

## Log-Normal Transformation

To enhance the distribution patterns, log-normal procedures were performed on the non-statistically valid results for PCBs and lead. Ouyang et al. (2003b) state that in general, a normal distribution requirement in kriging analysis may not be so critical, but when the data set is too skewed or contains outliers, some kind of transformation is needed. If the data are not Gaussian, statistical transformations (e.g., log, Box-Cox) can be used to transform them so that they do follow a Gaussian distribution. However, with the exception of the log-transform, it is not possible to directly back-transform the data to the original scale without bias (Krivoruchko and Gotway 2004).

The log-normalized cross-validation statistics (Tables 6 and 7) for PCBs and lead for both lakes are excellent indicating that the parameters used for creating the models are satisfactory and estimate the surfaces very well. The MPE, ASE and SRMSPE statistics are all within acceptable limits.

## PCBs (Log-Normal)

The log-normal PCB results for Lake Erie (Fig. 7) are similar in pattern to the statistically non-valid results (Fig. 3), however, overall concentration levels are lower. The SRMSPE value of 1.005 is very close to optimal. Higher concentrations are found in proximity to major urban centres and improved (lower) values (<TEL) are found in areas where currents and land use combine to produce smaller concentrations.

The bathymetry of Lake Ontario appears to have a great influence on the log-normalized PCB results (Fig. 8). Higher concentrations are found in the deep lake basins, with lower levels found in sill areas on the lake bottom. Less than TEL values are found along the



Fig. 6. Lead kriging results for Lake Ontario (inset: time-averaged circulation in Lake Ontario—isobaths every 50 m). Source: modified after Beletsky et al. (1999).

| Contaminant | MPE    | ASE   | SRMSPE |
|-------------|--------|-------|--------|
| PCBs        | 0.0048 | 0.269 | 1.005  |
| Pb          | 0.0115 | 0.236 | 1.059  |

 TABLE 6. Log-transformed kriging cross-validation results

 for Lake Erie

 TABLE 7. Log-transformed kriging cross-validation results

 for Lake Ontario

| Contaminant | MPE    | ASE   | SRMSPE |
|-------------|--------|-------|--------|
| PCBs        | 0.0131 | 0.530 | 0.9920 |
| Pb          | 0.0153 | 0.311 | 0.9831 |

northern shoreline away from large population centres and areas less affected by sediments from the Niagara River. An area of shallower (inshore) water exists along the northern shore while the deep water basins are much further offshore. This contrasts with the southern shore where the deep water areas are not as far offshore. The SRMSPE value of 0.9920 is very close to optimum.

## Lead (Log-Normal)

Although the initial kriging results of the lead distribution in Lake Erie sediment were within the statistical limits of the models, a log-normal analysis was performed. The results (Fig. 9) are similar in pattern to the original (Fig. 5) with the exception that the pocket of higher-than-PEL concentrations has disappeared. Additionally, larger portions of the lake also have <TEL concentrations than were determined in the non-log-transformed kriging estimate. There was only one original data point with a value (104  $\mu$ g/g) that was slightly above the PEL of 91.3  $\mu$ g/g and the SRMSPE of 1.059 indicates that the estimation may be slightly underestimated.

Statistically the log-normal estimates of the lead distribution in Lake Ontario sediment (Fig. 10) are a great improvement on the non-log-transformed surface (Fig. 6). The predictions are, however, slightly overestimating the values as indicated by the SRMSPE value of 0.9831. This, however, is a great improvement on the former value of 0.9331. The major differences occur in the areas found to be above the PEL (all three deep lake basins and in the vicinity of Hamilton Harbour) and in the expansion of areas that are below the TEL. The <TEL regions along the northern shoreline of the lake are much larger which can be related to the deposition of lead in the deep lake basins through lake circulation patterns and the low level of industrial production in this area. The location of sills on



Fig. 7. PCB kriging results (log-transformation) for Lake Erie (inset: time-averaged circulation in Lake Erie—isobaths at 20 and 50 m). Source: modified after Beletsky et al. (1999).



Fig. 8. PCB kriging results (log-transformation) for Lake Ontario (inset: time-averaged circulation in Lake Ontario—isobaths every 50 m). Source: modified after Beletsky et al. (1999).



Fig. 9. Lead kriging results (log-transformation) for Lake Erie (inset: time-averaged circulation in Lake Erie—isobaths at 20 and 50 m). Source: modified after Beletsky et al. (1999).



Fig. 10. Lead kriging results (log-transformation) for Lake Ontario (inset: time-averaged circulation in Lake Ontario—isobaths every 50 m). Source: modified after Beletsky et al. (1999).

the lake bottom is also very evident in the well-delineated concentration highs shown in the overall results.

## Conclusion

Human activities have had a great influence on the types and quantities of contaminants that have entered the Great Lakes system in both historical and present times. The estimated patterns of sediment contamination that were developed in this study can be related to the location of urban/industrial areas, lake currents and lake bathymetry. Measures including the banning of some chemicals such as PCBs, had a positive effect on the contamination present in lake sediments. The longevity and persistence of these substances does, however, necessitate continued monitoring and remediation measures.

The kriging technique allows for improved and valid areal estimates of the level of sediment contamination in comparison with point measurements alone. The lakewide distribution allows for derivation of spatial estimates. Through the use of cross-validation techniques, a statistical confirmation of the estimates can be produced. The implementation of log-normal data conversion enables (when necessary) the production of statistically valid contamination estimates which can be back-transformed to the original values after processing.

The kriging analyses have led to an increased understanding of the data sets obtained for sediment contamination in lakes Erie and Ontario. The longer water retention time and greater depth of Lake Ontario, together with lake bathymetry influence the observed patterns of contamination. In addition, the lake continues to be subjected to loadings from upstream sources including Lake Erie. The location of urban and industrial areas is also important in the interpretation of the results in both lakes. Statistically valid estimates were obtained for all contaminants in both lakes either through normal ordinary kriging or after the implementation of log-normal data transformation procedures. These results concur with analyses of the point data. The spatial trends are similar, and therefore overall interpretation is comparable. Kriging analysis for contaminated sediment spatial distribution estimation provides an effective communication tool, and a possible additional means of influencing management options and decisions.

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