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Assessing Lead Contamination in Buffalo River Sediments

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ABSTRACT. The Great Lakes Water Quality Agreement between Canada and the United States has identified the Buffalo River as an Area of Concern. The watershed has a long history of heavy industrial activity that contributed to its overall pollution. Sediment core data collected by the New York State Department of Environmental Conservation in 2005 were used to determine lead sediment contamination in a section of the Buffalo River. The ordinary kriging spatial interpolation technique was used to generate surface and subsurface sediment contamination estimates. Due to the meandering nature of the river, two kriging models were used to analyze surface contamination: a global kriging model and a regional kriging model, consisting of three separate sections. The results show that both the global and regional kriging models display similar interpolated surfaces and do not vary significantly. Within the sediment, lead contamination in the surface layer is lower than at the various subsurface depths. In 2011, habitat restoration efforts commenced to remediate environmental damage due to years of pollution inputs from various sources. Sediment dredging operations were initiated that are expected to be completed in 2015. The goal of these operations is to remove heavily contaminated sediments and rehabilitate the Buffalo River. The kriging results provide area-wide estimates of contamination. When compared to the dredging plan, the results indicate that additional removal of contaminated sediments may need to be considered where no dredging has occurred or is not currently planned.

Keywords: spatial interpolation, Buffalo River, ordinary kriging, sediment contamination, lead, dredging

1. Introduction

Lead makes its way into the human body through water, air and food. It enters hydrologic systems mainly via runoff, groundwater inflows and atmospheric deposition (Smol, 2008). Once it enters a water system, it quickly becomes part of the suspended particles and sediments and most of it eventually makes its way into marine sediments (Harrison and Laxen, 1981). Further, it is mostly put into the atmosphere by smelting industries and was released by vehicles through the combustion of leaded-gasoline (Harrison and Laxen, 1981). After reaching a peak in 1972, its atmospheric emissions in the USA were reduced by 98% as a direct effect of the Clean Air Act of 1970, which required the removal of lead additives from gasoline (Smol, 2008). Lead is also present in the tap water of homes that have lead piping (Harrison and Laxen, 1981) and even in food at varying concentrations. A study conducted in Spain revealed that lead concentrations in chicken, pork and beef were 0.0069, 0.005 and 0.0019 μg/g respectively (Gonzalez-Weller et al., 2006). Similarly, a study conducted in Hong Kong found that lead concentrations in large shrimp can be as high as 6 μg/g (Cheung and Wong, 2006). Lead poisoning can lead to severe damage of the human central nervous system and if the exposure is prolonged, it can even result in death (Harrison and Laxen, 1981). Once it enters the body it attaches to red-blood cells, accumulating in soft tissues and in bone (Harrison and Laxen, 1981). Body burden is the amount of toxic chemicals present in an individual at a given point in time and for lead in an average human body (70 kg) is 150 mg (Botkin and Keller, 2005).

The distribution of lead in the environment and its toxic effects on living organisms are closely related to its physical and chemical forms (Harrison and Laxen, 1981). For instance, the formation of lead and organic matter complexes is dependent on pH, as the likelihood of it precipitating out of water increases between a pH of 7.5 and 8.5 (Harrison and Laxen, 1981). Hence, at pH values other than between 7.5 and 8.5, lead concentrations could be relatively low in sediments but relatively high in the water above. Another spatial characteristic of lead is that it tends to accumulate in deep clayish sediments in ocean waters showing concentrations between 50 and 150 μg/g and exhibits a decrease in concentration towards

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the coast (Harrison and Laxen, 1981).

Ordinary kriging is a well-tested method used to investigate the geographic distribution of pollutants in sediments. Forsythe et al. (2004, 2010a) and Forsythe and Marvin (2009) utilized this spatial analysis technique to examine sediment contamination in the Great Lakes of North America. Forsythe et al. (2010b, 2013), Gawedzki and Forsythe (2012), and Ouyang et al. (2002, 2003) conducted sediment contamination spatial analysis research using kriging in river environments.

2. Site Location and Data

The Buffalo River is listed as an Area of Concern (AOC) by the Great Lakes Water Quality agreement between Canada and the United States. Cayuga Creek, Buffalo Creek, and Cazenovia Creek are the three major streams in the watershed that comprise the AOC (US EPA, 2011). The impacted area is 10 kilometres in length and contains the 2.3 kilometre stretch of the City Ship Canal (US EPA, 2011). The river is located south of the City of Buffalo and flows westward, discharging into Lake Erie (Figure 1). In the past few years, New York State has identified three major contributors to environmental damage: ExxonMobil Corporation, Honeywell Corporation, and PVS Chemicals. These major companies had a heavy industrial presence in the vicinity of the Buffalo River and their discharges were directed into the river itself (Tokasz, 2010). There are 45 inactive hazardous waste sites within the AOC and contaminants of concern include polychlorinated biphenyls (PCBs), polycyclic aromatic hydrocarbons (PAHs), metals (including lead) and industrial organics. In addition, there are currently 33 Combined Sewer Overflow (CSO) outfalls within the watershed that discharge into the Buffalo River and three connections to the Buffalo sewer system from outside sewer districts that overflow into the river during storm events (US EPA, 2011). The Buffalo River played an important role in the development of Buffalo. Years of Industrial and municipal use, however, degraded the river and sediment, and subsequent economic changes left the region with a number of abandoned, contaminated properties and deteriorating facilities on or near the river (NYSDEC, 2010, 2013).

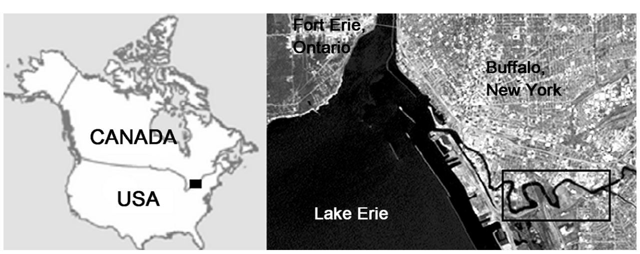

Figure 1. The Location of the Buffalo River (the hollow black box denotes study area extent).

Pollution within the Buffalo River has negatively affected the aquatic ecosystem. Potential habitat areas are imperfect and limited due to contamination as invasive plant and animal species threaten diversity and quality of the rivers habitat (US EPA, 2011). The Buffalo River has a low diversity of benthic macro-invertebrates that is dominated by pollution-tolerant species (US EPA, 2011). Nelson and Hites (1980) believe that the existence of organic compounds is partially responsible for the observed tumours in fish found in the Buffalo River. The AOC has fish consumptions advisories currently in place as recent research indicates an average of 34% deformities, fin erosions, lesions, and tumors (DELT) anomalies in fish, which range from a low of 14% for pumpkinseed to an extremely high 87% for brown bullhead (US EPA, 2011).

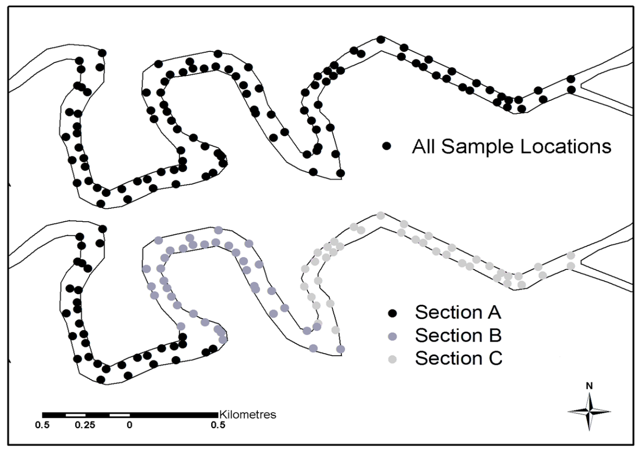

Figure 2. The 2005 sample locations at surface level for both global and regional kriging models.

Table 1. Lead Sediment Sampling Location Statistics for the Buffalo River (μg/g)

Depth (cm)	No. of Sites	Min	Max	Average	SDV*
Surface 0-30	111	8.1	2600	87.12	250.77
Subsurface 30-60	34	10.9	8510	409.34	1428.17
Subsurface 60-90	33	30.4	538	142.75	132.65
Subsurface 90-120	49	19.8	1100	127.02	183.46
Subsurface 120-150	50	30.4	1100	150.62	192.09

* Note: SDV = Standard Deviation.

In 2005, the New York State Department of Environmental Conservation collected sediment sample data for more than one hundred different pollutants in the Buffalo River. This study focuses on lead levels in the sediment at various depths. The depth intervals are categorized into two groups, surface and subsurface sediments. Surface sediments consisted of the top 30 cm in core depth, while the subsurface sediments ranged from a depth of 30 to 150 cm. The subsurface data were categorized into four separate groups: Depth 1 (30-60 cm), Depth 2 (60-90 cm), Depth 3 (90-120 cm), and Depth 4 (120-150 cm). A total of 182 sample cores were used in this analysis, which consisted of 111 surface and 166 subsurface samples. Each core sample could have any combination of data associated with it based on depth. The distribution of surface sample locations for both the global and regional kriging models can be seen in Figure 2. There are 33, 38 and 40 points in Sections A, B and C respectively for the regional data sets. The break points were based on the natural meanders in the river and to ensure that each section would contain more than 30 points. The distribution of subsurface sample points at each of the four depths can be seen in Figure 3. Table 1 displays the data characteristics for lead at all depths.

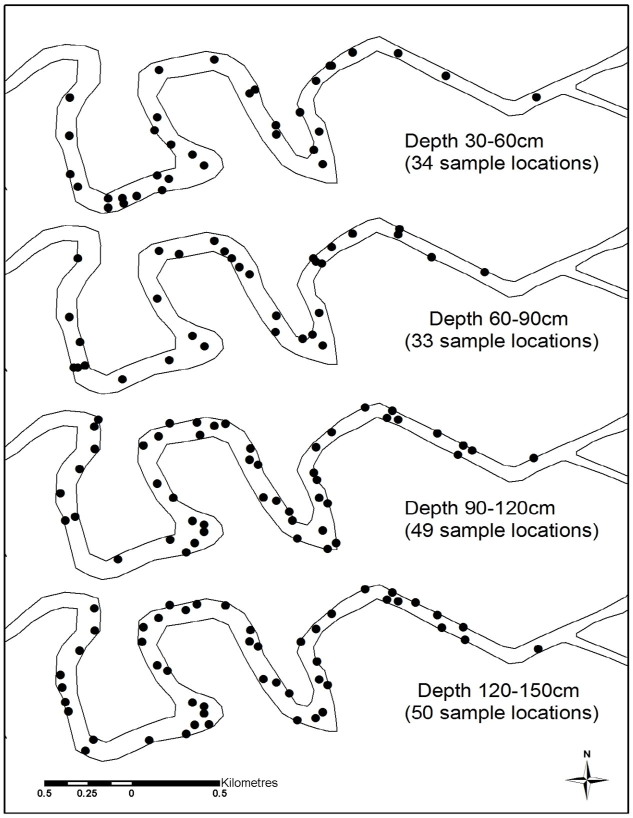

Figure 3. The 2005 sample locations at all subsurface levels.

3. Methodology

Ordinary kriging was selected as the spatial analysis technique for this research based on a number of factors. The data were not equally spaced and kriging performs well when the location of data points varies in both direction and distance. The number of points was also considered large enough (>30) for the results to be statistically valid. The ability to vary search ranges, ellipsoids and number of points to include also resulted in the selection of kriging as the most suitable spatial interpolation technique for these analyses.

The kriging geostatistical technique produces interpolation surfaces that are unbiased and exact (Clark, 1979; Wackernagel, 2003). In practical terms, unbiased means that the residuals' average is very close to zero, whereas exact means that estimated values are very close to the sampled values. Unlike other interpolation techniques, ordinary kriging provides a prediction error surface which allows for sound assessment of the predicted values.

There is no agreement on whether non-normal distributions should be transformed prior to fitting the semivariogram and performing kriging. Some authors argue that kriging, requires a normal distribution of values and thus, non-normal distributions need to be transformed (Houlding, 2000; Johnston et al., 2001). Forsythe et al. (2004) suggest that statistically insignificant models (average standard error > 20) may become significant if positively skewed data are log-normalized. When ASE values are higher than 20, predictions stray quite far from the original data point values. Similarly, Ouyang et al. (2003) argue that even though normality is not essential for kriging, the presence of pronounced skewness in a data set warrants some kind of transformation. Likewise, Clark (1979) argues that kriging as an interpolator does not require normality rather only kriging prediction errors rely on normality. Conversely, Wackernagel (2003) warns against the use of log-normal transformation as this procedure causes the mean to shift giving poor results. There is more agreement, however, with respect to the use of log-normal as the best choice for transforming non-normal data (Clark, 1979; Houlding, 2000; Wackernagel, 2003) as long as there are no zero values in the dataset. Thus, log-transformations of the lead dataset were done using base ten log. Only logged values were used for the fitting of theoretical semi-variograms. Log calculations and transformations were performed in SPSS and these values were then used in the ArcGIS Geostatistical Analyst environment.

Table 2. Kriging Log-Normalized Data Cross Validation Statistics for Lead

Depth (cm)	Model	MPE	RMSPE	ASE	SRMSPE
Surface 0-30	Exponential	0.00137	0.3436	0.3468	0.993
Subsurface 30-60	Exponential	-0.04087	0.5636	0.5575	1.008
Subsurface 60-90	Gaussian	0.00771	0.3728	0.3850	0.979
Subsurface 90-120	Gaussian	0.00744	0.3854	0.3741	1.030
Subsurface 120-150	Exponential	0.00309	0.4070	0.4031	1.022

The selected parameters that were deemed the most suitable for interpolating contamination in Buffalo River sediments using the ordinary kriging technique can be found in research conducted by Gawedzki and Forsythe (2012) and Forsythe et al. (2013). In the Geostatistical Analyst, the semivariogram model needs to be properly chosen to run the kriging interpolator. The most suitable model, among Spherical, Exponential, or Gaussian is selected by comparing the prediction error statistics. The ideal prediction error criteria are the mean prediction error (MPE) should be closest to zero, the root-mean square prediction error (RMSPE) should be as small as possible, the average standard error (ASE) should be the close to the RMSPE and not be greater than 20, and the standardized root-mean squared prediction error (SRMSPE) should be close to one (Forsythe and Marvin, 2009; Forsythe et al., 2010a, b; Gawedzki and Forsythe, 2012; Jakubek and Forsythe, 2004). The most suitable semivariogram models selected for each lead sediment contamination depth can be seen in Table 2. The results were based on experimentation. The models that performed better (i.e. were closest to the optimum values) in terms of MPE, RMSPE, ASE and SRMSPE were selected as they were the best performing models.

Although using the kriging spatial interpolation technique is useful in determining overall sediment contamination in the Buffalo River, looking at the raw results would not provide much meaning to the analysis. To assist in the interprettation of the kriged results, it is necessary to use comparative measures (Ouyang et al., 2003). Studies conducted by Forsythe and Marvin (2009), Forsythe et al. (2010a, b, 2013), Gawedzki and Forsythe (2012) and Jakubek and Forsythe (2004) all used Threshold Effect Level (TEL) and Probable Effect Level (PEL) measures established by the Canadian Council of Ministers of the Environment to assess contamination levels. The TEL for lead is 35 µg/g is and the PEL is 91.3 µg/g. The TEL indicates the concentration below which adverse biological effects are expected to occur rarely (< 25%), while the PEL indicates a concentration above which adverse biological effects are expect to occur frequently (> 50%). The range between TEL and PEL is known as the possible effect range (PER) and biological abnormalities are expected to occur occasionally within this range (Forsythe et al., 2013).

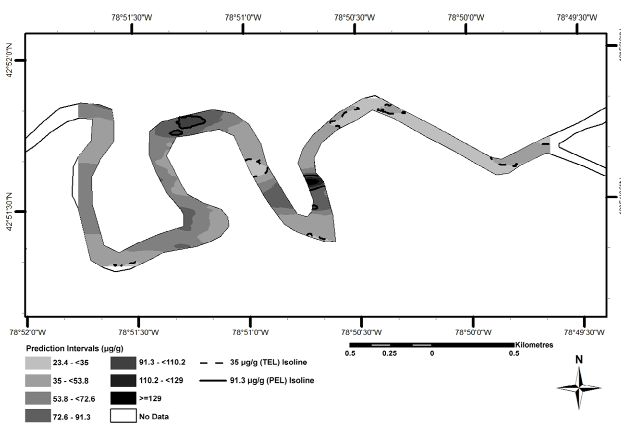

Figure 4. Kriged 2005 lead (log-normal) concentrations at surface level.

4. Results and Discussion

Two kriging models were produced to analyze lead surface sediment contamination in the Buffalo River. The global kriging model's interpolated results, which encompass the entire length of the study area, can be seen in Figure 4. The regional kriging model's interpolated results, which consist of three separate kriged sections combined together in one map, can be seen in Figure 5. The two models interpolate the surface sediment contamination of lead very similarly. Both maps show heavy contamination in the central section of the river where PEL isolines are present. This central section has been noted in previous research conducted by Gawedzki and Forsythe (2012) and Forsythe et al. (2013) as heavily contaminated with various compounds and metals. Also, both Figures 4 and 5 show the eastern section of the river having contamination levels mostly between the TEL and PEL with slight variations in the placement of PEL isolines. Similarily, the western section of the Buffalo River has lead contamination levels between the TEL and PEL visible in both the global kriging model and regional kriging model. The only difference between the two models that is evident is in the north-west section of the AOC as Figure 4 shows a PEL isoline and Figure 5 does not; however, high contamination levels are still accounted for in Figure 5 as contamination levels are displayed between 72.6 and 91.3 µg/g. The SRMSPE for the surface lead map of 0.993 indicates that the predicted surface is slightly overestimated. It should be noted that regional kriging maps could not be produced for subsurface sediments as there were not enough sample points (> 30 at each depth) to generate statistically valid predictions.

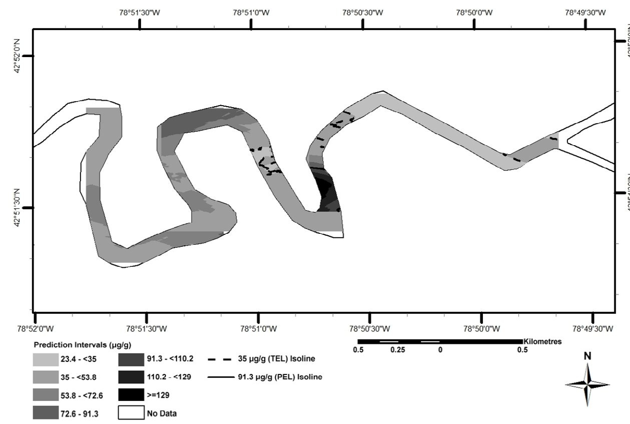

Figure 5. Kriged 2005 lead (log-normalized) concentrations at surface level (spliced map).

The distribution of lead contamination in sediments at four equal subsurface depths can be seen in Figure 6. The first subsurface depth of 30-60 cm has contamination levels that vary greatly when compared to the surface sediment contamination above. TEL and PEL isolines are scattered throughout the AOC at this depth. The eastern section of the river is heavily contaminated above the PEL. Although the central section of the river is contaminated above the PEL, the exact area of contamination is not the same as the subsurface sediment. The western section of the river is mostly contaminated above the TEL, but there is a small area where contamination is below the TEL. The SRMSPE for this depth of 1.008 indicates that the predicted surface is slightly underestimated. At a subsurface depth of 60-90 cm, the majority of the river is contaminated at a level above the PEL. The eastern and south-western section of the river is contaminated between the TEL and PEL. There are no TEL isolines visible at this depth. The SRMSPE for the surface lead map of 0.979 indicates that the predicted surface is slightly overestimated. Further, at the next depth of 90-120 cm, the central section again has contamination levels above the PEL. The eastern section shows contamination levels between the TEL and PEL with very small areas below the TEL. The western section of the river is contaminated mostly between the TEL and PEL with a small area that is contaminated above the PEL. The SRMSPE for this depth of 1.030 indicates that the predicted surface is slightly underestimated. At the final depth of 120-150 cm, contamination is above the TEL as there are no TEL isolines present. Again, the central section of the river is heavily contaminated above the PEL. Also, heavy contamination at this depth exists in the western section of the AOC in an area similar to the depth above. The eastern section of the river is contaminated between the TEL and PEL, similar to the two depths directly above. The S-RMSPE for this depth of 1.022 indicates that the predicted surface is slightly underestimated. The main pattern observed when comparing contamination levels between all surface and subsurface depths is that contamination is heaviest and consistent throughout the central sections of the Buffalo River AOC. Within the sediment, lead contamination in the surface layer is lower than at the various subsurface depths which corresponds with the findings of EML (2011).

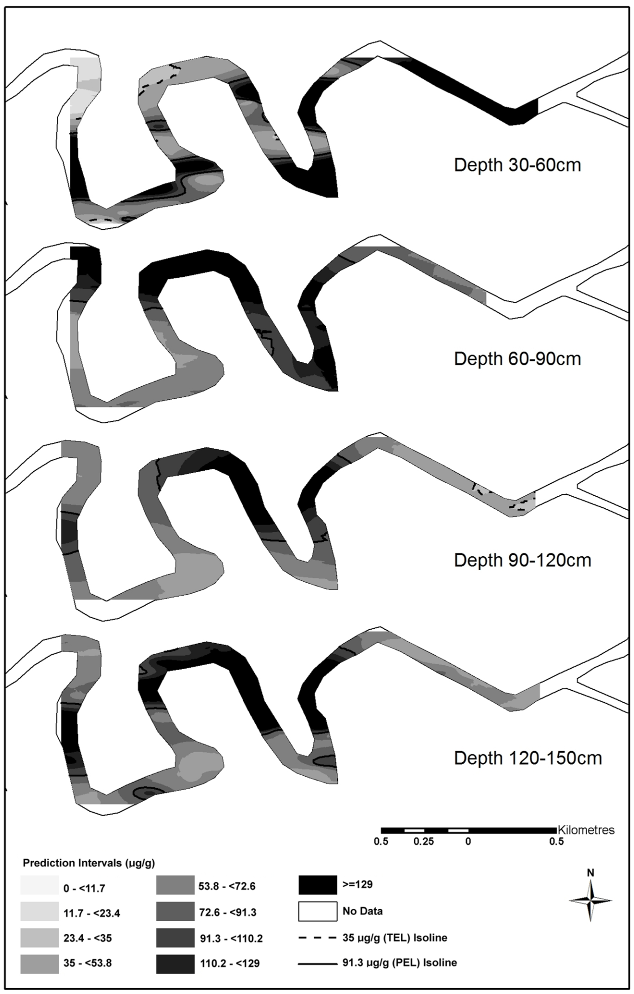

Figure 6. Kriged 2005 lead (log-normalized) concentrations at subsurface depths.

In 2011, habitat restoration efforts commenced to remediate environmental damage due to years of pollution inputs from various sources. Sediment dredging operations were initiated that are expected to be completed in 2015. The goal of these operations is to remove heavily contaminated sediments and rehabilitate the Buffalo River (NYSDEC, 2013; US Army Corps of Engineers, 2014). Approximately 373,100 cubic metres (488,000 cubic yards) of contaminated sediment has already or will be removed using a clam-shell bucket dredging system. This is equivalent to about 33,000 truck loads (NYSDEC, 2014a, b).

The kriging results provide area-wide estimates of contamination. When compared to the dredging plan from March 2013 (Figure 7), the results indicate that additional removal of contaminated sediments may need to be considered in areas where no dredging has occurred or is currently planned. This is particularly evident in the upper (eastern) portion of the river where the sediment is heavily contaminated from 30-60 cm in depth. In addition, the large meander bend in the south central portion of the river is contaminated at all subsurface depths at levels above the TEL and above the PEL for the 30-60 cm depth.

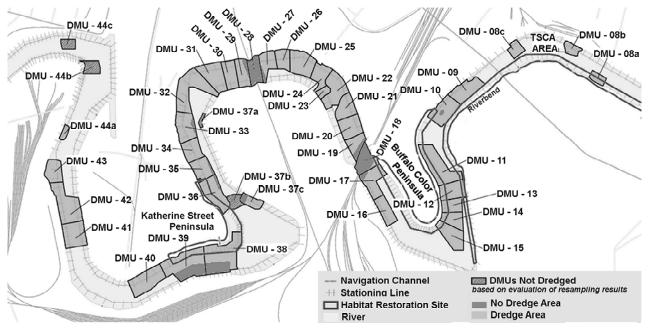

Figure 7. Buffalo River Dredging Plan (Source: modified after (NYSDEC, 2014b)).

5. Conclusions

Proper identification of heavily contaminated hotspots in Buffalo River sediments is important as remedial actions such as dredging are costly. This is why both a regional and global kriging models were developed to compare results. There may be added value to use a regional kriging model for a meandering river such as the Buffalo River; however, the results show that there is almost no difference in the interpolated lead contamination results of the two models. Using a global kriging model would be sufficient for determining the overall contamination levels in Buffalo River sediments.

This research produced estimates of lead contamination for the entire study site based on sediment sample points. This is useful as it provides a basis for assessing overall contamination patterns. Kriging is a spatial interpolation technique that is well-suited for producing area-wide estimates. In addition, spatial interpolation (i.e. kriging) enables and assists in the identification of area-wide patterns of contamination. This is not possible with point measurements.

Lead contamination differs between depths. There does not appear to be a sequential contamination pattern that exists among the depth intervals. Contamination levels are generally between the TEL and PEL throughout the AOC at all depths. Heavily contaminated areas, above the PEL, are mainly found in the central section of the river, with some smaller areas in the western section. There are only a few areas where contamination levels are not a concern and below the TEL.

Future research could be conducted to find the cause of this heavy contamination. There may be a geomorphological explanation as to why the central section of the river is the most contaminated; however, it is more likely a result of past industrial activity with discharges directly into the river.

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