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Lake Michigan sediment lead storage and history of loads

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ABSTRACT

Dated sediment box cores collected in 1994–1996 from 52 locations in Lake Michigan were analyzed to assess storage, trends, and loading history of lead. The results of this study provide information of historic lead loads to the lake for a time period (pre-1960) for which no reliable lead measurements exist. The information can be utilized by those wishing to model lead and to access lead loading trends. Anthropogenic lead storage in the lake's sediments totaled 143,000 t as of 1994. Storage of acid-extractable total (anthropogenic + background) lead totaled 171,000 t between 1850 and 1994. The date of 1850 is the time at which lead loads increased above background loads (219 t/y) to the lake. Anthropogenic loads peaked between 1959 and 1962 at 2440 t/y and were 1170 t/y between 1994 and 1995, illustrating that at the time of collection in 1994, loads were decreasing from previous highs. The load in 1994 to 1995 was equivalent to the load during the time frame of 1922 to 1925. Largest lead loads were to southeastern Lake Michigan in a region downwind of Chicago, illustrating the impact of large populated areas utilizing coal and gasoline on lead loads to the lake. Loading trends were impacted by coal and gasoline consumption, increased industrial activity during World War II, the Clean Air Act of 1970, and the phase-out of leaded gasoline.

Introduction

Researchers are continually lowering the level of lead at which significant and permanent adverse neural effects are found in children and adults (Needleman, 2004). A significant drop in children's blood levels in the United States accompanied the reduction in leaded gasoline (Needleman, 2004). Long term documentation of lead levels in Lake Michigan is desirable to understand the history of lead contamination and the events that impacted the loading of lead to the environment. Contaminants such as lead could not be or were not measured until the 1960's for Lake Michigan, United States, leaving a large blank in the history of lead loads to the lake. However, the record of contamination from 1850 until the time of collection is recorded within selected lead-210 dated sediments. With careful interpretation of such core results, the long term documentation of the history of lead contamination within the Lake Michigan basin can be documented and related to coal consumption, lead gasoline consumption, secondary lead smelting, and population within the basin; as well as, events that impacted the production of lead or use of lead-containing materials (wars, economic depression, and remediation measures).

Dated sediment cores have been used to reconstruct the history of contaminant loads to various water bodies, including reservoirs (Audry et al., 2004; Biester et al., 2007), lakes (Bookman et al., 2008; Drevnick et al., 2012; Engstrom and Swain, 1997; Evenset et al., 2007; Kamman and Engstrom, 2002; Muir et al., 2009; Rossmann, 2010; Yang and Rose, 2005), peat lands (Coggins et al., 2006; Ettler et al., 2008; Shotyk et al., 1996), bays (Yang and Rose, 2005; Fung and Lo, 1997; Sanders et al., 2006), and seas (Leonardo et al., 2006; Yang and Rose, 2005). The use of dated cores for this reconstruction is not as straightforward as it might appear. For all water bodies, sediment resuspension and transport, catchment area versus lake area, location relative to sources, and post-depositional mobility within the sediment associated with diagenesis can add complexity to the interpretation of the dated profiles of a contaminant (Edgington and Robbins, 1976; Drevnick et al., 2012; Engstrom and Swain, 1997; Rossmann, 2010). To properly interpret a dated sediment core, each of these complexities must be understood for the location of collection within the context of a contaminant's known history. Within this paper, we will examine in detail the history of lead contaminant loads to Lake Michigan using the technique described by Rossmann (2010) and applied to mercury. This examination includes a careful review of the impact of sediment resuspension, location relative to sources, and post-depositional mobility on the interpretation of historic lead loads represented by the cores.

Methods

Lake Michigan samples were collected between 1994 and 1996 from 52 stations that could be box cored with a Soutar-type box corer

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(Robbins et al., 1999) (Fig. 1). In total, 118 stations were occupied on a grid pattern from which sediment samples were collected that covered the entire main lake (Pfeiffer and Rossmann, 2010). In Fig. 1, the regions of the main lake without stations are those that could not be box cored. See Pfeiffer and Rossmann (2010) for locations of all stations. They were non-depositional areas with lag deposits of sand and gravel or eroded glacial clays. Four subcores were removed from each box core (Edgington and Robbins, 1997). The same subcore from each box core that was used by Robbins et al. (1999) for radionuclide (Pb-210 and Cs-137) measurements and by Rossmann (2002, 2010) for mercury analyses was analyzed for acid-extractable lead. Subcores were sectioned in 1 cm intervals from top to bottom. Each interval was placed in a pre-weighed polyethylene bottle, kept frozen until freeze-dried, and archived in the laboratory (Robbins et al., 1999; Rossmann, 2002).

Homogenized freeze-dried samples averaging 0.100 g were extracted with 25 mL 10% v/v Seastar ultrapure HNO_3 in Teflon vessels by microwave digestion (Rossmann, 1994; Uscinowicz and Rossmann, 1998; Pfeiffer and Rossmann, 2010). Lead concentrations were measured by inductively coupled plasma mass spectrometry (Pfeiffer et al., 2005; Perkin-Elmer, 1993). Blanks, replicates, and standard reference materials were run with each set of samples digested.

Quality assurance

Method detection limit was based on the standard deviation of the blanks and the number of blanks analyzed (Rossmann, 2002). A total of 192 blanks were analyzed. These had a mean concentration of 0.0586 ng/g with a standard deviation of 0.284 ng/g for a 0.1 g sample. All samples were above the 0.680 ng/g method detection limit based on the blanks. Lead was analyzed for all 1790 core segments. Standard Reference Material SRM8704 Buffalo River Sediment was extracted with each set of samples digested for a total of 124 samples (NIST, 2000). With each recovery within the acceptable range of 80%–120%, the mean recovery was 99.3%. Duplicate extracts were used to document reproducibility of the results. The percent relative difference ($N = 2$) or percent relative standard deviation ($N > 2$) for duplicate

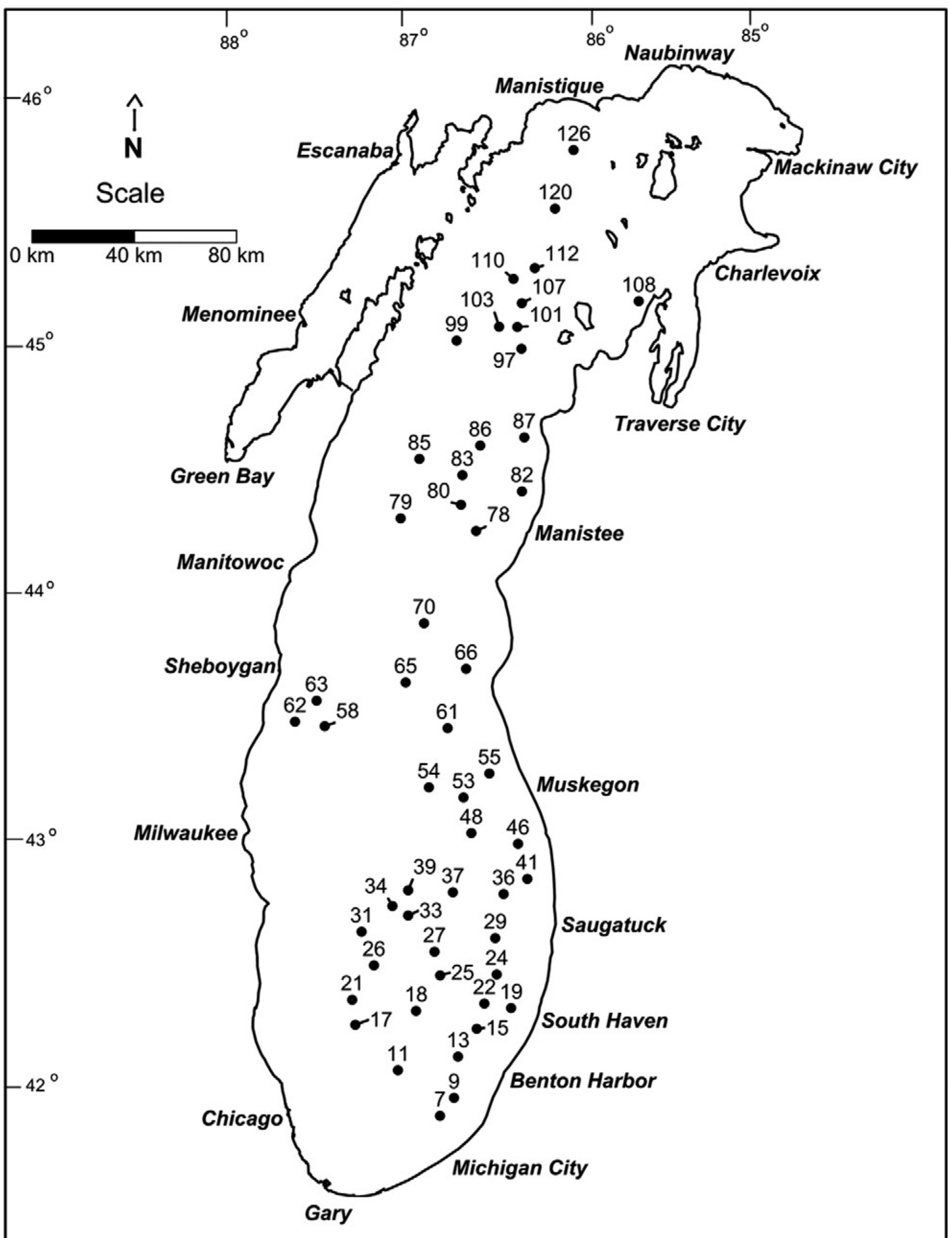

Fig. 1. Sampling locations for box cores collected from Lake Michigan between 1994 and 1996.