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Global and regional contributions to total mercury concentrations in Lake Michigan water

Xiaomi Zhang [a], Kenneth R. Rygwelski [b,*], Mark D. Rowe [b,c], Ronald Rossmann [d], Russell G. Kreis Jr. [b]

[a] Trinity Engineering Associates Inc., Large Lakes Research Station, 9311 Groh Road, Grosse Ile, MI 48138, United States
[b] U.S. Environmental Protection Agency, Office of Research and Development, National Health and Environmental Effects Research Laboratory, Mid-Continent Ecology Division, Large Lakes and Rivers Forecasting Research Branch, Large Lakes Research Station, 9311 Groh Road, Grosse Ile, MI 48138, United States
[c] University of Michigan — Cooperative Institute for Limnology and Ecosystem Research (CILER), 4840 S. State Rd, Ann Arbor, MI 48108-9719, United States
[d] Visiting Scientist, Large Lakes Research Station, 9311 Groh Road, Grosse Ile, MI 48138, United States

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ABSTRACT

The Lake Michigan mercury mass balance model, LM2-Mercury, which was calibrated to a comprehensive data set collected from Lake Michigan during 1994–1995, was applied to predict long-term total mercury concentrations in lake water for different mercury loading and air concentration scenarios. The model predictions (volume-weighted, lakewide average total mercury concentrations) appear to be comparable to the available independent measurements from 2005 through 2013. The forecast based on the constant condition scenario, where conditions representative of 1994–1995 were held constant, shows that total mercury concentrations in the lake are near steady-state. The model was used to investigate the relative importance of different global versus regional impact scenarios on total mercury concentrations in Lake Michigan. The results suggest that mercury from global sources could contribute between 30% and 70% of total mercury water concentrations in Lake Michigan. Results for declining global emission scenarios modeled, based on both high and low global contribution estimates and information on mercury emission trends from current observations and the literature, indicate that total mercury concentrations in the water column in Lake Michigan will continue to decrease.

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Introduction

Mercury (Hg) is a toxic chemical that continues to be a concern due to its broad distribution in the environment through atmospheric deposition and its potential impact on wildlife and human health. It is persistent in multiple environmental compartments and bioaccumulates in the aquatic food chain (Knightes et al., 2009; Mason and Sullivan, 1997; Watras et al., 1998). States bordering Lake Michigan have fish consumption advisories to protect human health from the harmful effects of mercury. These advisories are based on species and length of the fish caught in the lake (http://www.great-lakes.net/humanhealth/fish/advisories.html#MI accessed on August 12, 2015).

During the past few decades, the United States Environmental Protection Agency (U.S. EPA), Environment Canada, and state agencies have taken regulatory actions to reduce mercury emissions and mercury usage within the Great Lakes region. They have funded projects that focus on the relationship between Hg sources and concentrations in the Great Lakes region's aquatic ecosystems. U.S. EPA Great Lakes National Program Office (GLNPO) initiated the Lake Michigan Mass Balance Project (LMMBP) in 1994 to intensively monitor and model mercury, atrazine, and PCBs (polychlorinated biphenyls) in the Lake Michigan ecosystem (McCarty et al., 2006; U.S. EPA, 1997). More recently, the Great Lakes Restoration Initiative (GLRI) identified mercury as one of the legacy pollutants listed in Focus Area 1 of the GLRI action plan (http://glri.us/pdfs/glri_actionplan.pdf accessed on March 5, 2015).

The relationship between mercury sources and observed concentrations in an aquatic system is often not straightforward. Studies have shown that atmospheric input including wet and dry deposition and gaseous mercury absorption is the primary source of mercury to Lake Michigan (Vette et al., 2002; Zhang et al., 2014). Atmospheric input to the lake originates from sources within the Lake Michigan region as well as from long distance sources around the world due to mercury's long residence time in the atmosphere. Although local anthropogenic sources such as emissions from coal-fired power plants account for a significant share of mercury inputs to the lake, the share of mercury inputs from Asia in the global sources is likely growing as emissions in Asia have been growing. The long range atmospheric transport of mercury can make it more difficult for policy makers and resource managers to effectively regulate mercury emissions within the lake region. This is especially true if long range transport of mercury to the region overwhelms regional mercury sources. In this case, efforts to control mercury within the local area may not have much of an impact on reducing environmental exposure concentrations in lake water.

It is a significant challenge to quantitatively estimate the long-term impact of regional and global mercury emissions on the mercury

* Corresponding author.
E-mail address: rygwelski.kenneth@epa.gov (K.R. Rygwelski).

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concentrations in a large surface water system such as Lake Michigan. Using a global three-dimensional atmospheric chemistry transport model (GEOS-Chem) combined with ecosystem-scale fate and transport models, Selin et al. (2010) and Sunderland et al. (2009) demonstrated an approach to quantitatively assess the relationship between the potential contributions of atmospheric deposition by emission sources (global and regional) and impact on the Hg exposure projections for different and relatively small aquatic ecosystems in U.S. Northeast and Southeast regions and ocean basins. We used a similar approach but it is less extensive than theirs. The scope of our study is limited to assessing the connection between the total mercury concentration in Lake Michigan water and regional and global emission sources.

Data from recent monitoring programs (Gay and Risch, 2012; Krabbenhoft and Dove, 2012 and Dove et al., 2012; Marvin et al., 2012; including Krabbenhoft's recent August 2015 personal communication on tHg lake water concentration) suggest that mercury concentrations in water, sediment, and air compartments of Lake Michigan have been steadily decreasing over the past two decades. However, it remains uncertain how much of this decline is due to regional mercury emission reduction programs or to efforts taken to reduce mercury emissions around the world. Previous studies (Jeremiason et al., 2009; Mason and Sullivan, 1997; Zhang et al., 2014) assessed mercury cycling dynamics in multiple compartments of the Lake Michigan aquatic system but did not address the relative impact of global and regional sources of mercury on the concentration of mercury in Lake Michigan. As an initial effort to address this issue, we constructed long-term (sixty-two year) total mercury forcing data sets for Lake Michigan that reflect different scenarios for contributions from global and regional sources. These forcing data sets were used as inputs to the LM2-Mercury water quality model (Zhang et al., 2014), in order to simulate long-term total Hg concentrations in Lake Michigan. The LM2-Mercury water quality model described in our previous paper (Zhang et al., 2014) was calibrated to model mercury species cycling in Lake Michigan and is able to predict Hg^0, Hg^{2+}, and MeHg in Lake Michigan water and sediments. However, in this paper, only total mercury was simulated due to a lack of long term observational data for the mercury species. The results from these simulations will be used in a detailed investigation on the relationship between model-predicted mercury exposure concentrations in Lake Michigan and their impact on mercury body burdens in 5 to 6 year old lake trout and will be the focus of another paper in preparation.

The main objectives of this paper are as follows: 1) investigate the relative contributions of global and regional sources to total mercury concentrations in Lake Michigan, and 2) use the resultant contributions of global and regional sources within LM2-Mercury to make forecasts of possible long-term total mercury concentration trends in Lake Michigan.

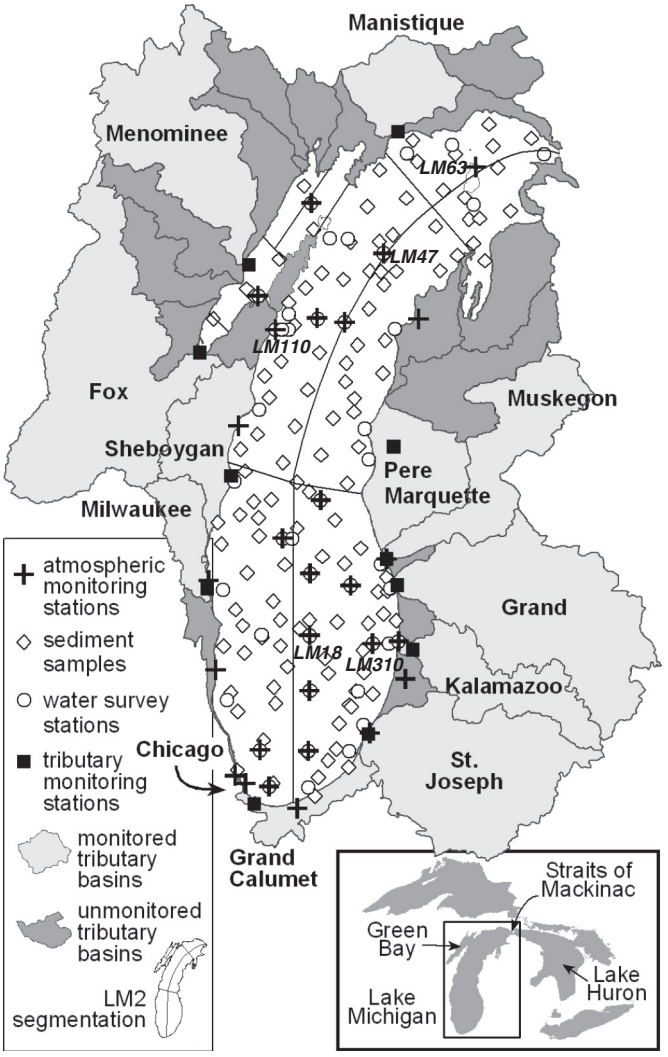

Fig. 1. Lake Michigan watershed and sampling sites (1994–1996).
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Methods and calculations

Model description

LM2-Mercury is a time varying, process-based mercury mass balance model that was previously developed and calibrated to simulate the transport and fate of total mercury (tHg) and mercury species in Lake Michigan (Zhang et al., 2014). The model uses data collected for air, water, and sediment collected from Lake Michigan (Fig. 1) during 1994–1996 (McCarty et al., 2004). Fig. 2 illustrates the processes associated with tHg fate and transport in Lake Michigan. The major external mercury loads to the model are from the atmosphere via wet and dry deposition, absorption of reactive gaseous mercury (RGM), and tributaries. The dry deposition referred in this paper only represents the mercury removed from the atmosphere by particle deposition. Physical and biogeochemical processes in the model include transport fields generated from the hydrodynamic model POMGL — Princeton Ocean Model for the Great Lakes (Schwab and Beletsky, 1998), air–water exchange, water–sediment cycling, and mercury partitioning. Based on the principle of conservation of mass and using the finite segment modeling approach, a general time-dependent finite differential equation was solved for a modeled state variable in a given segment. The time step used in model simulations was 3 h. Details of the model description

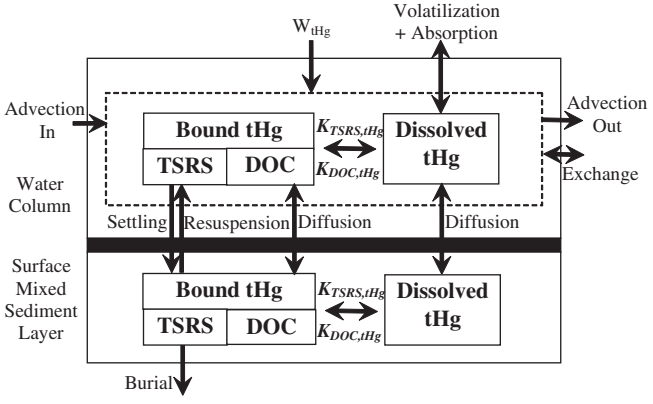

Fig. 2. Conceptual framework for total mercury (tHg), where: TSRS = total suspended and resuspendable solids; DOC = dissolved organic carbon; $K_{TSRS,tHg}$ = partitioning of total Hg between dissolved phase and particulate (TSRS) phase; $K_{DOC,tHg}$ = partitioning total Hg between dissolved phase and particulate (DOC) phase; W = sum of all loads.
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