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Comparison of mercury mass loading in streams to atmospheric deposition in watersheds of Western North America: Evidence for non-atmospheric mercury sources

Joseph Domagalski^{a,*}, Michael S. Majewski^a, Charles N. Alpers^a, Chris S. Eckley^b, Collin A. Eagles-Smith^c, Liam Schenk^d, Susan Wherry^e

^a U.S. Geological Survey, California Water Science Center, 6000 J Street, Placer Hall, Sacramento, CA 95819, United States

^b U.S. Environmental Protection Agency, Office of Environmental Assessment, EPA-Region 10, 1200 6th Ave., Suite 900, Seattle, WA 98101, United States

^c U.S. Geological Survey, Forest and Rangeland Ecosystem Science Center, 3200 SW Jefferson Way, Corvallis, OR 97331, United States

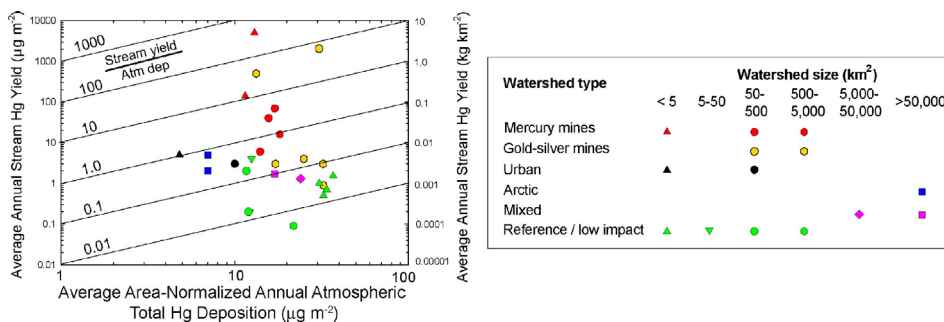
^d U.S. Geological Survey, Oregon Water Science Center, 2795 Anderson Ave., Suite 106, Klamath Falls, OR 97603, United States

^e U.S. Geological Survey, Oregon Water Science Center, 2130 SW 5th Ave., Portland, OR 97201, United States

HIGHLIGHTS

- Watersheds have geologic, anthropogenic, and global atmospheric sources of Hg.
- This study investigated sources of stream Hg loads in the Western U.S. and Canadian-Alaskan Arctic.
- Abandoned mines increased annual stream load relative to deposition.
- Watersheds in urban areas had > Hg loads/unit area than vegetated areas.
- River loads are attenuated in watersheds with forests and other natural vegetation.

GRAPHICAL ABSTRACT



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ABSTRACT

Annual stream loads of mercury (Hg) and inputs of wet and dry atmospheric Hg deposition to the landscape were investigated in watersheds of the Western United States and the Canadian-Alaskan Arctic. Mercury concentration and discharge data from flow gauging stations were used to compute annual mass loads with regression models. Measured wet and modeled dry deposition were compared to annual stream loads to compute ratios of Hg stream load to total Hg atmospheric deposition. Watershed land uses or cover included mining, undeveloped, urbanized, and mixed. Of 27 watersheds that were investigated, 15 had some degree of mining, either of Hg or precious metals (gold or silver), where Hg was used in the amalgamation process. Stream loads in excess of annual Hg atmospheric deposition (ratio > 1) were observed in watersheds containing Hg mines and in relatively small and medium-sized watersheds with gold or silver mines, however, larger watersheds containing gold or silver mines, some of which also contain large dams that trap sediment, were sometimes associated with lower load ratios (<0.2). In the non-Arctic regions, watersheds with natural vegetation tended to have low ratios of stream load to Hg deposition (<0.1), whereas urbanized areas had higher ratios (0.34–1.0) because of impervious surfaces. This indicated that, in ecosystems with natural vegetation, Hg is retained in the soil and may be transported subsequently to streams as a result of erosion or in association with dissolved organic carbon. Arctic watersheds (Mackenzie and Yukon Rivers) had a relatively elevated ratio of stream load to atmospheric deposition (0.27 and

* Corresponding author.

E-mail address: joed@usgs.gov (J. Domagalski).

0.74), possibly because of melting glaciers or permafrost releasing previously stored Hg to the streams. Overall, our research highlights the important role of watershed characteristics in determining whether a landscape is a net source of Hg or a net sink of atmospheric Hg.

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1. Introduction

Mercury (Hg) is a globally distributed contaminant in aquatic systems that can impact various species through bioaccumulation (Selin, 2009) with potential human health effects, especially from fish consumption (Ullrich et al., 2001; Selin, 2009). Within the United States, Hg is currently the seventh most common contaminant listed under the federal Clean Water Act as impairing rivers and streams, and is the most prevalent chemical for which a related management plan, known as a Total Maximum Daily Load (TMDL), is either underway or planned (http://iaspub.epa.gov/waters10/attains_nation_cy.control?p_report_type=T, accessed June 11, 2015). TMDLs for Hg are designed to reduce levels in top trophic level fish. Understanding the sources or pathways, and forms of Hg in water bodies is critical for a successful TMDL.

Major sources or pathways of Hg to the aquatic environment include atmospheric deposition, erosion from geologic material, urban discharges, agricultural and industrial discharges, mining locations, and combustion (Wang et al., 2004; Pirrone et al., 2009). In Eastern North America, it is known that atmospheric deposition of Hg from power plant emissions is a major pathway to the landscape and streams (Wentz et al., 2014). The predominant form of atmospheric Hg (>95%) is gaseous elemental (Hg(0)) (Schroeder and Munthe, 1998; Fitzgerald, 1986). Gaseous oxidized Hg, and particulate bound Hg are also present, but at lower concentrations. Hg(0) has an atmospheric half-life of 6 months to one year (Schroeder and Munthe, 1998) whereas gaseous oxidized Hg and particle bound Hg have much shorter half-lives, on the order of days, and are rapidly deposited. The primary removal mechanism for Hg(0) is by oxidation to the gaseous oxidized form followed by deposition, either wet or dry. Gaseous oxidized Hg can also be reduced back to elemental Hg allowing re-entry to the atmosphere. These processes also occur at Hg mine and amalgamation sites. Evasion, wind erosion, and runoff are potential release mechanisms for Hg from these areas to streams.

Anthropogenic uses of Hg include industrial processes for chemical manufacturing, fluorescent light bulbs, and dental fillings, but these are in the process of being phased out. Within Western North America, one of the largest sources of Hg to the environment was the historical processing or recovery of gold and silver by amalgamation (Nriagu, 1994; Hylander and Meili, 2003). Although largely eliminated in North America, amalgamation is still used by artisanal miners in many developing countries (Telmer and Veiga, 2009). Amalgamation was used historically in California from the mid to late 19th century to the mid 20th century (Alpers et al., 2005). It has been estimated that about 4,535,000 kg of Hg were lost to the environment from placer mines and an additional 1,360,000 kg from hard rock mining, within California, as a result of its use in amalgamation (Churchill, 2000).

Previous studies, such as Brigham et al. (2009), have compared atmospheric Hg deposition to Hg transport in streams to provide insight on watershed processes that affect Hg transport. Their study included locations in the Western, Mid-Western, and Eastern U.S., and estimated that 3–44% of the atmospherically deposited Hg was exported from the watersheds on an annual basis. In this study, we compare annual riverine loads of total Hg (THg) at selected river locations in the Western U.S. and the Canadian-Alaskan Arctic to annual estimates of wet and dry deposition over the upstream watersheds. Land use in these watersheds ranges from largely pristine to having variable levels of anthropogenic disturbance including urbanization, agriculture, and mining. Climatic conditions for the selected watersheds also varied, especially with respect to rainfall. Comparisons of this sort provide a first step at

quantifying the relative importance of atmospheric and non-atmospheric Hg sources or pathways to stream loads, as well as a way to estimate the amount of Hg stored in watersheds. Stream flow typically transports considerably less THg from the watershed than is deposited by wet and dry depositional processes (Shanley et al., 2008; Brigham et al., 2009; Journey et al., 2012). Runoff contributions of the atmospherically deposited Hg to river loads, however, are variable and difficult to quantify. The amount of Hg transported to streams and rivers by runoff attributed to atmospheric deposition is dependent upon the annual rainfall amount, the amount of Hg retained by the soil and vegetation, and the density of impervious surfaces within the watershed (Shanley et al., 2008; Brigham et al., 2009; Journey et al., 2012; Tsai and Hoenicke, 2001).

Watersheds of Western North America have a variety of Hg pathways because of variable geographic location, geology, and land uses. Atmospheric deposition originates from a combination of local sources and trans-ocean atmospheric transport from Asia (Strode et al., 2008). Western North America also has a number of mining districts for precious metals (gold and silver), base metals, and Hg (Brobst and Pratt, 1973). Mercury deposits in the Western U.S. are found primarily in California, Nevada, and Oregon associated with either altered mafic volcanic rocks or with epithermal hydrothermal systems (Gustin et al., 2000; Rytuba, 2003). Gold and silver deposits occur throughout the Western U.S. (Long et al., 1998), and are large potential sources of Hg to water, because of discharges from historical mining that used the Hg amalgamation process for metal recovery.

A study of this nature has a considerable uncertainty, some of which is difficult to quantify. Measurements of Hg in wet deposition are available, but limited. The Mercury Deposition Network (MDN) (<http://nadp.sws.uiuc.edu/mdn/>, accessed June 17, 2015), maintains more than 100 sites throughout the continental U.S., Alaska, and small portions of southern Canada for the purpose of collecting and measuring precipitation samples for Hg concentrations and fluxes. The majority of the sites, however, are east of the Rocky Mountain States. Current monitoring methods are not able to provide spatially comprehensive information on the wet and dry deposition of Hg, so modeling has become an important tool to estimate atmospheric deposition of Hg over wide areas (Gbor et al., 2007; Ryaboshapko et al., 2007; Lin et al., 2006). Model estimates of Hg dry deposition range from <1 to 3 times the rate of wet deposition (St. Louis et al., 2001; Evers and Clair, 2005; Seigneur et al., 2004). These results suggest that atmospheric dry deposition of Hg can be more significant than previously thought (Risch et al., 2012; Zhang et al., 2012), but there is no national sampling network and fewer direct measurements have been made. This study relied on only one year of modeled dry deposition. We recognize that year-to-year variability of dry deposition almost certainly occurs, therefore, the dry deposition estimate is a limitation that needs to be pointed out. In addition, local sources of potentially high Hg emissions can contribute to stream loads and may not be captured by the Hg deposition network or the dry deposition model.

Uncertainty of stream loads can be calculated using statistical techniques, and the uncertainty usually decreases with longer term monitoring, especially monitoring that takes into account variation in stream flow throughout the year. Climatic effects must also be considered. Some locations in this study had only a few years of water sample collection for Hg analysis, whereas others had up to 20 years. If sample collection took place during drought or during years of above average rainfall, stream load estimates, although accurate, would not fully explain the longer-term transport processes. Effects of climate can be better understood for sites with longer periods of record, but are difficult to

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