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Mercury in western North America: A synthesis of environmental contamination, fluxes, bioaccumulation, and risk to fish and wildlife

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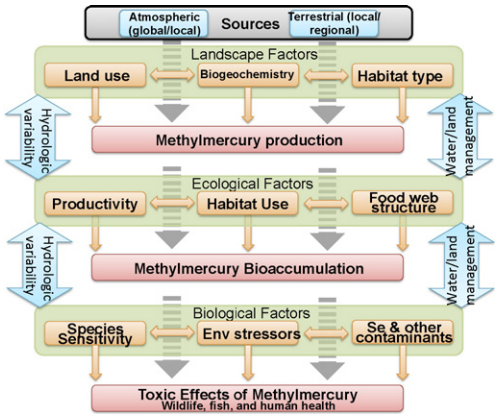
HIGHLIGHTS

- Western North America is a unique and diverse landscape.
- Mercury contamination is widespread from many different source types.
- The Western North America Mercury Synthesis summarizes mercury across this landscape.
- Landscape structure has an important influence on mercury processes and distribution.
- Biological mercury exposure is decoupled from inorganic mercury sources.

GRAPHICAL ABSTRACT



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ABSTRACT

Western North America is a region defined by extreme gradients in geomorphology and climate, which support a diverse array of ecological communities and natural resources. The region also has extreme gradients in mercury (Hg) contamination due to a broad distribution of inorganic Hg sources. These diverse Hg sources and a varied

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landscape create a unique and complex mosaic of ecological risk from Hg impairment associated with differential methylmercury (MeHg) production and bioaccumulation. Understanding the landscape-scale variation in the magnitude and relative importance of processes associated with Hg transport, methylation, and MeHg bioaccumulation requires a multidisciplinary synthesis that transcends small-scale variability. The Western North America Mercury Synthesis compiled, analyzed, and interpreted spatial and temporal patterns and drivers of Hg and MeHg in air, soil, vegetation, sediments, fish, and wildlife across western North America. This collaboration evaluated the potential risk from Hg to fish, and wildlife health, human exposure, and examined resource management activities that influenced the risk of Hg contamination. This paper integrates the key information presented across the individual papers that comprise the synthesis. The compiled information indicates that Hg contamination is widespread, but heterogeneous, across western North America. The storage and transport of inorganic Hg across landscape gradients are largely regulated by climate and land-cover factors such as plant productivity and precipitation. Importantly, there was a striking lack of concordance between pools and sources of inorganic Hg, and MeHg in aquatic food webs. Additionally, water management had a widespread influence on MeHg bioaccumulation in aquatic ecosystems, whereas mining impacts where relatively localized. These results highlight the decoupling of inorganic Hg sources with MeHg production and bioaccumulation. Together the findings indicate that developing efforts to control MeHg production in the West may be particularly beneficial for reducing food web exposure instead of efforts to simply control inorganic Hg sources.

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1. Introduction

The widespread distribution of environmental mercury (Hg) from both anthropogenic activities and natural sources makes it a globally prevalent threat to human health and natural resource conservation (Krabbenhoft and Sunderland, 2013). The cycling and pathways of inorganic Hg and methylmercury (MeHg) through the environment are varied and complex, and are strongly tied to processes that occur at local, regional, and global scales (Morel et al., 1998). Data syntheses at broad spatial extents are useful approaches for transcending site-specific variability, and for better understanding the emergent ecosystem properties that influence both Hg risk, as well as the processes that regulate it within and across landscapes (Chetelat et al., 2015; Evers and Clair, 2005; Wiener et al., 2012). As part of a series of papers associated with the Western North America Mercury Synthesis (WNAMS), environmental Hg contamination across this geographic area was synthesized, focusing on Hg in air, wet and dry atmospheric deposition, watersheds, soils, sediment, aquatic food webs, fish, and birds. Objectives of this synthesis were to compile diverse sets of Hg data in multiple environmental matrices from across the western region of North America, and conduct a comprehensive and integrated analysis of data to better understand the drivers of Hg contamination, distribution, and bioaccumulation across this diverse region.

1.1. Rational for concerns about mercury in the environment

More than 80% of the fish-consumption advisories posted in the United States and Canada are wholly or partially attributed to Hg (US EPA, 2011; Ontario Ministry of the Environment and Climate Change 2015). This toxic metal poses a potential health threat to humans, fish, and wildlife because of the high bioavailability, trophic biomagnification, extreme toxicity, and reproductive effects of MeHg (Driscoll et al., 2013; Lavoie et al., 2013; Tan et al., 2009). Most of the Hg in air, atmospheric deposition, soils, and parent rocks exists as inorganic forms, yet nearly all of the Hg bioaccumulated in fish and wildlife is MeHg (Bloom, 1992; Greenfield and Jahn, 2010; Van Walleghem et al., 2007), an organic compound produced primarily by Hg-methylating anaerobic bacteria present in surficial sediments, wetlands, and anoxic/suboxic bottom waters (Benoit et al., 2003; Gilmour et al., 2013; Parks et al., 2013).

In aquatic food webs, MeHg bioaccumulates in piscivorous fish to concentrations that can exceed those in water by a factor of 10^7 or more (Wiener et al., 2003). Aquatic food webs are important pathways for biotic exposure to MeHg, and its concentration in the diets or tissues of piscivorous fish and wildlife commonly exceed benchmark levels associated with deleterious effects on their health and reproduction (Ackerman et al., 2016b; Depew et al., 2012; Depew et al., 2013; Evers et al., 2007; Sandheinrich et al., 2011). Methylmercury in the diets of reproducing female birds and mammals readily passes to the developing egg or embryo (Ackerman et al., 2016a), life stages that are much more sensitive to MeHg exposure than the adult (Ackerman et al., 2013; Heinz et al., 2009; Wiener et al., 2003).

In humans, the consumption of fish is the primary pathway for MeHg exposure, and persons who frequently consume sport or commercial fishes are potentially at risk to the deleterious effects of MeHg exposure (Driscoll et al., 2013; Knobeloch et al., 2006; Mergler et al., 2007). Methylmercury is highly neurotoxic, adversely affecting both the adult and developing brain (Clarkson and Magos, 2006; McKelvey and Oken, 2012). Toxicological concern about MeHg in wildlife has focused largely on piscivores, because consumption of fish has long been considered the principal pathway for harmful MeHg exposure (Scheuhammer and Sandheinrich, 2008). However, during the past decade high concentrations of Hg (present as MeHg) have been reported in songbirds, shorebirds, and bats that feed on aquatic invertebrates (Eagles-Smith et al., 2009a; Jackson et al., 2015; Yates et al., 2014), including endangered species (Ackerman et al., 2012). Most of these species with elevated MeHg levels are trophically linked to food webs in Hg-methylating environments, including wetlands, streams, lakes, or hydric soils, and have a diet that includes emergent insects with aquatic larval stages or riparian consumers of emergent insects, such as spiders (Cristol et al., 2008; Jackson et al., 2015).

1.2. Western North America: a diverse region with varied Hg sources and substantial natural resources

Extreme geomorphological variation is a defining feature of the western region of North America. This variation drives substantial diversity in land cover, climate, and ecosystem processes, and differentiates the region from the rest of the continent. Areas with both the lowest (Mohave Desert, 30–60 mm y^{-1}) and highest (Pacific Northwest temperate rain forests, 3300–4670 mm y^{-1}) annual precipitation in North America reside in the West, and extremes in climate typically occur over very short distances because of complex mountain topography and elevation gradients that result in a high diversity of biomes and ecosystem types (Fig. 1). Mercury is uniquely superimposed across these diverse ecosystems in the West from widespread historic Hg, gold (Au), and silver (Ag) mining activities, as well as from industrial emissions and trans-Pacific transport and deposition (Jaffe and Strode, 2008; Obrist et al., 2008; Rytuba, 2003; Schmeltz et al., 2011). Thus, the distinct patterns of soil development, plant dynamics, fish and wildlife distribution, and carbon and nutrient geochemistry associated with western landscapes can differentially influence the occurrence,

Fig. 1. Map of major land cover classes across western North America derived from National Land Cover Database (http://www.mrlc.gov/nlcd2011.php), and North American Land Change Monitoring System Land Cover Atlas (http://www.cec.org/tools-and-resources/map-files/land-cover-2010).

transport, methylation, and bioaccumulation of Hg. These patterns also form a complex landscape mosaic that serves as the framework for natural resource management in the West, which is largely dominated by the need to balance: (1) limited water availability with growing urban, agricultural, energy, and ecological needs (e.g., water storage and transport, and flood control); (2) fire management with forest and shrubland health; and (3) grazing and resource extraction (e.g., timber harvest and mining) with habitat conservation and water quality. Mercury contamination adds another potential layer of complexity to these resource management challenges because many factors associated with addressing these challenges are intractably linked to processes that also influence Hg cycling. As a result, different approaches to address these challenges may either exacerbate or mitigate Hg availability, and a better understanding of the landscape and process-based drivers of these interactions could inform decision-making strategies for addressing resource management challenges within the context of also trying to minimize the potential impacts of Hg.

Natural resource management may have a particularly strong influence on Hg cycling and availability in the West, because a large proportion of the region is publicly owned or managed. In fact, more than 50% of the land area in the Western United States is publicly owned or managed, much of it in the federal domain (Table 1). As a result, nearly half of the 2.1 million lakes, 6.2 million km of rivers and streams, and 153,000 km^2 of wetlands are owned or managed by the U.S. federal government and the citizens of the United States.

Extensive management of aquatic and terrestrial habitats for both conservation and resource extraction purposes has implications for Hg cycling in the West because of potential alterations to ecosystem processes that influence inorganic Hg transport and loading, and MeHg production and bioaccumulation. Water management in particular is a defining characteristic of western resource conservation that is inherently linked to the Hg cycle. It is among the continent's most complex and pervasive resource management issues, and has greatly influenced western land use, development, and conservation. The spatial and

Table 1
Land area, lake and reservoir number and area, river length, and wetland area in the western U.S. that is located within public land boundaries or is managed by federal agencies.

	Land[a]	Lakes, reserviors and playas[b]		Stream and rivers[b]	Wetlands[c]
Federal agency	Total area (km^2)	Number	Total area (km^2)	Total length (km)	Total area (km^2)
Bureau of Land Management	996,755	251,532	23,494	1,328,322	17,639
U.S. Forest Service	749,098	183,969	4869	1,259,375	6426
U.S. Fish and Wildlife Service	400,236	457,686	30,760	329,199	49,961
National Park Service	307,248	75,034	5310	235,699	5803
U.S. Department of Defense	69,983	7374	1001	86,218	1892
U.S. Bureau of Reclamation	8821	4102	7823	6397	407
Bureau of Indian Affairs	1095	780	733	2133	21
U.S. Army Corps of Engineers	–	75	5444	–	–
Other Federal Agency	7923	604	82	8076	17
Total Federal	2,541,160	981,156	79,516	3,255,420	82,167
Total in Western US	4,920,453[d]	2,086,723	100,660	6,161,081	152,904

[a] 1:1,000,000-Scale Federal Lands of the United States, 2014 (https://www.sciencebase.gov/catalog/item/552d6eabe4b0b22a157f563c).
[b] National Hydrography Dataset (High Resolution/1:24 k) (http://nhd.usgs.gov/data.html).
[c] National Land Cover Database, 2011 (http://www.mrlc.gov/nlcd2011.php).
[d] Gorte et al., 2012. Federal Land Ownership: Overview and Data. Congressional Research Service 7–5700, RA2346.

temporal inconsistency of water availability, and the need to store and transport water throughout the West, spurred the construction of nearly 23,000 dams and a network of 214,000 km of water conveyance (Graf, 1999; Sabo et al., 2010). As a result, approximately 76% of the total flow in western rivers is appropriated for societal uses (particularly agriculture), with anthropogenic water draws from several watersheds exceeding their annual flow (Sabo et al., 2010). Such widespread hydrologic engineering in the west has transformed the landscape and dramatically altered the physical, chemical, and biological characteristics of river systems (Friedl and Wuest, 2002; Ward and Stanford, 1995). The ecological impacts of these activities are myriad (Bunn and Arthington, 2002; Johnson and Olden, 2008; Miller et al., 1989; Moyle and Mount, 2007), and include alterations to many of the biogeochemical and bioaccumulation processes that directly influence Hg cycling (Friedl and Wuest, 2002; Hall et al., 2005).

1.3. Conceptual model of Hg drivers and cycling in western North America

Fig. 2 provides a conceptual model of the processes and factors involved in Hg cycling, and how they relate to resource management in the West (Fig. 2). Inorganic Hg is delivered to the environment in the West from a variety of global, local, and regional sources. Landscape factors (such as land use, biogeochemistry, and habitat) interact with the deposited Hg in various ways, resulting in localized MeHg production and transport. Once MeHg is produced, ecological factors such as ecosystem productivity, habitat use, and food web structure result in variable MeHg bioaccumulation. Once MeHg bioaccumulates into organisms, biological factors such as species sensitivity, exposure to other environmental stressors, and exposure to selenium and other contaminants result in deleterious effects to fish, wildlife, and human health.

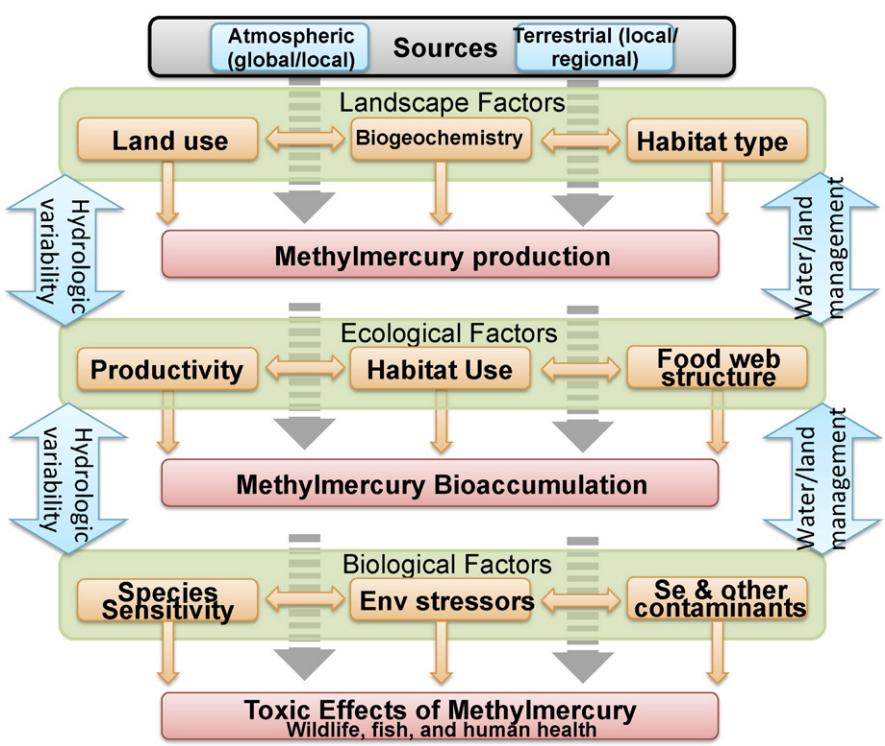

Fig. 2. Conceptual model of mercury cycling and bioaccumulation in western North America. Pink boxes represent key endpoints in the cycle that are controlled by the factors that influence Hg at different scales of organization.

Importantly, several of these factors that regulate MeHg production and bioaccumulation can be altered through hydrologic variability and water and land management activities. Thus, if those two processes are the key linkages resulting in Hg risk to environmental health, then there may be opportunities to influence their magnitude through resource management actions. The Western North America Mercury Synthesis sought to conduct a first-order assessment of Hg distribution and cycling at the landscape scale in order to facilitate a better understanding of the coarse drivers of various processes of Hg cycling across this diverse region, and to identify key data gaps that currently obstruct predictive modeling of Hg across the landscape. This paper synthesizes the key contributions from the individual papers published as part of the WNAMS effort, and presence them within the context of the conceptual model described above (Fig. 2).

2. Landscape patterns of mercury in western North America

2.1. Atmospheric emissions and deposition patterns

Emission of inorganic Hg into the atmosphere largely occurs through three primary mechanisms – 1) direct natural emissions, 2) direct anthropogenic emissions, and 3) secondary re-emission from terrestrial (and aquatic) substrates; on a global scale these three mechanisms account for 5–13%, 29–33%, and 56–65% of total atmospheric Hg emissions, respectively (Agnan et al., 2016). Natural releases are largely from volcanic (Hinkley, 2003) and hydrothermal activity (Waring, 1965), as well as flux from geologically-enriched soils (Coolbaugh et al., 2002). Volcanic and hydrothermal Hg emissions for the conterminous United States (most of which are in the West) are approximately 3000 kg y^{-1} (Engle et al., 2006a) and estimates from the Fremont ice core in Wyoming indicate that global volcanic releases have accounted for approximately 6% of inorganic Hg deposition over the past 270 years (Schuster et al., 2002). Although difficult to quantify without substantial uncertainty, natural Hg emission fluxes from undisturbed, geogenically enriched substrates in the Western United States may be as much as 10,000–20,000 kg y^{-1} (Gustin et al., 2008), making this a substantial source of inorganic Hg to the atmosphere.

Direct anthropogenic Hg sources from industrial and energy facilities in Mexico and the western regions of the US and Canada release approximately 11,000 kg of Hg to the atmosphere annually (Environment Canada, 2013; US EPA, 2011; Naturales, 2011), which represents roughly one-fifth of total direct anthropogenic emissions across the Continent. Western states account for 19% of the total U.S. anthropogenic Hg emissions (US EPA, 2011), whereas emissions from western Canadian Provinces comprise 55% of Canadian releases (Environment Canada, 2013). The higher proportional emissions in western Canada are largely due to Hg from electricity production (primarily coal-fired power plants), which accounted for 82% of all emissions from this source-type in Canada. Conversely, only 15% of emissions from coal-fired power plants in the U.S. originated in the West. Because U.S. coal-fired power plant emissions exceed those in Canada, one of the biggest distinctions with regard to emissions across western North America is the substantially diminished release from electricity production industries in the West compared to the rest of the Continent (Fig. 3). In contrast to electricity generation, 60% of Continental emissions from cement production and 53% of mining/metal processing emissions originate in the West (Fig. 3), and together they account for 40% of western Hg emissions. Despite this, the magnitude of Hg emissions associated with electricity production from coal-fired power plants still make it the largest category of releases (42%) in the West (Fig. 3 insert). The difference in total emission magnitude between the western and eastern regions of North America is primarily due to the lower density of Hg emission sources in the West, and not because of larger individual emitters in the east (Fig. 4). Currently, the largest single source of Hg emissions in western North America is associated with a cement manufacturing facility in southern California (610 kg y^{-1}; Fig. 4) that is the 3rd largest emission source in North America (Fig. 4).

Anthropogenic releases of inorganic Hg to the atmosphere have declined more than 4-fold in the United States since 1990 (Weiss-Penzias et al., 2016). In response, approximately half of the monitoring stations across the US and Canada showed decreasing trends in Hg concentrations of wet deposition, and no sites had positive trends (Weiss-Penzias et al., 2016) between 1997 and 2013. Similarly, gaseous elemental Hg concentrations decreased by $-1.5 \pm 0.2\%$ y^{-1}. However, over more recent time periods (2007/2008–2013) when more monitoring stations were operational, most sites (64–70%) had no significant temporal trends (Weiss-Penzias et al., 2016). Yet, at those sites with significant linear trends, a higher percentage (17–30%) had increasing Hg concentrations compared to decreasing concentrations (6–13%). Additionally, the wet deposition monitoring stations in the central-western U.S. were unique in that they showed some of the largest increases in Hg concentrations over the more recent time period from 2007 to 2013 (Weiss-Penzias et al., 2016). These findings indicate that Hg wet deposition concentrations in the Western region of the continent do not appear to respond similarly to linked larger-scale emissions reduction, air concentrations, and wet deposition in the midwestern and eastern U.S. and globally (Zhang et al., 2016). This eastern versus western discordance remains unresolved, but it is interesting that despite the fact that sulfate concentrations in precipitation (which reflect sulfur dioxide emission from combustion sources such as coal-fired power plants, that are important sources of Hg) have declined similarly across both western and eastern monitoring sites (Weiss-Penzias et al., 2016) Hg-sulfate trends appear unequally linked. These results highlight the greater importance of non-local sources in the western portion of the continent, and the effects of rising Hg emissions and long-range transport in the free troposphere from regions outside of the US and Canada.

2.2. Landscape storage, transport, and re-emission

Upon deposition, inorganic Hg can become incorporated into soils and sequestered into plant tissues via stomatal and non-stomatal uptake pathways (Stamenkovic and Gustin, 2009). In fact, except in areas of geologic enrichment, approximately 80% of terrestrial Hg originates from exogenic (i.e., atmospheric) inputs (Fiorentino et al., 2011; Grimaldi et al., 2008; Pena-Rodriguez et al., 2014), and soils contain more than 90% of Hg stored in ecosystems (Corbitt et al., 2011; Hararuk et al., 2013; Obrist et al., 2012). Vegetation patterns influence Hg retention in soil due to the binding of Hg with the functional groups of soil organic matter. The precipitation gradients and geomorphic variation of western North America create diverse landscapes ranging from densely forested regions, to grassland valleys, to mostly barren deserts. This diversity has a strong influence on terrestrial Hg pools and deposition of atmospheric Hg. Obrist et al. (2016) synthesized data on vegetation and soil Hg concentrations in the Western U.S., and estimated that foliage retained 13,000 kg (44 μg m^{-2}) of Hg across the region, which is greater than estimates of combined annual wet and dry depositional loads (Lyman et al., 2007). As such, plant-derived Hg deposition may be the predominant depositional pathway that drives the soil Hg distribution across the region. In fact, Obrist et al. (2016) found that ecosystem and climatic properties associated with plant productivity (soil organic carbon, precipitation, and Normalized Difference Vegetation Index) were the factors most strongly linked with soil total Hg (THg) concentrations. Additionally, foliar Hg mass and (to a much lesser extent) wet THg deposition sources were more strongly associated with soil THg concentrations than were distance to coal-fired power plants or Hg, Au, or Ag mines. Cumulatively, these findings suggest that landcover and associated plant productivity are among the most important landscape drivers of soil inorganic Hg across the Western U.S. (Obrist et al., 2016). The marked gradients in these landscape drivers are largely driven by the pronounced variation in precipitation through the West, indicating that precipitation is an important, but largely

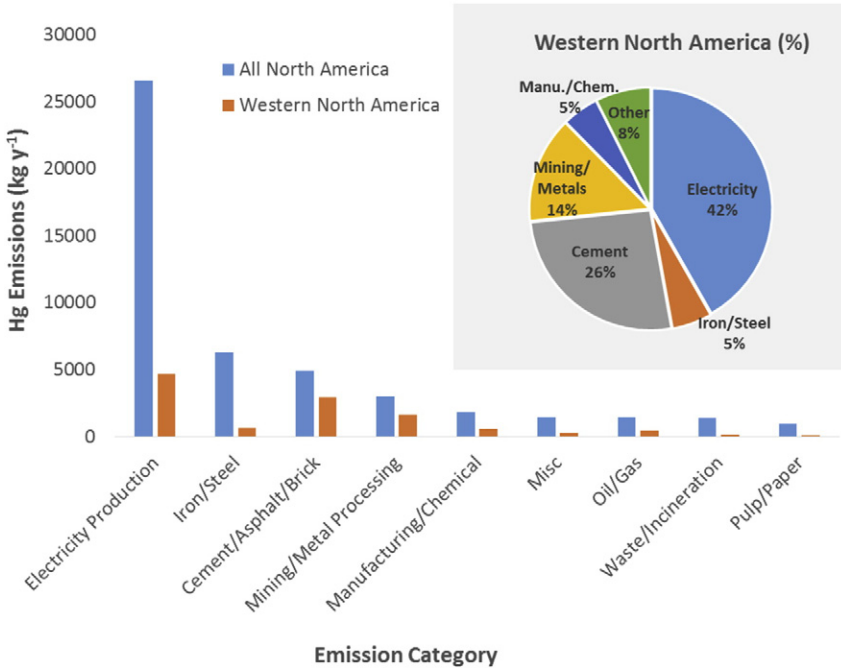

Fig. 3. The bar graph shows the total amount of Hg emitted from all North American sources (Canada, US, Mexico) and for only western North America (western states and provinces and Mexico). The inserted pie chart shows the relative proportion of Hg emission by sector type in western North America. The data originate from 2011 emission inventories from the US and Mexico and a 2013 inventory from Canada.

indirect contributor to soil Hg concentrations. As a result, soil Hg concentrations are generally highest in productive areas like the Marine West Coast Forest and Northwestern Forested Mountain ecoregions that contain high plant biomass, and lowest in the more barren and arid portions of the West such as the Temperate Sierra and Southern Semi-arid Highland ecoregions (Obrist et al., 2016). These patterns also translate to aquatic environments where forested waterbodies contain among the highest THg concentrations in bed sediments (Fleck et al., 2016).

Vegetation patterns not only influence soil Hg concentrations, but also the secondary re-emission, and thus, bi-directional terrestrial-atmospheric flux of inorganic Hg previously deposited on the Earth's surface from both natural and anthropogenic sources. An estimated 35,100 kg y^{-1} of Hg are emitted from soils to the atmosphere in the

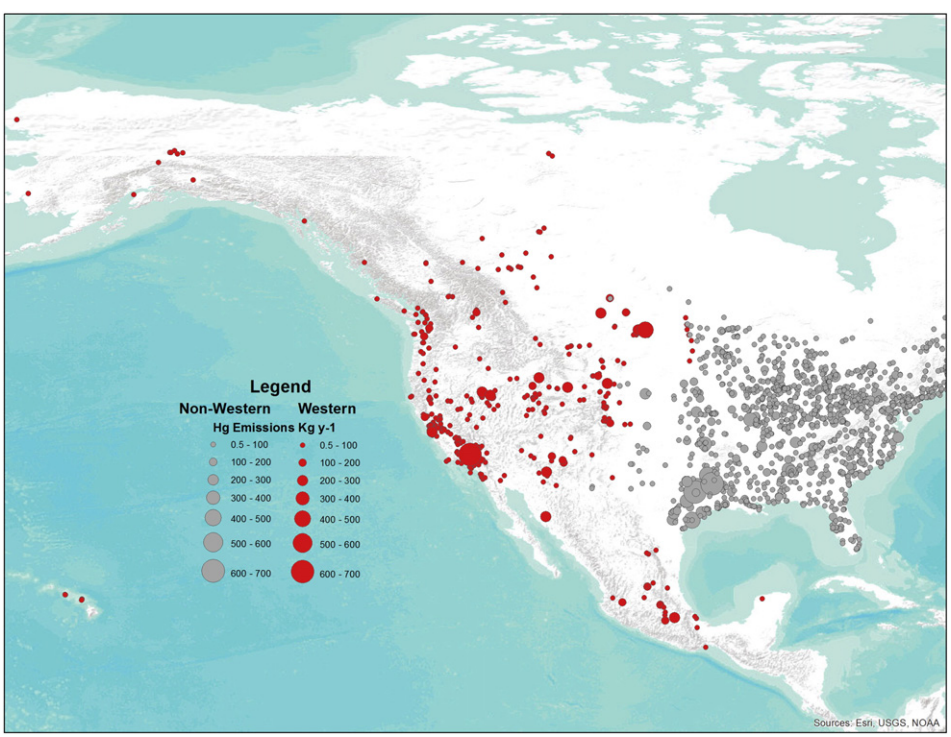

Fig. 4. Map of Hg emission sources >0.5 kg y^{-1} in western North America (red circles) and for the rest of the continent (grey circles). The data originate from 2011 emission inventories from the US and Mexico and 2013 data from Canada.

Western conterminous US (Eckley et al., 2016), which is approximately 4 times higher than annual anthropogenic emissions. However, 14,600 kg Hg y^{-1} are also taken up by vegetation, resulting in a net ecosystem flux of 20,500 kg Hg y^{-1} to the atmosphere (Eckley et al., 2016). This net emission from the terrestrial surface is similar, though slightly higher than estimated total wet deposition (19,400 kg y^{-1}). If dry Hg deposition is also considered, the total atmospheric inputs in the region likely exceed emissions, and the overall net-flux would favor net soil accumulation. However, the flux rates are highly variable throughout the West, with the heavily vegetated areas generally exhibiting higher Hg sequestration rates than the sparsely vegetated regions. For example, estimated wet Hg deposition is approximately 2.8 and 2.1 times greater than net ecosystem emissions in the Northwest Forested Mountain and Marine West Coast Forest ecoregions, respectively, whereas the land surface in the Mediterranean California ecoregion emits roughly 7 times more Hg to the atmosphere than is received as wet deposition (Eckley et al., 2016). Uncertainty in dry deposition rates convolutes estimates for the other more arid ecoregions (Deserts, Southern Semi-Arid Highlands, and Temperate Sierras), where total deposition is roughly equivalent to, but potentially slightly greater than emissions. The limited vegetation in these ecoregions suggests that large proportion of the Hg deposited in many areas of the West is likely not sequestered in vegetation and can be re-emitted or continue to cycle in the environment. These high emission/re-emission rates, particularly from sparsely vegetated areas could obscure or add a time-lag to the effects of reduced anthropogenic emissions on observed air Hg concentrations. This may be a contributing reason, along with other factors, why the very large reductions in the Canadian and US emission inventories have resulted in only small changes in measured atmospheric gaseous elemental mercury (GEM) concentrations (Weiss-Penzias et al., 2016).

2.3. Landscape disturbance influences on mercury

Although vegetation and soil pools are potentially long-term repositories of Hg within western ecosystems, wildfire is a defining feature of western landscapes that releases stored Hg to the atmosphere via biomass combustion and heat transfer to the underlying soil (Biswas et al., 2007; Engle et al., 2006b; Friedli et al., 2001; Woodruff and Cannon, 2010). The amount of Hg released from the landscape during wildfires depends upon the area burned, the amount of Hg stored in vegetation and soil, and the burn intensity or fire severity. Less Hg is released from soils during low and moderate severity, than high severity fires because of insufficient subsurface soil heating (Webster et al., 2016). As a result, low severity fires are predicted to release Hg at a rate of 18–34 μg Hg kg-fuel^{-1}; whereas high severity fires are estimated to release 125–780 μg Hg kg-fuel^{-1}. Overall, Hg released from the soil is the largest contributor to forest fire emissions, followed by Hg released from duff, foliage, woody fuel, litter, and branches. An average of 759,000 ha of land in the western U.S. and Alaska is burned by wildfires each year (Littell et al., 2009). When accounting for burn severity, 3100 ± 1900 kg Hg y^{-1} is estimated to be released from forest fires in the western continental U.S. alone (Webster et al., 2016). On average, low severity burning occurs across a little over half the burn area of a typical wildfire, whereas 19% of the area is subjected to high intensity burning. As a result, this new emission estimate is nearly 5 times lower than those that previously did not take fire severity into consideration (Wiedinmyer and Friedli, 2007). However, current trends of wildfire on the North American landscape are following global patterns of increasing occurrence and severity (Kasischke et al., 2010; McCoy and Burn, 2005; Westerling and Bryant, 2008; Westerling et al., 2006), thus the fire-based contribution of Hg in the West could increase concomitantly, which could offset some of the benefits of reduced anthropogenic emissions.

Although patterns of atmospheric transport, deposition, and sequestration are important aspects of global and regional Hg budgets and cycling, there is uncertainty surrounding their nexus to biological exposure and risk associated with MeHg. Inorganic Hg must first be mobilized to aquatic environments where anaerobic microbial communities convert it to MeHg under appropriate biogeochemical conditions (Gilmour et al., 2013). Landscape disturbance is a major component of mobilizing Hg to aquatic environments, and can include wildfire, resource extraction, and land development. Western North America contains several areas of natural Hg enrichment, as well as Au and Ag deposits where Hg was used in the extraction process (Fig. 5; Rytuba, 2000). Historical Hg mining in the West produced an estimated 130 million kg of Hg (Rytuba, 2003), of which approximately 30 million kg remains in and around mined regions. Additionally, at least 13 million kg of Hg are thought to have been released from Au and Ag mining operations throughout the West where Hg amalgamation was used, resulting in Hg contamination of downstream sediments and riverbanks (Alpers et al., 2005; Hunerlach and Alpers, 2003; Singer et al., 2013).

The impact of mining activity on inorganic Hg transport to aquatic systems has been widely reported (Gray et al., 2004; Hylander and Meili, 2003; Malm, 1998; Wang et al., 2004), and many of the highest surficial bed sediment THg concentrations across western North America are associated with gold, silver, and mercury mines (Fleck et al., 2016). Additionally, sediment cores from lakes in western North America with historic mining activity in their watersheds illustrate unique accumulation patterns in comparison to lakes without direct mining impact (Drevnick et al., 2016). In mine-impacted lakes, sediments commonly transition from background levels to rapid increases that span orders of magnitude upon initiation of mining, followed by a rapid decline and return to a new steady state following mine closure and clean-up (Drevnick et al., 2016). Conversely, sediments from lakes that have not been influenced by mining show gradually increasing Hg accumulation rates over time without the distinctive and dramatic peaks associated with mining influences. In riverine environments, the influence of mining on Hg loading and transport is highly scale dependent. In sites across the West, the proportional importance of inorganic Hg stream load relative to atmospheric Hg deposition was positively correlated with mining density in streams with small (<500 km^2) watersheds, whereas there was no relationship with mine density in watersheds larger than 500 km^2 (Domagalski et al., 2016). This suggests that the direct influence of mining on aqueous Hg loads is often localized, and the mining signal is rapidly lost in large catchments that are disproportionately influenced by the surrounding terrestrial environment and receive a greater aggregate amount of atmospheric Hg deposition. However, it is important to note that sediment-bound Hg from legacy mining operations can be transported hundreds of km and broadly distribute in sediments of environments that are well downstream of intensive past mining activity, such as the Sacramento-San Joaquin River Estuary (Heim et al., 2007; Singer et al., 2013), where its availability for biological uptake will be constrained by the Hg methylating activity of the environment.

Landscape disturbance associated with development is generally more widespread than mining activities across the West, and can also influence Hg transport to aquatic systems. Landscape disturbances can mobilize soils and vegetation, which contain substantial ecosystem pools of Hg (Obrist et al., 2016), as well as reduce soil permeability to influence erosion. Terrestrial vegetation and associated soil organic matter also contribute to watershed Hg cycling through delivery of dissolved organic matter (DOM) that exerts both direct and indirect controls on the transport and reactivity of Hg in aquatic systems. As a result, THg accumulation rates in sediments from lakes without direct point sources across western North America are positively correlated with the extent of watershed development, and inversely correlated with distance to major urban area (Drevnick et al., 2016). Additionally, Hg accumulation rates in lake sediments have generally been increasing across western North America since the preindustrial period. Rates of change in heavily disturbed watersheds increased dramatically between 1930 and 1960, and their accumulation rates are now approximately 5 times higher than during pre-industrial times. In contrast,

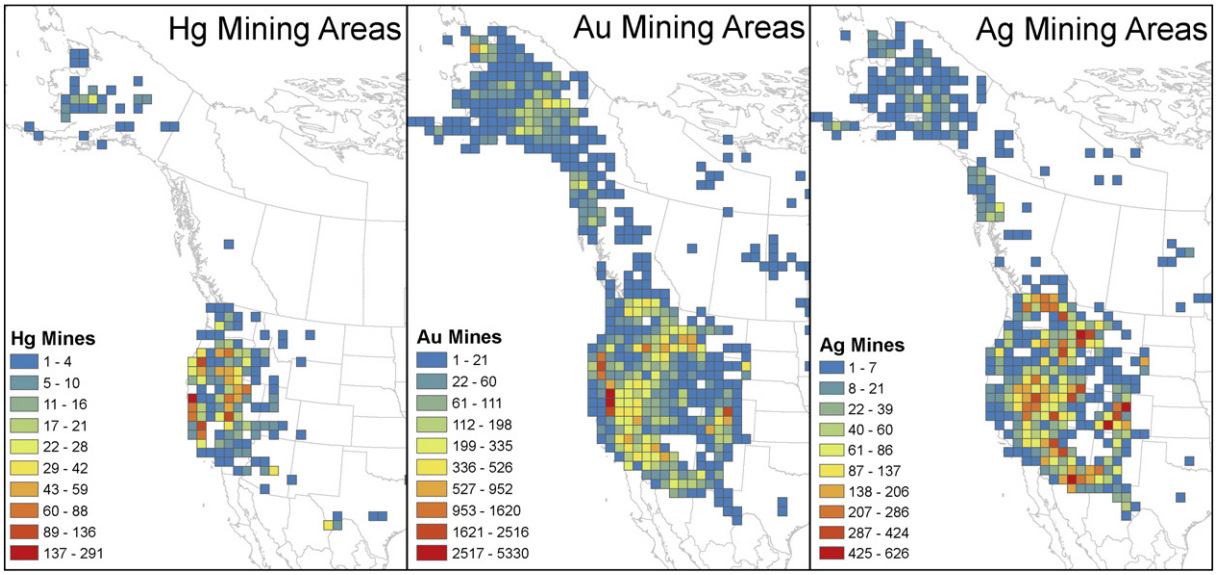

Fig. 5. Distribution of mercury (Hg), gold (Au), and silver (Ag) mines across western North America. Grids represent 100 km × 100 km areas and the grid color indicates the number of past-producing mines within each grid cell.

there has been less than a two-fold increase in Hg accumulation rates during the same time period from lakes in moderate, low, and undisturbed watersheds (Drevnick et al., 2016). Thus, the combination of increasing atmospheric deposition over time in concert with Hg storage in terrestrial habitats and watershed disturbance has substantially increased sediment THg concentrations in lakes, which are at their highest levels in modern history. Importantly, <1% of Hg deposited in little-disturbed watersheds from areas of dense vegetation, such as in the Northwest Forested Mountain ecoregion, is delivered to lakes (Drevnick et al., 2016), which is consistent with the findings of Obrist et al. (2016); Eckley et al. (2016), and Domagalski et al. (2016) that these areas are likely large "sinks" for Hg. Thus, forest conservation and management may be an important and little-recognized component for addressing future Hg risk in those areas.

The diversity of Hg sources, watershed characteristics, and disturbance regimes throughout western North America has resulted in substantial heterogeneity in bed sediment THg concentrations that ranges five orders of magnitude across the West (Drevnick et al., 2016; Fleck et al., 2016). This expansive gradient not only allows for a better understanding in how inorganic Hg is distributed across the region, but also facilitates a coarse evaluation of whether the patterns in THg distribution likewise confer risk to environmental health through exposure to MeHg. Methylmercury is the form of Hg most strongly related to toxicological risk. Methylmercury production primarily occurs in sediments and anoxic waters, and is facilitated by a suite of drivers that influence both the activity of Hg(II)-methylating microbes and the availability of Hg(II) to those organisms. Importantly, MeHg concentrations are typically only loosely correlated with inorganic Hg concentrations across broad spatial scales (Krabbenhoft and Sunderland, 2013) and often not correlated at smaller spatial scales (Heim et al., 2007). In fact, across more than 1500 unique sites throughout the West, sediment MeHg concentrations were only weakly correlated with sediment THg concentrations ($r^2 = 0.25$) and substantial variability among habitat types (Fleck et al., 2016), indicating that the relationship is not only weak, but also inconsistent across habitats. This spatial decoupling of inorganic Hg and MeHg demonstrates that local processes are important in driving MeHg concentrations and that inorganic Hg sources may have a relatively limited influence on the more toxic and bioaccumulative form of Hg when biogeochemical conditions do not support efficient MeHg production.

The primary drivers of Hg-methylating microbial activity include: temperature, a suitable redox environment, pH, and the presence of appropriate electron donors (e.g. organic carbon), and acceptors (e.g. sulfate, iron, etc.), whereas the primary drivers of inorganic Hg availability include dissolved organic matter quantity and character, sulfur chemistry and speciation, and the chemical form of inorganic Hg (Benoit et al., 1999; Boening, 2000; Gilmour et al., 2013; Hsu-Kim et al., 2013; Ullrich et al., 2001). Western North America contains a unique and diverse assemblage of Hg-sensitive habitats, each with a distinctive combination of those drivers that facilitate MeHg production. These habitats include highly productive forested watersheds of Alaska and the Pacific Northwest (Eckley et al., 2015; Gray et al., 2012; Schuster et al., 2011; Tsui et al., 2010), subalpine lakes and wetlands (Eagles-Smith et al., 2014; Eagles-Smith et al., 2016b; Landers et al., 2008), seasonal wetlands of the Great Basin and arid southwest (Bonzongo et al., 1996; Chen et al., 1996; Johnson et al., 2015), wetlands of the Prairie Pothole region of the northern Great Plains (Hoggarth et al., 2015), estuarine wetlands along the Pacific coast (Ackerman et al., 2007; Ackerman et al., 2008a; Eagles-Smith and Ackerman, 2014), agricultural wetlands such as rice fields in California's Central Valley (Ackerman and Eagles-Smith, 2010; Marvin-DiPasquale et al., 2014; Windham-Myers et al., 2014), large river systems that traverse the broad expanses of the region (Peterson et al., 2007; Walters et al., 2010), and one of the world's largest networks of reservoirs (Willacker et al., 2016). Aquatic habitats in the region are also highly managed with regard to the timing and amount of water they receive or release. This management can have profound implications for MeHg production (Eckley et al., 2015; Gray et al., 2014). Further, the heterogeneous distribution of historic Hg, Au and Ag mines can influence the availability of Hg to methylating microbial communities in mercury-sensitive environments well downstream of the point sources (Alpers et al., 2005; Gustin et al., 1994).

2.4. Importance of biological resources of western North America

The complex geography and varied ecological characteristics of the West also support diverse assemblages of fish and wildlife communities, many of which are threatened by habitat loss and other stressors including MeHg exposure. Western fish communities are characterized by remarkably high endemism (Warren and Burr, 1994) and when coupled with limited water availability or redirection of water

resources, they are disproportionately imperiled compared with the Eastern portion of the continent (Warren and Burr, 1994; Wilcove et al., 1998). Many avian species are similarly jeopardized in the West because of their reliance on diminishing aquatic (primarily wetland) resources. As a result, a third of native fish species in the Western US are imperiled compared to only 7% in Eastern states (Warren and Burr, 1994), and 27 of the 46 (59%) threatened and endangered bird species in the continental U.S. and Alaska breed in the western United States (USFWS, 2015). Moreover, greater than 50% of these bird species of greatest conservation concern forage in aquatic habitats, where there is a higher likelihood of exposure to MeHg (Ackerman et al., 2016b). Despite these threats, the West hosts fisheries and wildlife communities that are of profound ecological and economic importance (Gresh et al., 2000; Schindler et al., 2003). Commercial and sport fishing are an integral part of the economy and culture for western North American states, provinces, and territories. There are an estimated 8.5 million anglers in the western U.S. and Canada that spend nearly $12 billion per year on sport fishing (Southwick Associates, 2012). Coastal commercial catches were a reported 3.0 metric tons valued at $2.6 billion from U.S. Pacific states in 2011 (NMFS, 2012), and coastal fisheries added $6 billion into the Canadian economy in 2015 (http://www.dfo-mpo.gc.ca/international/issue-enjeu-eng.htm). Fishing plays an important cultural and subsistence role for many subpopulations, particularly Native American and aboriginal groups in the U.S. and Canada (e.g., Tonn et al., 2015). Although commercial wildlife harvest is much less widespread than fish, waterfowl serve many consumptive purposes and can accumulate high Hg concentrations where food web MeHg concentrations are elevated, resulting in potential threats to subsistence and recreational hunters (Cristol et al., 2012; Vest et al., 2009). Despite the limited commercial harvest, the recreational and associated economic value of wildlife is high. In the U.S. alone, waterfowl hunting and bird watching annually generate approximately $110 billion in economic growth (Carver, 2013), and birds also provide substantial and diverse ecosystem services (Whelan et al., 2008). Thus, MeHg exposure and bioaccumulation in western ecosystems can have profound economic, environmental, and human health implications.

2.5. Mercury bioaccumulation in western North America

Fish are useful bioindicators of MeHg contamination because they are an important vector of MeHg to both wildlife (Ackerman et al., 2015) and humans (Jewett and Duffy, 2007; Shilling et al., 2010). Mercury concentrations in fish are regularly monitored by federal, state/provincial, and local agencies, and this widespread use of fish as bioindicators allows for broad scale spatiotemporal assessments of Hg contamination and risk across the landscape. Mercury contamination in fishes of western North America is widespread, but exhibits substantial heterogeneity across the landscape among fish taxa and locations (Eagles-Smith et al., 2016a). Despite this variability, distinctive patterns still emerge over the landscape. Perhaps the most striking is the lack of concordance between inorganic Hg sources and storage in comparison to Hg bioaccumulation in fishes. Whereas landscape deposition and terrestrial pools of inorganic Hg are greatest in productive forested areas that receive substantial precipitation (Eckley et al., 2016; Obrist et al., 2016; Weiss-Penzias et al., 2016), hotspots of Hg bioaccumulation were most pronounced in the more arid regions of the West, such as the Great Basin, arid southwest, and Mediterranean California (Eagles-Smith et al., 2016a).

Elucidating the mechanisms driving these contrasts in Hg patterns between matrices requires further study, but could be associated with some combination of higher lability of Hg in arid regions that contain less long-term carbon storage than forested areas, greater overland flow and sediment delivery in more arid regions associated with limited vegetation, or more efficient MeHg production in arid regions due to differences in wetting and drying cycles, higher overall temperatures, aquatic biogeochemistry, and proportional inputs of wet versus dry Hg deposition. Indeed, a very small proportion of Hg deposited on forested landscapes of the West is delivered to lakes and deposited in sediments (Drevnick et al., 2016), suggesting that watershed Hg delivery rates could play a role in these contrasts. However, the delivery of inorganic Hg to aquatic systems (as inferred from bed sediment THg concentrations) is not correlated with fish THg concentrations across western North America, whereas sediment MeHg concentrations showed a weak positive correlation with fish THg levels (Eagles-Smith et al., 2016a). This illustrates that net production of MeHg is likely more important than inorganic Hg sources for determining fish Hg bioaccumulation at a landscape scale. Yet, sediment MeHg concentrations still only accounted for a small proportion of variation in fish THg concentrations, indicating that point-in-time estimates of sediment MeHg concentrations alone are likely not reflective of the dynamic processes associated with MeHg cycling and bioaccumulation through food webs.

2.6. Resource management and mercury in western North America

More informative metrics for evaluating Hg exposure in the West might be those associated with aquatic habitat characteristics and management of water resources. The influence of wetlands on promoting MeHg production and bioaccumulation transcends geography and has been documented for decades (St. Louis et al., 1994). Mercury concentrations in wetlands across western North America are highly variable, but contain some of highest concentrations in fish reported across the region (Eagles-Smith et al., 2016a). Wetlands in the West have diminished extensively over the past century (Dahl, 2011), and those that remain are generally subject to highly managed hydrologic control (US EPA, 2016). The limited abundance of wetland habitats in the West relative to their past distribution enhances their conservation importance, but also potentially makes them areas of enhanced Hg risk for fish and wildlife communities that rely on them. Thus, these are areas where efforts to develop management techniques that minimize MeHg exposure while still providing suitable habitat for target species may be most beneficial. Rivers and lakes (including reservoirs) are more widespread habitats across the West, and fish THg concentrations are generally higher in riverine environments of the region than lakes (Eagles-Smith et al., 2016a), even though sediment THg concentrations illustrate the opposite pattern (Fleck et al., 2016). Hydrology, its management, and the associated influence on aquatic ecosystem biogeochemistry likely play large roles in dictating these patterns. The strongly seasonal flow regimes of Western rivers results in pulsed hydrology, intermittent inundation, and short-term extreme ranges in primary productivity, all of which are directly linked to MeHg production. As a result, MeHg production potential in representative riverine environments of the West increases with both the frequency of floodplain inundation over decadal time scales, as well as the duration of floodplain inundation events (Singer et al., 2016). This has important ecological implications because floodplain habitats are zones of high biological productivity and are critical foraging habitats for many fish and wildlife species (Sheldon et al., 2002; Ward et al., 1999). Therefore, floodplains and off-channel habitats may have a disproportionate influence on Hg exposure patterns in lotic food webs in the West.

Arguably, no aquatic habitat is more representative of western North America than are reservoirs. The creation of reservoirs facilitated the widespread modern settlement of the West and created a landscape with vast networks of water storage and conveyance. The creation of reservoirs has long been associated with pulsed increases in MeHg production and bioaccumulation during the decade following inundation (Bodaly et al., 2007; Hall et al., 2005), but the hydrology of reservoirs and the associated riverine systems remains highly managed throughout their operation. This management of flows and storage carries the potential to profoundly influence Hg cycling. Indeed, across the West, fish THg concentrations average between 1.5 and 2.6-fold higher in reservoirs than in natural lakes (Willacker et al., 2016). More importantly, management of reservoir hydrology can have profound influences on

fish THg concentrations because of cyclical soil drying and rewetting and the associated changes in redox conditions and organic matter decomposition. For example, fish THg concentrations in reservoirs that have minimum water levels occuring in May, June, or July are up to 11 times higher than in reservoirs that reach with their lowest water storage levels in August through April (Willacker et al., 2016). Additionally, the magnitude of between-year differences in maximum reservoir storage is directly correlated with fish THg concentrations, whereas the magnitude of within-year differences from the lowest to highest water levels does not influence fish THg concentrations (Willacker et al., 2016). This is an important aspect of mercury bioaccumulation in the West because by their nature, reservoirs avail resource managers with potential opportunities for control measures. Whether those measures would be mutually consistent with designated reservoir operations would need to be ascertained based upon site-specific conditions and additional research, as well as a determination by appropriate regulatory bodies on the relative importance of the different outcomes. Regardless, the existence of these possibilities suggests that effective approaches may be feasible for mitigating Hg risk in reservoirs through reducing MeHg production as opposed to simply source control of inorganic Hg, which is likely ineffective or unattainable in many circumstances.

Although the results of this synthesis highlight the disconnect between inorganic Hg sources and the environmental health risks associated with MeHg, the importance of Hg, Au, and Ag mining across western North America cannot be overlooked. As with reservoirs, the legacy of mining is a defining feature of the region, and one that has resulted in both localized and more widespread distribution of inorganic Hg. Certainly the Hg mobilized from mining activity can become methylated and then bioaccumulate through down-gradient food webs, but it is less clear whether the presence of mining-derived Hg near a water body or within a watershed necessarily leads to elevated MeHg bioaccumulation. California's Sierra Nevada range provides a representative landscape for evaluating these questions because streams in the region drain a landscape that was subjected to historic placer and lode Au mining activity (Clark, 1970). Fish THg concentrations in Sierra Nevada streams are influenced by both geospatial variables describing the landscape, as well as mine density, urban development, and sediment MeHg concentrations (Alpers et al., 2016). The mining impact on fish THg concentrations at the local watershed scale is clearly apparent, but only in combination with landscape attributes that influence MeHg production. Importantly, although fish THg concentrations are typically moderately elevated in mining-affected streams, unaffected streams contain fish ranging from very low to elevated THg concentrations for a given fish length (Alpers et al., 2016). These findings again point to the overriding importance of MeHg production in driving environmental health risks despite the presence or absence of defined inorganic Hg point sources. Further, evidence using Hg stable isotopes indicates that food web components in wetlands and streams that drain mining-influenced areas do not show clear source attribution to the mine-derived inorganic Hg in the sediments (Donovan et al., 2016). This points to a more complex conceptual model of MeHg bioaccumulation in mining-impacted waterbodies, whereby the MeHg burden of biota reflect a mixture of sources that may be related to the lability of the available inorganic Hg that enters the system, and not necessarily directly related to mine sources alone. The cumulative evidence across the landscape suggest that mining sources of inorganic Hg may be most important with respect to food web bioaccumulation when they are directly connected to habitats that tend to promote MeHg production (Ackerman et al., 2016b; Eagles-Smith et al., 2016a; Singer et al., 2016; Willacker et al., 2016), but similar exposure levels can be attained in sites without a direct nexus to Hg, Au, or Ag mining. Thus, although efforts to control and reduce inorganic Hg source loadings could likely achieve some reductions in biological MeHg exposure in the West, avenues for addressing MeHg production and bioaccumulation processes may be more effective.

2.7. Wildlife exposure to mercury in western North America

In addition to their utility for estimating MeHg availability across aquatic ecosystems, fish also can be informative for evaluating potential exposure to wildlife and estimating toxicological risk to humans, wildlife, and fish themselves. However, doing so requires alternative analytical and statistical approaches (Ackerman et al., 2015; Jackson et al., 2016; Lepak et al., 2016). Additionally, wildlife-specific data are ultimately the most informative and accurate means for determining actual exposure to wildlife species across the landscape (Ackerman et al., 2016b). As with fish, avian MeHg exposure is widespread across western North America (Ackerman et al., 2016b; Jackson et al., 2016). Importantly, independent datasets indicate that there is substantial concurrence in the spatial patterns of avian MeHg exposure relative to those in fish, suggesting high confidence of risk estimates for some areas. In particular, the San Francisco Bay area and Central Valley of California, the Great Basin (including portions of Nevada, Idaho, and Oregon), and areas of Arizona appear to be consistent biological hotspots for MeHg exposure (Ackerman et al., 2016b; Eagles-Smith et al., 2016a). Avian MeHg exposure was also elevated in other areas that either did not contain particularly elevated fish Hg concentrations, or where fish data are limiting. Some of those locations include the Aleutian Islands, the North Slope of Alaska, and east-central Alaska, and the Puget Sound region of Washington. Large-scale ecological attributes are important factors influencing bird Hg concentrations, and Hg concentrations differ among bird species, foraging guilds, habitat types, locations, and ecoregions (Ackerman et al., 2016b). In particular, foraging guild played a predominant role in influencing bird Hg concentrations, with piscivores and carnivores exhibiting the highest Hg concentrations and herbivores and granivores exhibiting the lowest Hg concentrations (Ackerman et al., 2016b). Habitat type also is among the most important ecological factors and bird Hg concentrations in the West are highest in ocean and salt marsh habitats and lowest in terrestrial habitats (Ackerman et al., 2016b).

2.8. Toxicological risk of mercury in western North America

Ultimately, elevated Hg concentrations confer the potential for toxicological impairment and risk to fish, as well as to the wildlife and humans that consume them. Across the region, 34%, 30%, and 4% of fish exceed toxicological benchmarks associated with biogeochemical impairment within fish (0.2 μg/g ww in whole body), the US EPA fish tissue residue criterion (0.3 μg/g ww in muscle), and the US Food and Drug Administration action level (1.0 μg/g ww in muscle), respectively (Eagles-Smith et al., 2016a). However, these exceedance rate estimates are aggregated across all size classes and fish species, and do not account for unbalanced sampling of different water bodies, therefore they are likely biased estimates of toxicological risks in the region. However, more robust modeled assessments indicate that elevated exceedance probabilities (the probability that a fish of a given size sampled from a given watershed would exceed a benchmark of interest) for certain benchmarks and size classes are widespread (Lepak et al., 2016). In fact, for freshwater fish greater than 20 cm in length, the median probabilities that THg concentrations exceed levels associated with potential fish health effects (biogeochemical, behavioral, and reproductive impairment) across watersheds analyzed in the West ranges between 0.08 and 0.73 depending upon the fish size considered (Lepak et al., 2016). The spatial heterogeneity of Hg bioaccumulation in the region and differences in sampling design and resulting species composition results in higher probabilities in some watersheds, and lower in others. Additionally, when spatially aggregated into 1-degree grid cells across the West, the mean THg concentrations in fish from at least 25% of all grid cells equate with moderate toxicological risk to piscivorous birds (equating to 1–2 μg/g ww in blood; Jackson et al., 2016). These findings are in rough concordance with exposure estimates of the western North American avian community, for which 28% of birds sampled exceed

moderate risk benchmarks and 8% of individuals have blood-equivalent Hg concentrations that put them at high risk for potentially impaired reproduction (Ackerman et al., 2016b).

Mercury contamination in fish and some wildlife has a strong nexus to human health through direct consumption of subsistence, recreational, and commercial harvests. The public health issues associated with this are complex because fish consumption confers many health benefits. This creates a challenge to those agencies responsible for issuing consumption advisories because of the balance associated with protecting the public from Hg exposure risks while also recognizing the nutritional value of fish consumption. For freshwater fish greater than ~30 cm in length, the median probabilities that fish would exceed advisory guidelines recommending consumption of no more than 1 meal per week (>0.11 μg/g wet weight) and 1 meal per month (>0.22 μg/g wet weight) across the watersheds analyzed in the West was 0.76, and 0.46, respectively (Lepak et al., 2016). Exceedance probabilities for recommendations of no fish consumption range between 0.02 (>15 cm) and 0.06 (>~46 cm), and were limited to relatively few watersheds (Lepak et al., 2016). Importantly, different fish species accumulate Hg at various rates that adds uncertainty to these estimates of exceedance probabilities, but also provides anglers and subsistence fishers with potential choices in reducing Hg exposure, and public health officials with species-specific flexibility in developing consumption recommendations and guidelines. Human exposure studies suggest that inland populations of the West experience moderate Hg exposure (Mahaffey et al., 2009), whereas coastal populations in the West are among the highest exposure groups in the United States (Mahaffey et al., 2009). This is has been attributed in part to geographic patterns in types of fish consumed between populations. Marine fishes are also important vectors for human exposure, including both commercial and recreational harvests. Spatial variation in coastal marine fish THg concentrations in the Pacific Ocean, ranging from southern California to Alaska, also indicates that a high proportion of fish exceed health risk benchmarks (Davis et al., 2016). Fifty-four percent of the sites sampled had at least one species with an average THg concentration exceeding the 0.30 μg/g ww EPA criterion for human consumption, and 15% of locations had at least one species with an average concentration exceeding the 1.0 μg/g ww U.S. FDA action level (Davis et al., 2016). The highest fish Hg concentrations were found along the coast of Northern California and the Puget Sound, and there were no detectible temporal trends over the nearly 30 year time period covered by the dataset. These findings, in combination with limited wildlife data, indicate that Hg may be broadly elevated in coastal waters throughout the eastern Pacific. Extensive studies on Hg accumulation and effects in birds of the San Francisco Bay Estuary indicate that a large proportion of Forster's terns are at high risk of reproductive impairment and several lines of evidence indicate that Hg might be impairing health and reproduction in these and other species (Ackerman et al., 2008b; Ackerman et al., 2014; Eagles-Smith et al., 2009a; Eagles-Smith et al., 2009b). Moreover 99% of northern elephant seals along the California coast exceeded the commonly used clinical neurotoxicity threshold of 0.21 μg/g whole blood for marine mammals (Peterson et al., 2015). Cumulatively, these findings indicate that western North American coastal environments may be important areas for biological Hg exposure, but robust information mirroring freshwater environments in the West is lacking.

Regardless of the environmental setting, the cycling and bioaccumulation of Hg is extremely complex and strongly tied to the surrounding hydrology, geology, biogeochemistry, and ecology. Further complicating these processes is the potential interaction with selenium (Se). The amount and distribution of Se in western North America generally exceeds many areas in the East, thus is particularly germane for this synthesis. Selenium is an important component of Hg risk because of its potential to mitigate Hg toxicity. Selenium is abundant in marine waters and there are multiple geologic sources of selenium (i.e., black shales, coals, petroleum source rocks, and phosphorites) throughout the West that are mobilized by human activities including agricultural irrigation of selenium enriched soils (e.g., San Joaquin Valley, Colorado River watershed), oil refining (e.g., San Francisco Estuary), and phosphate, copper and uranium mining (e.g., Blackfoot River watershed) (Presser et al., 1994). These sources result in large gradients of selenium across diverse habitats (i.e., estuaries, rivers, reservoirs) that may influence the relative toxicity of MeHg to fish and their consumers (Karimi et al., 2013; Peterson et al., 2009). Although the potential for Se to mitigate Hg toxicity has been recognized for some time (Ganther et al., 1972; Parizek and Ostadalova, 1967; Burk et al., 1974; Naganuma and Imura, 1980; Ralston et al., 2008), this antagonism between Se and Hg has recently received consideration by scientists and regulators as a potential means to better evaluate the toxicological risks of tissue Hg levels in nature (Peterson et al., 2009). In an assessment of Hg and Se concentrations and molar ratios in 468 fish representing a variety species throughout the Western U.S., Petersen et al. (2009) reported that ~98% of the fish examined had molar quantities of Se that exceeded those of Hg. Substantial uncertainty and intense scientific debate still exist regarding the toxicological meaning of Se molar quantities that exceed those of Hg, but some argue that applying molar ratios to risk evaluations may refine them. Still, many questions remain unanswered concerning the precise molar ratios in different tissues, species, and consumers (wildlife or humans) that provide protection to MeHg exposure. Further, it is not well known how these molar ratios vary depending on exposure gradients for MeHg and Se, for different tissue types or species, and how they influence the trophic transfer of MeHg in either wildlife or human consumers. Thus, it is still unclear whether Se could be acting as a protective mechanism for MeHg contamination in western North America.

3. Conclusions

Mercury is broadly distributed throughout western North America, and a diverse suite of sources, biogeochemistry, and bioaccumulation processes contributes to the extreme heterogeneity in Hg contamination across the landscape. Anthropogenic Hg emissions to the atmosphere are substantially lower in the West than in the East, primarily because of the lower density of coal-burning power plants in the western U.S. However, the western region contains a greater area of geogenically enriched soils and has more historic Au, Ag, and Hg mines, though impacts on Hg contamination from past mining activities appear to be relatively isolated to the watersheds in which they are located. The largest pools of inorganic Hg include vegetation and soils, thus forested areas of the West generally act as net sinks of Hg deposition, whereas less vegetated areas can be net emitters of Hg to the atmosphere. Additionally, land disturbance, including wildfire, appears to be a key contributor to inorganic Hg mobilization and transport through the region.

Importantly, inorganic Hg sources and sediment inorganic Hg concentrations are poorly correlated with MeHg concentrations in aquatic sediments. Instead, local biogeochemical conditions appear to be the predominant drivers of MeHg contamination, which is the form of Hg that biomagnifies through food webs and is most toxic to fish, wildlife, and humans. Ecosystem management can have a strong influence on biogeochemical drivers associated with MeHg cycling, and as such is particularly important in the West where a large proportion of land and water resources are publicly managed. For example, reservoirs are dominant features on the western landscape that control large expanses of ecosystem hydrology and food web structure. Management of reservoir water-level fluctuations has a pronounced effect on fish MeHg bioaccumulation that may influence human and wildlife exposure risk. Additionally, species, animal size, foraging guild, and habitat type are strong drivers of MeHg exposure in fish, birds, and humans. This complexity contributes to the broad heterogeneity in MeHg concentrations of fish and birds, as well as the decoupling of inorganic Hg loading and sources from MeHg bioaccumulation. Effective management of the

environmental health risks associated MeHg in the West will require looking beyond simply controlling inorganic Hg sources control, and will necessitate development of management tools associated with controlling the net production of MeHg, and ultimately its entry into, and bioaccumulation through, food webs.

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