



Mercury cycling in the U.S. Rocky Mountains: a review of past research and future priorities

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Abstract Mercury cycles at levels three- to five-fold higher today than the pre-Industrial era, resulting in global contamination of ecosystems. In the western United States (U.S.), mercury mobilization has led to widespread production of methylmercury (MeHg), a potent, bioaccumulating neurotoxin, which has resulted in fish consumption advisories across all states. Mountain regions are particularly sensitive to continued mercury contamination as they receive higher rates of atmospheric deposition, compared to lower elevations, and have aquatic ecosystems on the landscape conducive to MeHg production. In this paper, we focus on the U.S. Rocky Mountain region and synthesize: (1) current knowledge regarding the mercury cycle; (2) impacts of climate change on the mercury cycle connected to hydrology and wildfire; and (3) future research priorities for informing mercury research and regulation. Studies on the

interactions between mercury contamination and climate change in mountain ecosystems is still nascent. We use the findings from this synthesis to summarize the following research needs: (1) quantify sources of mercury in wet and dry deposition, as these pathways dictate mercury exposure and toxicity, and are shifting with climate change; (2) investigate MeHg in mountain aquatic ecosystems, which are important pathways of human mercury exposure and provide food resources and habitat to local wildlife; and (3) examine the disproportionate impact of mercury contamination on indigenous communities through community-led research. Although we focus on the Rocky Mountains for this review, the findings are applicable to semi-arid mountain ecosystems globally and must be prioritized to promote the health of ecosystems and people everywhere.

Keywords Methylmercury · Bioaccumulation · Climate change · Hydrology · Wildfire · Semi-arid mountains

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Introduction

Mercury is a geologically sourced, bioaccumulating trace metal, and its release, global transport, and exposure have been greatly accelerated by human activities (UNEP 2018). Catastrophic poisoning events, and the global extent of contamination, have provided international motivation to curb the

extraction, use, and distribution of mercury. These efforts have been primarily led by the United Nations' Minamata Convention on Mercury (UN Environment 2019). Now signed and ratified by 147 parties, this treaty has made historic progress in expanding our understand of the mercury cycle, and mitigating exposure. However, mercury contamination remains a serious global threat to human and ecosystem health due to its continued use, release, and persistent nature (Chen et al. 2018; UNEP 2018). When emitted into the atmosphere, mercury can be transported long distances and deposited onto remote landscapes (Selin 2009). Following initial deposition to the Earth surface, mercury may be subsequently reemitted to the atmosphere by evasion, or mobilized via water to aquatic ecosystems. This cycle can repeat several times before mercury is permanently sequestered in sediments (Amos et al. 2014). In aquatic ecosystems, mercury can be converted to methylmercury (MeHg), a bioaccumulating neurotoxin, that is responsible for widespread contamination in wildlife and fish consumption advisories in all fifty states of the United States (U.S.). The mercury cycle is highly manipulated by human activities with 3–5 times more mercury cycling today than during the pre-Industrial era; coal combustion, artisanal gold mining, and other industrial practices are the major activities that have led to the rapid mobilization and bioavailability of this element (Li et al. 2020; UNEP 2018).

The increased cycling of mercury in the biosphere and associated harmful human health impacts have provided motivation for research and regulation over the past several decades. The long atmospheric residence time of mercury allows for long range transport to remote regions such as high latitude and altitude ecosystems (Selin 2009). With concentrations of mercury above background levels predicted to persist for centuries, due to reemission from legacy pools (Amos et al. 2013), it is important to understand the response of remote regions to continued mercury contamination, especially in the context of ongoing global change. Over the past 10 years, mercury research in remote ecosystems has largely focused on Arctic, as well as mountain systems in the Himalaya and eastern U.S. (Blackwell and Driscoll 2015; Chai et al. 2022; Demers et al. 2007; Gerson et al. 2017; Tripathee et al. 2019; Zhang et al. 2019a, b). In mountain ecosystems of the western U.S., research on mercury cycling has historically been limited and

relatively narrow in scope. This paucity of studies marks an important knowledge gap. Mountain ecosystems make up a large portion of the western U.S. and are experiencing increasing mercury deposition, as well as climate change, which likely alter baseline mercury cycling (Eagles-Smith et al. 2016a, b; Mast et al. 2005; Packer et al. 2020). As such, we seek to assess the state of research regarding mercury cycling in mountain ecosystems of the western U.S. to identify unknowns and priorities for future research.

In this Synthesis and Emerging Ideas paper, we focus on the U.S. Rocky Mountains region (henceforward, “Rocky Mountains”). The consequences of elevated mercury deposition to the Rocky Mountains are still poorly understood despite this region covering over one-third of the conterminous western U.S. land area. In general, processes in semi-arid mountain ecosystems, such as the Rocky Mountains, remain inadequately constrained with regards to mercury cycling. Specifically, studies are lacking on mercury uptake, release, and evasion in shrub/grassland and forest environments; storage in high elevation soils; and transformations in aquatic ecosystems such as mountain wetlands, reservoirs, and lakes. We synthesize the state of knowledge regarding mercury cycling in the Rocky Mountains with relevant comparisons to other mountain regions, evaluate how mercury cycling processes may evolve with continued climate change, and highlight important areas for future research.

Mercury cycling in the U.S. Rocky Mountains

Background

The Rocky Mountains of the U.S. are $> 800,000 \text{ km}^2$ in total area, spanning $\sim 3,000 \text{ km}$ from New Mexico to the Canadian Border; they cross Colorado, Utah, Wyoming, Idaho, and Montana (Fig. 1). The region is characterized by extreme gradients in climate, elevation, and land cover, which drive patterns of mercury cycling (Eagles-Smith et al. 2016a). The Rocky Mountains range from ~ 1500 – 4300 m in elevation and are comprised of desert, grassland, shrubland, and forested land covers. Approximately 70% of the annual water supply to the region is tied to mountain snowpack, with over 200 reservoirs greater than 0.1 km^3 storage located within the Rocky Mountains

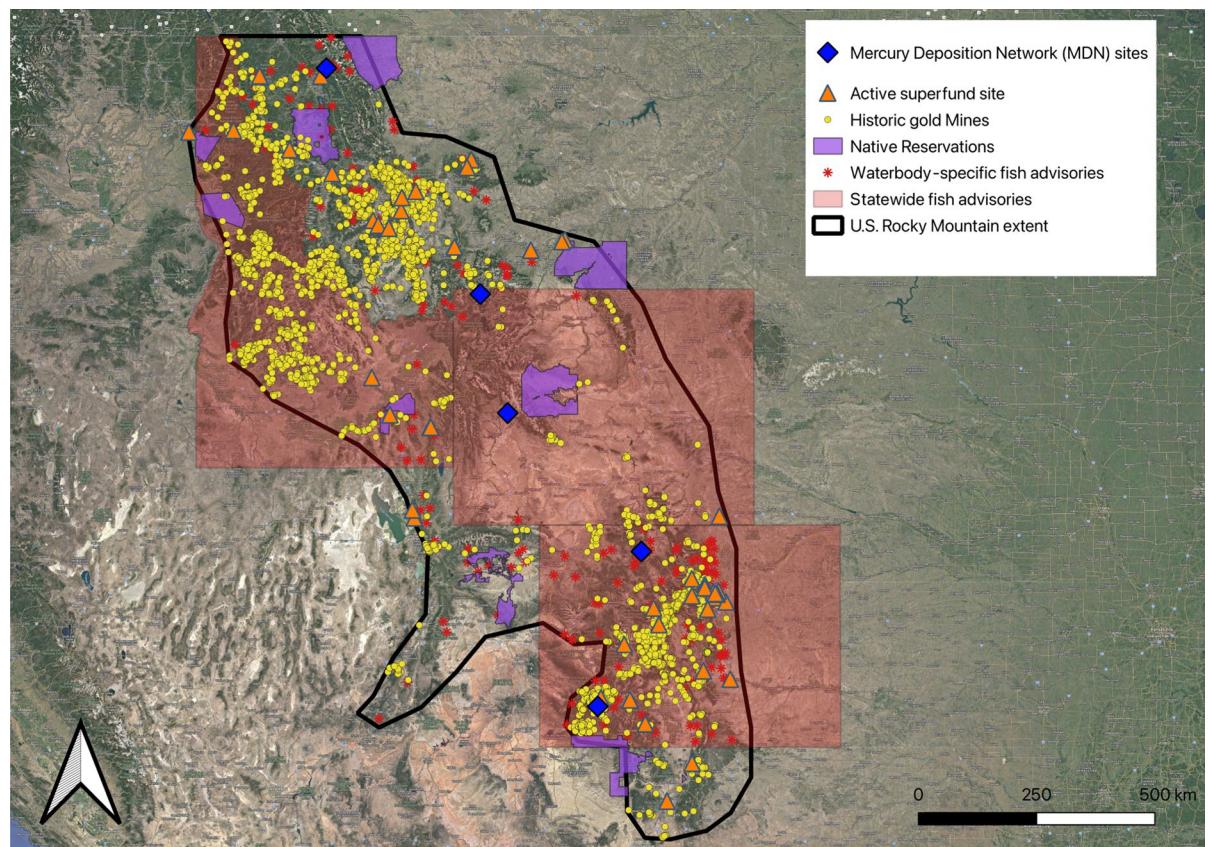


Fig. 1 Map of study area showing MDN sites (blue diamond), active superfund sites (orange triangle), historic gold mines locations (yellow circle), Native American Reservations (purple shading), waterbody-specific fish advisories (red star),

and statewide fish advisories (red shading) within the Rocky Mountains. Statewide fish advisories in Idaho, Wyoming, and Colorado are for all waterbodies for specific species of fish (see manuscript text for more information)

alone (Lehner et al. 2011). Ongoing climate change has the potential to shift mercury cycling in ecosystems and landscapes of the Rocky Mountains with consequences that are both local and regional in scale. In the following sections, we summarize the research investigating sources, storage, transport, and transformations of mercury within the Rocky Mountains and how it is impacted by climate change (Table 1). We focus on two specific drivers associated with a changing climate that are having a profound effect on ecosystems of the mountainous western U.S.: shifts in hydrologic regimes and wildfire activity.

Sources and atmospheric deposition

Mercury is transferred from the atmosphere to the Earth surface in oxidized (Hg(II)) and elemental

(Hg(0)) forms, and deposited through both wet and dry atmospheric deposition (Selin 2009). Globally, dry deposition dominates the atmospheric flux, accounting for 60–90% of terrestrial atmospheric mercury deposition (Zhou et al. 2021). Wet atmospheric mercury deposition occurs during periods of precipitation and fog and is typically comprised of soluble Hg(II) dissolved in water or adsorbed on the surface of water particles. Dry mercury deposition occurs primarily from the uptake of atmospheric Hg(0) by plants and deposition of particle-bound Hg(II) to foliar and land surfaces (Li et al. 2020; Selin 2009). Studies from across the western U.S., including sites in the Rocky Mountains, show that most of atmospheric mercury deposition in this region is derived from the well-mixed global pool of Hg(0), as opposed to local sources (Olson et al. 2020; Selin

Table 1 Total mercury and methylmercury concentrations for previously studied ecosystem compartments in the Rocky Mountains

| Compartment | Total mercury | Methylmercury | Description | Region | References |
|-------------|---|--|----------------------------------|---|--|
| Atmosphere | $16.1 \pm 45 \text{ pg m}^{-3}$ (PBM), $20.4 \pm 28 \text{ pg m}^{-3}$ (GOM), $1.9 \pm 0.9 \text{ ng m}^{-3}$ (GEM) | | Average values from 2008 to 2018 | Salt Lake City, Utah (AmNet UT97) | Zhang et al. (2016), https://nadp.slh.wisc.edu/sites/amnet-UT97/ |
| Soil | Conifer forests: $58.4\text{--}208 \text{ ng g}^{-1}$ ($\mu=108 \text{ ng g}^{-1}$); deciduous forests: $25.2\text{--}37.5 \text{ ng g}^{-1}$ ($\mu=31.7 \text{ ng g}^{-1}$) | | O horizon | Wyoming Rocky Mountains | Biswas et al. (2007) |
| | $<10\text{--}1320 \text{ ng g}^{-1}$ ($\mu=30 \text{ ng g}^{-1}$) | $0.17\text{--}0.43 \text{ ng g}^{-1}$ | A horizon | U.S. Rocky Mountains | Olson et al. (2022)* |
| | $<10\text{--}520 \text{ ng g}^{-1}$ ($\mu=25 \text{ ng g}^{-1}$) | | C horizon | U.S. Rocky Mountains | Olson et al. (2022)* |
| | $40.4\text{--}118.1 \text{ ng g}^{-1}$ ($\mu=81.7 \text{ ng g}^{-1}$) | $0.32\text{--}1.50 \text{ ng g}^{-1}$ ($\mu=0.89 \text{ ng g}^{-1}$) | Wetland soils | Wetlands near Great Salt Lake, UT | Fleck et al. (2016)* |
| | $5.7\text{--}24,732.4 \text{ ng g}^{-1}$ ($\mu=776.9 \text{ ng g}^{-1}$) | $0.01\text{--}77.00 \text{ ng g}^{-1}$ ($\mu=2.97 \text{ ng g}^{-1}$) | Lake sediments | U.S. Rocky Mountains | Fleck et al. (2016)* |
| | $1.59\text{--}466 \text{ ng g}^{-1}$ ($\mu=111.1 \text{ ng g}^{-1}$) | $0.1\text{--}1.9 \text{ ng g}^{-1}$ ($\mu=0.6 \text{ ng g}^{-1}$) | Stream sediment | U.S. Rocky Mountains | Fleck et al. (2016)* |
| | $29.0\text{--}45.8 \text{ ng g}^{-1}$ ($\mu=39.2 \text{ ng g}^{-1}$) | $0.09\text{--}0.12 \text{ ng g}^{-1}$ ($\mu=0.104 \text{ ng g}^{-1}$) | Reservoir sediment (0–9 cm) | CO (Narraguinep Reservoir) | Gray et al. (2014) |
| Water | $0.27\text{--}14.09 \text{ ng L}^{-1}$ | $0.01\text{--}0.73 \text{ ng L}^{-1}$ | Lake surface water | 90 high-altitude lakes in the western U.S | Krabbenhoft et al. (2002) |
| | $0.5\text{--}13.5 \text{ ng g}^{-1}$ | $0.04\text{--}0.048 \text{ ng L}^{-1}$ | Alpine stream | U.S. Rocky Mountains | Mast et al. (2005), Packer et al. (2020), Shanley et al. (2008) |
| | filtered = $0.17\text{--}0.43 \text{ ng L}^{-1}$, particulate = $0.22\text{--}0.83 \text{ ng L}^{-1}$ | filtered = $0.005\text{--}0.114 \text{ ng L}^{-1}$, particulate = $0.003\text{--}0.102 \text{ ng L}^{-1}$ | Reservoir surface water | Idaho | Baldwin et al. (2022) |
| Fish | $30.3\text{--}3992 \text{ ng g}^{-1}$ wet weight ($\mu=670 \text{ ng g}^{-1}$) | | Salmonidae family in lake | U.S. Rocky Mountains | USGS, unpublished data |
| | | $36.6\text{--}488 \text{ ng g}^{-1}$ | Centrarchids in reservoir | Idaho | Baldwin et al. (2022) |
| Dragonflies | $5\text{--}1769 \text{ ng g}^{-1}$ ($\mu=219 \text{ ng g}^{-1}$) | | Aeshnidae family | U.S. Rocky Mountains | Eagles-Smith et al. (2020)* |

*Rocky Mountain sites extracted from larger dataset

and Jacob 2008). There are periods—typically in the spring—of greater sourcing directly from east-Asian industrial activities. Long-distance transport of mercury from Asia occurs across the Pacific in the free troposphere (Weiss-Penzias et al. 2006; Lin et al. 2012; Huang and Gustin 2015).

Regulation in the U.S. through the 2011 Mercury and Air Toxins Standards (MATS), in addition

to control technologies for other pollutants such as sulfur dioxide and nitrogen oxides, have resulted in a $> 75\%$ decrease in mercury emissions from U.S. coal-fired utilities (Zhang et al. 2016). As a result of these emission declines, wet atmospheric mercury deposition has been decreasing over the past several decades in the eastern U.S., which is downgradient

of major mercury emission sources in the Midwestern U.S. (Fig. 2; Olson et al. 2020).

This decreasing trend of wet atmospheric mercury deposition, however, is not mirrored in the western U.S. where National Atmospheric Deposition Program (NADP) sites show mostly non-significant, increasing mercury concentrations in wet deposition since 2008 (Weiss-Penzias et al. 2016). Specifically in the Rocky Mountains, NADP sites show increasing mercury concentrations since 2000 in wet deposition with the highest concentrations occurring between 2010 and 2015 (Fig. 2). Additionally, wet deposition rates are significantly higher at sites

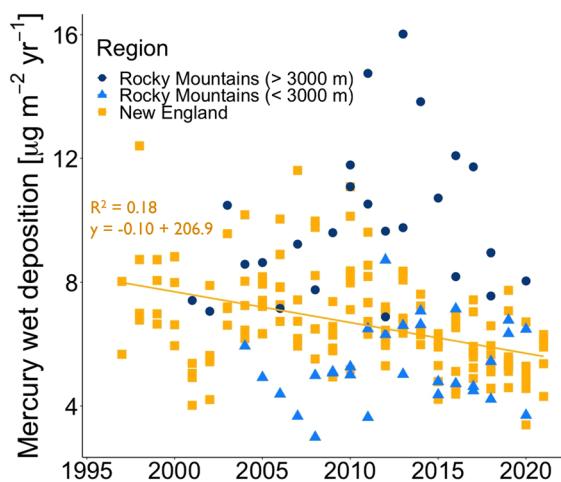


Fig. 2 National Atmospheric Deposition Program (NADP) mercury wet deposition rates for eight sites in New England (yellow squares), two sites below 3000 m elevation in the Rocky Mountains (light blue triangles), and two sites above 3000 m elevation in the Rocky Mountains (dark blue circles). The lower elevation Rocky Mountain data, and log-transformed higher elevation Rocky Mountain and New England data, are normally distributed (Shapiro–Wilk test, $p>0.05$). The New England sites show significant decreasing trends in atmospheric mercury deposition (Pearson correlation coefficient, $p<0.01$) likely due to decreases in regional mercury emissions. Rocky Mountain sites, alternatively, show non-significant increasing trends at both the high elevation sites (Pearson correlation coefficient, $p=0.19$) and lower elevation sites (Pearson correlation coefficient, $p=0.33$) likely due to the augmenting global pool of mercury. Additionally, mercury wet deposition rates increase with elevation with significantly higher deposition rates at Rocky Mountain sites over 3000 m in elevation compared to sites below 3000 m (t -Test, $p<0.01$). These higher rates of atmospheric deposition are likely due to the location of higher elevation sites within the free troposphere, which has higher atmospheric Hg concentrations compared to lower elevation air masses (Huang and Gustin 2015)

above 3000 m elevation compared to lower elevation sites ($9.90\pm2.44\text{ ng m}^{-2}\text{ yr}^{-1}$ versus $5.36\pm1.28\text{ ng m}^{-2}\text{ yr}^{-1}$, $p<0.01$; Fig. 2). These contrasting patterns are likely due to East Coast monitoring sites, such as in New England, falling within the planetary boundary layer (<2 km elevation), which is primarily influenced by local mercury sources. The Rocky Mountain sites, alternatively, fall within the free troposphere (>2 km elevation), which reflects global background mercury concentrations. As a result, higher elevation sites in the Rocky Mountains, that reflect the augmenting global pool of atmospheric mercury, show increasing patterns, whereas lower elevation sites in New England show decreasing trends due to reductions in regional mercury emissions (Lin et al. 2012; Weiss-Penzias et al. 2016; Olson et al. 2020).

Wet atmospheric mercury deposition rates range from 2.9 to $16.0\text{ }\mu\text{g m}^{-2}\text{ yr}^{-1}$ in the Rocky Mountains, as calculated from five active Mercury Deposition Network sites (MDN; Fig. 2). The highest wet mercury deposition occurs at the two Colorado sites which are both located over 3,000 m elevation (Fig. 2). These rates are comparable to other mountain regions globally with rates varying from 1.75 to $8.20\text{ }\mu\text{g m}^{-2}\text{ yr}^{-1}$ in the Tibetan Plateau (Chai et al. 2022; Gu et al. 2020; Huang et al. 2012) and from 4.2 to $13.0\text{ }\mu\text{g m}^{-2}\text{ yr}^{-1}$ in the Adirondacks and Green Mountains of the eastern U.S. (Gerson et al. 2017; Shanley et al. 2008).

Direct dry deposition (the flux of mercury in the absence of precipitation) is more difficult to measure and often estimated by using atmospheric mercury species concentrations combined with model estimated deposition velocities, eddy covariance techniques, or by sampling vegetation litterfall and throughfall (Wright et al. 2016). The Atmospheric Mercury Network (AMNet) includes measured concentrations of atmospheric mercury species with model calculations of dry mercury deposition. This network is sparse; all sites west of the Mississippi were discontinued by 2018, and only two of those sites were located within the Rocky Mountains in Utah. The estimated dry deposition at these two Utah AMNet sites ranged from 9.5 to $14.0\text{ }\mu\text{g m}^{-2}\text{ yr}^{-1}$. These rates are comparable to wet deposition rates, but do not include mercury fluxes via litterfall and throughfall, suggesting that dry deposition dominates in the region, consistent with global patterns

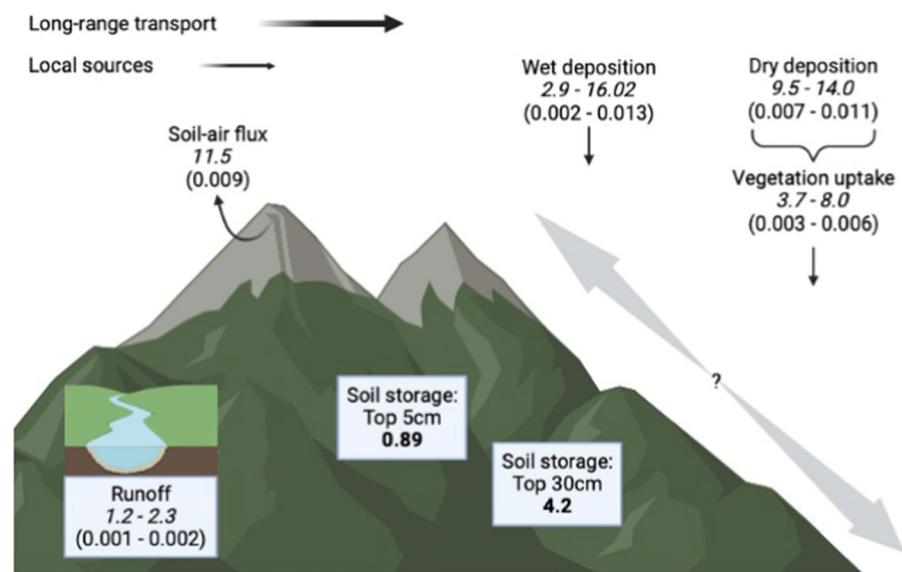
(Fig. 3; Zhang et al. 2016). Comparisons of direct dry deposition to other mountain regions are challenging due to the lack of direct and accurate measurements; however, one study found mercury fluxes up to $35.3 \mu\text{g m}^{-2} \text{ yr}^{-1}$ in the Tibetan Plateau (Chai et al. 2022; Sun et al. 2021) and fluxes ranging from 5.2 to $16.9 \mu\text{g m}^{-2} \text{ yr}^{-1}$ in eastern U.S. mountain ecosystems (Shanley et al. 2008; Wright et al. 2016).

Eckley and colleagues (2016) estimated vegetation uptake of mercury (a proxy for dry deposition) across the western U.S. They used previously published litterfall and throughfall data from other regions in the U.S., as well as studies from Europe and China, and found that uptake of mercury by vegetation varies by ecoregion. Their estimates ranged from $1.6 \pm 0.1 \mu\text{g m}^{-2} \text{ yr}^{-1}$ in desert ecosystems to $10.9 \pm 0.1 \mu\text{g m}^{-2} \text{ yr}^{-1}$ in marine West Coast forests (Eckley et al. 2016). As far as we are aware, there have been no studies investigating rates of vegetation-derived deposition specifically in the Rocky Mountains. However, modeled estimates from the Great Plains (comparable to Rocky Mountains foothills vegetation) average $3.7 \pm 0.1 \mu\text{g m}^{-2} \text{ yr}^{-1}$, estimates from northwestern U.S. forested mountains (comparable to montane and subalpine vegetation of the Rocky Mountains) average $8.8 \pm 0.1 \mu\text{g m}^{-2} \text{ yr}^{-1}$, and measurements from the Alaskan tundra (comparable to high alpine vegetation of the Rocky Mountains) average $8.0 \mu\text{g m}^{-2} \text{ yr}^{-1}$ (Fig. 3; Eckley et al. 2016; Olson et al. 2019). In the absence of local data, these

rates help to constrain atmospheric deposition rates in the Rocky Mountains. Extensive dry deposition measurements across the Rocky Mountains, however, are needed to better quantify mercury inputs to this region. Increasing monitoring efforts is especially urgent in the context of increasing wildfire intensity and frequency; wildfire mobilizes mercury from vegetation and surface soils for subsequent deposition back to the land surface (Kumar et al. 2018).

Studies from the Tibetan Plateau and eastern U.S. show that mercury cycling varies significantly along mountain elevation gradients due to shifts in atmospheric mercury deposition and vegetation cover (Blackwell and Driscoll 2015; Gerson et al. 2017; Li et al. 2022; Zhang et al. 2013a, b). Steep elevation gradients in the Rocky Mountains also likely play an important role in atmospheric mercury deposition but have not yet been investigated. Precipitation generally increases with elevation in the Rocky Mountains, from $\sim 190 \text{ mm yr}^{-1}$ at 1600 m to $\sim 1500 \text{ mm yr}^{-1}$ at 3500 m, suggesting that mercury inputs through wet atmospheric deposition likely increase with elevation as well (assuming continual transport of mercury into the area; Heindel et al. 2020; USDA n.d.). Alternatively, particulate deposition through dust generally decreases with elevation in the Rocky Mountains, likely due to greater contributions of dust from urban and agricultural practices at lower elevations, and atmospheric suspension of soil due to climate induced decreases in soil moisture. This pattern could

Fig. 3 The mercury cycle in the Rocky Mountains ($\sim 807,075 \text{ km}^2$). Fluxes are in italics ($\mu\text{g m}^{-2} \text{ yr}^{-1}$), annual area fluxes are in parentheses (Gg yr^{-1}), and soil pools are bolded (Gg). Gray arrows represent uncertainty along mountain elevation gradients



potentially result in a negative correlation between direct dry deposition of particulate mercury with altitude (Heindel et al. 2020). Changes in precipitation and temperature with elevation also drive dramatic shifts in plant communities in the Rocky Mountains, going from, for example, Tallgrass prairie in the plains, to open *Pinus ponderosa* forests in the foothills, to more dense mixed stands of *Pseudotsuga menziesii* and *Pinus contorta* in the montane and subalpine, to sparse krummholz and open tundra in the alpine. Differences in plant community structure play an important role in determining patterns of mercury dry deposition through plant uptake and transfer to soils (see above) but these impacts have not yet been quantified in the Rocky Mountains. The absence of these data was corroborated by the synthesis study of Eagles-Smith et al. (2016a). They reported that currently, data on mercury uptake by shrubs, grasslands, and herbaceous plant functional groups is lacking relative to forested ecosystems, thereby making it challenging to properly characterize mercury cycling in areas where these plant communities dominate (Gerson et al. 2022; Zhou et al. 2021).

Soil storage and evasion

Across the ~ 807,000 km² of the Rocky Mountains, approximately 4.2 Gg of mercury is stored in the upper 0.3 m of soil, with approximately 0.89 Gg in the top 5 cm (derived from Olson et al. 2022; Fig. 3). Mercury concentrations are highest in the soil O horizon when present ($70\text{--}200\text{ ng g}^{-1}$) followed by the A horizon ($30.2\pm61\text{ ng g}^{-1}$) and the C horizon ($25.5\pm32\text{ ng g}^{-1}$). The presence and depth of an O horizon is highly heterogenous across the Rocky Mountains and is driven by changes in elevation, temperature, aspect, slope, vegetation cover, and disturbance history (Hoffman et al. 2014). The concentration of mercury within an O horizon also varies markedly depending on the type of parent litter and period of decomposition. Some areas may have an older, thinner O horizon composed of high mercury concentration material (e.g., moss, lichen); in contrast, other areas may be composed of newer, thicker O horizon comprised of lower mercury concentration material (e.g. deciduous leaves; Pokharel and Obrist 2011). Soil mercury concentrations in the A horizon are broadly driven by soil organic matter content, land cover, and ecoregion (Olson et al.

2022). Although mercury concentrations in the O horizon are typically higher than the A horizon, mercury pools are typically larger in the A horizon due to higher soil bulk density (Olson et al. 2022). Across the U.S., the soil A horizon has significantly higher mercury concentrations compared to the C horizon ($34.0\pm0.5\text{ ng g}^{-1}$ versus $27.0\pm0.4\text{ ng g}^{-1}$, $p<0.01$). Olson and colleagues (2022) attributed this difference to enrichment of surface soils by external inputs such as atmospheric deposition. Yet, this pattern is not evident in the Rocky Mountains where there is no significant difference between the A and C horizons ($p>0.05$; Olson et al. 2022). The lack of horizontal variation in non-aquic soils may be a result of lower organic carbon content, lower precipitation rates, and higher incident solar radiation that drives photoreduction in surface soils; however, further analysis would be needed to determine the driving influences.

Watersheds in the Rocky Mountains with historic mercury, gold, or silver mining typically have elevated mercury concentrations that can exceed 100,000 ng g⁻¹ total mercury and 20 ng g⁻¹ MeHg in soils (Fleck et al. 2016). Within the Rocky Mountains, there are over 7,300 historic sites where gold was mined either as a primary, secondary, or tertiary commodity (Fig. 1; Mason et al. 1996). Most of the mines are located within central Colorado, western Montana, and central Idaho which coincides with the highest density of waterbody-specific fish advisories (Fig. 1). Fewer downstream impacts are associated increasing watershed size and greater natural vegetation land cover (Domagalski et al. 2016).

In mountains of the eastern U.S. and China, researchers have found that soil mercury concentrations are positively correlated with altitude due to shifts in land cover, atmospheric mercury deposition, and soil storage capacity (Gerson et al. 2017; Zhang et al. 2013a, b). Studies from the central Himalaya, however, have found an inverse relationship between total mercury concentrations and elevation associated with decreasing soil carbon content (Tripathee et al. 2019). Soil mercury concentrations and pools along elevational gradients in the Rocky Mountains have yet to be investigated marking an important knowledge gap regarding the factors that drive mercury soil storage in this region, and how these factors may change in the future with ongoing global change.

Across the western U.S., an estimated 35,100 kg yr⁻¹ of mercury is emitted from soils to the

atmosphere, primarily in the elemental Hg(0) form due to its high volatility. Fluxes vary widely across the west ranging from $7.7 \pm 0.2 \text{ } \mu\text{g m}^{-2} \text{ yr}^{-1}$ in the Great Plains to $29.7 \pm 1.9 \text{ } \mu\text{g m}^{-2} \text{ yr}^{-1}$ in the Mediterranean ecosystems of California (Eagles-Smith et al. 2016a; Eckley et al. 2016). Comparing estimates of mercury inputs and losses across the entire western U.S. (inclusive of the Rocky Mountains) indicates that, on average, this region is a mercury sink (Eagles-Smith et al. 2016a). Within the Rocky Mountains, the source-sink behavior of mercury likely varies across elevation gradients, land and plant cover, variability in atmospheric deposition rates, leaching in runoff, and evasion to the atmosphere. In Rocky Mountain National Park, researchers found that less than 20% of atmospherically deposited mercury was lost in annual runoff, suggesting that the alpine zone acts as a sink for mercury (Mast et al. 2005; Shanley et al. 2008). However, this estimate was made without soil flux measurements and intense solar radiation at high elevations likely promotes high evasion rates (Eckley et al. 2016). Soil evasion measurements using dynamic flux chambers from northwestern forested mountains average $11.5 \pm 0.4 \text{ } \mu\text{g m}^{-2} \text{ yr}^{-1}$ and provide an estimate for rates in forested montane and subalpine regions of the Rocky Mountains (Eckley et al. 2016). However, measurements from across a diverse subset of Rocky Mountain land covers will be necessary to better constrain the overall source or sink nature of mercury.

Transport, transformations, and bioaccumulation

In the Rocky Mountains, most of the mercury transport in runoff occurs in the spring when snowmelt flushes surface soils (Mast et al. 2005; Packer et al. 2020). Mercury is transported in both dissolved and particulate phases but is dominated by the dissolved phase, particularly later in the summer (Mast et al. 2005). Studies from Rocky Mountain National Park, Colorado and Provo River, Utah found stream concentrations of total mercury ranging from $> 8 \text{ } \text{ng L}^{-1}$ during snowmelt and $< 1 \text{ } \text{ng L}^{-1}$ during baseflow with total annual mercury fluxes of 1.2 to $2.3 \text{ } \mu\text{g m}^{-2} \text{ yr}^{-1}$ (Fig. 3; Mast et al. 2005; Shanley et al. 2008).

If deposited or transported into areas of permanent or temporary saturation, inorganic mercury can be readily transformed into MeHg. Across the western U.S., MeHg concentrations in aquatic sediments have

large spatial variability that are driven by landscape and land-use characteristics. Importantly, Fleck and colleagues (2016) found some of the areas with the highest MeHg concentrations occurred in areas with relatively low total mercury concentrations, particularly in areas of the Rocky Mountains. The production of MeHg in aquatic regions of the Rocky Mountains, however, has received little attention despite over 6000 km^2 of lakes and ponds, 3000 km^2 of reservoirs, 700 km^2 of streams, and 650 km^2 of wetlands.

There is evidence, however, from dragonfly larvae used as biosentinels that lakes, streams, and wetlands in the Rocky Mountains have MeHg concentrations at levels of concern for human and ecosystem health (Eagles-Smith et al. 2020). Eagles-Smith and colleagues (2020) conducted a survey of > 450 sites spanning 100 U.S. National Park service units to create integrated impairment indices for fish, wildlife, and humans based on mercury concentrations of dragonfly larvae. Data extracted from the seven sites sampled within the Rocky Mountains shows 10% of samples were below any of the deleterious effect benchmarks, 16% had low hazard risk, 35% had moderate hazard risk, 22% had high hazard risk, and 5% had severe hazard risk. The percentage of sites within the high hazard and severe hazard risk categories in the Rocky Mountains was higher than in the U.S. as a whole, where only 11 and 1% of sites fell into those categories, respectively (Eagles-Smith et al. 2020). Despite the Rocky Mountains having areas with high- to severe-risk for MeHg contamination, we did not find any studies that quantify the processes driving these concentrations, such as studies on mercury methylation and demethylation rates. Indeed, methylmercury production in lake, stream, and wetland environments of the Rocky Mountains has received little to no attention, with most studies previously reporting total mercury MeHg concentrations from high altitude lake ecosystems (e.g., Krabbenhoft et al. 2002). This knowledge gap is important to address, as these aquatic regions act as gateways for the transport of water from high elevations downstream, and provide habitat and forage for local wildlife.

Despite limited investigation of MeHg production in mountain regions, the atmospheric deposition and methylation of mercury across the western U.S. has resulted in widespread mercury contamination and mercury bioaccumulation within fish populations of the Rocky Mountains (Lepak et al. 2016).

There are over 200 waterbody-specific fish consumption advisories for mercury in the region (Fig. 1). These advisories likely underestimate the extent of the mercury contamination, as waterbody-specific advisories are limited to sites where fish mercury concentrations have been tested. In addition, there are state-wide consumption advisories for specific fish species and fish lengths for all locations in Idaho (<8 meals per month of Smallmouth (*Micropterus dolomieu*) and Largemouth (*Micropterus salmoides*) bass), Colorado (<1 meals per month of Smallmouth Bass (*Micropterus dolomieu*) <38 cm, Largemouth Bass (*Micropterus salmoides*) >38 cm, Tiger Muskie (*Esox masquinongy*); and <2 meals per month of Cutthroat Trout (*Oncorhynchus clarkia*) and *Micropterus dolomieu* >38 cm), and Wyoming (avoid *Oncorhynchus clarkia* >38 cm, *Micropterus* >30 cm, Black Crappie (*Pomoxis nigromaculatus*) >25 cm, Burbot (*Lota lota*) >51 cm, Channel Catfish (*Ictalurus punctatus*) >51 cm, Sauger (*Sander canadensis*) and Walleye (*Sander vitreus*) >30 cm, and Northern Pike (*Esox Lucius*) and *Esox masquinongy*; Fig. 1). These advisories are for general populations and more stringent recommendations exist for pregnant people and children.

Of the waterbody-specific advisories in the Rocky Mountains, over half of the locations are found above 1500 m in elevation, with the majority in constructed waterbodies such as reservoirs. Alternatively, lotic, or moving freshwater environments, generally, have lower mercury bioaccumulation in fish. Day et al. (2020) found that only 13% of over 2,300 samples exceeded fish health benchmarks in their study of the Upper Colorado River Basin. This pattern suggests that high elevation reservoirs may be particularly important hot spots for mercury bioaccumulation and exposure with implications for downstream ecosystems and human populations.

In addition to mercury bioaccumulation in aquatic food webs, evidence from other mountain and steppe regions suggest that terrestrial bioaccumulation of mercury is also an area of concern. The impacts, however, of MeHg bioaccumulation on behavior, reproduction, and survival is poorly understood for most terrestrial taxa in mountainous regions (Rimmer et al. 2010; Rodenhouse et al. 2019). Although terrestrial ecosystems typically produce low concentrations of MeHg, aquatic MeHg can pass into terrestrial food webs and enhance mercury bioaccumulation (Cristol

et al. 2008; Janssen et al. 2023). Additionally, terrestrial food webs can have higher trophic levels compared to aquatic food webs resulting in greater MeHg bioaccumulation in top consumers (Bartrons et al. 2015; Janssen et al. 2023). Elevated mercury concentrations, specifically in terrestrial mountain food webs, have been observed across trophic levels from arthropods (Rimmer et al. 2010) to birds (Ackerman et al. 2016; Jackson et al. 2016; Sauer et al. 2020) to top predators (Ma et al. 2023). These studies generally found organism tissue MeHg concentrations to be highest in mid- to high-elevation zones because of elevated atmospheric mercury deposition and increased MeHg bioavailability (Rodenhouse et al. 2019; Sauer et al. 2020; Townsend et al. 2014).

The human impact of mercury exposure is widespread throughout the world, causing a variety of neurological health consequences, primarily through consumption of fish and shellfish (USEPA 2018). Many of the fish species with advisories are popular for anglers who feed themselves and their families with locally caught fish. Indeed, for many communities and families experiencing financial hardship, eating locally caught fish is an essential protein source (Quimby et al. 2020). Additionally, catching and consuming fish is a sovereign right for the Tribal Nations marking an essential social practice and source of economic sustenance for indigenous communities (Cantzler and Huyn 2016). Thus, the widespread mercury contamination of fish populations across the U.S. marks a stark environmental justice issue (Barbo et al. 2023; Dai et al. 2023; Chiapella et al. 2021; Eagles-Smith et al. 2016b; Houde et al. 2022; Roe 2003). The disproportionate impact of mercury contamination on indigenous peoples has been studies most extensively in the Arctic where adverse health outcomes have been observed across all life stages (see Basu et al. 2022). The effect of mercury contamination on native communities outside the Arctic is much less studied. One 2003 study examined the disproportionate impact of mercury contamination in food sources for indigenous communities across the U.S. and found 59 reservations are at moderate risk, 70 at high risk, and 19 at severe risk for mercury exposure. Additionally, across 655 watersheds containing a native community (> 10% native population), the mean fish mercury concentration was 0.32 ppm, just above the EPA's guidance value for safe fish consumption at the time (Roe 2003). There are eleven reservations with native

communities within the Rocky Mountains (Fig. 1), and indigenous peoples make up ~ 2–10% of the total population within each of the states that include the Rocky Mountains (Fig. 1). These communities are likely to be impacted by continued mercury contamination; however, information is not currently available regarding the disproportionate exposure of MeHg to these groups. This deficiency marks an important need for community-driven research, education, and outreach to better understand the scope of this issue and effective means for counteracting mercury exposure while maintaining cultural traditions.

How does climate change impact mercury cycling in the Rocky Mountains?

Background

Climate change is impacting high elevation ecosystems more rapidly and intensely than lowland regions (Kittel et al. 2015; Hock and Rasul 2019). Since these ecosystems are highly sensitive to mercury contamination, it is important to consider how future change will impact mercury transport, bioavailability, and

toxicity. In the Rocky Mountains, climate change is causing increased warming (McGuire et al. 2012), drought conditions (Tague & Dugger 2010), and growing season length (Hu et al. 2010), all of which have important implications for mercury cycling in local and distant ecosystems. Here, we focus specifically on the effects of shifting hydrology and wildfire on mercury mobilization and ecosystem exposure (Fig. 4).

Shifts in hydrology

Drought and warming temperatures are causing shifts in hydrology that impact the availability and transport of inorganic mercury, as well as the potential for MeHg production in the Rocky Mountains through a variety of mechanisms (Fig. 4). As the Rocky Mountains warm, snowmelt is occurring earlier and more precipitation is falling as rain than snow (Halofski and Peterson 2018; Larson et al. 2011). Clow (2010) found that between 1978 and 2007, increasing springtime air temperatures and declining snowpack shifted snowmelt 2 to 3 weeks earlier in the Colorado Rocky Mountains. Hydrologic simulation experiments using reconstructed

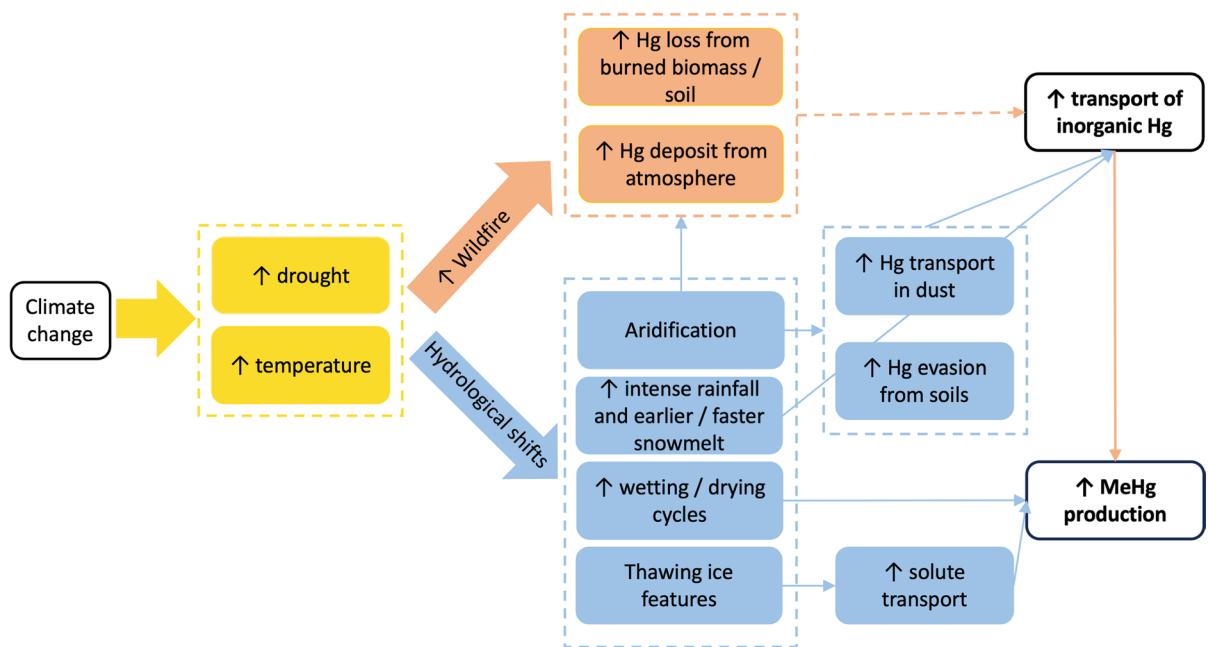


Fig. 4 Schematic illustrating the climate change driven impacts of changing wildfire (red) and hydrology (blue) on inorganic mercury transport and MeHg production in the Rocky Mountains

snowpacks, also from the Colorado Rocky Mountains, predicted an earlier melt-out of 31 days on average, spanning the years 2001–2014 (Badger et al. 2021). Additionally, more precipitation is occurring as rain than snow across the Northern Hemisphere where snow occurs, with periods of heavy precipitation intensifying (McCabe and Wolock 2010; Rocca et al. 2014). In mountain environments, more precipitation as rain is also causing well documented increases in rain-on-snow events (Cache et al. 2023; Musselman et al. 2018). These changes increase erosion and transport of sediment downstream, and these responses are predicted to worsen with continued climate change (Cache et al. 2023; Pelletier 2009). Using a landscape evolution model, Cache and colleagues (2023) demonstrated that under the most extreme climate scenario (RCP8.5), sediment yield in a small Swiss Alps catchment increased by 6% due to more precipitation falling as rain and intensification of heavy precipitation events. Increased flushing and erosion of surface soils results in greater export of soil-bound mercury downstream to lentic ecosystems where anoxic conditions and availability of nutrient substrates favor conversion of inorganic mercury to MeHg (Halofsky and Peterson 2018; Sun et al. 2022).

Earlier snowmelt, decreasing snowpack, coupled with subsequent drought and more intense periodic rainfall lead to more extreme wetting and drying cycles that can accelerate MeHg production. Reservoirs are particularly sensitive to this phenomenon; earlier, more intense spring runoff causes reservoir stage to decrease earlier and fluctuate more dramatically among years (Cohen et al. 2020). Across the western U.S., a 3.2-fold increase in fish mercury concentrations was observed across – 30 to +50% variations in interannual reservoir water levels (Willacker et al. 2016). Fish in reservoirs that experienced their lowest water stage at the beginning of the summer (May, June, or July) had fish mercury concentrations up to 11-fold higher than in reservoirs with water minimums at other times of the year (Willacker et al. 2016). Elevated MeHg production under these conditions is likely driven by accelerated decomposition of organic matter in littoral sediments experiencing water-level fluctuations. This wet-dry cycle enhances mercury methylation by liberating inorganic mercury into bioavailable forms during low stage, as well as

increasing reducing conditions and dissolved organic carbon needed for microbial methylation during high stage conditions (Eckley et al. 2017).

Changes in selective water withdrawal can also impact temperature and oxygen conditions within reservoirs. These changes have implications for MeHg production in reservoirs with past studies documenting increased MeHg production and uptake in aquatic food webs at the Hells Canyon Complex (Snake River, Idaho-Oregon) because of increased thermal stratification and anoxia (Baldwin et al. 2022). Climate and land-use changes are driving widespread increases in seasonal anoxia and thermal stratification, which combined with increased wetting and drying cycles has the potential to exacerbate MeHg production across a variety of mountain aquatic ecosystems (Jane et al. 2021). This marks an important knowledge gap and area for future research to better quantify the impact of climate-driven hydrologic shifts on MeHg production in reservoirs, as well as other natural water bodies, within the Rocky Mountains.

Thawing ice features in high elevation regions may also impact hydrology and the potential for MeHg production in mountain regions across the globe. Over the past three decades, chemistry in high elevation streams from multiple sites in the Rocky Mountains, western Canada, the European Alps, the Icelandic Shield, and the Himalayas demonstrate consistent and widespread patterns of increasing sulfate and base cation concentrations or fluxes (Crawford et al. 2019). In the Rocky Mountains, despite decreasing trends in atmospheric sulfate deposition, sulfate concentrations in runoff have increased by 300% over the past 30 years. This trend is likely the result of accelerated weathering of pyrite associated with thawing ice features (Crawford et al. 2019). It is unknown whether the MeHg production in the Rocky Mountains is sulfate limited; however, it is possible that increases in sulfate export could stimulate MeHg production by sulfate reducing bacteria in downstream aquatic environments, such as mountain reservoirs and wetlands (Jeremiason et al. 2006).

Aridification also impacts mercury cycling in the Rocky Mountains by changing inputs and losses of dust-bound mercury through wind erosion, and soil mercury evasion (Duniway et al. 2019; Huang et al. 2020; Overpeck and Udall 2020; Scott and Black 2020). Aridification and land use changes are

increasing desertification and dust storms in many regions of the globe, particularly the Asian and African continents (Han et al. 2021; Yang et al. 2022; Zhang et al. 2019a, b; Zhang et al. 2013a, b; Zhu et al. 2022). The western U.S. receives a significant percentage of annual dust loads from these regions with 49–77% coming from Asia and 15–34% coming from Africa (Duncan et al. 2007; Zhang et al. 2013a, b). Multiple studies from western Chinese mountain ecosystems, the Atlantic Ocean, and Antarctic snowpack, demonstrate that wind-transported dust from Asia and Africa are important sources of mercury to downwind regions, such as the western U.S. (Huang et al. 2020; Witherow and Lyons 2008).

In addition to receiving dust-bound mercury from afar, the western U.S. is also experiencing aridification and intensifying dust storms that can transport dust-bound mercury to the Rocky Mountains (Duniway et al. 2019; Overpeck and Udall 2020). Historical data from glaciers and high elevation lake sediments cores in the Rocky Mountains demonstrate that dust is an important source of mercury to mountain ecosystems (Carling et al. 2017; Mast et al. 2010). Future studies that determine the concentration of mercury in dust, rates of deposition, and how these factors are shifting with global change are needed to better understand the relative importance of this mercury source. Inputs through dust, though, likely play an important role in the mercury cycle in the Rocky Mountains, as work from the Arctic demonstrates how elevated dust increases mercury concentrations in vegetation with implications for litterfall, soil, and local wildlife (Olson et al. 2019).

With continued drought, the Rocky Mountains may also become a global source of mercury through wind erosion and soil-air evasion (Eckley et al. 2016; Goudie 2018; Scott and Black 2020). To our knowledge, no studies have investigated mercury export in aeolian erosion from this region, particularly in the context of the total mercury transport flux. Future work must determine if the Rocky Mountains are a sink or source for dust-bound mercury and what the global and regional implications are for mercury transport and bioaccumulation. Aridification also impacts mercury evasion from soils back to the atmosphere by reducing waterbody and vegetation extent, thereby exposing more bare soil surfaces (Bodner and Robles 2017; Hannoun and Tietjen 2022). In general, bare soils receive greater solar

radiation and have drier surfaces, two factors known to be positively correlated with greater soil-air mercury fluxes (Eckley et al. 2016). Eckley and colleagues (2016) demonstrated across the western U.S. that sparsely vegetated regions have larger net ecosystem mercury emissions compared to forested and other heavily vegetated regions. This suggests that continued aridification of the west may contribute to greater net losses of mercury from these ecosystems over time.

Increased wildfire activity

Warmer air temperatures and increased drought are driving more frequent and intense wildfires across the western U.S (Abatzoglou and Williams 2016). Ash released from wildfires can have a variety of consequences for local mercury cycling. When biomass is burned, mercury previously stored in above ground plant tissues such as grasses, shrubs, and trees, as well as surface soils (<5 cm), is released back to the atmosphere in elemental and oxidized forms that act as a substantial release of mercury from terrestrial ecosystems (Homann et al. 2015; Webster et al. 2016). Remobilized mercury is then available for further transformations and uptake by organisms once it is redeposited onto the landscape (Kumar and Wu 2019; Li et al. 2022). Webster and colleagues (2016) report that across the western U.S., $\sim 3100 \pm 1900 \text{ kg yr}^{-1}$ of mercury is released annually from wildfires; this value is likely to increase because of more frequent and intense wildfires. Additionally, enrichment of mercury in terrestrial ecosystems, due to increasing atmospheric mercury emissions, is projected to increase mercury wildfire emissions across North America by 19% in 2050 (Kumar et al. 2018). The amount of mercury released from an ecosystem during wildfire depends on the vegetation structure and fire severity. In general, across forests of the western U.S., Aspen forests (*Populus tremuloides*) tend to release the lowest amount of mercury during a burn event, averaging 0.9 g ha^{-1} , while Hemlock-Sitka Spruce forests (*Picea sitchensis*) release the most averaging, 7.8 g ha^{-1} (Webster et al. 2016). In the Wyoming Rocky Mountains, wildfire was found to release $3.6\text{--}12.9 \text{ g ha}^{-1}$ of mercury in deciduous forests and $7.4\text{--}25.3 \text{ g ha}^{-1}$ in coniferous forests (Biswas et al. 2007).

Wildfire in grasslands is also likely an important vector for mercury loss within the Rocky Mountains. Long-term records demonstrate increasing wildfire activity in these ecosystems over the past 30 years (Donovan et al. 2017). The quantity of mercury stored and released in grassland ecosystems, however, has received little attention. One study from the Rocky Mountains reported a loss of ~ 4.1 g ha^{-1} of mercury during wildfire (Biswas et al. 2007). This research suggests that grasslands can act as a source of mercury during wildfire, similar to forested regions, but additional studies are needed. In addition to vegetation cover, soil development also impacts the amount of mercury released during wildfire with soil O horizons releasing more mercury than A horizons (Homann et al. 2015). Since the Rocky Mountains are characterized by extreme gradients in soil development and vegetation cover with changes in elevation and aspect, better measurements of mercury release from wildfire across these different regions will be critical in assessing the mercury sink/source nature of the region.

Most of the mercury released during wildfire is in its particulate, oxidized form with a relatively short residence time. Thus, a large fraction of ash is redepositing on the landscape close to the source (Seigneur et al. 2004). When ash falls to the surface of Earth, it acts as a vector for the movement of mercury into an ecosystem where it can have a variety of fates and consequences. Ash-bound mercury typically has a relatively low methylation potential, resulting in low bioavailability (Ku et al. 2018). However, wildfire ash has also been shown to leach labile organic matter which provides an important energy source to mercury methylating microbes, thereby indirectly increasing MeHg production (Li et al. 2022). Li and colleagues (2022) showed that wildfire ash efficiently sorbs inorganic mercury onto its surface, helping to store mercury within ecosystems.

These various fates of ash-bound mercury illustrate the uncertainties regarding the impact of increased wildfire activity on mercury contamination, specifically for the Rocky Mountains. In addition to local wildfires, the Rocky Mountains also intercept smoke plumes from more distant wildfires such as those occurring in California (Martin et al. 2013; Brey et al. 2018). These distal sources of smoke may increase inputs of mercury into the Rocky Mountains through wet and dry atmospheric deposition. It

will be important for future studies to quantify the sources, concentrations, and species of mercury in smoke plumes to better predict how continued wildfire activity will impact mercury transport, bioavailability, and exposure of biota.

Summary of research opportunities

With continued atmospheric mercury deposition to the Rocky Mountains, there is a need for research that addresses important knowledge gaps both for this region, as well as semi-arid mountains globally. We examined these gaps in the text above and summarize research priorities here (Table 2).

First, it is important to quantify the inputs and losses of mercury through atmospheric deposition and evasion, particularly in the context of increasing wildfire activity and aridification. Quantifying net mercury budgets is critical to constrain the source or sink nature of the Rocky Mountains and to improve understanding of the role of this region in the global mercury cycle. Constraining the sources and pathways of mercury inputs through wet and dry deposition is also important as the proportions of each are likely to change with increased wildfire and wind erosion, as well as shifts in global primary emission sources, which will dictate mercury exposure and toxicity. It will be particularly critical to characterize and quantify the role of vegetation in mediating the atmosphere-land exchange of mercury.

Second, it is important to constrain how the mercury cycle shifts along elevation gradients to determine the zones with the highest contamination risk, and how higher elevation sites may impact downstream regions. Assessing mercury inputs, storage, and transformations with changes in precipitation and vegetation cover along elevation gradients in the Rocky Mountains will help determine the factors controlling mercury cycling in western U.S. mountain ecosystems. Additionally, it will strengthen our ability to compare the Rocky Mountains to other mountain regions of the world where research has provided a better understand of mercury dynamics along mountain elevation gradients, such as in the Himalaya and eastern U.S.

Third, methylmercury production in mountain aquatic regions such as reservoirs, wetlands, and streams needs to be assessed to determine

Table 2 Summary of knowledge gaps and future research priorities

| Knowledge gap | Future research priority for western mountain regions |
|--|--|
| Concentrations and flux of atmospheric mercury dry deposition | Increase number of AMNet sites. Collect litterfall and throughfall data. Assess the impact of plant cover along elevation gradients on dry deposition rates |
| Concentrations and pools of mercury in soils along mountain elevation gradients | Conduct studies similar to past work done in the eastern U.S. and China to determine patterns of total and methylmercury concentrations and pools along elevation gradients. This will provide insights into regions with the greatest risk for mercury exposure to humans and wildlife |
| Rates of mercury evasion from soils along mountain elevation gradients | Use consistent, reproducible methods (e.g., dynamic flux chambers) for measuring mercury evasion rates across different land covers and elevation zones |
| Methylmercury production in mountain reservoirs, lakes, and wetlands | Measure concentrations and mercury methylation efficiencies in different aquatic landforms to determine regions that are hotspots for MeHg production and exposure to local wildlife and downstream ecosystems. Use dragonflies as biosentinels to put ecosystems into larger contamination risk index |
| The disproportionate impact of mercury contamination on indigenous communities | Use community-driven research, education, and outreach to better understand the scope of mercury exposure and effective measures for counteracting health concerns while maintaining cultural traditions |
| Impacts of climate change on mercury cycling in mountain regions | Measure mercury concentrations in runoff and use stable isotopes to determine mercury source (atmospheric versus terrestrial) |
| Rates of mercury transport downstream as a result of earlier and faster snowmelt | Measure concentrations and methylation rates in aquatic mountain regions to determine areas that act as MeHg hotspots. Conduct wetting/drying incubation experiments. Measure soils <i>in situ</i> following wetting/drying events and compare to baseline conditions |
| Methylmercury production in reservoirs, lakes, and wetlands that are experiencing increasing wetting/drying cycles | Collect dust from persistent snowpack/glacial regions to determine mercury inputs via dust over time. Collect bulk deposition and air samples in areas exposed to dust plumes |
| Mercury transport in dust from increased aridification and dust storms | Combine measurements of soil evasion rates across moisture and plant cover gradients with model predictions of aridification |
| Mercury evasion from soils with increased aridification | Collect air samples within wildfire plumes to determine source, concentrations, and species of mobilized mercury. Use these data to determine the bioavailability of mobilized mercury and source/sink nature of regions that are burning |
| The sources, concentrations, and species of mercury transported in wildfire plumes | |

contamination risk for humans and local wildlife. Reservoirs are an important area of focus since they make up a large percentage of waterbodies in the Rocky Mountains and are susceptible to MeHg production due to increases in wet-dry cycles and low stage conditions. Additionally, reservoirs are important pathways of human exposure to mercury through fish consumption. Investigating MeHg production in streams and wetlands is also important. These aquatic expanses provide important food resources and habitat to local ecosystems, as well as pathways for water

supply downstream. Shifts in hydrologic conditions due to climate change in these areas will likely exacerbate net MeHg production.

Fourth, there is a need to examine the disproportionate impact of mercury contamination on indigenous communities within the Rocky Mountains—and U.S., more broadly—through community-led research, education, and outreach. Such efforts will require integration of indigenous representatives into scientific studies; such inclusion must occur at the beginning of the research process to ensure that the

priorities and standards of indigenous communities lead and are represented in the research.

Finally, it is important to assess the impact of climate change on mercury cycling in mountain regions. Shifts in hydrology and wildfire connected to global change have the potential to increase the availability of mercury, production of MeHg, and exposure to humans and wildlife in mountain regions making this a critical area of research. Quantifying the impacts of climate change on mercury cycling will require a combination of long-term observations and modeling efforts to understand the consequences of different climate-related forcings (Table 2).

Although we have focused on existing research and knowledge gaps in the U.S. Rocky Mountains region, the topic of mercury cycling is applicable to other semi-arid mountain ecosystems of the world. Indeed, due to the ubiquity of mercury in environments globally, it is truly a topic that concerns everyone, and must be prioritized in the research agenda to promote the health of ecosystems and people everywhere.

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Declarations

Competing interests The authors have no relevant financial or non-financial interests to disclose.

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