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# Conifer density within lake catchments predicts fish mercury concentrations in remote subalpine lakes☆

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ABSTRACT

Remote high-elevation lakes represent unique environments for evaluating the bioaccumulation of atmospherically deposited mercury through freshwater food webs, as well as for evaluating the relative importance of mercury loading versus landscape influences on mercury bioaccumulation. The increase in mercury deposition to these systems over the past century, coupled with their limited exposure to direct anthropogenic disturbance make them useful indicators for estimating how changes in mercury emissions may propagate to changes in Hg bioaccumulation and ecological risk. We evaluated mercury concentrations in resident fish from 28 high-elevation, sub-alpine lakes in the Pacific Northwest region of the United States. Fish total mercury (THg) concentrations ranged from 4 to 438 ng/g wet weight, with a geometric mean concentration (±standard error) of 43 ± 2 ng/g ww. Fish THg concentrations were negatively correlated with relative condition factor, indicating that faster growing fish that are in better condition have lower THg concentrations. Across the 28 study lakes, mean THg concentrations of resident salmonid fishes varied as much as 18-fold among lakes. We used a hierarchal statistical approach to evaluate the relative importance of physiological, limnological, and catchment drivers of fish Hg concentrations. Our top statistical model explained 87% of the variability in fish THg concentrations among lakes with four key landscape and limnological variables: catchment conifer density (basal area of conifers within a lake's catchment), lake surface area, aqueous dissolved sulfate, and dissolved organic carbon. Conifer density within a lake's catchment was the most important variable explaining fish THg concentrations across lakes, with THg concentrations differing by more than 400 percent across the forest density spectrum. These results illustrate the importance of landscape characteristics in controlling mercury bioaccumulation in fish.

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## 1. Introduction

Mercury (Hg) is a globally distributed pollutant that poses considerable risks to human and wildlife health (Scheuhammer and Sandheinrich, 2008). Under appropriate biogeochemical conditions in aquatic ecosystems, inorganic Hg is microbially converted to the highly toxic and bioavailable organic form, methylmercury (MeHg), which biomagnifies through food webs where it can reach toxicologically relevant concentrations in top predators. As a result, the ecological risk of Hg is driven by a combination of both inorganic loading, as well as ecological parameters that facilitate MeHg production and bioaccumulation. Inland lakes are particularly vulnerable to Hg contamination because they are recipients of transported Hg, and their organic rich, anoxic sediments can promote MeHg production.

Lakes receive Hg from various sources, including atmospheric deposition, watershed runoff, and glacial meltwater (Rudd, 1995; St. Louis et al., 1994). Over the past 150–200 years since the advent of the industrial revolution, approximately 80 percent of global emissions have come from anthropogenic sources, largely fossil fuel combustion (Mason et al., 1994), but also gold mining, and manufacturing (Pirrone et al., 2010). Mercury also is re-emitted to the atmosphere from forest fires and agricultural burning (Friedli et al., 2003). As a result, atmospheric deposition of Hg in remote environments has increased by approximately 3-fold (Lindberg et al., 2007). Importantly, there is substantial variability in the spatial patterns of anthropogenic Hg releases and deposition at

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local, regional, and global scales, associated with coal-fired power plants, industrial activity, and mining (Engstrom et al., 2007), as well as in the propensity of inorganic Hg to be methylated across the landscape. Therefore, it can be difficult to determine how much of the variability in Hg concentrations among systems can be attributed to Hg loading relative to in situ MeHg production.

Because of their isolation, remote high elevation lakes represent unique environments for evaluating the bioaccumulation of atmospherically deposited Hg through freshwater food webs, as well as for evaluating the relative importance of Hg loading versus landscape influences on Hg bioaccumulation. The increase in Hg deposition to these systems over the past century (Phillips et al., 2011), coupled with their limited exposure to direct anthropogenic disturbance make them useful indicators for estimating how changes in Hg emissions may propagate changes in Hg bioaccumulation and ecological risk. However, catchment structure of sub-alpine lakes can vary substantially in terms of forest density, which has been shown to influence fish Hg concentrations in lower elevation lakes with greater catchment areas (Drenner et al., 2013), thus it may be important to account for tree density in subalpine lake catchments because conifers can adsorb Hg from the atmosphere, resulting in increased deposition rates.

In this study, we evaluated Hg bioaccumulation in fishes of high elevation, sub-alpine lakes in the Wallowa–Whitman National Forest in northeastern Oregon and western Idaho (Fig. 1). Our goals were to (1) assess the magnitude of Hg contamination in a collection of small-catchment lakes, (2) quantify the spatial variability in fish Hg concentration across these lakes, and (3) determine the physiological, limnological, and landscape factors that are most strongly related to fish THg concentrations.

## 2. Methods

### 2.1. Site description and field collection

The Wallowa–Whitman National Forest in northeastern Oregon and western Idaho occupies more than 9000 $km^2$ of land, including nearly 2500 $km^2$ of designated wilderness. The forest is comprised of varied landscapes including low-elevation grasslands, coniferous forests, and alpine meadows. High-elevation habitats are found in the Elkhorn Mountains, the Wallowa Mountains and Eagle Cap Wilderness, and the Hells Canyon National Recreation Area. Throughout the region there are small (<0.5 $km^2$), high-elevation (>2000 m) lakes within isolated catchments that have naturally reproducing populations of brook trout (*Salvelinus fontinalis*) and rainbow trout (*Oncorhynchus mykiss*) as a result of historical fish stocking for recreational purposes (Figure S1). These lakes are solely catchment or groundwater fed, and Hg sources are largely limited to atmospheric deposition because they do not have inflowing streams, and are not known to have mercury-enriched geologic deposits. The catchments associated with these lakes generally are small (<5 $km^2$), and comprised of a combination of open, rocky terrain, and temperate, subalpine coniferous forest.

Between July 11 and September 1, 2011 we sampled brook trout, rainbow trout, cutthroat trout (*Oncorhynchus clarkii*), and lake trout (*Salvelinus namaycush*) from 28 different lakes within 5 geographic regions (North Elkhorn, South Elkhorn, North Eagle Cap, South Eagle Cap, and Seven Devils) of the Wallowa–Whitman National Forest (Table S1, Figure S1). Visits to lakes were randomized over that time-frame to prevent sampling date from confounding regional Hg assessments. Fish were sampled using both hook and line and 30 m × 2 m variable-mesh gillnets and were only retained if they exceeded lengths of stocked fish so we could control for any stocking effect. Upon collection fish were immediately wrapped in clean, solvent-rinsed aluminum foil and sealed in individually labeled polyethylene bags. Each bagged fish was kept cool on ice or snow until delivery to the laboratory (within 48 h), where they were stored at −20 °C until processing. At 21 of the lakes, we also collected water samples in 125 mL acid-cleaned amber glass bottles for water chemistry analysis.

### 2.2. Fish sample processing and tissue preparation

In the laboratory, we thawed each fish sample to room temperature, and then measured standard length to the nearest millimeter on a fish board and mass to the nearest 0.1 g on a digital balance. We calculated the body condition of each fish using the Relative Condition Factor ($\boldsymbol{K}_n$), which accounts for potential changes in shape as fish grow (Anderson and Neumann, 1996). The relative condition factor was calculated as:

$$\boldsymbol{K}_n = \boldsymbol{W}/\boldsymbol{W}',$$

Where:

$\boldsymbol{W}$ = fish whole body mass, in g and,
$\boldsymbol{W}'$ = the predicted length-specific mean mass from a predictive length-mass regression model calculated for each species.

To determine W′ for each species, we used $\log_{10}$-transformed standard length (mm) and $\log_{10}$-transformed wet mass (g) data for all fish of each species, from all lakes in which they were captured (brook trout linear regression: n = 230, $r^2$ = 0.96, intercept = −11.646, slope = 3.099; rainbow trout linear regression: n = 85, $r^2$ = 0.91, intercept = −10.696, slope = 2.913; cutthroat trout linear regression: n = 11, $r^2$ = 0.97, intercept = −10.520, slope = 2.854). From each fish, we dissected 5–10 g of skinless axial muscle, rinsed the muscle tissue in deionized water, blotted it dry with a clean, lint-free wipe, and weighed it on an analytical balance to the nearest 0.0001 g. We then dried each muscle sample in a convection oven at 50 °C for 48 h, or until a constant mass was achieved. We subsequently removed the samples from the drying oven and allowed them to cool to room temperature in a desiccator. Once cool, we measured the dry mass of each muscle tissue sample to the nearest 0.0001 g, and ground them to a fine powder in a stainless steel tissue mill. The homogenized samples were then stored in a dark desiccator until chemical analyses.

### 2.3. Mercury determination

We determined total mercury (THg) concentrations on skinless axial muscle of each fish sample because most (90–95%) of mercury in fish muscle tissue is in the methylmercury (MeHg) form (Bloom, 1992). Total Hg concentrations were determined following EPA method 7473 (EPA, 2000) on a Milestone tri-cell DMA-80 Direct Mercury Analyzer (Milestone Inc, Monroe, Connecticut, USA) at the USGS Forest and Rangeland Science Center's Contaminant Ecology Laboratory in Corvallis, Oregon. Briefly, we used an integrated sequence of drying (250 °C for 30s), thermal decomposition (650 °C for 90s), catalytic conversion, and then amalgamation, followed by cold vapor atomic absorption spectroscopy. Unless stated otherwise, we analyzed and report all muscle tissue samples on a dry weight basis in order to control for variable moisture content among individuals. However, to facilitate comparison to other studies and to allow conversion to wet weight concentrations we determined moisture content in each sample. Moisture content in the fish muscle tissue ranged from 74.7 to 85.8 percent with a mean (±standard error) of 79.8 ± 0.1%.

Quality-assurance measures included analysis of two certified reference materials (either dogfish muscle tissue [DORM-3;