Date: 29 November 2011

Descriptive Title: Mercury cycling, bioaccumulation, and risk across western North America: a landscape scale synthesis linking long-term datasets.

Short Title: Western Mercury Synthesis

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Project Summary:

Mercury (Hg) is a serious environmental problem that is impacting ecological and human health on a global scale. However, local and regional processes are largely responsible for producing methylmercury, which drives ecological risk. This is particularly true in western North America where the combination of diverse landscapes, habitat types, climates, and Hg sources may disproportionally impact the region relative to other areas in North America. Even with decades of regional Hg research and monitoring, there is still no holistic synthesis of the spatiotemporal patterns of Hg in abiotic and biotic resources across the region, nor has there been a formal, simultaneous analysis of the landscape, ecological and climatological factors that drive Hg cycling, bioaccumulation, and risk of Hg in western North America.

Through a compilation of decades of data records on Hg, we will conduct a tri-national synthesis of Hg cycling and bioaccumulation throughout western North America in order to quantify the influence of land use, habitat, and climatological factors on Hg risk. With public land comprising more than 60% of the total surface area in the region, this knowledge is critical for more effectively managing resource to reduce Hg impacts. We have developed an interdisciplinary team of scientists and policy experts, representing three countries, to accomplish these goals across such an expansive area.

Proposed Start and End Dates: February 2012 – January 2014
Proposed Data Release Date: March 2014
Is this a resubmission? No
Conflicts of Interests with Reviewers: None

Problem Statement

Mercury (Hg) pollution is a serious environmental problem that impacts both ecosystem and human health on regional and global scales. Data from long term monitoring, sediment cores, ice cores, and museum specimens indicate that anthropogenically-derived Hg deposition has increase substantially since the 19^{th} century (Lamborg et al. 2002, Shuster et al. 2003), and Hg concentrations in abiotic and biological components of ecosystems have increased concomitantly (Kamman and Engstrom 2002). This has raised concern regarding potential impacts to ecological communities because the organic compound methylmercury (MeHg) is highly bioaccumulative and highly toxic (Clarkson and Magos 2006). Mercury is a relatively unique contaminant because the risk of deleterious environmental effects is strongly related to (1) **Landscape factors** that control Hg cycling and microbial MeHg production, (2) **Ecological factors** that control bioaccumulation, and (3) **Biological factors** that control its toxicity and effects on organisms, including fish, wildlife, and humans (Figure 1).

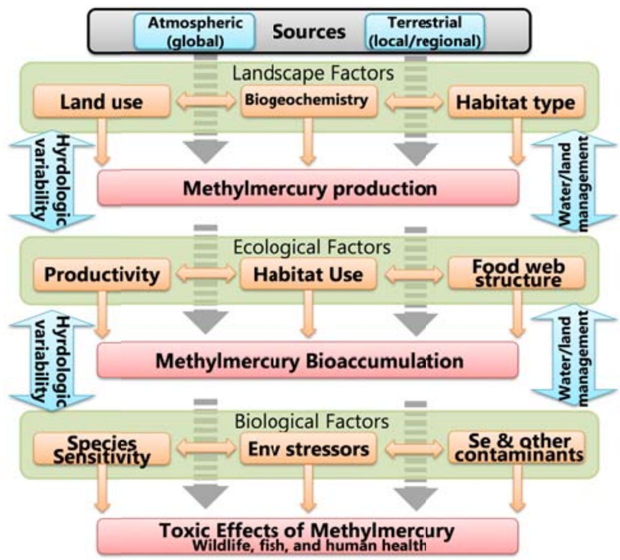

Figure 1. Conceptual model of the interactions among hierarchical factors that regulate environmental mercury cycling and risk.

The ecological risk of Hg toxicity is thus strongly related to MeHg production, which is controlled by multiple biogeochemical parameters that interact in complex ways with the landscape. Important landscape factors include land cover and use (Roué-LeGall et al. 2005), human and natural disturbances (UNEP 2008, Witt et al. 2009), habitat type (Heim et al. 2007, Ackerman and Eagles-Smith 2010), water inundation and associated wetting and drying cycles (Ricklbe et al. 2010), organic matter and nutrient cycling and inputs from upland terrestrial habitats (Chen et al. 2005, Rypel 2010). Once produced, MeHg bioaccumulation is driven by food web structure (Rolfhus et al. 2011), habitat use (Eagles-Smith et al. 2009), and population dynamics (Eagles-Smith et al. 2008). As MeHg bioaccumulates through the food web, its toxicity and impacts to ecological communities are driven by variation in sensitivity among species and lifestages, the simultaneous effects of other ecological stressors (e.g. predation pressure), and potential interactions with other contaminants (e.g. selenium). Additionally, climate variability and water availability are dynamic and critical drivers of the above landscape parameters. Thus, future changes in climate and precipitation dynamics could strongly influence the cycling, distribution, bioaccumulation, and effects of Hg through alterations in hydrology, land use, and habitat structure.

The western region of North America (Figure 2) is an expansive area with a diverse landscape, a diverse array of habitat types and land uses, and a broad range of climates. Compared to the central and eastern regions of North America, the western region may be disproportionally impacted by Hg because of the extensive mining legacy, the distribution pattern of local emitters, and increasing Asian Hg emissions. This combination of sources, habitat types, geography, and climates greatly complicate our understanding of Hg cycling and the bioaccumulation of MeHg across western landscapes, impeding management actions to mitigate the threats of Hg to western North American ecosystems. Importantly, public land comprises greater than 60% of the total land area in the west, making land management efforts to reduce Hg risk potentially more plausible than in central and eastern North America. Several decades of research and monitoring of Hg in western North America have provided an opportunity for regional synthesis of the existing data to facilitate a more quantitative understanding of Hg cycling, bioaccumulation, and ecological risk at landscape and regional scales. This is important because effective resource conservation related to Hg contamination will require information derived from integrated datasets across broad abiotic and biological compartments to predict (1) the cycling of Hg through the landscape under specific and changing climatic conditions, (2) the bioaccumulation of Hg within habitat-specific and regional food webs, and (3) spatiotemporal patterns in Hg risk to key indicator species.

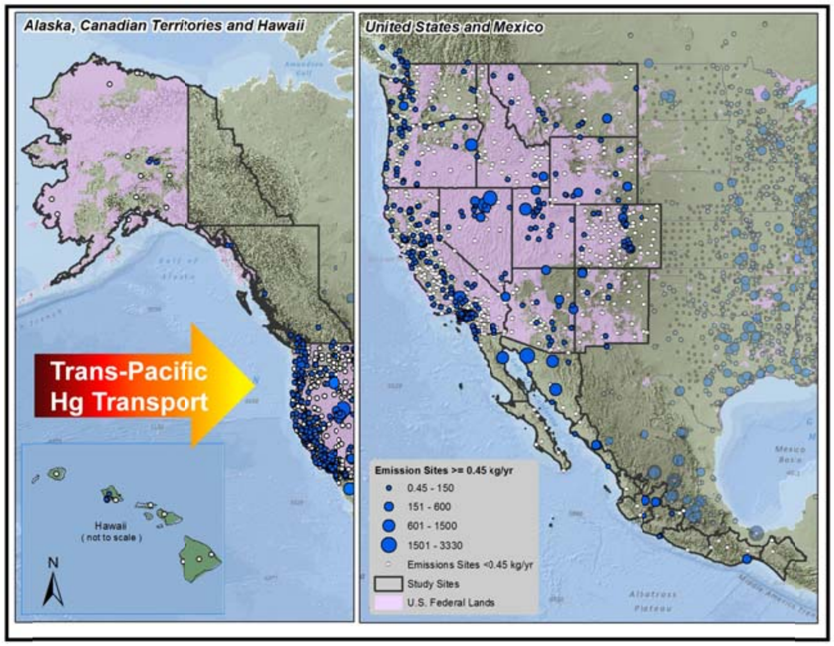

Figure 2. Map of synthesis region of Western North America. Dots represent known mercury sources and the size of the circle reflects total annual emissions (kg/year). Shaded region identifies US federally-owned and managed public lands within the region.

We propose to compile and combine existing datasets on Hg sources, biogeochemistry, bioaccumulation, ecological, and human-health effects across western North America, and to use a landscape scale approach to understand how Hg cycling varies across this region, and to examine how the factors that drive those changes vary geographically. We will place particular emphasis on land use relative to Hg cycling, as well as how the linkages between abiotic and biological processes interact to influence Hg biomagnification in food webs across the region.

This proposed project will benefit from the experience gained during two recent regional syntheses of mercury in North America, the first encompassing northeastern United States and eastern Canada (Evers and Clair 2005) and the second the Laurentian Great Lakes regions (Evers et al. 2011a, Wiener et al. 2012, http://www.briloon.org/mercuryconnections). In addition to compiling broad regional datasets from existing studies and monitoring programs, these two highly successful synthesis efforts also yielded a more holistic understanding of the unique relations among watershed processes, Hg bioaccumulation, and ecosystem risk for their respective regions (Evers et al. 2011b). They also highlighted regional differences in how Hg is mobilized and bioaccumulated in various ecosystem compartments, suggesting that a regional perspective can enhance development of management and policy strategies to reduce Hg risk.

The proposed western regional Hg synthesis will in many ways mirror the approaches used in the two previous synthesis efforts (e.g. quantifying spatial and temporal patterns in multiple abiotic and biotic compartments, determining ecological risk, and linking Hg dynamics with landscape processes at a regional scale), with some important new components. Specifically, we are synthesizing data across a much larger geographic area, allowing for a greater set of landscape scale questions to be addressed. Additionally, with the vast diversity of habitat and ecosystem types across western North America, as well as the high proportion of publicly owned and managed lands, our emphasis will be on habitat and landscape factors that control Hg cycling and bioaccumulation, as well as the land management options available to reduce MeHg exposure and risk. Finally, the density and distribution of current and historic mining in the west presents an opportunity to examine the relative importance of legacy mining sources versus global emissions sources to Hg cycling across the region.

Proposed Activities

General approach:
The Hg issue in western North America is multi-faceted, requiring interdisciplinary collaboration to address it in a holistic fashion. Our approach for this synthesis is to conduct detailed analyses within four subgroups, each compiling key related datasets across the region. The four subgroups include (1) Sources, (2) Biogeochemistry, (3) Fish and wildlife, and (4) Policy and human health. Each dataset will be analyzed within the subgroups to evaluate spatial and temporal variability of Hg in relation to a hierarchical set of geological, ecological, and climatological factors, as well as key questions pertinent to natural-resource management and policy. Overlapping components of each dataset will also be merged to provide linked analyses among the four groups.

To accomplish these goals, we will target the following key questions among the subgroups:

1. **1.** What are the spatial patterns of Hg contamination in atmospheric deposition, water, sediment, and aquatic and terrestrial biota in western North America and how do they overlap?
    a. Within this question, we will assess the quantitative relationship among these variables to guide an evaluation of the importance of source type related to spatial distribution of Hg in wildlife.

2. **2.** What are the temporal patterns (seasonal, annual, and decadal) of Hg contamination in wet and dry deposition, water, sediment, and wildlife in the western North America?
    a. Beyond simply describing the patterns within the datasets, we will evaluate them using a spatiotemporal approach within the context of changes in climate, Hg emissions, land use/cover, and policy.

3. **3.** What landscape, ecosystem, and human factors are associated with spatial and temporal patterns in concentrations of total Hg or MeHg in water, sediment, and wildlife in western North America?
    a. A specific goal within this question is to evaluate the relative effect of Hg source and habitat type on MeHg production and bioaccumulation.
    b. We will also examine habitat linkages and controls to the availability of Hg to the methylation process, the uptake of MeHg into the base of the food web, and the changes in pH and dissolved organic matter as the primary controls on Hg methylation and movement in each unique habitat of the west.

4. **4.** Which (and what fraction of) aquatic and terrestrial environments in western North America have concentrations of MeHg in food webs high enough to pose a risk to fish and wildlife in upper trophic levels or to humans who consume fish from these waters?
    a. We will use datasets of habitat-specific (riparian, inland wetland, coastal wetland, forest, etc.) aquatic and avian indicator species because not all species are found in all habitats.
    b. The identification of Hg-sensitive landscapes within the region is expected to be an important outcome of these analyses.

5. **5.** How are Hg cycling and distribution expected to change over the coming decades with anticipated changes in the timing, amount, and type of precipitation, as associated hydrologic changes in watersheds?

6. **6.** With the large proportion of publicly-owned land throughout the west (>60%), are there land management options that can be tested or employed to reduce Hg risk?
    a. Use patterns on federal lands include: grazing, agriculture, timber harvest, prescribed fires, fish stocking, mining, wildlife refuges, and reservoirs. Activities associated with these practices likely alter Hg dynamics, or the factors that control Hg dynamics. Thus, resource managers may have tools available to directly address Hg issues on public lands.

7. **7.** What are the policy options and implications for Hg management in western North America?

Compilation of Hg Data:

The extensive multi-media data to be used in this synthesis will be compiled from several Hg monitoring and research efforts that have been implemented across western North America during recent decades. Collectively, these datasets contain substantial information on Hg in abiotic and biotic matrices across multiple habitat types. Many of these datasets are publically available through State and Federal water quality agencies, whereas others are associated with more proprietary research efforts.

The following list exemplifies some of the more extensive datasets that will be included in the proposed regional synthesis effort.

a) State, provincial, and tribal fish-contaminant monitoring programs (>50,000 records)
b) U.S. National Park Service – Western Airborne Contaminants Assessment Project (WACAP; >2,000 records)
c) NOAA – Mussel Watch (1,000 records)
d) National Water Quality Monitoring Council (http://acwi.gov/monitoring)
e) USGS National Water-Quality Assessment Program (>5,000 records)
f) USGS Environmental Mercury Mapping, Modeling, and Analysis (>10,000 records)
g) US EPA sediment and water monitoring data (>10,000 records)
h) National Atmospheric Deposition Program, Mercury Deposition Network (>4,500 records)
i) US EPA assessment of Hg and Se in steam fish throughout the Western US (>3,000 records)
j) US Fish and Wildlife Service Environmental Contaminants Program Database (25,000 records)
k) USGS Contaminant Exposure and Effects Database (>700 records)

In total, we estimate at least 200,000 data records on Hg in air, water, sediment, and biota will be incorporated in this effort from these information sources, as well as more targeted research by project investigators and participants.

Addressing the key questions proposed, will require sophisticated spatial modeling that incorporates remote sensing, land use, and climatic data. We will use a hierarchical grid cell approach similar to the analyses conducted in the northeastern U.S. and Great Lakes regions (Evers et al. 2007, 2011b), using 30x30 minute grid cells that will serve as a basic scale for broad spatial assessments, but allowing for finer or coarser spatial resolution as required due to data density, landscape complexity, or the scale of the question. Temporal variability and time trends will be evaluated within a dynamic linear modeling framework that incorporates an explicit recognition of sequential structure in time series data. We will incorporate historical changes in Hg emissions, land cover, and climate from available data, including Landsat remote sensing imagery from NASA and USGS (1972-present; http://landsat.usgs.gov/), and GEOS-MERRA

(1979-present; http://gmao.gsfc.nasa.gov/research/merra/intro.php) and PRISM spatial climate products (1900-present; http://www.wcc.nrcs.usda.gov/climate/prism.html).

The evaluation of spatial and temporal variation in Hg in fish can be complicated by confounding factors, such as species and fish length, time of sampling, and type of water body. Given this, the statistical analyses of fish Hg data will use mixed-effects models to partition variance and covariance components among multiple levels, as exemplified by the analysis of Hg in game fish from lakes in the Great Lakes region (Monson et al. 2011).

Participants

The broad scope of this synthesis requires disciplinary experts with extensive experience integrating their Hg expertise across topical compartments. We have assembled a tri-national team of scientists and policy experts that meet these requirements and have strong publication records. Additionally, two of the Principal Investigators (D. Evers and J. Wiener), who previously led Hg syntheses in the Northeast and Great Lakes regions have substantial experience in organizing and coordinating efforts of this magnitude and scope.

Participant	Subgroup(s)	Expertise
[1†] **Collin Eagles-Smith**, U.S. Geological Survey, Corvallis, OR https://profile.usgs.gov/ceagles-smith	PI; Fish and Wildlife	Bioaccumulation in food webs; toxicological effects
[1] **David Evers**, Biodiversity Research Institute, Gorham, ME http://www.briloon.org/about-bri/the-people-of-bri/staff/leadership/david-evers	PI; Fish and Wildlife	Bioaccumulation and effects in birds
[1] **Mark Marvin-DiPasquale**, U.S. Geological Survey, Menlo Park, CA http://profile.usgs.gov/mmarvin	PI; Biogeochemistry	Microbial biogeochemistry and Hg methylation
[1] **Elsie Sunderland & graduate student**, Harvard University, Cambridge, MA http://www.people.fas.harvard.edu/~esunderl/ems/Home.html	PI; Biogeochemistry/ Sources	Modeling of Hg in ecosystems to assess risks to human health
[1] **James Wiener**, University of Wisconsin – La Crosse, La Crosse, WI	PI; Fish and Wildlife	Bioaccumulation and ecotoxicology of Hg in fish
[1] **George Aiken**, U.S. Geological Survey, Boulder, CO http://sofia.usgs.gov/people/aiken.html	Biogeochemistry	Organic matter geochemistry
[1] **Joel Blum**, University of Michigan, Ann Arbor, MI http://www.lsa.umich.edu/eeb/directory/faculty/jdblum/	Biogeochemistry/ Sources	Isotope geochemistry
[1] **Jacob Fleck**, U.S. Geological Survey, Sacramento, CA	Biogeochemistry	Hg hydrologic transport
[1] **David Krabbenhoft**, U.S. Geological Survey, Middleton, WI http://wi.water.usgs.gov/professional-pages/krabbenhoft.html	Biogeochemistry	MeHg production and biogeochemistry
[1] **Allison Luengen**, University of San Francisco, San Francisco, CA http://www.usfca.edu/facultydetails.aspx?id=4294969506	Biogeochemistry	Phytoplankton MeHg accumulation
[1] **Josh Ackerman**, U.S. Geological Survey, Davis, CA http://www.werc.usgs.gov/person.aspx?personid=40	Fish and Wildlife	Bioaccumulation and effects in birds

[1]**Allyson Jackson (graduate student)**, Oregon State University, Corvallis, OR	Fish and Wildlife	Hg bioaccumulation and effects in birds
[1]**Robin Stewart**, U.S. Geological Survey, Menlo Park, CA http://wwwrcamnl.wr.usgs.gov/tracel/people/robin_stewart.html	Fish and Wildlife/ Biogeochemistry	Bioaccumulation in aquatic food webs
[1*]**Madeline Turnquist**, Biodiversity Research Institute, Gorham, ME http://www.briloon.org/about-bri/the-people-of-bri/staff/associate-wildlife-research-biologists/madeline-turnquist	Fish and Wildlife	Hg bioaccumulation
[1]**Charles Alpers**, U.S. Geological Survey, Sacramento, CA https://profile.usgs.gov/cnalpers/	Sources/ Biogeochemistry	Hg transport and sources
[1]**Mae Gustin**, University of Nevada, Reno, NV http://www.ag.unr.edu/gustin/	Sources/ Biogeochemistry	Hg emission, geologic sources
[1]**Dan Jaffe**, University of Washington, Seattle, WA http://faculty.washington.edu/djaffe/	Sources	Hg modeling and atmospheric transport
[1]**Heather Morrison**, Environment Canada, Toronto, ON, CA	Policy	Hg modeling and policy
[1]**Gustavo Ochoa**, National Institute of Ecology, Mexico City, MX	Policy	Hg risk and policy
[1]**Leigh Woodruff**, U.S. EPA, Boise, ID	Policy	Hg transport, sources, and policy
[2]**Robert Gerlach**, Division of Environmental Health, Anchorage, AK http://dec.alaska.gov/eh/vet/	Policy	Hg policy
[2]**Kristi Morris**, National Park Service, Denver, CO	Policy	Hg deposition and policy
[2]**Dave Schmeltz**, U.S. EPA, Washington D.C.	Policy/Sources	Hg emissions, sources, and policy
[2]**Jay Davis**, San Francisco Estuary Institute, Richmond, CA http://www.sfei.org/user/9	Fish and Wildlife	Bioaccumulation in fish
[2]**Darell Slotten**, University of California, Davis, CA	Fish and Wildlife	Hg bioaccumulation in aquatic food webs
[2]**Chris Eckley**, U.S. EPA, Seattle, WA	Fish and Wildlife/ Sources	Sources and environmental transport
[2]**Angela Matz**, U.S Fish and Wildlife Service, Fairbanks, AK	Policy/Fish and Wildlife	Hg bioaccumulation
[2]**Phil Johnson**, U.S Fish and Wildlife Service, Anchorage, AK	Policy/Fish and Wildlife	Hg bioaccumulation

[1]Confirmed participant

[2]Data contributor and collaborator, who may or may not attend the workshops

*Technical liaison

[†]Responsible for meeting Data and Information Policy requirements

Timetable of Activities

To assess the feasibility of this synthesis effort, a scoping workshop with prospective key partners was held in October 2011. This effort brought the principal investigators and some participants together to assess interest and feasibility, to outline key questions and approaches, and to begin identifying available datasets. Much of the difficulty in conducting an effort that focuses on topics ranging from atmospheric Hg deposition to Hg bioaccumulation spanning the spatial and temporal scales described above lies in compiling an integrated database that will facilitate model development. Thus, we will focus considerable effort prior to our first group meeting at the Powell Center on database development and group correspondence to maximize the efficiency of our working sessions and refine our questions and approaches.

Feb – Aug 2012:	Secure complete suite of datasets from States, Provinces, Federal agencies, and, other project participants. Integrate datasets into a working database available to project participants. Establish initial goals within subgroups and ensure compatibility among subgroups. Conduct preliminary analyses of data.
Sept 2012:	First meeting – 3 days, plus 2 days for travel. Devote 1.5 days to subgroup meetings (breakout sessions) and 1.5 days to a full meeting with all participants. Discuss preliminary analyses, interpret preliminary findings and revise approaches as necessary. Expand analyses within and among subgroups and assign writing tasks.
Oct - June 2013:	Continue analyses of data and begin preparation of manuscripts. Communicate results and interpretation via email, and conduct conference calls, as needed, to resolve final analytical and interpretive details. Complete draft manuscripts and presentations.
July 2013:	Second meeting – 3days, plus 2 days for travel. Summarize progress with presentations to working groups, discuss manuscript drafts, begin revisions, and identify and begin conducting additional statistical analyses and modeling, as needed.
Aug - Jan 2014:	Complete draft manuscripts, circulate for colleague and institutional review, and submit for publication in refereed journals.

Anticipated Results and Benefits

As with the other regional Hg syntheses, we anticipate a series of papers targeted for a dedicated issue of a major international environmental health journal, such as *Environmental Science and Technology, Environmental Pollution*, or *Ecotoxicology*. We anticipate that the biogeochemistry, fish and wildlife, and sources subgroups will each produce two manuscripts on spatial and temporal patterns of Hg in the west, and on habitat/landscape factors in relation to Hg cycling. The policy subgroup will produce at least one manuscript on mercury, human health, and subsistence foods, and one on the implications of source regulation versus land management options. Finally, the group will product at least one integrated manuscript identifying factors that control Hg sources, MeHg production, and bioaccumulation. A spatially-explicit approach will be used to overlay areas of concern in order to preliminarily identify conservation priorities in relation to Hg risk. The results of this cumulative work will greatly assist resource managers and understand risks posed by Hg to their trust resources, as well as potential tools available to address that risk.

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