HHWP/Hetch Hetchy FY11/12 Proposal
Division of Resources Management and Science
Yosemite National Park

Mapping and Attributing Sources of Mercury Bioaccumulation in the Tuolumne River Watershed

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Abstract, Background, and Objectives

Abstract

Recent fish sampling from the Hetch Hetchy Reservoir, and Sequoia-Kings Canyon National Park, where atmospheric mercury deposition is the most likely source, has found fish containing mercury concentrations exceeding state and federal regulatory thresholds for fish consumption. Unfortunately, sample sizes and spatial extent for these studies were small (Davis et al. 2010). The SFPUC conducted additional fish sampling in Hetch Hetchy Reservoir in 2009, and the findings from this work are consistent with previous investigations (city SFPUC report – Hetch Hetchy Reservoir Fish Bioaccumulation Study, Preliminary Data Summary, October 2010, under internal review). Leveraging ongoing studies already investigating the effect of ozone on Hg deposition, we propose to quantify the degree of mercury contamination in representative natural lakes and three reservoirs, and identify most likely sources of that contamination. This information would help the SFPUC and park managers determine whether mitigation of sources of ozone and mercury precursors has potential to reduce mercury loading to some of the West's most valued and pristine watersheds.

Background/Introduction

Mercury (Hg) is a heavy metal that in its elemental form, Hg (0), is volatile at room temperature and is thus distributed globally via the atmospheric pool. Gold mining, coal burning, and other anthropogenic activities that expose and volatilize this metal to the atmosphere have likely tripled the amount of Hg in the atmosphere available for deposition to landscapes, though concentrations are still relatively low (typically 1-2 ng m^{-3}) compared to other pollutants (NAS 2009) At such low concentrations, Hg (0) is thus notoriously hard to measure and has little if any direct toxicity of Hg (0) to humans who inhale it. At higher air concentrations, inhalation (e.g., indoor spills) of Hg (0) can be significantly toxic to the nervous system, kidneys, and lungs.

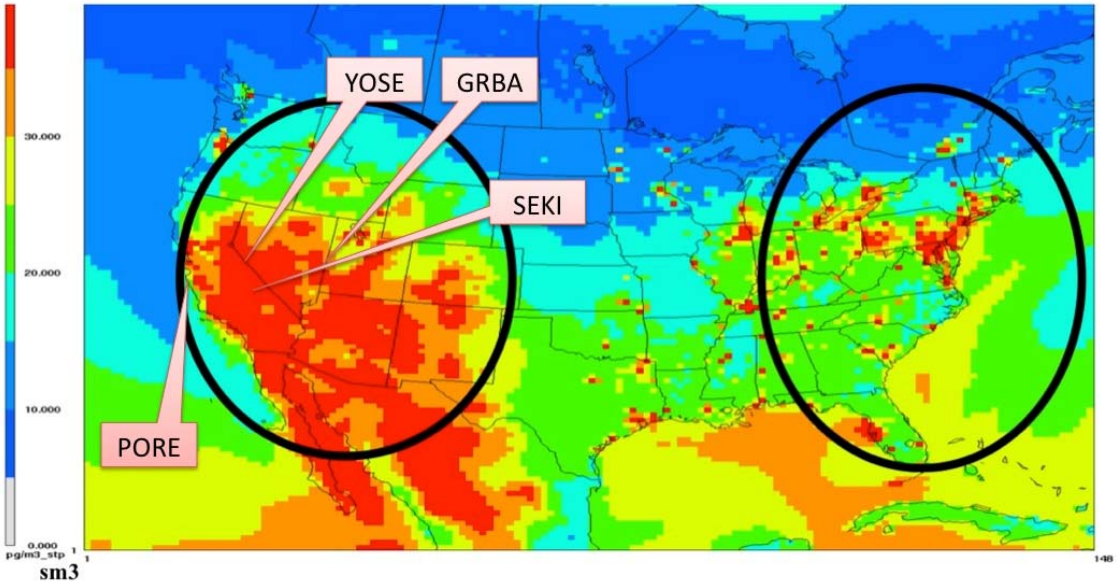
Despite its volatility and global ubiquity as an atmospheric pollutant, the greatest risk to humans posed by mercury is not from inhalation of Hg (0), but from the ingestion of fish or other high trophic-level organisms that have bioaccumulated the methylated form of mercury (MeHg) in aquatic environments (http://www.epa.gov/hg/). Though MeHg is most harmful when ingested and is, like Hg (0), a neurotoxin, it is also a teratogen, and has been linked to cardiovascular problems. Due to its tendency to bioaccumulate, its toxicity can be further enhanced for children who breastfeed from mothers with significant body MeHg body burdens (NAS 2009)

Oxidation of elemental Hg to divalent or reactive mercury, Hg(II), mediates the transfer from atmospheric Hg(0) to MeHg that accumulates in fish. In contrast with Hg(0), which is volatile and often bidirectional at ambient temperatures, Hg(II) deposition is less reversible, and facilitates net transfer to aquatic and terrestrial surfaces from the air. As a result, Hg(II) often comprises the bulk of the total mercury to be found in water and sediment associated with aquatic and lacustrine environments, and it is the primary mercury-containing precursor from which MeHg is formed (Wiener et al. 2006).

In the atmosphere, Hg (II) is thought to form when oxidants such as ozone are present. Hg from natural sources is emitted almost entirely in the Hg(0) form.
Some anthropogenic sources, however, directly emit the Hg (II) and particulate mercury (Hg(p)) forms. Rates of deposition for Hg(p) are a factor of 2 higher than Hg(0), and rates of deposition for RGM are up to 10 times higher than Hg(0), (Gustin et al. 1997, Zhang et al. 2009). In the eastern US, the resulting Hg(II) deposition occurs mostly as wet deposition (6:1 wet dry ratios were measured in the southeastern US) (Bullock et al. 2007, 2009, Prestbo and Gay 2009).

In the West however, most deposition occurs via dry depositionRegional atmospheric chemistry modeling suggests that deposition is elevated over vast swaths of Great Basin and Sierra Nevada landscapes (Gustin et al. 2003, Bullock et al. 2007). More recent modeling for the western continental US, constrained by observations at Mt. Bachelor in Oregon confirms that oxidation of Hg(0) to RGM and modeling based on those concentrations suggests that subsequent deposition is enhanced in the presence of elevated ozone, and that this enhancement is likely a primary cause of elevated RGM (and often total) deposition in the West (Weiss-Penzias et al. 2003, Swartzendruber et al. 2006, Bullock et al. 2007, Lindberg et al. 2007).

In Nevada's Great Basin, more recent observations have found that diurnal variations of elevated ozone correlate well with diurnal variation and elevation in RGM concentrations, and inversely with Hg(0) concentrations (Weiss-Penzias et al. 2009). These observations, combined with back-trajectory analysis, imply that oxidants at the mid-levels of the free troposphere could produce observed gaseous oxidized Hg. Enhanced deposition may therefore result. Nearby in the Sierra Nevada, ozone research and monitoring in YOSE (Burley and Ray 2007), and ozone monitoring in SEKI (www.nps.gov/air) indicated that some high elevation sites in YOSE and SEKI share exposure these free troposphere, oxidant-rich air masses. Ongoing ozone measurements in YOSE show the same diurnal variation and absolute ozone values as the Weiss-Penzias (2009) Reno site (Tarnay et al. unpublished data 2010, Weiss-Penzias et al. 2006a).



Map of proposed NAAMEX flight areas in the Western and Eastern US with locations of proposed parks (YOSE, SEKI, GRBA, PORE), adapted from http://research.uwb.edu/jaffegroup/publications/NAAMEX_Whitepaper_v14.pdf.

The map colors correspond to RGM at the surface calculated with the CMAQ model. High concentrations of RGM in the Western US are a result of tropospheric oxidation (in the model) and high RGM concentrations in the Eastern US are from direct industrial emissions (R.Bullock personal communication).

To verify that this regional RGM deposition pattern indeed occurs and is correlated to ozone, the NPS air resources division has funded researchers to deploy both active and passive measurements of speciated (i.e., Hg(0), RGM and Hg(p)) concentrations, as well as speciated Hg deposition to surrogate surfaces (Lyman et al. 2009, Lyman et al. 2010). This would occur at exposed sites transecting elevational gradients in YOSE, SEKI, and GRBA. Two additional coastal sites (including PORE) are also included in this research to quantify differences between relatively "pristine" air masses arriving on westerly winds off the Pacific and air masses influence by regional oxidants from inland sources. At these coastal sites, Asian sources may also be important contributors to deposition, and this work—the observations at these coastal sites would also characterize the importance of Asian sources for California and Nevada parks (Jaffe et al. 2005, Weiss-Penzias et al. 2006b, Strode et al. 2008). Finally, there is also potential for this work to coincide with yet-to-be funded continent-wide observations of speciated mercury oxidants targeted constraining higher resolution Hg chemistry and deposition models (NAAMEX 2010, http://research.uwb.edu/jaffegroup/modules/NAAMEX/)

Objectives

The objectives of this study are to:

- Expand on previous SFPUC work to quantify magnitude and extent of MeHg bioaccumulation at two trophic levels within lake, river and reservoir ecosystems (i.e., fish and zooplankton)

- Measure and quantify indicators of atmospheric deposition across time (i.e., sediment cores) to confirm source hypothesis
- Qualitatively identify the extent to which regional vs. global air pollutants may be contributing to bioaccumulation. This objective leverages ongoing Hg speciation and deposition measurements and uses relative proportion of RGM in deposition as indictor of a regional enhancement of deposition from the globally available elemental Hg pool.

This project would be a collaborative effort between NPS and SFPUC.

Methods

Overall Approach: The overall approach for this project is simply to:

- find where fish have bioaccumulated levels of Hg high enough to be of concern to those eating the fish and
- compare patterns of RGM deposition with concentrations of total Hg in fish, zooplankton, and sediment cores and look for correlations.

Three target species of fish (brown trout, brook trout, and rainbow trout), water chemistry measurements (dissolved oxygen, conductivity, pH, and temperature) and duplicate water samples will be collected from Kibbie Lake, Vernon Lake, Wilma Lake, Tilden Lake, Benson Lake, McCabe Lake, Dog Lake, Spillway Lake, and Evelyn Lake, Cherry Reservoir and Eleanor Reservoir, and a section of the Tuolumne River below the O'Shaughnessy Dam. Fish samples from Hetch Hetchy were collected and analyzed for Hg in 2009; additional fish samples will not be recollected from this reservoir.

In addition, triplicate sediment cores and duplicate zooplankton samples will be collected from Dog Lake, Kibbie Lake, and Benson Lake, and Cherry Reservoir, Eleanor Reservoir, and Hetch Hetchy Reservoir. Sampling will occur between August 16 and September 20, 2011. Sites will be accessed via foot, and gear and samples will be transported via pack stock to all sites except Dog Lake.

Brown Trout (BR)	Brook Trout (BK)	Rainbow Trout (RN)
Hetch Hetchy Reservoir		Hetch Hetchy Reservoir
Cherry Reservoir (15)		Cherry Reservoir (15)
		Eleanor Reservoir (15)
Below O'Shaughnessy (15)		Below O'Shaughnessy (15)
Upper McCabe (15-30)		Upper McCabe Lake (15-30)
Spillway Lake (15-30)	Spillway Lake (15-30)	Spillway Lake (15-30)
	Dog Lake (15-30)	Dog Lake (15-30)
	Benson Lake (15-30)	Benson Lake (15-30)
	Kibbie Lake (15-30)	Kibbie Lake (15-30)
	Evelyn Lake (15-30)	Evelyn Lake (15-30)

		Vernon Lake (15-30)
		Wilma Lake (15-30)
		Tilden Lake (15-30)

Sampling locations were chosen to: 1) provide a representative sample across the Tuolumne River watershed's elevational gradient and to be geographically distributed throughout the watershed; 2) determine the concentrations of Hg in representative samples of brook, brown, and rainbow trout species. Species presence was determined using stocking records and survey data; 3) target lakes with the greatest likelihood of high MeHg loads due to proximity to wetlands, water depth, and exposure to high levels of ozone; and 4) accessible via trail. Mercury analysis of the fish, zooplankton, sediment cores and water samples would be conducted by SFPUC. Sediment cores will be analyzed for mercury. If we are able to collect adequate samples and laboratory analysis determines there is a mercury deposition gradient, then the sediment cores would be radiodated by a contracted laboratory.

We will sample the target species during a 24 hour period using gill nets set out from the shore and in the deepest part of the lake. A map of net placement and approximate depth will be constructed with a reference numbers for each net. Individual fish will be removed from the nets and they will be rinsed in ambient water to remove any foreign material from the external surface. The specimens will then be grouped by species and general size class and placed on clean aluminum foil to prevent contamination. All fish will be inspected carefully to ensure that their skin and fins have not been damaged by the sampling equipment, and damaged specimens will be discarded. Total body length (the length from the anterior-most part of the fish to the tip of the longest caudal fin ray when the lobes of the caudal fin are compressed dorsoventrally) will be measured in mm.

Data will be recorded into field notebooks including: date, time, crew initials, lake name, species, total length, net reference number, water temperature, air temperature, gear type and a unique number identifying the sample throughout field sampling, storage, transport, laboratory processing and analysis procedures. Sample ID will include: First 4 letters of lake name-2 letter species code-sample #-net #-date and a matching label will be made out of write in the rain paper to include in with the fish.

> KIBB-RN-03-05-080811 indicates that the fish is the third rainbow trout collected from net number 5 on Kibbie Lake August 8, 2011

Based on the catch, three size-classes will be calculated for each target species caught in an individual lake, and a minimum of 15 samples will be collected to best represent that size-class distribution.

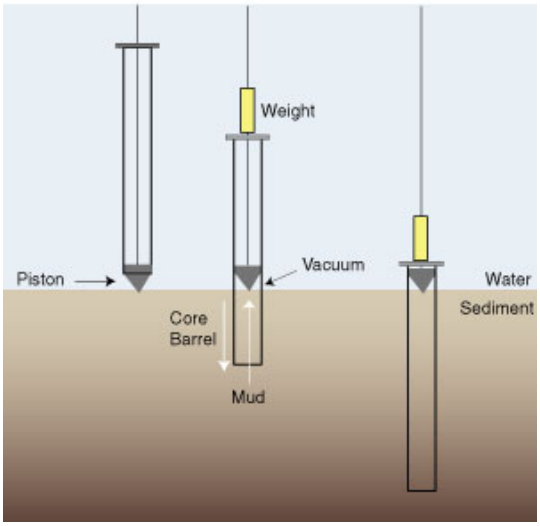
After initial processing, each fish will be individually wrapped in extra heavy duty aluminum foil. The sample identification label will be taped to the outside of each aluminum foil package; each individual fish will be placed into a waterproof plastic bag and sealed. All of the packaged individual specimens will be placed in one large waterproof plastic bag in the same shipping container on dry ice for transport. Samples

will be frozen during transport and will be kept frozen in the lab until analysis. An SFPUC chain of custody document will follow each sample throughout the process to insure the integrity of the sample and provide tracking through the SFPUC Laboratory Information Management System (LIMS).

Sample preparation will take place in the laboratory. Sex will be determined through internal examination of the gonads and will be recorded. A skinless 4cm x 4cm plug of tissue will be removed from the fish for mercury analysis. Otoliths and scales will be collected for aging the fish. The remaining fish will be rewrapped in aluminum foil and will be kept frozen.

A gravity corer will be used to collect a 20 cm x 5 cm core of sediments from each lake. The corer will be lowered into the sediments from the rubber raft. The raft will be anchored at three points to prevent boat movement during core collection. The piston coring head will be lowered into the lake using the extensions until just above the bottom. The weight of the corer drives the barrel into the sediment and then the messenger is sent down the line to close the stop-ball to form the vacuum. After collection, the sediment core will be lifted into the boat.

The corer is lowered to just above the sediment-water interface, with the piston at the bottom of the core barrel. The piston is then fixed in one place by a cable while a weight pushes the core barrel into the sediment. Because the core barrel is moving, but the piston isn't, a vacuum develops between the piston and the mud. This vacuum allows the mud to rise easily into the core barrel despite the friction which would otherwise cause the mud to drag along the sides of the barrel.



Once the cores have been collected they will be brought to shore and extruded and sectioned in one centimeter increments into small Ziploc bags. Each sample will be labeled similar to that described in the fish section.

> KIBB-Sedi-02-03-080811 indicates that the sample was the second sample, third section from the top collected from Kibbie Lake August 8, 2011

Samples will be kept on ice during transport to the trailhead. All sampling equipment will be cleaned with laboratory grade detergent and rinsed with de-ionized water between each site. All sampling equipment will be acid washed before each day's use.

Duplicate water samples will be collected from just below the surface of each lake. At Kibbie, Dog, and Benson, samples will be collected from the epilimnion and hypolimnion near the thermocline and near the bottom will be collected from each intensive survey lake. Discrete samples will be collected using an all Teflon Kemmerer that has been acid washed and rinsed with de-ionized (DI) water prior to each day's use. The equipment will be stored in cleaned containers to avoid contamination and clean handling (clean hands / dirty hands) techniques used between sampling areas. Samples will be transferred from the Kemmerer directly into pre-cleaned certified 250 ml glass amber bottles and double bagged prior to storage. All data, LIMS tracking labels, and chain of custody procedures will be followed. Field/travel blanks will be used throughout each step of the sampling process to test for the possibility of contamination. Field blanks, prepared with ultra-pure deionized water prior to deployment, will follow the field sampling effort each day. Each sample will be labeled similar to that described in the fish section.

> KIBB-Water-02-080811 indicates that the sample was the second sample collected from Kibbie Lake August 8, 2011

Water chemistry parameters (conductivity, dissolved oxygen, pH, and temperature) and depth will be measured using a Hydrolab multiprobe. Measurements will be taken in the epilimnion and hypolimnion. The depth to thermocline will be used to determine where zooplankton vertical tows will commence for sampling from the intensive lakes.

Duplicate zooplankton samples will be collected using a 243 µ mesh plankton net with a 300mm opening. Most lakes have one large basin with a zone of deoxygenated water in summer. This lake type should be sampled as follows. Determine the depth at which dissolved oxygen is less than 0.5 ppm. This depth will hereafter be referred to as the "critical depth" because zooplankton will not occur below it. Retrieval of the plankton net should begin at that depth. Other lakes are so shallow that dissolved oxygen occurs throughout the water column and there is no critical depth.

As a precaution, tie net, bucket, and brass stopper together with a safety string to prevent loss of parts. Lower the net very slowly and carefully for the last 3 feet. Upon reaching the proper depth, pause for at least 30 seconds and then raise the net at a rate of approximately 4 feet per second. A hand reel with revolving handles on both sides will greatly facilitate smooth, uninterrupted retrieval. Raise the net out of the water in one motion until the plankton bucket is just above the surface. While hanging on to the net with one hand, splash lake water on the outside of the net to dislodge plankton that may still adhere to the inside of the net. After washing all plankton into the bucket, detach the bucket and wash down its sides with a 125-ml wash bottle. Remove stopper, and allow sample to drain into a Ziploc bag while washing the inside of the bucket with a squeeze

bottle. Samples will be frozen in the field and kept frozen until analysis. Each sample will be labeled similar to that described in the fish section.

> KIBB-Zoo-02-080811 indicates that the sample was the second sample collected from Kibbie Lake August 8, 2011

Four atmospheric monitoring stations would be set up within the Tuolumne River watershed. Stations would be set up at the O'Shaughnessy Dam, above Yosemite Creek at Road Marker T14, Siesta Lake, and Tioga Pass entrance station. These sites were chosen to optimize our ability to distinguish between regional and global Hg sources. Passive and surrogate air samplers would be deployed for one year to measure Hg air concentrations and dry deposition, respectively. The Tekran 2537A/1130/1135 system would be deployed for one month to measure continuous speciated air Hg concentrations. The atmospheric monitoring portion of this study would be conducted through a cooperative agreement with the University of Nevada, Reno and would be covered by a separate research permit (YOSE-2011-SCI-0044)

Statement of Work

A. The NPS would:
   1) Obtain research permits, complete NEPA compliance, and complete MRA
   2) Identify and collect sediment cores, zooplankton, and fish samples from 10 lakes and river sections within Yosemite for total mercury analysis
   3) Develop and administer a cooperative agreement with the University of Nevada, Reno for conducting the atmospheric monitoring component of this study including supplying and maintaining equipment, collecting and analyzing data, and providing a final report
   4) Develop and administer a contract with an outside laboratory to analyses and radiodate the sediments
   5) Oversee a literature review related to mercury contamination and reservoir management
   6) Perform data analysis and draft interim and final reports in collaboration with SFPUC
   7) Collaborate with SFPUC to ensure that the project is completed in a timely manner

B. The SFPUC would:
   1) Collect sediment cores, water, zooplankton, and fish samples from Cherry and Eleanor Reservoirs and sediment cores from Hetch Hetchy Reservoir for total mercury analysis
   2) Assist with collection and transport of samples from natural lakes and river sections
   3) Prepare and analyze water, zooplankton and fish samples for total mercury
   4) Perform data analysis and draft interim and final reports in collaboration with NPS
   5) Collaborate with NPS to ensure that the project is completed in a timely manner

Schedule/Milestones/Dates

Phase 1 (March 1, 2011 to June 30, 2011)

Obtain research permits, identify sampling sites, complete NEPA compliance, and complete MRA

Phase 2 (July 1, 2011 to Aug 31, 2011)

Deploy passive and surrogate air samplers to measure Hg air concentrations and dry deposition, respectively, over one year period of time
Deploy Tekran 2537A/1130/1135 system to measure continuous speciated air Hg concentrations for one month
Collect water, fish, zooplankton samples and sediment cores

Phase 3 (Sep 1, 2011 to Nov 30, 2011)

Continue to collect water, fish, zooplankton samples and sediment cores
Analyze water, fish, zooplankton samples, and sediment cores for total mercury concentrations and radiodate core samples

Phase 4 (Dec 1, 2011 to June 30, 2012)
Complete data analysis of water samples, fish, zooplankton, and sediment cores
Conduct literature review
Prepare interim report for submittal June 30, 2012

Phase 5 (July 1, 2012 to Nov 30, 2012)
Complete data analysis from atmospheric monitoring
Complete and submit final report Nov 30, 2012

Project Products and Deliverable

- Quantification of the magnitude and extent of Hg accumulation in fish and zooplankton
- Quantification of indicators of Hg deposition across time (sediment cores)
- Quantification of current rates of Hg deposition, and qualitative description of the extent to which global versus regional sources are contributing to deposition
- Literature review related to mercury contamination and reservoir management
- A final report (November 2012)

Budget

Section 1: NPS Personal Services			
Position Title & Grade (& employee name if known)	**Rate**	**X PPs**	**Total Cost**
Aquatic Ecologist GS11	$3,360	4	$13,440
Biological Science Technician GS05 (2)	$3,500	3	$10,500
Biological Science Technician GS07	$2,460	3	$7,380
Interpretive Ranger GS09	$2,588	1	$2,588
RMS Technical Support	$1,566	Lump	$1,566
Section 1 Subtotal			**$35,474**

Section 2: Contract Services		
Firm	**Service Description**	**Total Cost**
Contracted Lab (TBD)	Sediment Radiodating	$21,000
University of Nevada, Reno	Mercury Deposition Monitoring & Analysis	$55,000
Graduate Student Institution (TBD)	Literature Review, Data Synthesis	$20,000
Section 2 Subtotal		**$96,000**

Section 3: Equipment, Materials, and Other Costs			
Equipment/Materials	**Cost**	**X Units**	**Total Cost**
Vehicle Costs	$1,000	3	$3,000
Equipment and Supplies	$3,000	1	$3,000
Travel	$1,000	1	$1,000
Section 3 Subtotal			**$7,000**
Subtotal of Sections 1, 2, & 3			$138,474
5% of Sections 1, 2, 3 Subtotal for NPS Administrative Costs			$6,924
Total Funding Requested (subtotal + 5%)			**$145,398**

NPS In Kind Support	FY11	FY12	Total
Leland Tarnay, GS-11 Physical Scientist, 2pp/year, YOSE	$6,745.00	$6,865.00	$13,610.00
Katy Warner, GS-7 Physical Science	$7,365.00		$10,299.00
Mae Gustin, University of Nevada, Reno (investigating mercury deposition and sources of Hg in Yosemite and three other western parks)	$52,494.00		$52,494.00
Ozone monitoring in Yosemite (collocated with and leveraged for RGM measurements and source apportionment)	$14,376.00		$14,376.00
TOTAL CONTRIBUTED	**$80,980.00**	**$6,865.00**	**$87,845.00**

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