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## State Water Resources Control Board

August 7, 2017

Thomas Mumley, Ph.D.  
Assistant Executive Officer  
San Francisco Bay Regional  
Water Quality Control Board  
1515 Clay Street, Suite 1400  
Oakland, CA 94612

**SUBJECT: REQUEST FOR EXTERNAL PEER REVIEW OF THE SCIENTIFIC BASIS OF THE PROPOSED PLAN AMENDMENT TO ESTABLISH THE STATEWIDE IMPLEMENTATION PROGRAM FOR MERCURY IN RESERVOIRS**

Dear Dr. Mumley,

This letter responds to the attached April 10, 2017 request for external scientific peer review for the subject noted above. The review process is described below. All steps were conducted in confidence. Reviewers' identities were not disclosed.

To begin the process for selecting reviewers, I contacted the University of California, Berkeley (University) and requested recommendations for candidates considered qualified to perform the assignment. This service is supported through an Interagency Agreement co-signed by CalEPA and the University. The University was provided with the request letter and attachments. No additional material was asked for. The University interviews each promising candidate.

Each candidate who was both qualified and available for the review period was asked to complete a Conflict of Interest (COI) Disclosure form and send it to me for review, with Curriculum Vitae. The cover letter for the COI form describes the context for COI concerns that must be taken into consideration when completing the form. "As noted, staff will use this information to evaluate whether a reasonable member of the public would have a serious concern about [the candidate's] ability to provide a neutral and objective review of the work product."

In subsequent letters to candidates approved as reviewers, I provided the attached January 7, 2009 Supplement to the CalEPA Peer Review Guidelines, which, in part, serves two purposes: a) it provides guidance to ensure confidentiality through the course of the external review, and b) it notes reviewers are under no obligation to discuss their comments with third-parties after reviews have been submitted. We recommend they do not. All outside parties are provided opportunities to address a proposed regulatory action, or potential basis for such, through a well-defined rulemaking process.

Later, I sent letters to reviewers to initiate the review. These letters provided access instructions to a secure FTP site where all material to be reviewed was placed, including a Table of Contents for them. Attachment 2 to the request memorandum was highlighted as the focus for the review. Each reviewer was asked to address each topic, as expertise allows, in the order given. Five weeks were provided for the review. I also asked reviewers to direct enquiring third-parties to me after they have submitted their reviews.

Reviewers' names, affiliations, curriculum vitae, initiating letters and reviews are being sent to you now with this letter.

Approved reviewers:

- 1) Janina Benoit, Ph.D.  
Professor and Chair of Chemistry  
Wheaton College  
26 East Main Street  
Norton, MA 02766  
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- 2) Cynthia C. Gilmour, Ph.D.  
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- 3) Daniel A. Jaffe, Ph.D.  
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- 4) Robert P. Mason, Ph.D.  
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If you have any questions, or require clarification from the reviewers, please contact me directly.

Regards,



Gerald W. Bowes, Ph.D.  
Manager, Cal/EPA Scientific Peer Review Program  
Office of Research, Planning and Performance  
State Water Resources Control Board  
1001 "I" Street, 16<sup>th</sup> Floor  
Sacramento, California 95814

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FAX: (916) 341-5284

Email: [GBowes@waterboards.ca.gov](mailto:GBowes@waterboards.ca.gov)

Attachments:

- (1) April 10, 2017 Request by Thomas Mumley for Scientific Peer Review
- (2) Letters to Reviewers Initiating the Review
  - (1) Janina Benoit, Ph.D.
  - (2) Cynthia C. Gilmour, Ph.D.
  - (3) Daniel A. Jaffe, Ph.D.
  - (4) Robert P. Mason, Ph.D.
- (3) January 7, 2009 Supplement to Cal/EPA Peer Review Guidelines
- (4) Curriculum Vitae
  - (1) Janina Benoit, Ph.D.
  - (2) Cynthia C. Gilmour, Ph.D.
  - (3) Daniel A. Jaffe, Ph.D.
  - (4) Robert P. Mason, Ph.D.
- (5) Reviews
  - (1) Janina Benoit, Ph.D.
  - (2) Cynthia C. Gilmour, Ph.D.
  - (3) Daniel A. Jaffe, Ph.D.
  - (4) Robert P. Mason, Ph.D.

cc: Karen Larsen, Deputy Director, Division of Water Quality, State Water Board  
[Karen.Larsen@waterboards.ca.gov](mailto:Karen.Larsen@waterboards.ca.gov)

Rik Rasmussen, Manager, Water Quality Standards and Assessment, State Water Board  
[Rik.Rasmussen@waterboards.ca.gov](mailto:Rik.Rasmussen@waterboards.ca.gov)

Stacy Gillespie, Office of Chief Counsel, State Water Board  
[Stacy.Gillespie@waterboards.ca.gov](mailto:Stacy.Gillespie@waterboards.ca.gov)



EDMUND G. BROWN JR.  
GOVERNOR

MATTHEW RODRIGUEZ  
SECRETARY FOR  
ENVIRONMENTAL PROTECTION

## State Water Resources Control Board

**TO:** Gerald W. Bowes, Ph.D.  
Manager, Cal/EPA Scientific Peer Review Program  
Office of Research, Planning, & Performance  
State Water Resources Control Board

A handwritten signature in blue ink, appearing to read "T. Mumley".

**FROM:** Thomas Mumley, Ph.D.  
Assistant Executive Officer  
San Francisco Bay Regional Water Quality Control Board

**DATE:** April 10, 2017

**SUBJECT: REQUEST FOR EXTERNAL SCIENTIFIC PEER REVIEW OF DRAFT PROPOSED RULE FOR THE MERCURY RESERVOIR PROVISIONS TO ESTABLISH A MERCURY TMDL AND IMPLEMENTATION PROGRAM FOR RESERVOIRS**

Pursuant to the requirements of California Health and Safety Code Section 57004, State Water Resources Control Board (State Water Board) staff request by transmittal of this memorandum that you identify and assign reviewers to provide external scientific peer review of proposed "Amendment to the Water Quality Control Plan for Inland Surface Waters, Enclosed Bays, and Estuaries of California—Mercury TMDL and Implementation Program for Reservoirs" (Mercury Reservoir Provisions). The scientific basis of the Mercury Reservoir Provisions is contained in the "Draft Staff Report for Scientific Peer Review for the Amendment to the Water Quality Control Plan for Inland Surface Waters, Enclosed Bays, and Estuaries of California, Mercury Reservoir Provisions— Mercury TMDL and Implementation Program for Reservoirs" (Staff Report).

Attachment 1 summarizes the Mercury Reservoir Provisions. Attachment 2 describes the scientific conclusions to be evaluated by the scientific peer reviewers. Attachment 3 lists the individuals involved in developing the Mercury Reservoir Provisions and Staff Report. Attachment 4 list references for the Staff Report.

Additionally we request that the scientific peer reviewers evaluate conclusions 17 and 18 of Attachment 2. While these conclusions are not part of the proposed rule and are not subject to the statutory mandate of Health and Safety Code Section 57004, we are seeking scientific review of a weight of the evidence method that would be employed to assess compliance with mercury water quality objectives. The basis for these conclusions is in the Staff Report.

FELICIA MARCUS, CHAIR | THOMAS HOWARD, EXECUTIVE DIRECTOR

1001 I Street, Sacramento, CA 95814 | Mailing Address: P.O. Box 100, Sacramento, Ca 95812-0100 | [www.waterboards.ca.gov](http://www.waterboards.ca.gov)

**Expected Dates of State Water Board Hearing**

We anticipate that the State Water Board will consider adoption of the Mercury Reservoir Provisions and Staff Report in 2018.

**Expected Date the Documents will be Available for Review**

April 11, 2017

**Length of Documents for Review**

The Mercury Reservoir Provisions are approximately 50 pages (which include about 20 pages of text and 30 pages of tables), and the Staff Report is approximately 660 pages (not including figures and tables). The references cited in the documents for review will be made available to the assigned reviewers.

**Suggested Areas of Expertise for Reviewers**

The Staff Report is comprehensive and encompasses numerous disciplines. We recommend that the scientific peer reviewers have expertise in (a) mercury cycling and controls in reservoirs and lakes, (b) mercury fate and transport including biochemistry, geochemistry, and aquatic chemistry, and (c) biostatistics. Each of the conclusions will require all of the above expertise.

**Additional Information**

To assist in the selection of reviewers, the following information is provided as attachments to this memorandum:

1. Summary of the Mercury Reservoir Provisions
2. List of focused scientific conclusions for the peer reviewers to evaluate
3. List of scientists involved in development of the Mercury Reservoir Provisions and Staff Report
4. List of references

**Contact Information**

The staff contact for the statewide mercury control program for reservoirs is Carrie Austin, who can be reached at (510) 622-1015 or via e-mail at [carrie.austin@waterboards.ca.gov](mailto:carrie.austin@waterboards.ca.gov). Please feel free to call me or Carrie if you have any questions about this request. Thank you for your assistance.

cc: Karen Larsen, Deputy Director, Division of Water Quality, State Water Board  
Rik Rasmussen, Manager, Water Quality Standards and Assessment, State Water Board  
Stacy Gillespie, Office of Chief Counsel, State Water Board

**Attachment 1**  
**Summary of the**  
**Mercury Reservoir Provisions**

The proposed rule is “Amendment to the Water Quality Control Plan for Inland Surface Waters, Enclosed Bays, and Estuaries of California—Mercury TMDL and Implementation Program for Reservoirs” (Mercury Reservoir Provisions). The purpose of the Mercury Reservoir Provisions is to guide management actions that result in levels of mercury in reservoir fish that are safe to eat by humans and wildlife.

The Mercury Reservoir Provisions include: 1) a total maximum daily load (TMDL) for mercury-impaired reservoirs, 2) an implementation program for achieving TMDL targets in mercury-impaired reservoirs, and 3) an implementation program for managing discharges of mercury-contaminated sediments from dredged or fill materials in watersheds of all reservoirs.

The TMDL will apply to reservoirs that do not meet water quality standards due to elevated levels of methylmercury in fish tissue affecting the commercial and sport fishing (COMM), wildlife habitat (WILD), and/or rare, threatened, or endangered species (RARE) beneficial uses. The analysis provided in the draft Staff Report being submitted to scientific peer review is based on 74 reservoirs that were identified as impaired by mercury on the 2010 Clean Water Act Section 303(d) List. The final Mercury Reservoir Provisions and Staff Report will include additional reservoirs identified as impaired by mercury on the 2016 303(d) List, and possibly additional reservoirs identified in Staff Report as impaired by mercury using a method described in the Staff Report (Appendix L, Assessment of Compliance with the Proposed Water Quality Objectives) to determine whether a water quality standard is attained based on weight of evidence of available data and information.

The TMDL, developed in accordance with Clean Water Act Section 303(d)(1)(A), includes numeric targets for methylmercury concentrations in reservoir fish to protect the health of humans and wildlife that consume reservoir fish. The TMDL targets are designed to protect the health of people who consume fish at a recreational (i.e., not subsistence) level and wildlife that consume fish. The three TMDL targets are equal to the respective mercury water quality objectives for sport fish, prey fish, and prey fish for California least tern.

The TMDL includes load allocations that apply to mercury discharges from mine sites, runoff from non-urbanized areas, atmospheric deposition, and in-reservoir production of methylmercury. TMDL waste load allocations apply to mercury discharges from municipal and industrial wastewater treatment facilities. These allocations apply to mercury sources discharging to and upstream of the mercury-impaired reservoirs.

The Mercury Reservoir Provisions implementation program for achieving TMDL targets includes requirements for the control of mercury discharges from mercury, gold, and silver mine sites, and from urban storm water runoff in areas with historic mines. The Mercury Reservoir Provisions would require nonpoint sources to control discharges of inorganic mercury by minimizing the erosion of mining waste and mercury-contaminated sediments in the watersheds. Controlling mercury discharges from the mine sites is initially focused on those sites that are close to the mercury-impaired reservoirs and are highly erosive and discharging highly contaminated sediment. In addition, the Mercury Reservoir Provisions require erosion and sediment control practices to minimize discharges of mercury for activities that disturb mercury-contaminated soils in areas with historic mines.

## Attachment 1 - Summary of the Mercury Reservoir Provisions

Most NPDES permitted municipal and industrial wastewater treatment facilities that discharge to reservoirs or reservoir tributaries would be required to maintain their current wastewater treatment efficiencies, but some may need to improve their treatment efficiencies to attain their waste load allocation.

The Mercury Reservoir Provisions include a phased implementation program for reservoir pilot tests. During the first phase, expected to last ten years, reservoir owners and operators would evaluate management practices to reduce in-reservoir methylmercury production and bioaccumulation (i.e., develop and implement work plans for reservoir pilot tests, and provide report on reservoir water quality and fisheries management practices). After the first phase, the State Water Board will conduct a program review of the Mercury Reservoir Provisions and evaluate the reservoir management pilot test results to determine second phase implementation requirements.

In addition to the TMDL implementation program, the Mercury Reservoir Provisions includes requirements applicable to all reservoirs statewide for managing discharges of mercury-contaminated sediments from dredged or fill materials in watersheds that are downstream of mercury, gold, and silver mine sites.

**Attachment 2**  
**Description of Scientific Conclusions**  
**to be Evaluated by Scientific Peer Reviewers**

**A. Conclusions to be Evaluated by Scientific Peer Review**

The statutory mandate for external scientific peer review (California Health and Safety Code Section 57004) states that the reviewers are to provide a written evaluation of the scientific basis of the proposed rule.

Accordingly, we request that the scientific peer reviewers prepare the evaluation for each of the following conclusions that constitute the scientific basis of the proposed rule. The proposed rule is “Amendment to the Water Quality Control Plan for Inland Surface Waters, Enclosed Bays, and Estuaries of California—Mercury TMDL and Implementation Program for Reservoirs” (Mercury Reservoir Provisions). Explanatory statements are provided for each conclusion to help focus the review. The following numbered conclusions are based on information provided in the “Draft Staff Report for Scientific Peer Review for the Amendment to the Water Quality Control Plan for Inland Surface Waters, Enclosed Bays, and Estuaries of California, Mercury Reservoir Provisions— Mercury TMDL and Implementation Program for Reservoirs” (Staff Report).

Additionally we request that the scientific peer reviewers evaluate conclusions 17 and 18 of Attachment 2. While these conclusions are not part of the proposed rule and are not subject the statutory mandate of Health and Safety Code Section 57004, Water Board staff is seeking scientific review of a weight of the evidence method that would be employed to assess compliance with mercury water quality objectives. The basis for these conclusions is in the Staff Report.

Conceptual Model for Mercury Methylation and Bioaccumulation

**1. Many factors—not just the amount of inorganic mercury in water and sediment— influence methylmercury concentrations in reservoir fish.**

The conceptual model describes the mercury methylation process and subsequent bioaccumulation of methylmercury, and it identifies factors that affect both. Many factors influence methylmercury concentrations in reservoir fish because there are many successive steps in mercury cycling, from mercury sources to methylation to bioaccumulation in fish. The conceptual model also identifies specific effects reservoirs have on mercury methylation and bioaccumulation.

These factors that affect methylation and bioaccumulation are the scientific foundation of the California-specific linkage analysis (see next section).

Location of relevant information for review: Review should focus on Staff Report Chapter 4 (Conceptual Model: The Mercury Cycle and Bioaccumulation) and Staff Report Appendix A (Importance of Primary and Secondary Production in Controlling Fish Methylmercury Concentrations).

California-Specific Linkage Analysis

**2. The three most important factors that control fish methylmercury concentrations in California reservoirs are: the ratio of aqueous methylmercury concentration to chlorophyll-a concentration, aqueous total mercury concentration, and annual reservoir water level fluctuations.**



- 3. Inorganic mercury sources alone are not the primary driver of mercury impairments in California reservoirs. Multiple factors drive reservoir fish methylmercury levels: amount of mercury, methylmercury production, and bioaccumulation.**
- 4. Inorganic mercury levels in many reservoirs would need to be lower than natural background to achieve the TMDL targets<sup>1</sup> and mercury water quality objectives if no other factors are addressed.**

A linkage analysis was developed to establish the quantitative relationships between fish methylmercury concentrations and the environmental factors that control methylmercury production, bioaccumulation, and biomagnification in California reservoirs. The linkage analysis assessed more than 70 factors identified by the conceptual model and includes statistical analyses and model development based on data collected from California reservoirs.

The linkage analysis concludes there is no single limiting factor that controls fish methylmercury concentrations. The ratio of aqueous methylmercury to chlorophyll-a explains 52 percent of the variability in fish methylmercury concentrations. Reservoir sediment mercury has the second strongest correlation and explains 24 percent of the variability. When multiple factors are considered, the combination of three factors—aqueous total mercury concentration, the ratio of aqueous methylmercury concentration to chlorophyll-a concentration, and annual reservoir water level fluctuations—explain more variability in fish methylmercury concentrations than any other combination of factors. Together these three factors explain greater than 85 percent of variability in reservoir fish methylmercury concentrations.

Location of relevant information for review: To address conclusions 2 and 3, drivers of mercury impairment in reservoirs and factors that control fish methylmercury concentrations are discussed in Staff Report Chapter 2 (TMDL Targets), Chapter 5 (Linkage Analysis), Appendix A (Importance of Primary and Secondary Production in Controlling Fish Methylmercury Concentrations) and Appendix B (Methods to Develop Statistical Models to Explain and Predict Methylmercury Concentrations in Predatory Fish in California Reservoirs). To address conclusion 4, Chapter 2 (TMDL Targets) discusses methylmercury concentrations in fish tissue. The TMDL targets are used in the linkage analysis (Staff Report Chapter 5) to derive an aqueous methylmercury goal. The aqueous methylmercury goal, in turn, guides the development of a TMDL allocation and implementation requirements for in-reservoir methylmercury production (Staff Report Chapters 7, 8, and 9).

#### Mercury Source Assessment

- 5. Mercury sources are not evenly distributed across the State and no one source type is responsible for all reservoir impairments.**

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<sup>1</sup> Note: the proposed TMDL targets are equal to the proposed mercury water quality objectives (see Draft Part 1 of the *Water Quality Control Plan for Inland Surface Waters, Enclosed Bays, and Estuaries of California—Tribal and Subsistence Fishing Beneficial Uses, Mercury Water Quality Objectives, and Program of Implementation*). The State Water Board received external scientific peer review comments on the mercury water quality objectives on September 28, 2016.

- 6. The most important anthropogenic sources to impaired reservoirs are historic mine sites and atmospheric deposition from global and local (California) industrial emissions.**
- 7. Reducing watershed mercury sources is not expected to result in substantial reductions in reservoir sediment mercury concentrations and fish methylmercury concentrations in many reservoirs.**
- 8. Global industrial emissions are the predominant anthropogenic source to about 20 percent of mercury-impaired reservoirs.**

The source assessment provides an inventory and description of inorganic mercury sources. In addition, the source assessment identifies current source discharge concentrations, with a focus on sources that have particularly elevated mercury concentrations and are substantial contributors of mercury to reservoirs. The source assessment incorporates a concentration-based approach supported by the conceptual model and linkage analysis. Coincidentally, a concentration-based approach also better enables us to evaluate the feasibility of source reductions for many of the sources.

The assessment determined that mercury sources are not evenly distributed across the state. Some impaired reservoirs and their watersheds are dominated by mercury mining sources, while others are dominated by gold mining sources, atmospheric deposition, or watershed soils. The majority of California's urban areas are downstream of reservoirs. Hence, urban runoff and wastewater facility discharges are sources to only a couple of the impaired reservoirs.

Also, many reservoirs do not have sediment mercury concentrations elevated above modern background levels, even though some of these reservoirs are downstream of historic mines. This indicates that sediment mercury concentrations in these reservoirs are dominated by naturally-occurring mercury in watershed soils and mercury from atmospheric deposition.

Location of relevant information for review: Review of conclusions 5 through 8 should focus on Staff Report Chapter 6 (Source Assessment) and Appendices C through G. Specifically, sources of mercury are discussed throughout Chapter 6. Historic mines and atmospheric deposition are discussed in Chapter 6 sections 6.3 and 6.4, respectively, and in Appendices C (Mines Data) and D (Description of REMSAD Atmospheric Mercury Deposition Model and Emission Inventories). Conclusion 7 is discussed in Chapter 6 section 6.2 and 6.8. Conclusion 8 is within Chapter 6 sections 6.4 and 6.8 and in Appendix D (Description of REMSAD Atmospheric Mercury Deposition Model and Emission Inventories).

#### Potentially Controllable Processes and Predictions for Improvement

- 9. There are a variety of mercury source control options and reservoir water chemistry and fisheries management practices that may be effective for reducing fish methylmercury concentrations.**
- 10. A combination of source control actions and reservoir and fish management practices—versus source control alone—will be needed to achieve both timely and measurable fish methylmercury reductions in most of California's mercury impaired reservoirs.**
- 11. Actions to reduce fish methylmercury levels may need to vary for each reservoir because of the many combinations of different mercury sources (e.g., some are natural or global and therefore not regulated by state and federal agencies),**

**competing factors that control methylmercury production, and reservoir operational constraints. Reservoir-specific characteristics and operational requirements and mandates may not allow for all methylmercury management tools to be used in all reservoirs. Even so, there should be a possible solution to mercury impairment for every reservoir.**

The large number of factors that control mercury methylation and bioaccumulation complicates resolving the mercury impairment in California reservoirs. However, the large number of factors also increases the number of possible tools that may be available to reduce reservoir methylmercury levels. There are a variety of mercury source control options and reservoir water chemistry and fisheries management practices that may be effective for reducing fish methylmercury concentrations. The assessment of potentially controllable processes in the Staff Report provides examples of actions that can reduce fish methylmercury concentrations along with predictions for their effectiveness in mercury-impaired reservoirs in California.

The assessment then uses these predictions, along with key conclusions of the conceptual model, linkage, and source assessment chapters, to predict fish methylmercury reductions. The assessment conclusions and predictions help determine where and how relatively quick improvements may be possible, and guide the development of TMDL allocations and implementation requirements that will effectively reduce fish methylmercury concentrations in California reservoirs.

Location of relevant information for review: Review of conclusions 9, 10, and 11 should focus on Staff Report Chapter 7 (Assessment of Allocation and Implementation Options) and Appendix H (Supporting Information for the Assessment of Allocation and Implementation Options).

#### TMDL and Load Allocations

**12. The TMDL loading capacity and allocations, combined with reservoir water chemistry and fisheries management pilot tests and implementation actions identified in the proposed program of implementation), are adequate to achieve the proposed mercury water quality objectives and TMDL numeric targets for methylmercury in reservoir fish.**

**13. The allocations are adequate for both current and future mercury sources to the mercury-impaired reservoirs.**

U.S. EPA regulations allow TMDLs and allocations to be expressed as loads, concentrations, or other appropriate measure. A TMDL establishes the allowable loadings or other quantifiable parameters for a water body. Based on the findings and conclusions of the assessments described above, we propose load allocations for nonpoint sources (a–c) and waste load allocations for point sources (d) as follows:

- (a) Total mercury concentrations of suspended sediments (i.e., particulate mercury) in runoff from mine sites, mining waste downstream of mine sites, and non-urbanized upland areas (watershed soils). The load allocations correspond to the mercury regions in California described in the source assessment, as follows:
  - i. Mercury mineralized zone (400 mg/kg [dry weight, annual median]);
  - ii. Mercury-enriched areas (0.3 mg/kg [dry weight, annual median]); and
  - iii. Trace mercury areas (0.1 mg/kg [dry weight, annual median]).

- (b) Annual mercury mass deposited onto California for atmospheric deposition, including urban and nonurban areas. The total mercury load allocations for atmospheric deposition are:
- i. 1,400 kg/yr for deposition from natural sources;
  - ii. 230 kg/yr for deposition from anthropogenic sources within California; and
  - iii. 1,600 kg/yr for anthropogenic sources outside of California.
- (c) Methylmercury concentration in reservoir water for in-reservoir methylmercury production. The load allocation for methylmercury in reservoir water is no detectable methylmercury (annual geometric mean) in the water column with a detection limit not exceeding 0.009 ng/L. This detection limit is analytically achievable using U.S. EPA Method 1630 if a laboratory uses extra attentiveness in sample handling, reagent selection, and equipment preparation.
- (d) Total mercury concentrations in effluent for wastewater facility mercury discharges. Waste load allocations are assigned based on facility type and whether the facility design discharge flow is large or small compared to other reservoir inputs.

The load and waste load allocations for mercury sources were developed to reduce the inputs of mercury to reservoirs caused by anthropogenic activities by feasible means. The load allocation for in-reservoir methylmercury production was developed to direct implementation of management practices for reservoir water chemistry (i.e., minimize transformation of mercury to methylmercury) and bioaccumulation.

The load and waste load allocations are calculated in a way that enables them to be applied to current and future sources of mercury to the 74 reservoirs identified on the 2010 Clean Water Act Section 303(d) List as impaired by mercury. In addition, the allocations will apply to:

- Any new point source and nonpoint source discharges to the 74 reservoirs; and
- Point source and nonpoint source discharges to reservoirs identified in the future as having fish with methylmercury levels exceeding the water quality objectives.

Location of relevant information for review: TMDL load allocations for nonpoint sources (suspended sediment from mine sites, atmospheric deposition, and methylmercury concentration in reservoir water) are described in Staff Report Chapter 8 (Allocations, TMDL, and Loading Capacity) and Mercury Reservoir Provisions Chapter IV.C and Provisions Table 4 (item nos. 1 through 5 and 7). Waste load allocations for point sources are described in Staff Report Chapter 8 (Allocations, TMDL, and Loading Capacity) and Mercury Reservoir Provisions Chapter IV.C and Provisions Table 4 (item no. 5) and Table 5.

#### Margin of Safety, Seasonal Variations, and Critical Conditions

##### **14. The Reservoir Mercury TMDL incorporates an adequate margin of safety.**

The Staff Report describes an implicit margin of safety involves using conservative assumptions (more likely to be over-protective than under-protective) throughout the analysis for developing load and waste load allocations.

Water Board staff recommended mercury allocations for watershed and global sources. However, the linkage analysis and other assessments indicate that source control alone is insufficient to achieve the mercury water quality objectives for fish methylmercury in all mercury-

impaired reservoirs. As a result, the Mercury Reservoir Provisions contain a methylmercury allocation of non-detect for reservoir water and requirements for the development of potential water chemistry and fisheries management practices to reduce methylmercury bioaccumulation in the reservoir food web in addition to load and waste load allocations to mercury discharges in the watershed. The combination of allocations assigned to in-reservoir, watershed, and global mercury sources, plus the development and implementation water chemistry and fishery management practices, provide multiple methods to address fish mercury contamination; this combination of methods provides an implicit margin of safety.

In addition, federal regulations and U.S. EPA guidance direct TMDLs to account for seasonal variations and critical conditions. This program accounts for seasonal and inter-annual variations in inorganic mercury loads and concentrations and the critical condition of anoxia needed for methylmercury production.

Location of relevant information for review: Margin of Safety is discussed within Staff Report Chapter 8 section 8.3 (Loading Capacity, TMDL, and Allocations, Margin of Safety), Chapter 7 section 7.8 (Assessment of Allocation and Implementation Options), and Appendix H (Supporting Information for the Assessment of Allocation and Implementation Options).

#### Implementation and Monitoring

- 15. The Mercury Reservoir Provisions requirements for inorganic mercury controls are adequate to reduce anthropogenic discharges of inorganic mercury to reservoirs.**
- 16. During the first phase of the implementation program for the impaired reservoirs, the Mercury Reservoir Provisions require reservoir water chemistry and fisheries management practices pilot tests. Implementing reservoir pilot tests to develop and evaluate and water chemistry and fisheries management practices in a phased approach is an adequate approach to reduce reservoir fish methylmercury levels. This phased approach includes State Water Board review of the Mercury Reservoir Provisions prior to full scale implementation of effective and feasible management practices.**

A portion of the Mercury Reservoir Provisions relies on the control of inorganic mercury discharges to the reservoirs. Mercury from these sources is predominately attached to sediment; therefore, control of mercury-contaminated sediment will reduce inorganic mercury loading to the reservoir. The Mercury Reservoir Provisions include requirements for the control of mercury discharges from mercury, gold, and silver mine sites. The Mercury Reservoir Provisions would require nonpoint sources to control discharges of inorganic mercury by minimizing the erosion of mining waste and mercury-contaminated sediments in the watersheds. Controlling mercury discharges from the mine sites is initially focused on those sites that are close to the mercury-impaired reservoirs and are highly erosive and discharging highly contaminated sediment. In addition, the Mercury Reservoir Provisions require erosion and sediment control practices to minimize discharges of mercury for projects that disturb soils in areas with historic mines.

Since most NPDES permitted municipal and industrial wastewater treatment facilities are not major discharges of mercury to the reservoirs, most NPDES permitted municipal and industrial wastewater treatment facilities would be required to maintain their current wastewater treatment efficiencies. A few facilities may need to improve their treatment efficiencies to attain their waste load allocation.

As previously noted, a combination of source control actions and reservoir and fish management practices—versus source control alone—will be needed to achieve both timely and measurable fish methylmercury reductions in most of California’s mercury impaired reservoirs.

Many potential reservoir water chemistry and fisheries management practices described in the Staff Report have been employed elsewhere in the world to reduce methylmercury levels in fish but not in California, or if in California, not for the purpose of reducing reservoir fish methylmercury concentrations.

Consequently, the Mercury Reservoir Provisions includes a phased approach for impaired reservoirs. During the first phase, expected to last ten years, reservoir owners and operators would develop and evaluate management practices to reduce in-reservoir methylmercury production and bioaccumulation (i.e., pilot tests, work plans, and reports of reservoir water quality and fisheries management practices). The water chemistry and fisheries management pilot tests can be conducted in a coordinated manner in representative reservoirs. The Staff Report contains recommendations for pilot tests and monitoring efforts.

After the first phase, the State Water Board will conduct a program review of the Mercury Reservoir Provisions and evaluate the pilot test final reports concerning the reduction of fish methylmercury levels. In the second phase, the State Water Board would require each owner and operator to implement effective, long-term reservoir water quality and fisheries management practices and continued cleanup of mine sites.

Location of relevant information for review: To address Conclusion 15, the discussion of inorganic mercury controls is in Staff Report Chapter 7 (Discussion of Assessment of Allocation and Implementation Options), Appendix H (Supporting Information for the Assessment of Allocation and Implementation), Chapter 9 (Implementation Plan), Appendix I (Supporting Information for Implementation Plan), and Chapter 10 (Water Quality Objectives Assessment). Requirements for the reduction of anthropogenic discharges of inorganic mercury to reservoirs are in Mercury Reservoir Provisions Chapter IV.F, Chapter V, and Table 4.

To address Conclusion 16, the Staff Report discusses reservoir pilot tests and associated studies throughout Chapter 9 (Implementation), Chapter 10 (Monitoring), and Appendix L (Assessment of Compliance with the Proposed Water Quality Objectives). Requirements for pilot tests for water chemistry and fisheries management practices are in Mercury Reservoir Provisions Chapters IV.B, Chapter IV.F, Chapter V, and Chapter VI.

#### Assessment of Compliance with the Proposed Water Quality Objectives

- 17. The upper 90th confidence limit of the mean is an appropriate statistical method to determine compliance with water quality objectives based on an annual average fish methylmercury concentration. In addition, it is appropriate to use consistent fish trophic levels and sizes, sample type and quantity, and sampling locations when determining compliance with water quality objectives.**

The Staff Report describes a method that can be considered for assessing attainment with the proposed mercury water quality objectives. The “Water Quality Control Policy for Developing California’s Clean Water Act Section 303(d) List” (SWRCB 2015) (Listing Policy) prescribes the

factors for adding or removing a water body from the Clean Water Act Section 303(d) list of impaired waters. One factor, the “Situation-Specific Weight of the Evidence Listing Factor” may be used to list a water segment as not meeting a water quality standard (i.e., impaired) if other Listing Policy factors do not result in the listing of the water segment but information demonstrates nonattainment. Likewise, the Listing Policy contains a “Situation-Specific Weight of Evidence Delisting Factor” to be used to delist a water segment when all other delisting factors do not result in the delisting of a water segment but information indicates attainment of a water quality standard.

The proposed mercury water quality objectives (see footnote 1) are based on annual average fish tissue mercury concentrations. The Staff Report evaluates various methods to assess compliance with these long term average objectives, including the binomial distribution (specified in the Listing Policy) the arithmetic mean.

The Staff Report describes a weight of the evidence method that could be used to satisfy the situation specific weight of the evidence factors for listing and delisting. This method is based on statistical averaging using the upper 90th confidence limit of the mean with a minimum data set of nine. The Staff Report also proposes guidance for consistent sample collection, including fish trophic levels and sizes, sample type and quantity, and sampling locations when using a situation specific weight of the evidence factors for listing and delisting.

Location of relevant information for review: Review should focus on Chapter 10 (Water Quality Objectives Assessment) and Appendix L (Assessment of Compliance with the Proposed Water Quality Objectives).

**18. Biosentinel fish monitoring provides a means to evaluate relatively rapid changes to biotic methylmercury levels.**

For the reservoir pilot tests, the Staff Report recommends that the initial fish monitoring to evaluate the effectiveness of a management practice being tested use “biosentinel” fish rather than larger sport fish to determine whether there are statistically significant differences in fish methylmercury levels. Biosentinel fish are young (up to 1-year-old) prey fish with high site fidelity. The rationale for biosentinel fish is that they provide more precise measurements of bioaccumulation than the larger sport fish, because biosentinels accumulate all their methylmercury during the test period whereas sport fish have accumulated methylmercury over several years and not only during the test period. The biosentinel fish are slightly different than the TMDL targets (e.g., narrower length range). Subsequent fish monitoring would include the larger sport fish to assess compliance with the TMDL fish tissue targets.

Location of relevant information for review: Fish monitoring recommendations for fisheries management actions are discussed in the Staff Report Chapter 9 (Implementation Plan) section 9.8.6.

**B. The Big Picture**

Reviewers are not limited to addressing only the specific conclusions presented above in Section A, and are asked to contemplate a broader perspective.

- (a) In reading the Staff Report and Mercury Reservoir Provisions, are there any additional scientific conclusions that are part of the scientific basis of the proposed rule not

## Attachment 2 - Scientific Conclusions to be Evaluated by Scientific Peer Reviewers

described above in Section A? If so, please comment on these with respect to the statute language given above.

- (b) Taken as a whole, is the scientific portion of the proposed rule based upon sound scientific knowledge, methods, and practices?

Reviewers also should note that some proposed actions may rely significantly on professional judgment where available scientific data are not as extensive as desired to support the statute requirement for absolute scientific rigor. In these situations, the proposed course of action is favored over no action.

The preceding guidance will ensure that reviewers have an opportunity to comment on all aspects of the scientific portion and basis of the proposed State Water Board action. At the same time, reviewers also should recognize that the State Water Board has a legal obligation to consider and respond to all feedback on the scientific portions of the proposed rule. Because of this obligation, reviewers are encouraged to focus feedback on the scientific issues that are relevant to the central regulatory elements being proposed in the Mercury Reservoir Provisions.



### Attachment 3

#### **Individuals Involved in Development of the Amendment to the Water Quality Control Plan for Inland Surface Waters, Enclosed Bays, and Estuaries of California, Mercury Reservoir Provisions— Mercury TMDL and Implementation Program for Reservoirs**

Health and Safety Code Section 57004 requires all Cal/EPA organizations to submit for external scientific review the scientific portion and basis of all proposed policies, plans and regulations. Additionally, Health and Safety Code Section 57004, subdivision (c), states, “No person may serve as an external scientific peer reviewer for the scientific portion of a rule if that person participated in the development of the scientific portion and basis of the rule.”

Accordingly, the following is an accounting of external participants who were involved with development of the Mercury Reservoir Provisions and Staff Report. The first five scientists provided informal review comments for an earlier version of Staff Report Appendix A, Importance of Primary and Secondary Production in Controlling Fish Tissue Mercury Concentrations. They did not review any other portion of the Staff Report. The sixth scientist (Willits) was a paid consultant to review the statistical analysis in Chapter 5, Linkage. There were no other external participants.

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## State Water Resources Control Board

June 13, 2017

Janina Benoit, Ph.D.  
Professor and Chair of Chemistry  
Wheaton College  
26 East Main Street  
Norton, MA 02766

**SUBJECT: REQUEST FOR EXTERNAL PEER REVIEW OF THE SCIENTIFIC BASIS OF THE PROPOSED PLAN AMENDMENT TO ESTABLISH THE STATEWIDE IMPLEMENTATION PROGRAM FOR MERCURY IN RESERVOIRS**

Dear Professor Benoit,

The purpose of this letter is to initiate the external peer review.

The State Water Resources Control Board will receive reviewers' comments and curriculum vitae from me after the review has concluded, and not be a party to the process.

Documents for review are being provided through a secure FTP site. Sections I and II below give instructions for accessing the FTP site and list the documents on the site.

You can access this site through the five week period of review. The URL, username and password are as follows:

I. <https://ftp.waterboards.ca.gov/>

User name: PRFTP  
Password: Water123

II. **List of Documents at FTP site:**

- A. April 10, 2017 memorandum signed by Thomas Mumley, Ph.D: **“Request for External Scientific Peer Review of Draft Proposed Rule for the Mercury Reservoir Provisions to Establish a Mercury TMDL and Implementation Program for Reservoirs**

**Attachment 1:** Summary of the Mercury Reservoir Provisions

**Attachment 2:** Conclusions to be Evaluated by Scientific Peer Reviewers. These 18 conclusions are the focus for the review. As expertise allows, comment on topics in the order listed.

**Attachment 3:** Individuals Involved in the Development of the Amendment to the Water Quality Control Plan for Inland Surface Waters, Enclosed Bays, and Estuaries of California, Mercury Reservoir Provisions – Mercury TMDL and Implementation Program for Reservoirs

**Attachment 4:** List of References

- B. **Staff Report for Scientific Peer Review**
- C. **Staff Report Appendices**
- D. **Staff Report References**
- E. **Staff Report Tables**
- F. **Staff Report Figures**
- G. **January 7, 2009 Supplement to the CalEPA Peer Review Guidelines.** This Supplement provides guidance to ensure the review is kept confidential through its course. The Supplement notes reviewers are under no obligation to discuss their comments with third-parties after reviews have been submitted. We recommend they do not. All outside parties are provided opportunities to address a proposed regulatory action through a well-defined regulatory process. Please direct third parties to me.

**Please send your reviews to me on July 20, 2017 to ensure I receive all on the same day.**

Questions about the review should be for clarification, in writing – email is fine, and addressed to me. My responses will be in writing also. I subsequently will forward all reviews together to Thomas Mumley with reviewers' Curriculum Vitae. All this information will be posted at the State and Regional Water Boards' Scientific Peer Review website.

Your acceptance of this review assignment is most appreciated.

Sincerely,



Gerald W. Bowes, Ph.D.  
Manager, Cal/EPA Scientific Peer Review Program  
Office of Research, Planning and Performance  
State Water Resources Control Board  
1001 "I" Street, MS-16B  
Sacramento, California 95814

Telephone: (916) 341-5567

Email: [gerald.bowes@waterboards.ca.gov](mailto:gerald.bowes@waterboards.ca.gov)



## State Water Resources Control Board

June 13, 2017

Cynthia C. Gilmour, Ph.D.  
Senior Scientist  
Smithsonian Environmental Research Center  
647 Contees Wharf Road  
Edgewater, MD 21037

**SUBJECT: REQUEST FOR EXTERNAL PEER REVIEW OF THE SCIENTIFIC BASIS OF THE PROPOSED PLAN AMENDMENT TO ESTABLISH THE STATEWIDE IMPLEMENTATION PROGRAM FOR MERCURY IN RESERVOIRS**

Dear Dr. Gilmour,

The purpose of this letter is to initiate the external peer review.

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Manager, Cal/EPA Scientific Peer Review Program  
Office of Research, Planning and Performance  
State Water Resources Control Board  
1001 "I" Street, MS-16B  
Sacramento, California 95814

Telephone: (916) 341-5567

Email: [gerald.bowes@waterboards.ca.gov](mailto:gerald.bowes@waterboards.ca.gov)

## State Water Resources Control Board

June 13, 2017

Daniel A. Jaffe, Ph.D.  
Professor, Atmospheric Sciences  
University of Washington-Bothell  
18115 Campus Way NE  
Bothell, WA 98011-8246

**SUBJECT: REQUEST FOR EXTERNAL PEER REVIEW OF THE SCIENTIFIC BASIS OF THE PROPOSED PLAN AMENDMENT TO ESTABLISH THE STATEWIDE IMPLEMENTATION PROGRAM FOR MERCURY IN RESERVOIRS**

Dear Professor Jaffe,

The purpose of this letter is to initiate the external peer review.

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Gerald W. Bowes, Ph.D.  
Manager, Cal/EPA Scientific Peer Review Program  
Office of Research, Planning and Performance  
State Water Resources Control Board  
1001 "I" Street, MS-16B  
Sacramento, California 95814

Telephone: (916) 341-5567

Email: [gerald.bowes@waterboards.ca.gov](mailto:gerald.bowes@waterboards.ca.gov)

## State Water Resources Control Board

June 13, 2017

Robert P. Mason, Ph.D.  
Professor  
Department of Marine Science  
University of Connecticut  
1080 Shennecossett Road  
Groton, CT 06340

**SUBJECT: REQUEST FOR EXTERNAL PEER REVIEW OF THE SCIENTIFIC BASIS OF THE PROPOSED PLAN AMENDMENT TO ESTABLISH THE STATEWIDE IMPLEMENTATION PROGRAM FOR MERCURY IN RESERVOIRS**

Dear Professor Mason,

The purpose of this letter is to initiate the external peer review.

The State Water Resources Control Board will receive reviewers' comments and curriculum vitae from me after the review has concluded, and not be a party to the process.

Documents for review are being provided through a secure FTP site. Sections I and II below give instructions for accessing the FTP site and list the documents on the site.

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Manager, Cal/EPA Scientific Peer Review Program  
Office of Research, Planning and Performance  
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Sacramento, California 95814

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Email: [gerald.bowes@waterboards.ca.gov](mailto:gerald.bowes@waterboards.ca.gov)

**Supplement to Cal/EPA External Scientific Peer Review Guidelines –  
“Exhibit F” in Cal/EPA Interagency Agreement with University of California  
Gerald W. Bowes, Ph.D.**

**Guidance to Staff:**

1. Revisions. If you have revised any part of the initial request, please stamp “Revised” on each page where a change has been made, and the date of the change. Clearly describe the revision in the cover letter to reviewers, which transmits the material to be reviewed. The approved reviewers have seen your original request letter and attachments during the solicitation process, and must be made aware of changes.
2. Documents requiring review. All important scientific underpinnings of a proposed science-based rule must be submitted for external peer review. The underpinnings would include all publications (including conference proceedings), reports, and raw data upon which the proposal is based. If there is a question about the value of a particular document, or parts of a document, I should be contacted.
3. Documents not requiring review. The Cal/EPA External Peer Review Guidelines note that there are circumstances where external peer review of supporting scientific documents is not required. An example would be "A particular work product that has been peer reviewed with a known record by a recognized expert or expert body." I would treat this allowance with caution. If you have any doubt about the quality of such external review, or of the reviewers' independence and objectivity, that work product – which could be a component of the proposal - should be provided to the reviewers.
4. Implementation review. Publications which have a solid peer review record, such as a US EPA Criteria document, do not always include an implementation strategy. The Cal/EPA Guidelines require that the implementation of the scientific components of a proposal, or other initiative, must be submitted for external review.
5. Identity of external reviewers. External reviewers should not be informed about the identity of other external reviewers. Our goal has always been to solicit truly independent comments from each reviewer. Allowing the reviewers to know the identity of others sets up the potential for discussions between them that could devalue the independence of the reviews.
6. Panel Formation. Formation of reviewer panels is not appropriate. Panels can take on the appearance of scientific advisory committees and the external reviewers identified through the Cal/EPA process are not to be used as scientific advisors.
7. Conference calls with reviewers. Conference calls with one or more reviewers can be interpreted as seeking collaborative scientific input instead of critical review. Conference calls with reviewers are not allowed.

## Guidance to Reviewers from Staff:

### 1. Discussion of review.

Reviewers are not allowed to discuss the proposal with individuals who participated in development of the proposal. These individuals are listed in Attachment 3 of the review request.

Discussions between staff and reviewers are not permitted. Reviewers may request clarification of certain aspects of the review process or the documents sent to them.

Clarification questions and responses must be in writing. Clarification questions about reviewers' comments by staff and others affiliated with the organization requesting the review, and the responses to them, also must be in writing. These communications will become part of the administrative record.

The organization requesting independent review should be careful that organization-reviewer communications do not become collaboration, or are perceived by others to have become so. The reviewers are not technical advisors. As such, they would be considered participants in the development of the proposal, and would not be considered by the University of California as external reviewers for future revisions of this or related proposals. The statute requiring external review of science-based rules proposed by Cal/EPA organizations prohibits participants serving as peer reviewers..

### 2. Disclosure of reviewer Identity and release of review comments.

Confidentiality begins at the point a potential candidate is contacted by the University of California. Candidates who agree to complete the conflict of interest disclosure form should keep this matter confidential, and should not inform others about their possible role as reviewer.

Reviewer identity may be kept confidential until review comments are received by the organization that requested the review. After the comments are received, reviewer identity and comments must be made available to anyone requesting them.

Reviewers are under no obligation to disclose their identity to anyone enquiring. It is recommended reviewers keep their role confidential until after their reviews have been submitted.

### 3. Requests to reviewers by third parties to discuss comments.

After they have submitted their reviews, reviewers may be approached by third parties representing special interests, the press, or by colleagues. Reviewers are under no obligation to discuss their comments with them, and we recommend that they do not.

All outside parties are provided an opportunity to address a proposed regulatory action during the public comment period and at the Cal/EPA organization meeting where the proposal is considered for adoption. Discussions outside these provided avenues for comment could seriously impede the orderly process for vetting the proposal under consideration.



4. Reviewer contact information.

The reviewer's name and professional affiliation should accompany each review. Home address and other personal contact information are considered confidential and should not be part of the comment submittal.

**Janina M. Benoit**  
**Chemistry Department, Wheaton College**  
**26 East Main Street**  
**Norton, MA 02766**  
**Phone: (508) 286-3966 Email: jbenoit@wheatonma.edu**

**Professional Employment:**

2012-present Professor, Chemistry Department, Wheaton College, Norton MA  
2007-2012 Associate Professor, Chemistry Department, Wheaton College, Norton, MA  
2001-2007 Assistant Professor, Chemistry Department, Wheaton College, Norton, MA  
2000-2001 Clayton Fellow, Princeton Environmental Institute, Princeton University

**Education:**

Ph.D. The University of Maryland, College Park, MD, 2000,  
Marine, Estuarine and Environmental Science  
M.S. The University of Connecticut, Storrs, CT, 1990,  
Oceanography  
B.A. Connecticut College, New London, CT, 1985,  
Major in Botany, Minor in Chemistry

**Teaching Experience:**

The courses I've taught in the Wheaton College Chemistry Department core curriculum include General Chemistry (Chem 153, *Chemical Principles*), Introductory Analytical Chemistry (Chem 232, *Aqueous Equilibria*), and Advanced Analytical Chemistry (*Instrumental Analysis*, Chem 332). I have developed lab exercises and taught lab sections in all of these courses. I've also regularly taught *Current Problems in Environmental Chemistry* (Chem 303), a core course in the Environmental Science major. My contributions to the general curriculum include *Chemistry and Your Environment* (Chem 103), an environmentally-themed introductory course for non-science majors, and *First-Year Seminar* (FYS). I am a Co-coordinator of the Environmental Science Program.

**Professional Societies**

American Chemical Society  
Society of Wetlands Scientists  
Society for Environmental Contamination and Toxicology

## Publications:

- Kirkpatrick, M., J. Benoit, W. Everett, J. Gibson, M. Rist and N. Fredette. 2015. The effects of methylmercury exposure on behavior and biomarkers of oxidative stress in adult mice. *Neurotoxicology*. 50:170-178.
- Benoit, J.M., D.A. Cato, K.C. Denison, and A.E. Moreira. 2013. Seasonal Mercury Dynamics in a New England Vernal Pool. *Wetlands*. 33:887-894.
- Benoit, J.M., D.H. Shull, R.M. Harvey and S.A. Beal. 2009. Effect of bioirrigation on sediment-water exchange of methylmercury in Boston Harbor, Massachusetts. *Environmental Science and Technology*. 43:3669-3674.
- Shull, D.H., J.M. Benoit, C. Wojcik, and J.R. Senning. 2009. Infaunal burrow ventilation rates and pore-water transport in muddy sediments. *Estuarine, Coastal and Shelf Science*. 83:277-286.
- Benoit, J.M., D.H. Shull, P. Robinson and L.R. Ucran. 2006. Infaunal burrow densities and sediment monomethyl mercury distributions in Boston Harbor, Massachusetts. *Marine Chemistry*. 102:124-133.
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### **Conference Presentations:**

- Herzog, A. and J. Benoit. 2017. Mesocosm studies on the effects of increased wet-dry cycles on methylmercury production in vernal pools. 11<sup>th</sup> International Conference on Mercury as a Global Pollutant, Providence, RI, July 2017

- Benoit, J., J. Barrows, A. Herzog and D. Cato. 2016. Vernal pools as methylmercury sources to the forest: Export via emergent insects. Annual Meeting of the Society for Wetlands Scientists, Corpus Christi, TX, June 2016.
- Kirkpatrick, M, J. Benoit, W. Everett, J. Gibson and M. Rist. 2013. The effect of methylmercury exposure on biomarkers of oxidative stress and locomotor behaviors in adult mice. 11<sup>th</sup> International Conference on Mercury as a Global Pollutant, Edinburgh, Scotland, July 2013.
- Muller, L., and J.M. Benoit. 2012. SPEC & Lab Buddies: Creating a Community of Learners in Chemistry. ACS Biennial Conference of Chemical Education, University Park, PA, July 2012.
- Benoit, J.M. 2012. Visual Communication in 200- and 300-Level Chemistry Courses. (Part of the panel: Teaching Visual Communications Across the Chemistry Curriculum). 11<sup>th</sup> International Writing Across the Curriculum Conference, Savannah, GA, June 2012.
- Benoit, J.M., K.C. Denison and N. Sacha. 2011. Seasonal Mercury Cycling in a New England Vernal Pool. 10<sup>th</sup> International Conference on Mercury as a Global Pollutant, Halifax, Nova Scotia, Canada, July 2011.
- Benoit, J.M. and D.H. Shull. 2009. Impact of Bioirrigation on Benthic Methylmercury Flux from Coastal Marine Sediments. American Chemical Society, Washington, DC, August 2009. (Invited speaker).
- Benoit, J.M., D.H. Shull, R. Harvey, and S. Beal. 2008. The Effect of Bioirrigation on Sediment-Water Exchange of Methylmercury in Boston Harbor, Massachusetts, USA, Geological Society of America Joint Annual Meeting. Houston, Texas, October 2008.
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- Baron, G. and J.M. Benoit. 2006. Mercury Concentrations in Hair of Autistic and Typical Young Adults and the Relationship with Fish Consumption Levels. Eighth International Conference on Mercury as a Global Pollutant, Madison, WI, August 2006.
- Bonzagni, A., H. Magruder and J.M. Benoit. 2006. Mercury Uptake from Fish Fertilizer by Spinach Plants. Eighth Annual Northeast Student Chemistry Research Conference (NSCRC), Cambridge, MA, April 2006.

## **CURRICULUM VITAE**

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### **Education**

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1980 B.A., Cornell University, Biochemistry

### **Professional Experience**

2004- Senior Scientist, Smithsonian Environmental Research Center  
2003-2004 Associate Director, The Academy of Natural Sciences, Estuarine Research Center.  
2002-2004 Curator, The Academy of Natural Sciences, Estuarine Research Center. 1996-2001  
Associate Curator, ANSERC.  
1997-present Participating Faculty, Marine, Estuarine, and Environmental Sciences Program,  
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1990-1996 Assistant Curator, ANSERC.  
1988-1990 Patrick Scholar, ANSERC.  
1988-1993 Associate of the Division of Applied Sciences, Harvard University, Cambridge, MA.  
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1986-1990 Adjunct Assistant Professor, Marine Sciences Research Center, State University of  
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1980-1985 Graduate Research and Teaching Assistant, University of Maryland, Department of  
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**Research Interests** Trace metal biogeochemistry, particularly mercury: mechanisms and control of  
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Microbiology (Microbial Ecology), American Society of Limnology and Oceanography, Estuarine  
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## Publications (68)

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- Jonas, R.B., C.C. Gilmour, D.L. Stoner, M.M. Weir, and J.H. Tuttle. 1984. Comparison of methods to measure acute metal and organometal toxicity to natural aquatic microbial communities. *Appl. Environ. Microbiol.* 47:1005-1011.

### **Recent and Upcoming Presentations and Posters at Scientific Meetings**

2013 Mason, R.P., T.A. Hollweg, A. Schartup, P. Balcolm and C.C. Gilmour. Examining the role of sulfur in the biogeochemical cycling of mercury and methylmercury in coastal sediments through measurements and models. 245th ACS National Meeting, New Orleans, April 2013.

*Presentations at the 11<sup>th</sup> International Conference on Mercury as a Global Pollutant, Edinburgh, UK, July 2013:*

- Gilmour, C.C., Graham, A.M., A. L. Bullock, A.M. Maizel, A. Somenahally, A. Johs, J.M. Parks, M. Podar, J. Smith and D.A. Elias. Phylogenetic distribution of the ability to methylate Hg among Bacteria and Archaea. Oral presentation.
- Harris, R.C., M. Amyot, P.J. Blanchfield, B. Branfireun, B. Dimock, C.C. Gilmour, J. Graydon, B. Hall, A. Heyes, H. Hintelmann, J. Hurley, C.A. Kelly, D.P. Krabbenhoft, M. Paterson, C. Podemski, J.W.M. Rudd, K. Sandilands, V.L. St. Louis, and M. Tate. Simulations of the Response of a Lake and its Food Web to Hg Additions During the METAALICUS Experiment. Oral presentation.
- Maglio, M., D.P. Krabbenhoft, M. Tate, J. DeWild, J. Ogorek, C. Thompson, G.Aiken, W.Orem, J. Kline, J. Castro, and C.C. Gilmour. Drivers of geospatial and temporal variability in the distribution of mercury and methylmercury in Everglades National Park. Oral presentation.

- Maizel, A.M. Graham, A.M, R.J. Reash, P. Chu, G.F. Riedel, and C.C. Gilmour. Mercury chemistry and mitigation strategies in leachate from a wet magnesium-lime FGD landfill. Poster presentation.
- Parks, J.M. A. Johs, M. Podar, R. Bridou, R.A. Hurt, S.D. Smith, S. J. Tomanicek, Y. Qian, S.D. Brown, C.C. Brandt, A.V. Palumbo, J.C. Smith, J.D. Wall, D.A. Elias, and L. Liang. New approaches for determining the molecular basis of bacterial mercury methylation. Poster presentation.
- Ghosh, U, J. Gomez Eyles, S.Kwon, C.C. Gilmour, G. Riedel, J.T. Bell, A. Maizel, C. A. Menzie, S. S. Brown. Black carbon sorption of Hg and MeHg and application in sediment remediation. Oral presentation.
- Mason, R.P. A.T. Schartup, P. Balcom and C.C. Gilmour. Modeling the role of sulfur in the biogeochemical cycling of mercury and in influencing methylmercury production in coastal sediments. Oral presentation.
- Bullock, A.L., A.M. Maizel, A.M. Graham, and C.C Gilmour. Methodology for assessing extents and rates of microbial Hg methylation in pure culture.
- Bridou, R., S. D. Smith, A. Kucken, S. Fels, J.M. Parks, A. Johs, R. Hurt, M. Podar, C.C. Gilmour, L. Liang, D. A. Elias, Judy D. Wall. Genetic tools for the determination of genes and enzymes involved in Hg<sup>2+</sup> methylation in *Desulfovibrio desulfuricans* strain ND132. Oral presentation.
- Smith. S.D., R. B. Bridou, R.A. Hurt, L. Liang, D.A. Elias, A.O. Summers and J.D. Wall. Biochemistry and regulation of mercury methylation by sulfate reducing bacteria. Poster presentation.
- Graham. A.M., A. L. Bullock, G.R. Aiken, and C.C. Gilmour. Controls on the bioavailability of Hg-DOM-sulfide nanoparticles/clusters to Hg-methylating bacteria. Poster presentation.
- Elias, D.A. et al. What could be the genetic mechanisms behind mercury methylation and why has it eluded us for so long? Oral presentation.
- Heyes, A., J.T. Bell, M. Gonsior, G.S. Riedel, G.F. Riedel, and C.C. Gilmour. Mercury cycling in first order watersheds of the Chesapeake Bay. Oral presentation.

### **Selected Professional Activities**

- Chesapeake Bay Program, Scientific and Technical Advisory Committee, At-large appointee, 2006-2011.
- EPA Science Advisory Board, Member, Ecological Processes and Effects Committee 1999-2005.
- SETAC Mercury Monitoring and Assessment Workshop. Sept. 2003.
- Technical Advisory Team, Fourth International Conference on Mercury as a Global Pollutant, Hamburg, Germany, Aug. 1996; Sixth ICMGP Minimata Japan, Oct. 2001; Seventh ICMGP Ljubljana Slovenia, June 2004.
- Development of the “Mercury Strategy for the Bay-Delta Ecosystem: A Unifying Framework for Science, Management, and Ecological Restoration” for CALFED, 2001-2003. With J. Wiener and D. Krabbenhoft.
- Advisory Committee, CALFED San Francisco Bay Mercury Study, 1999-2002.
- Maryland Sea Grant Scientific Advisory Committee, 1991-1993; 2001- 2006.
- Peer Reviewer, EPA Draft Mercury Research Strategy, 1999.

EPA Science Advisory Board, Consultant to:

Review of ORD's 2000 *Research Strategy for Mercury*

Ecological Processes and Effects Committee, Blackstone River Initiative. March, 1998.

Review of EPA's Mercury Report to Congress, 1997.

**Reviewer for Journals and other Publications:**

Appl. Environ. Microbiol., Aquatic Microbial Ecology, Arch. Environ. Tox. and Chem., Biogeochemistry, Bioscience, Biotechnol. Prog., Can. J. Fish. Aquat. Sci., Chemosphere, Encyclopedia of Microbiology, Environmental Indicators, Environ. Poll., Environ. Sci. Technol., Environ. Tox. Chem., Est. Coastal Shelf Sci., Estuaries, FEMS Microbial Ecology, Fresenius J. Analytical Chem., Geochim. Cosmochim. Acta, J. Great Lakes Res., Limnol. Oceanogr., Marine Chem., Mar. Ecol. Prog. Ser., Mar. Environ. Res., Microbial Ecol., Sci. Tot. Environ., Wat. Air Soil Poll., Wat. Resources Res., Wat. Poll. Res. J. Canada.

**Reviewer for Funding Agencies:**

Environmental Protection Agency (STAR), EPA/NSF Water and Watersheds, Electric Power Research Institute, Hudson River Foundation, Maryland Power Plant Topical Research Program, National Sciences and Engineering Council (Canada), National Science Foundation (Biological and Chemical Oceanography; EGB, Ecology; Atmospheric Chemistry, Polar Programs; International Programs), National Undersea Research Program (NURP/NOAA), Quebec Hydro, Ontario Hydro, Sea Grant (various state programs), USGS Water Resources Research Centers (various state programs).

**Students, Student Committees and Teaching:**

Master's committee, Sarah Downey, U. Maryland, MEES, Andrew Heyes, advisor. MS. 2010.

Doctoral committee, Terill Hollweg, U. Connecticut, Marine Sciences, Robert Mason, advisor. PhD 2010.

Doctoral committee, Marvourneen Dolor, U. Maryland, MEES, G. Helz Advisor, PhD 2009.

Co-advisor, Liz Kerin, U. Maryland, MEES, M. Suzuki, advisor. MS. Aug. 2007.

Doctoral committee, Carrie Miller, U. Maryland, MEES, Robert Mason, advisor. Ph.D. 2006.

Master's committee, J.R. Flanders, UC Santa Cruz. Russ Flegal, advisor. MS spring 2004.

Master's committee, Michael Rearick, U. Maryland, MEES, Robert Mason, advisor. MS Spring 2004.

Co-Advisor, Janina Benoit, U. Maryland, MEES, Robert Mason, advisor. Ph.D. May 2000. "Sulfide Controls on Mercury Methylation by Sulfate-Reducing Bacteria."

Doctoral committee; Jennifer Jay, Mass. Inst. Technology, Ph.D. 1999. Profs. Harry Hemond and Francois Morel, advisors.

Master's committee, Michael Ewell, University of Maryland, Center for Marine Biotechnology, M.S. 1995. Prof. R. Wiener, advisor.

Doctoral committee, Frederic Chanania, George Mason University, 1993.

Assoc. Prof. Robert Jonas, advisor.

Doctoral committee, Elizabeth A. Henry, Harvard University, Ph.D. 1992.

Prof. Ralph Mitchell, advisor.  
Master's committee, Maureen Leavitt, Univ. Mass. Boston, M.S. 1988.  
Asst. Prof. Michael P. Shiaris, advisor.  
Plus >20 undergraduate interns.

**Postdoctoral Scientists**

Andrew Graham 2010-2012  
Carl Mitchell 2008-2010  
Sonja Fajgervold 2007-2008  
Andrew Heyes 1995-2000

## Biographical Information for Professor Dan Jaffe

Dr. Jaffe is a Professor of Environmental Science at the University of Washington in the Department of Atmospheric Sciences (UWS) and in the School of STEM at the UW Bothell Campus (UWB). He is expert on atmospheric chemistry, mercury, ozone, urban and regional smog and long range transport of pollutants and is the author of more than 150 peer-reviewed publications on ozone, aerosols, mercury and other air pollutants. Dr. Jaffe is widely recognized as an expert on global transport of pollutants, especially transport from Asia to the U.S. and has several papers on the influence of background sources on regional and urban air quality. He recently participated on the panel for the National Academy of Science's study on The Significance of Intercontinental Transport of Air Pollutants. His research has been supported by the NSF, NOAA, EPA, NASA, NPS, EPRI, API and other organizations. He is the Principal Investigator for the Mt. Bachelor Observatory in Central Oregon, which is the only high elevation research station on the west coast of the U.S. and is a Principal Investigator for the NOMADSS project (<http://www.eol.ucar.edu/projects/sas/>). Data from Mt. Bachelor and further information about Dr. Jaffe's research group can be found at <http://www.atmos.washington.edu/jaffegroup>

### Professional Positions Held

Professor of Science and Technology (University of Washington-Bothell) and Atmospheric Sciences (University of Washington-Seattle), September 1997-current.

Professor of Chemistry--University of Alaska Fairbanks, Department of Chemistry/Geophysical Institute, June 1993 - September 1997.

### Education

B.S. Chemistry, February 1979, Massachusetts Institute of Technology

M.S. Chemistry, December 1983, University of Washington

Ph.D. Chemistry, June 1987, University of Washington; graduate work in inorganic, analytical and atmospheric chemistry, atmospheric sciences, environmental sciences and policy.

### Selected publications:

1. N.L. Wigder, D.A. Jaffe, F.A. Saketa, Ozone and Particulate Matter Enhancements from Regional Wildfires Observed at Mount Bachelor during 2004-2011. *Atmos Environ.*, DOI:10.1016/j.atmosenv.2013.04.026, 2013.
2. Timonen, H., Ambrose, J. L., and Jaffe, D. A.: Oxidation of elemental Hg in anthropogenic and marine airmasses, *Atmos. Chem. Phys.*, 13, 2827-2836, doi:10.5194/acp-13-2827-2013, 2013.
3. Ambrose J. L., Lyman S.N., Huang J., Gustin M.S. and Jaffe D.A. Fast Time Resolution Oxidized Mercury Measurements during the Reno Atmospheric Mercury Intercomparison Experiment (RAMIX), *Envir. Sci. Tech.* DOI: 10.1021/es303916v, 2013.
4. Wigder, N. L., D. Jaffe, F. L. Herron-Thorpe, and J. K. Vaughan. Influence of Daily Variations in Baseline Ozone on Urban Air Quality in the United States Pacific Northwest *J. Geophys. Res.*, doi:10.1029/2012JD018738, 2013.
5. Jaffe, D.A., Wigder, N.L., Ozone production from wildfires: A critical review. *Atmos, Envir.*, doi:10.1016/j.atmosenv.2011.11.063, 2012.
6. Qiao X., Tang Y., Jaffe D., Chen P., Xiao W. and Deng G. Surface Ozone in Jiuzhaigou National Park, Eastern Rim of the Qinghai-Tibet Plateau, China. *J. Mt. Sci.* 9: 687-696, DOI: 10.1007/s11629-012-2449-8, 2012.

7. Smith, D.J., Jaffe, D.A., Birmele, M.N., Griffin, D.W., Schuenger, A.C., Hee, J., Roberts, M.S. Free tropospheric transport of microorganisms from Asia to North America. *Microbial Ecology* 64(4):973-985, DOI 10.1007/s00248-012-0088-9, 2012.
8. Jaffe, D.A. and Wigder, N.L., Ozone production from wildfires: A critical review. *Atmos, Envir.*, doi:10.1016/j.atmosenv.2011.11.063, 2012.
9. Lyman S.N. and Jaffe D.A. Formation and fate of oxidized mercury in the upper troposphere and lower stratosphere. *Nature Geosciences*, DOI: 10.1038/NGEO1353, 2011.
10. McDonald-Buller E.C. Allen D.T., Brown N., Jacob D.J., Jaffe D., Kolb C.E., Lefohn A.S., Oltmans S., Parrish D.D., Yarwood G., and Zhang L. [Establishing Policy Relevant Background \(PRB\) Ozone Concentrations in the United States](#). *Envir.Sci. Tech.* DOI: 10.1021/es2022818, 2011.
11. Fischer E.V., K. D. Perry, and D. A. Jaffe. Optical and chemical properties of aerosols transported to Mount Bachelor during spring 2010, *J. Geophys. Res.*, 116, D18202, doi:10.1029/2011JD015932, 2011.
12. Ambrose, J.L., Reidmiller D.R. and Jaffe D.A., Causes of high O<sub>3</sub> in the lower free troposphere over the Pacific Northwest as observed at the Mt. Bachelor Observatory, *Atmospheric Environment*. doi:10.1016/j.atmosenv.2011.06.056, 2011.

### **Synergistic activities**

Professor Jaffe teaches courses in chemistry, environmental and atmospheric chemistry and global environmental issues. He strives to integrate research and active learning elements into his teaching so as to bring the excitement of science to his students. He has been an active participant in a number of U.S. and international task forces/panels on global pollution including the task force on Hemispheric Transport of Air Pollutants (HTAP), Arctic Monitoring and Assessment Program (AMAP), EPA Region X air toxics group and the Columbia River Gorge Commission. He is frequently quoted in the media based on his research and expertise on air pollution and mercury.

### **Graduate advisors: Drs. Norman Rose and Robert Charlson (U.Washington)**

**Post-doctoral advisor: None**

### **Graduate students advised (major professor):**

Pao Baylon, Harald Beine, Bianca Cerundolo, Emily Fischer, Richard Honrath, Jennifer Kelly, Robert Kotchenruther, Alexander Mahura, Crystal McClure, Heather Price, David Reidmiller, David Smith, Phil Swartzendruber, Barbara Trost, Nicole Wigder, Zhiyong Zhang, Matt Zukowski.

### **Post-doctoral fellows advised:**

Jesse Ambrose, Isaac Bertschi, Duli Chand, Brandon Finley, Lynne Gratz, Will Hafner, Jack Herring, Seth Lyman, Peter Weiss-Penzias, William Simpson, Julie Snow.

### **Current collaborators:**

Alex Guenther (NCAR), Mae Gustin (U. Nevada), Daniel Jacob (Harvard), Lyatt Jaeglé (U. Washington), Kevin Perry (U.Utah), Noelle Eckley Selin (MIT), Staci Simonich (Oregon State U.), Joel Thornton (U.Washington), Lin Zhang (Harvard),



## BIOGRAPHICAL SKETCH

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**ROBERT P. MASON**  
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Groton, CT 06340  
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Robert.mason@uconn.edu

### a. Professional Preparation.

University of Natal, Durban, South Africa (RSA), Chemistry, B.S., 1979  
University of Cape Town (UCT), RSA, Analytical Chemistry, M.S., 1983  
University of Connecticut (UConn), Chemical Oceanography, Ph.D., 1991  
"The Chemistry of Mercury in the Equatorial Pacific Ocean"

### b. Appointments.

8/2005-	Professor, Dept. Marine Sciences & Chemistry, University of Connecticut
7/2004-8/2005	Professor, CBL, UMCES, Solomons, MD
1999-2004	Associate Professor, CBL, UMCES, Solomons, MD
1994-1999	Assistant Professor, CBL, UMCES, Solomons, MD
1992-1994	Post-doctoral Investigator, Ralph Parsons Laboratory, MIT
1987-1991	Research Assistant, Department of Marine Sciences, UCONN
1984-1987	Research Scientist, Sea Fisheries Research Institute, RSA
1981-1983	Research Assistant, Department of Analytical Science, UCT, RSA
1980	Development Technologist, Hulett's Sugar, RSA

### c. 10 Recent Relevant Products.

1. Chen, C.Y., M.E. Borsuk, D.M. Bugge, T.A. Hollweg, P.H. Balcom, D.M. Ward, J. Williams & **R.P. Mason**. 2014. Benthic and pelagic pathways of methylmercury bioaccumulation in estuarine food webs of the northeast United States. *PLOS One* 9(2): Article #: e89305.
2. Driscoll, C.T., C.Y. Chen, C.R. Hammerschmidt, **R.P. Mason**, C.C. Gilmour, E.M. Sunderland, B.K. Greenfield, K.L. Buckman & C.H. Lamborg. 2012. Nutrient supply and mercury dynamics in marine ecosystems: A conceptual model. *Environmental Research* 119: 118-131.
3. Schartup, A.T., **R.P. Mason**, P.H. Balcom, T.A. Hollweg & C.Y. Chen. 2013. Methylmercury production in pristine and anthropogenically impacted sediments. *Environ. Sci. Technol.* 47: 695-700.
4. Schartup, A.T., P.H. Balcom & **R.P. Mason**. 2014. Sediment-porewater partitioning, total sulfur, and methylmercury production in estuaries. *Environ. Sci. Technol.* 48: 954-960.
5. Driscoll, C.T., **R.P. Mason**, H.M. Chan, D.J. Jacob and N. Pirrone. 2013. Mercury as a global pollutant: sources, pathways and effects. *Environ. Sci. Technol.* 47: 4967-4983.
6. **Mason, R.P.**, A.L. Choi, W.F. Fitzgerald, C.R. Hammerschmidt, C.H. Lamborg & E.M. Sunderland. 2012. Mercury biogeochemical cycling in the ocean and policy implications. *Environ. Res.* 119: Art # 03.013.
7. Schmeltz D., D.C. Evers, C.T. Driscoll, R. Artz, M. Cohen, D. Gay, R. Haeuber, D.P. Krabbenhoft, **R.P. Mason**, K. Morris & J.G. Weiner. 2011. MercNet: A national mercury monitoring network to assess responses to changing mercury emissions in the United States. *Ecotoxicol.* 20: 1713-1725.
8. Selin, N.E., E.M. Sunderland, C.D. Knightes & **R.P. Mason**. 2010. Sources of mercury exposure for US seafood consumers: Implications for policy. *Environ. Health Persp.* 118: 137-143.
9. Ndu, U., **R.P. Mason**, H. Zhang, S. Lin & P. Visscher. 2012. Effect of inorganic and organic ligands on the bioavailability of methylmercury as determined by using a *mer-lux* bioreporter. *Appl. Environ. Microbiol.* 78: 7276-7282.
10. Soerensen, A.L., **R.P. Mason**, P.H. Balcom & E.M. Sunderland. 2013. Drivers of surface ocean

## BIOGRAPHICAL SKETCH

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mercury concentrations and air-sea exchange in the western Atlantic Ocean. *Environ. Sci Technol.* 47: 7757-7765.

### d. Synergistic Activities.

1. Studies using stable mercury isotopes have examined redox processes as well as methylation and demethylation of Hg in aquatic systems, and the bioaccumulation of Hg and methylmercury (MeHg) in both bacteria and plankton. Studies through collaboration with Dartmouth have examined the sources and cycling of Hg and MeHg in estuaries, both pristine and impacted, from Maine to Maryland. Other studies have focused on Long Island Sound, the Delaware River, and in Labrador (Lake Melville). Studies of the mechanisms controlling bioavailability of Hg to methylating bacteria have led to a better understanding of the controlling factors, and to better models for Hg fate and transport. The coastal studies are providing insights into the importance of coastal methylation as a source of methylmercury to ocean fish.
2. Recent studies in the vicinity of Bermuda, on the New England shelf, and in the equatorial Pacific have further documented the importance of air-sea exchange and led to comparisons of measurements and models developed at Harvard University that have improved the understanding of sources and sinks of mercury to the ocean, and led to more refined mercury models.
3. On-going studies are examining the photochemical redox chemistry of inorganic Se and the abiotic formation and degradation of organic Hg and Se compounds. The rate information gained from these studies will be incorporated into a global Se model that has been developed and initially tested to examine the importance of various transformations in the ocean biogeochemical cycling of Se.

### e. Collaborators & Other Affiliations.

(i) **Recent Collaborators, besides UConn:** Celia Chen & Brian Jackson, Dartmouth College; Charley Driscoll, Syracuse Univ; Dave Evers, Biodiversity Inst.; Nick Fisher, Stony Brook; Jenny Jay, UCLA; Chad Hammerschmidt, Wright State; Carl Lamborg, Rachel Stanley & Mak Saito, WHOI, William Landing, FSU; Nicole Pirrone & Ian Hedgecock, CNR Institute, Rende, Italy; Noelle Selin, MIT; Elsie Sunderland, Anna Choi & Daniel Jacob, Harvard University; David Streets, Argonne National Lab; Melanie Witt, Oxford University; Ralph Ebinghaus, Institute for Coastal Research, Germany; Jim Weiner, Univ Minnesota.

(ii) **Graduate Advisor:** William Fitzgerald, Univ. of CT; **Postdoctoral Advisors:** Francois Morel, Princeton Univ. and Harold Hemond, MIT

(iii) **Thesis Advisor: Degrees completed:** Jani Benoit, Ph.D., Wheaton College, MA; Guey-Rong Sheu, Ph.D., Taiwan National Univ.; Joy Leaner, Ph.D., CSIR, South Africa; Eun-Hee Kim, Ph.D, post-doc, Univ. Korea; Carrie Miller, Ph.D., Oak Ridge National Lab; Terill Hollweg, Ph.D., NOAA Silver Springs; Udonna Ndu, Ph.D., post-doc, Rutgers University; Amina Traore, Ph.D., post-doc, Harvard; Susan Gichuki, Ph.D., Chemistry, teaching; Angie Lawrence, M.S., Baltimore Aquarium; Nicole Lawson, M.S., Not Currently Working; Kelly McAloon, M.S., VERSAR Inc., Virginia; Auja Sveinsdottir, M.S, Alcoa, Iceland; Mike Rearick, M.S, Los Alamos Lab, NM; Lindsay Whalin, M.S, Regional Water Quality Board, CA.; Christine Bergeron, M.S, completed PhD at UVA; Veronica Ortiz, MS, working at UConn.

**Current students:** Brian DiMento, Kati Gosnell, Emily Seelen; all Ph.D. Mar. Sci., Nashaat Mazrui, PhD, Chemistry

(iv) **Postgraduate-Scholar Sponsor:** Sofi Jonsson (current); Maria Andersson, working in Sweden; Ogus Yitergerhan, working in Turkey; Bian Liu, Columbia Univ; Eun-Hee Kim, Univ. of Korea; Fabien Laurier, US Climate Change Group; Elka Porter, U Baltimore; Andrew Heyes, U Maryland (CBL); Jean Michel Laporte, Syngenta Agro SAS, France; Sandra Andres, Position Unknown; Jenny Lee, Position Unknown.

**Total Graduate Students as Major Advisor: 21; Total Postdoctoral Sponsored: 11**

# Review of the Staff Report for Scientific Peer Review (April 2017) for the Statewide Mercury Control Program for Reservoirs

Janina Benoit, Wheaton College

19 July 2017

## I. Evaluation of Conclusions 1 - 11

### A. Conclusion 1

Evaluation:

**Conclusion 1** is supported in Chapter 4 and Appendix A. Overall, Chapter 4 provides a thorough literature review, and it largely accomplishes the stated goal of: “identifying factors that affect mercury methylation and bioaccumulation” (p. 4-1, 1<sup>st</sup> paragraph). Those two processes are widely recognized as key to controlling fish MeHg concentrations. A number of important factors influencing methylation are reviewed: sediment inorganic Hg (HgI) concentration and organic matter content, water column HgI and DOC concentrations, bioavailability of HgI, and type of landscape. In addition, factors affecting bioaccumulation are described, including lake/watershed characteristics (e.g., MeHg concentration in water, MeHg and total Hg (HgT) in sediment, forest cover, water column DOC and pH) and food web dynamics (e.g., primary productivity and food chain length). Reservoir stratification and turnover are described, and the impacts of those processes on MeHg production and bioaccumulation are explained. The chapter uses appropriate support from the literature to illustrate that sediment and water HgI concentrations alone cannot explain MeHg concentrations in fish. An understanding of the variables discussed in the chapter can provide an underpinning for modelling MeHg bioaccumulation and developing approaches to reduce MeHg in fish.

The review presents a conceptual model that is largely summarized in figures 4.2 and 4.3. In this model, inorganic mercury settles from the water column to sediments (p. 4-4, 4<sup>th</sup> paragraph), where it is converted to MeHg (p. 4-4, 1<sup>st</sup> – 3<sup>rd</sup> paragraphs). Subsequently, MeHg is taken up by algae (Figure 4.3), and ultimately biomagnifies through the food web causing elevated levels in top predatory fish. As a result, MeHg in water is a strong predictor of MeHg in fish (p. 4-2, 4<sup>th</sup> bullet point and p. 4-4, 4<sup>th</sup> paragraph). This description is valid as far as it goes, but a weakness in the conceptual model is that it doesn't strongly link sediment MeHg production to water concentrations. The accumulation of MeHg in anoxic hypolimnetic waters is mentioned in section 4.3.2 (p. 4-18, 3<sup>rd</sup> paragraph), but a more thorough consideration of factors influencing MeHg transport from sediment to water would strengthen the model. Furthermore, it isn't clear if initial bioaccumulation occurs in the pelagic or benthic environment (or both). Section 4.2.2 describes biodilution, the process whereby higher phytoplankton density leads to lower MeHg concentration (p. 4-12, 2<sup>nd</sup> paragraph, and Appendix A). Some further discussion of other factors

that control MeHg concentrations at the base of the food web would provide a stronger linkage between MeHg production and bioaccumulation in fish.

Specific comments on Chapter 4:

p. 4-3, 3<sup>rd</sup> paragraph, line 4. Although fewer mercury methylating iron-reducing bacteria have been identified, they may methylate at rates comparable to SRB and may be important Hg methylators in iron-rich environments (e.g. Fleming, E.J. et al. 2006. *App Env Microbiol* 72:457-464).

p. 4-4, 3<sup>rd</sup> paragraph, lines 1-4. This citation doesn't illustrate a relationship between sediment HgI and MeHg, which is the focus of the section. It belongs in the discussion of stratification, low oxygen and MeHg (p. 4-18).

p. 4-6, 2<sup>nd</sup> paragraph. The last sentence in the paragraph is unclear. Since the reservoirs have different Hg sources, differences in bioavailability *are expected* among them.

Section 4.1.2. This section reviews factors that influence MeHg production, but the effect of pH on methylation isn't discussed until the 6<sup>th</sup> paragraph on p. 4-11. Perhaps that paragraph should be moved here.

p. 4-8, 1<sup>st</sup> paragraph. Sediments don't have to be suspended to be available for methylation. Peak methylation rates often occur just below the sediment water interface. Sedimentation "removes" HgI when lower Hg materials rapidly cover sediments with higher Hg concentrations.

p. 4-16, 2<sup>nd</sup> full paragraph. The second sentence is a little misleading because the mechanisms overlap and because fall turnover doesn't increase methylmercury production. It would be more correct to summarize that thermal stratification can cause low oxygen concentrations in sediments and bottom waters; therefore, it can lead to enhanced production and/or release of MeHg to the water column.

Section 4.3.2. This section would be clearer with some reorganization. Specifically, if the section on "Redox Potential and Sulfate Reduction" were inserted between the 3<sup>rd</sup> and 4<sup>th</sup> paragraphs on p. 4-18, all of the consequences of stratification would be discussed together before considering fall turnover.

p. 4-19, 2<sup>nd</sup> paragraph. Given the somewhat conflicting evidence given here, should epilimnetic sediments be considered significant sources of MeHg to the water column in California reservoirs?

p. 4-21, 2<sup>nd</sup> full paragraph. An additional reference showing increased fish MeHg concentrations due to water level fluctuations: Selch, T.M. et al. 2007. *Bull Environ Contam Toxicol* 79:36-40.

p. 4-21, 3<sup>rd</sup> full paragraph. An additional reference supporting stimulated methylmercury production due to water level fluctuations: Eckley, C.S. 2017. *Environ Pollut* 22:32-41.

## B. Conclusions 2-4

Evaluation:

**Conclusion 2, 3 and 4** are supported in Chapter 5 and Appendices A and B. Chapter 5 describes a quantitative linkage analysis aimed at identifying predictors of fish MeHg concentrations in California Reservoirs. First, associations between seventy reservoir variables and fish MeHg concentrations are determined using correlation analysis (section 5.1.2). Next, strong correlates from the first step are used in a multiple linear regression model to identify the combination of variables controlling reservoir fish MeHg (Model 1, section 5.1.3). Further regression analyses are used to determine target levels for water column MeHg (Models 2 and 3, section 5.2) and sediment HgT (Models A and B, section 5.3) concentrations.

Throughout the chapter, the term “aqueous” is used for water column HgT and MeHg concentrations. The description of the water data states that “the linkage analysis uses results for unfiltered samples collected throughout the water column...” (p. 5-7, 2 paragraph) The term “aqueous” normally refers to the dissolved phase, so it should be replaced with “water column” to be consistent with common scientific usage and to avoid confusion. This overlap occurs in some places in Chapter 4 as well, and care should also be taken there to indicate whether “aqueous” refers to filtered or unfiltered samples from the water column or pore water.

**Conclusion 2** is consistent with the results of multiple regression Model 1. This model explains 84% of the variability in standardized fish MeHg concentration across reservoirs with three variables: ratio of water column MeHg to chlorophyll concentration, water column HgT concentration and mean annual water level fluctuation. These variables have been seen as important predictors of fish MeHg in previous studies, as discussed in Chapter 4.

Although those three variables are identified as “most important”, two of them are not pursued for further evaluation in models 2 and 3. The reasons for not considering water level fluctuations are outlined on p. 5-11 (5<sup>th</sup> full paragraph). Further explanation should be provided for: 1) why water column HgT concentration isn't further evaluated, and 2) why sediment HgT concentration is pursued instead as a factor to control. All of the statistical methods show the strengths of relationships, rather than cause-and-effect. Therefore, it is essential to provide a reasonable and literature-supported mechanism for how sediment HgT influences (controls) fish MeHg levels. Overall, a better description of the insights gained from Model 1 is needed.

The first part of **conclusion 3** is supported by the correlation analysis (Table B.3, B.4 and discussion in 5.1.2), which shows that some of the strongest correlates with fish MeHg are factors not associated with HgT loading, e.g., MeHg:chlorophyll-a ratio, longitude and water level flux; all of which had  $r \geq 0.3$  (Table B.3). Correlations between Hg sources and fish MeHg concentrations vary from not significant (e.g. watershed development, facilities, mine density, upstream wetlands) to moderately significant (e.g., soil HgT and atmospheric Hg deposition to the watershed). Furthermore, internal Hg pools are strongly correlated with each

other, suggesting intensive internal cycling (Table B.4). Considering the results of the correlation analysis, it is reasonable to conclude that a variety of factors influence MeHg concentrations in reservoir fish.

The second sentence in **conclusion 3** is likely true, but it isn't fully supported by the linkage analysis in Chapter 5. Although sediment Hg concentration was strongly correlated with fish MeHg concentration, the linkage analysis doesn't directly include MeHg production or bioaccumulation as factors (Table B.3). The conclusion about the role these two processes makes sense in view of the literature review in Chapter 4, but doesn't follow from results of the modelling efforts in Chapter 5.

**Conclusion 4** is supported by two lines of evidence presented in Chapter 5 (and Appendix B). First, many California reservoirs currently have natural background sediment Hg levels, but are still impaired (i.e., fish MeHg levels are higher than the target). For example, among the mercury impacted lakes included in models A and B, 21% and 12% had sediment mercury levels at and below natural background levels, respectively (Table 5.6). Second, these same models predict that only a small percent of reservoirs (<5%) would fully recover if sediments mercury concentrations were reduced to natural background levels, if no other factors were addressed. The linkage analysis shows that sediment mercury reductions alone are not an effective approach for reaching the MeHg sport fish target.

The end of Chapter 5 analyzes the possibility of using light fertilization to boost primary productivity of oligotrophic reservoirs (reviewed in detail in Appendix A). Models 2 and 3 are used to predict how changes in chlorophyll would affect Hg fish concentrations, and it is determined that fertilization could lower MeHg is fish independent of any changes in Hg. It would be worthwhile to adapt these models to predict an optimal MeHg:chlorophyll ratio, considering that this ratio was the strongest predictor of fish MeHg concentration. Optimizing this ratio could increase the implementation options; for example, less substantial decreases in MeHg might be effective in more productive reservoirs.

Overall, the linkage analysis identifies important variables associated with (and presumably controlling) fish MeHg concentrations in California reservoirs. It uses multiple linear regression, a straightforward statistical method that is commonly used to ascertain important controlling factors in complex environmental systems. The apparent controlling factors are consistent with the literature review in Chapter 4 and can be explained based on known mercury cycling processes.

Specific comments on Chapter 5:

p. 5-6 3<sup>rd</sup> and 4<sup>th</sup> paragraphs. Both of these paragraphs refer to TL4 fish, but the second is probably about TL3 fish. There is also appears to be mistake in the explanation under the caption for Figure 5.2, where TL4 fish are parenthetically described as 150-500 mm.

p. 5-7, 2<sup>nd</sup> paragraph. Is this paragraph suggesting that bioaccumulation of MeHg *only* occurs in the hypolimnion after fall turnover? This point should be clarified.

p. 5-7, 4<sup>th</sup> paragraph. The term “modern” is used here to describe soil mercury levels and later to describe reservoir sediment levels. This term should be explained more fully here.

p. 5-8, 5<sup>th</sup> full paragraph. Some of the mercury sources listed here are expected to be sources of MeHg and other predominantly HgI. Perhaps these different types of sources should be considered separately.

p. 5-9, 3<sup>rd</sup> full paragraph. A little further explanation is needed of how the variables were chosen for the model. Table B.3 shows that a few non-significant correlates were included and a few significant correlates were not. What was the rationale?

p. 5-11, 3<sup>rd</sup> full paragraph. The statement that “water level fluctuations do not increase aqueous MeHg concentrations” seems unlikely in view of the literature. The lack of correlation may result from the nature of the dataset, and a stronger relationship would likely be observed for hypolimnetic samples.

p. 5-11, 4<sup>th</sup> full paragraph. Given the importance of benthic biota as food for aquatic organisms, what is the potential for MeHg transfer via this route? The role of the benthic food web in pelagic bioaccumulation should be briefly reviewed in Chapter 4 (as mentioned above).

p. 5-13, last paragraph. For the sake of comparison, it would be helpful to calculate the target water column MeHg concentration predicted from these BAF values.

### **C. Conclusions 5-7**

#### Evaluation

**Conclusions 5, 6 and 7** are supported in Chapter 6. This chapter assesses potential sources of inorganic Hg to California reservoirs, by considering the watersheds of 74 impaired 303(d)-listed reservoirs.

This assessment begins by determining the natural (BG) and modern (MBG) Hg concentrations in the reservoir watersheds. This determination uses surface soils to reflect modern Hg levels and deep core soils and sediments to reflect natural background. Since California has varying native Hg levels, representative BG and MBG concentrations are determined for each of three areas, defined as trace mercury, mercury-enriched and mineralized. The BG and MBG levels are compared to surface sediment concentrations in 44 reservoirs for which this data is available (Table 6.4). This analysis shows that a significant proportion of the reservoirs have sediment Hg concentrations at natural (15) or modern (13) background levels. Therefore, for 64% of the reservoirs the dominant source is likely background Hg in watershed soil. Although MBG Hg concentrations have resulted from industrial-era Hg deposition, the term “background” is used

because watersheds will continue to provide this legacy mercury to reservoirs for centuries. For reservoirs at or below MBG levels, source reductions in the watershed (e.g. mining and point sources) are not likely to reduce sediment Hg loads or lower MeHg concentrations in fish. This analysis is consistent with **conclusion 7**. The key assumptions behind this conclusion are that 1) the measured soil and sediment concentrations adequately represent typical levels in California, and 2) watershed Hg concentrations with modern backgrounds will remain high for a long period of time. In view of the large datasets presented in Tables 6.2, 6.3 and 6.4, the first assumption is likely valid. The second assumption is supported by the literature.

The remainder of Chapter 6 evaluates a number of potential sources of Hg to reservoirs including mining waste (section 6.3), atmospheric deposition (section 6.4), urban run-off (section 6.5), facility discharges (section 6.6), and other sources (section 6.7). Assessments of mining, urban run-off and facilities discharges use a geographical approaches to identify the presence of those sources within watersheds of the 74 impaired reservoirs. Mercury deposition is assessed using the USEPA's Regional Modeling System for Aerosols and Deposition (REMSAD, details in Appendix D). The other sources (groundwater, springs, coastal fog and anthropogenic erosion) are identified as possible contributors of Hg to reservoirs, although adequate data is not available to characterize the magnitude of the contributions. **Conclusion 5** is consistent with the overall assessment. Evidence for the "uneven distribution of mercury sources" is provided by the geologic data, which show that 40% of the reservoirs occur in the enriched zone (Figure 6.3), so they receive more Hg from soil than those in low Hg areas. Also, historical mining sites are not evenly distributed among the watershed, both in terms of type (mercury, gold, silver) and density (Figure 6.9). In fact, about 40% don't contain any historical mines in their watersheds at all (p. 6-12, 3<sup>rd</sup> bullet point). Although the majority of the 303(d)-listed reservoirs are upstream of urban areas and their associated facilities, one may receive substantial facilities discharges (p. 6-36, third bullet point). The REMSAD model identifies variability in both the magnitude of Hg deposition (Figure 6-17) and the relative contribution of local versus global sources across California (Figure 6-18). Thus, there is not a single source of Hg to all impaired reservoirs.

As stated in **conclusion 6**, the assessment indicates that historic mine sites and atmospheric deposition are the most important current anthropogenic sources of Hg to impaired reservoirs. The evaluation of historic mine sites as Hg sources uses a number of federal and state datasets (p. 6-18, bullet points) representing the locations of historical mercury, gold, and silver mining features, including prospects, productive mines, tailings, etc. These features are mapped (Figures 6-6 to 6-8) to identify the number, density, and production of historic mines in the watersheds of the 303(d)-listed reservoirs (Figure 6.9). In this way, it is determined that 48 (65%) of the reservoirs could be affected by mining waste, because they have at least one mining feature in their watersheds. Of those 48 reservoirs, 41 have more than 50 productive sites (p. 6-19, 2<sup>nd</sup> bullet point).



After historical mines are located within watersheds, their importance as a source is inferred from sediment Hg concentrations in the 44 reservoirs with sediment data. Of the 26 (60%) of those reservoirs that had elevated mine densities, 50% have elevated mercury levels, indicating mine waste as a probable Hg source (p. 6-20, last paragraph). The other half of the reservoirs have Hg levels below modern and even natural Hg levels. If this analysis is extrapolated more broadly, about one-third of impaired reservoirs in California likely receive mining waste as an Hg source. This possibility warrants the conclusion that historic mines are an important source of Hg to California reservoirs.

A second major source of Hg identified in the Chapter 6 is atmospheric deposition. The assessment of this source (section 6.4) first reviews historical mercury emissions both globally and in California to identify key sources and trends. Facilities mapping reveals the “clustered” nature of emissions sources in the state (Figure 6.15). Although some wet deposition data is available, it is deemed too limited for a state-wide assessment, so atmospheric Hg deposition is modelled with REMSAD. This model calculates wet and dry deposition of atmospheric pollutants, and it is also able to track emissions (p. 6-27, 4<sup>th</sup> paragraph). The tracking feature, called “tagging”, is useful for attributing Hg deposition to California emissions sources. First, the model is used to quantify anthropogenic Hg deposition from global, regional and local sources. Results of this simulation (Table 6.5) reveal that California anthropogenic sources account for about 10% of total Hg deposition in the state, whereas global anthropogenic sources account for about 60%. Atmospheric deposition is deemed to be a major source of Hg for reservoirs with few or no point sources or historic mining activity in their watersheds (p. 6-31, 2<sup>nd</sup> paragraph). Twenty-nine (62%) of the 47 303(d)-listed reservoirs fit those criteria, suggesting that atmospheric deposition is a primary source of Hg to a substantial proportion of California reservoirs.

**Conclusion 7** is also supported by output of the REMSAD model. Model-derived deposition maps characterize patterns throughout the state, including the patterns for total Hg deposition (Figure 6.17) and deposition attributed specifically to California sources (Figures 6.18 and 6.21). Those deposition maps show that global emissions dominate for much of the state, although there are hotspots where California emissions account for 20% or more of the total (Figure 6.23). Overlaying the REMSAD deposition patterns on the watershed map shows that 21 of the 303(d) listed reservoirs are in one of these hot spot areas. Further analysis reveals that among the 29 reservoirs where atmospheric deposition is the dominant anthropogenic source of Hg, 12 have a significant (>20%) contribution from California sources. For the remaining 17 reservoirs, Hg deposition is attributed to predominantly global anthropogenic sources (p. 6-31, 2<sup>nd</sup> paragraph). The finding that many reservoirs receive deposition from primarily global emissions is reasonable in view of the predominance of global deposition statewide and the magnitude of current emissions outside of North America (e.g. Figure 6.11 and section 6.4.3). Lowering local emissions will not diminish impairment in all deposition-dominated reservoirs in the state.

An important assumption underlying **conclusions 6 and 7** is that the REMSAD model reliably recreates anthropogenic Hg deposition. The REMSAD model has been peer-reviewed (p. D-2) and validated through comparison to wet and dry deposition rates in California and Nevada (p. D-3). Although the model may underestimate point source emissions (p. D-3), discrepancies between modelled and actual deposition rates aren't likely to change the conclusions about the significance of atmospheric deposition as a source of Hg or the relative contribution of California emissions to total Hg deposition

An important assumption of the overall assessment in Chapter 6 is that the 74 303(d)-listed reservoirs and their watersheds are characteristic of other reservoirs in California. This assumption seems logical given the broad geographic distribution of the reservoirs (Figure 6.1), their occurrence within both enriched and trace mercury areas (Table 6.4), the range of sediment mercury concentrations (Table 6.4), the variety of mine types and densities (Figure 6-9), and the broad range of atmospheric deposition in their watersheds (Figure 6-17).

Specific comments on Chapter 6:

p. 6-7, last paragraph. What was the cutoff date between modern and natural for dated sediment cores?

p. 6-9, last bullet point. Is this concentration determined from the data in Tables 6.3 and 6.4?

p. 6-20, last full paragraph, lines 2-5. This statement seems inconsistent with p. 6-11. The 7<sup>th</sup> bullet point on p. 6-11 says that of the 16 reservoirs with elevated Hg concentration, 13 are downstream of historic mine sites and 3 are in urban areas.

#### **D. Conclusions 9-11**

Support for **conclusions 9, 10 and 11** is provided by Chapter 7 and Appendix H. Chapter 7 discusses strategies that could potentially lower Hg concentrations in reservoir sediments, waters, and/or fish. The chapter provides examples of successful remediation from previous studies reported in the literature, and points out limitations and drawbacks associated with each strategy. It also predicts the success of each of the strategies for the 303(d)-listed reservoirs. The defining reservoir characteristics that are used for the prediction are summarized in Table H.1, and the chapter culminates with a summary of the remediation techniques that are likely to be successful in each of the reservoirs (Table 7.1).

The analysis in section 7.2 supports **conclusion 10** because it shows that source reductions alone will not be effective for reducing fish MeHg to the target in most of the reservoirs. It explains that reduced loading from mining sources will only be effective in a subset of reservoirs in mine-impacted watersheds, because many of these reservoirs already have sediment HgT levels below modern background levels (p. 7-11, 2<sup>nd</sup> paragraph). Furthermore, elevated fish MeHg levels are currently found in some reservoirs with background mercury levels, so reductions in mine waste

might not achieve the fish MeHg target even if initially high sediment levels were reduced by mine mitigation (e.g., 7-12 2<sup>nd</sup> full paragraph). Similarly, only a small number of reservoirs with atmospheric deposition as the dominant source are expected to recover if California emissions are reduced, because most receive Hg primarily from global emissions (Chapter 6). Reducing local emissions is expected to lower fish MeHg concentrations in only four of the 303(d)-listed reservoirs (p. 7-17, 1<sup>st</sup> bullet point). Overall, reduction of anthropogenic Hg sources in the state are expected to measurably reduce MeHg levels in fish in 40% of 303(d)-listed reservoirs, but rapid and significant decreases due to source reduction are only expected for 7 (9%) of them (Table 7.1). Given the limitations of source reductions as a means of lowering fish MeHg, a variety of reservoir-specific interventions will be needed to reach the MeHg sport fish target.

Chapter 7 supports **conclusions 9 and 11** by presenting a range of options and assessing the likelihood of their success in California reservoirs. Section 7.2 considers mercury source reductions, and determines that it could be effective for *some* reservoirs. For example, reduction of mining wastes may be effective in reservoirs that have high mine density in their watersheds and high sediment and fish HgT concentrations (p. 7-11, 1st paragraph). Ten of the 46 reservoir with sediment Hg data fit this description. Previous research shows that reductions in atmospheric deposition can lead to relatively rapid (years to decades) reduction in fish Hg levels in water bodies where atmospheric deposition is the primary source of Hg (reviewed p.7-15, last paragraph). Similarly, reductions in California emission are likely to lessen impairment in reservoirs that receive > 50% of atmospheric deposition from California sources (p. 7-17, 1<sup>st</sup> paragraph). Forestry practices that minimize the transport of DOC-bound mercury are likely to lead to reductions in fish MeHg concentrations in reservoirs in rural areas (p. 7-27, 3<sup>rd</sup> and 5<sup>th</sup> paragraphs), whereas lowering MeHg in MS4 discharges could reduce MeHg in the water column and fish in reservoirs in urban areas (p. 7-37, last paragraph). Overall, section 7.2 shows that source reduction measures can have a positive impact, but their applicability varies among reservoirs.

Chapter 7 further supports **conclusions 9 and 11** in its discussion of approaches that lower MeHg production by reducing methylation rates or sediment HgT concentrations (section 7.3). Hypolimnetic oxygenation has been shown to reduce MeHg in the water column in previous studies in California and elsewhere (reviewed p. 7-42 and 7-43). The analysis in section 7.3.1 suggests that this method could work in reservoirs that have strong thermal stratification that leads to bottom water anoxia; this characteristic applies to more than 50% of the 303(d) listed reservoirs (p. 7-44, 1<sup>st</sup> paragraph). Those same reservoirs could see reductions in fish MeHg levels from hypolimnetic nitrogen additions (p. 7-45, 3<sup>rd</sup> paragraph), because this approach functions by raising redox potential, thereby lowering net methylation and reducing MeHg flux. Sediment removal and capping can lower sediment HgT, hence fish MeHg, in highly contaminated reservoirs that have nearby sources that have been remediated (p. 7-46, bullet points). At least three of the 303(d) listed reservoirs are candidates for this approach (p. 7-47, 1<sup>st</sup> paragraph).

Among the strategies discussed in section 7.3, there are also limitations that make them applicable to only a subset of reservoirs. Oxygenation only applies to strongly stratified reservoirs with seasonally anoxic bottom water. Sediment capping or removal would only be effective after mine remediation and in reservoirs that have sediments Hg concentrations above the background in watershed soils (p.7-46, 3<sup>rd</sup> paragraph).

The analysis of fisheries management practices (section 7.4) is also consistent with **conclusions 9 and 11**. Light fertilization increases primary productivity and enhances biodilution, so it can lower fish MeHg concentrations (explained in detail in Appendix A). The linkage analysis used Models 2 and 3 to predict how doubling chlorophyll-a concentration would affect impairment of reservoirs with high fish MeHg and chlorophyll-a concentrations  $\leq 3$  ug/L (Table A.1). Although the two models gave somewhat different results, both indicate that all lakes with those characteristics would see at least a 25% improvement. Section 7.4 points out that 21 reservoirs have low enough chlorophyll-a levels to benefit from light fertilization (p. 7-52, 5<sup>th</sup> paragraph). Light fertilization is not a universal solution, because it would only be effective in oligotrophic lakes with sufficiently long residence times. Another approach that may be effective is altering stocking practices in the reservoirs where stocking is ecologically sound (p. 7-55, 3<sup>rd</sup> paragraph). The chapter also predicts that intensive fishing would be feasible for reducing fish MeHg concentrations in reservoirs that are oligotrophic, have elevated MeHg levels only in predatory fish and are not too large. It is suggested that this method could be effective in about half of the 74 303(d) listed reservoirs (p. 7-56, 2<sup>nd</sup> and 3<sup>rd</sup> paragraphs). Taken together, fisheries management practices might be applied in more than half of the impaired reservoirs, but specific strategies would need to vary among reservoirs.

Section 7.5 elucidates additional characteristics that necessitate reservoir-specific actions to reduce fish MeHg concentrations in reservoirs. This section underscores the complexity of processes leading to elevated fish MeHg levels, operational constraints that depend on reservoir uses, and differences in sediment and nutrient loads due to watershed characteristics. Table 7.1 summarizes the implementation options deemed to be appropriate for each of the 303(d)-listed reservoirs. Given the multiple options available for most reservoirs, it is reasonable to conclude that there will be a feasible strategy for every reservoir.

## **II. The Big Picture**

Chapters 4-7 (and appendices) consistently support Conclusions 1-11 through literature review, statistical evaluation of controlling factors, source attribution and assessment of implementation options. Taken as a whole, the conclusions are predicated on sound science. Inferences are drawn from the evaluation of up-to-date, peer-reviewed literature, viable statistical methods (correlation and regression models), and a validated deposition modelling tool (RMSAD). The analyses use a large and comprehensive dataset, and the report steers clear of drawing conclusions where sufficient data is not available.

The Staff Report shows that a variety of control options need to be applied on a reservoir-specific basis in order to meet the target sport fish MeHg concentration. This strategy is necessitated by the breadth of Hg sources, complex within-reservoir processes, and variable characteristics of reservoirs and their watersheds. Another conclusion that can be drawn from Chapters 4-7 is that *mitigation efforts will be most effective if they can adapt to an evolving understanding of reservoir processes and to fluctuations in environmental conditions brought about by climate change*. The need for additional surveys and pilot studies emerges throughout the chapters, and climate change impacts are addressed directly in section 7.7.5.



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July 27, 2017

Gerald W. Bowes, Ph.D.  
Manager, Cal/EPA Scientific Peer Review Program  
Office of Research, Planning and Performance  
State Water Resources Control Board  
1001 I Street  
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Dear Dr. Bowes –

Thank you for the opportunity to participate in peer review of the proposed “Mercury TMDL and Implementation Program for Reservoirs”

I commend the California Water Board Staff on a detailed and thoughtful evaluation of the problem, including a thorough summary of the available data. California has made significant progress toward understanding the magnitude of the mercury problem in reservoirs. The draft proposed “Mercury TMDL and Implementation Program for Reservoirs” is an ambitious program that should reduce MeHg risk to people and ecosystems in impaired California Reservoirs.

My review includes review of the conclusions presented in Attachment 2 of the review request (pages 1-5) plus a review of the staff report follow on pages 6-14, and references, as requested. My comments are made in the spirit of finding the best strategies to reduce risk to Californians, and not to take away from the excellent work done by the Water Board Staff in preparation of the draft Mercury Reservoir Provisions.

Sincerely,

Cynthia Gilmour  
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443-482-249

## **Review of Proposed Rule: “Mercury TMDL and Implementation Program for Reservoirs”**

Cynthia Gilmour  
Smithsonian Environmental Research Center  
July 27, 2017

### **Review of conclusions presented in Attachment 2 of the review request**

#### Conceptual Model

##### **1. Many factors, not just the amount of inorganic Hg in water and sediment, influence MeHg concentrations in reservoir fish.**

I agree with this general statement based on the data and analysis provided and based on the wider scientific literature on MeHg production and bioaccumulation. However, I am concerned with the overall conclusion of this report that Hg source control will be insufficient to reduce fish MeHg levels in most impaired reservoirs to target levels. The linkage analysis relies on several measures of Hg (for example sediment Hg concentration instead of Hg load) that may not adequately capture the amount of Hg available for MeHg production (see detailed discussion below).

My interpretation of the extensive literature on Hg remediation is that mercury source control should always be the first approach to reducing MeHg risk. For example, Sweden’s long experience with management of fish Hg levels showed that while some interventions (intensive fishing, liming) worked, they were short-term expensive fixes. Only reductions in Hg deposition to Sweden really improved the problem across large spatial scales. The strong spatial relationships between fish MeHg and mining sites in CA is obvious in the data presented here and in the Western Hg Synthesis [*Fleck et al., 2016*]. Despite the linkage analysis showing a strong relationship between fish MeHg and chl<sub>a</sub>:MeHg ratios, Occam’s Razor says the most obvious solution is usually best. I wonder if the approaches to remediation proposed here may rely too heavily on chemical alterations to reservoirs, to the detriment of emphasis (and resources) on Hg source control.

#### CA-specific linkage analysis

##### **2. The three most important factors that control fish methylmercury concentrations in California reservoirs are: the ratio of aqueous methylmercury concentration to chlorophyll-a concentration, aqueous total mercury concentration, and annual reservoir water level fluctuations.**

The analysis presented is an exhaustive look at the substantial available data. Congrats to the team for such an in-depth thoughtful look. I agree that the data set shows that trophic status (chl a concentrations) may be a knob that can be tweaked to reduce MeHg concentrations in fish. Is more fish in a reservoir, but with lower MeHg levels the appropriate goal?

But I'm not sure I agree that the three factors that came out of the analysis are the most important in control of fish MeHg levels. The analysis presented is appropriate to the available data, and the resulting conclusions are consistent with good statistical analysis. However the data have substantial limitations. The linkage analysis did not include several parameters that may be strongly related to MeHg production and MeHg in water or fish, including the degree of stratification or anoxia, the organic content of sediments, growth of submerged aquatic vegetation (the last two enhance microbial activity and MeHg production), DOC, and critically the loading rate of Hg to reservoirs. No doubt these data are unavailable, but their lack does present limitations on interpretation of the analysis.

The use of unfiltered (and often spatially and temporally scarce) water MeHg data may be problematic in the linkage analysis. The collection of unfiltered MeHg data is a common problem in monitoring programs, and CA should work to fix this in monitoring going forward. And last, the linkage analysis only included data from Hg-impaired reservoirs. Would an analysis that included all reservoirs may have shown stronger relationships between total Hg, MeHg and MeHg in fish?

The analysis conducted made appropriate use of available data, but could go further in acknowledging the limitations imposed by the data. Model 1 includes only a small subset of all CA reservoirs. The chl<sub>a</sub>:MeHg ratio is available for only ~40 reservoirs. The use of unfiltered MeHg data, and of MeHg and chl<sub>a</sub> data that may not have been taken at the same season, depth, or frequency in all reservoirs makes this model more uncertain than it might appear in the formal statistical analysis presented.

**3. Inorganic mercury sources alone are not the primary driver of mercury impairments in California reservoirs. Multiple factors drive reservoir fish methylmercury levels: amount of mercury, methylmercury production, and bioaccumulation.**

I disagree, based on data limitations in the linkage analysis. See my comments on the linkage analysis in #2 above.

**4. Inorganic mercury levels in many reservoirs would need to be lower than natural background to achieve the TMDL targets and mercury water quality objectives if no other factors are addressed.**

There are reservoirs that will never meet the TMDL targets, including reservoirs w/o mines upstream. See my response to #7.

Mercury Source Assessment

**5. Mercury sources are not evenly distributed across the State and no one source type is responsible for all reservoir impairments.**

Agree. The detailed analysis of sources shows there are several sources that contribute to Hg load to CA reservoirs. But mines (and re-emissions from mining areas) are the obvious driver of elevated Hg in most impaired CA reservoirs.



**6. The most important anthropogenic sources to impaired reservoirs are historic mine sites and atmospheric deposition from global and local (California) industrial emissions.**

Agree.

**7. Reducing watershed mercury sources is not expected to result in substantial reductions in reservoir sediment mercury concentrations and fish methylmercury concentrations in many reservoirs.**

Disagree. I think that the linkage model underestimates the benefit of mine site clean-up (source reduction), both in amount and timing. The available data include sediment Hg concentration, but not Hg loading. Critically, Hg in sediments becomes less available for MeHg production over time after deposition to sediments [Harris *et al.*, 2007]. Our estimate of the half-life of Hg bioavailability for methylation in sediments within the METAALICUS study was several months to a few years at most. Mercury becomes unavailable more rapidly than sedimentation, due to sorption into unavailable phases.

The relationship between sediment Hg and fish MeHg is significant but weak in the CA data set. But there doesn't seem to be a good measure of loading in the data set - there are data on the number and density of mines in reservoir watersheds, but not flux of Hg off of the sites or into reservoirs (data limitation). I suspect the relationship between Hg load and fish Hg would be stronger than the sediment Hg:fish MeHg relationship.

**8. Global industrial emissions are the predominant anthropogenic source to about 20 percent of mercury-impaired reservoirs.**

Not sure. Does the REMSAD model adequately capture re-emissions from contaminated soils in northern CA mining areas?

Potentially Controllable Processes and Predictions for Improvement

**9. There are a variety of mercury source control options and reservoir water chemistry and fisheries management practices that may be effective for reducing fish methylmercury concentrations.**

Agree.

**10. A combination of source control actions and reservoir and fish management practices—versus source control alone—will be needed to achieve both timely and measurable fish methylmercury reductions in most of California's mercury impaired reservoirs.**

I'm not sure I agree. But I do think the idea of testing a variety of approaches and evaluating results over the next decade is a good one. However, I'd make sure that several mine site clean ups... with a really complete evaluation of fluxes off of mine sites and into

reservoirs during the process... should be part of the pilot testing process. Go slow and low on any nitrate additions and chemical changes other than oxygenation.

**11. Actions to reduce fish methylmercury levels may need to vary for each reservoir because of the many combinations of different mercury sources (e.g., some are natural or global and therefore not regulated by state and federal agencies competing factors that control methylmercury production, and reservoir operational constraints. Reservoir-specific characteristics and operational requirements and mandates may not allow for all methylmercury management tools to be used in all reservoirs. Even so, there should be a possible solution to mercury impairment for every reservoir.**

There will be some reservoirs for which there is no reasonable way to reduce fish MeHg to CA targets. But reductions in fish MeHg should be achievable in the majority of impaired reservoirs. A key question for CA will be whether to try minimally tested interventions like nitrate amendment and fisheries alterations while waiting for clean-up of mine sites.

#### TMDL and Load Allocations

**12. The TMDL loading capacity and allocations, combined with reservoir water chemistry and fisheries management pilot tests and implementation actions identified in the proposed program of implementation), are adequate to achieve the proposed mercury water quality objectives and TMDL numeric targets for methylmercury in reservoir fish.**

If the loading targets for mining areas can be reached, I suspect that alone would move most reservoirs close to fish Hg targets. But I can't tell how the proposed Reservoir Mercury Control Program will force mine remediation. Appendix I states that the proposed TMDL "will not pose new economic costs or environmental impacts to address discharges from mercury and gold mines." ... and further explains that existing regulations already require clean-up. But mine remediation has just barely begun. How will the new TMDL force cleanup without additional spending??

I'm also concerned that the loading targets for mine areas are given as Hg concentrations on particles, rather than mass loading to reservoirs. Of course, the former is easier and less expensive to measure. But evaluation of the efficacy of mine clean up will require quantitative measurement of change in reservoir loading.

But overall I'm more concerned with the ability to get mine clean up done than the choice of TMDL targets for clean-up.

WRT other reservoir management tools, I don't think there is enough evidence to be assured that tools other than load reduction, bottom water oxygenation and water level control can reduce fish Hg levels to targets. Data on other controls are sparse. That's not to say that other approaches aren't worth trying in a phased, pilot study approach. But whether these measures along with source control will be adequate to meet fish MeHg

goals will have to be evaluated again in a decade. The linkage analysis is a good first step, but its conclusions will have to be tested as remediation proceeds.

**13. The allocations are adequate for both current and future mercury sources to the mercury-impaired reservoirs.**

Same answer as #12.

Margin of Safety, Seasonal Variations, and Critical Conditions

**14. The Reservoir Mercury TMDL incorporates an adequate margin of safety.**

It's too soon to know. See response to #12.

Implementation and Monitoring

**15. The Mercury Reservoir Provisions requirements for inorganic mercury controls are adequate to reduce anthropogenic discharges of inorganic mercury to reservoirs.**

See response to #12.

**16. During the first phase of the implementation program for the impaired reservoirs, the Mercury Reservoir Provisions require reservoir water chemistry and fisheries management practices pilot tests. Implementing reservoir pilot tests to develop and evaluate and water chemistry and fisheries management practices in a phased approach is an adequate approach to reduce reservoir fish methylmercury levels. This phased approach includes State Water Board review of the Mercury Reservoir Provisions prior to full scale implementation of effective and feasible management practices.**

As stated above, I don't believe that reservoir management w/o source control will be adequate to achieve target fish MeHg levels in most reservoirs. But other controls may help in the interim, especially bottom water oxygenation in stratified systems.

Assessment of Compliance with the Proposed Water Quality Objectives

**17. The upper 90th confidence limit of the mean is an appropriate statistical method to determine compliance with water quality objectives based on an annual average fish methylmercury concentration. In addition, it is appropriate to use consistent fish trophic levels and sizes, sample type and quantity, and sampling locations when determining compliance with water quality objectives.**

Agree.

**18. Biosentinel fish monitoring provides a means to evaluate relatively rapid changes to biotic methylmercury levels.**

To my knowledge, young of the year sport fish as monitors for MeHg bioaccumulation have been effective in some studies/locals and not others. Consistency in sample timing and location seem important. But year-to-year variability in year class size and growth rate can confound analysis. Monitoring of upper trophic level fish should not be abandoned or reduced if YOY monitoring programs are added.

## **Comments on Staff Report**

Cal Water Boards/Cal EPA

### **Summary comments**

The proposed “Mercury TMDL and Implementation Program for Reservoirs” is a highly ambitious program that should reduce MeHg risk to people and ecosystems in impaired California Reservoirs.

I commend the use of fish tissue Hg targets rather than water or sediment.

I found the distinctions between TMDL under review, the parallel subsistence fishers TMDL, and the separate Water Quality Objectives confusing, no doubt my lack of understanding of California law.

The linkage analysis conducted is a statistically appropriate and exhaustive look at available data. However, interpretation of results should go further in acknowledging the limitations imposed by the data, which I discuss in detailed comments below. I am not convinced by the analysis that chl<sub>a</sub>:MeHg ratio is the most important control factor for fish Hg in California lakes.

I feel that the major focus of the Mercury Reservoir Provisions TMDL implementation program should be on source reduction. I am concerned that reservoir management approaches other than source reduction will take away resources and attention from mine clean up and monitoring. However, I agree with the idea of a few focused pilot tests of reservoir management actions other than source reduction. I suggest that pilot tests should mainly focus on lower risk approaches (e.g. bottom water oxygenation, sediment hot spot cover or removal) that have already been tested in other lakes. Nutrient additions, nitrate addition for bottom water redox control, or intensive fishing bear especially careful monitoring of food web structure response.

All efforts toward remediation should include significant measurement and monitoring programs. For mines, this should include efforts to measure the efficacy of mine clean approaches up in reducing Hg runoff, and careful monitoring of changes in Hg load to reservoirs (particulate and filterable) relative to changes in MeHg in water and fish. For all remediation tests (including water chemistry or fisheries modifications), implementation should include detailed assessment programs of Hg and MeHg in water, sediment and biota through time, and careful monitoring of food web structure and composition especially if nutrient amendments are tried. I am concerned that there will not be enough funding/resources for adequate monitoring of remediation tests, increasing the risk of negative consequences of water chemistry and fisheries modification pilot studies. The work should have external expert panel oversight throughout, including design of proposed management efforts.

I would have like to seen more emphasis in this report on how source control efforts will be prioritized relative to lake chemistry and fisheries management tools, including how resources and spending will be allocated. Source control from mines is a long-term, expensive fix, with many jurisdictions and stakeholders. Appendix I states that “the

Reservoir Mercury Control Program will not pose new economic costs or environmental impacts to address discharges from mercury and gold mines.” ... and further explains that existing regulations already require clean-up. But mine remediation has just barely begun. I see a huge disconnect here. If the new TMDL implementation program does not force additional mine clean up through allocation of additional state, federal, local and private funds, the TMDL loading goal will never be met. Again, perhaps I am missing something in the law.

I recommend more deposition and air monitoring across the state in support of TMDL implementation. Current spatial coverage is relatively poor. Consider using new, much less expensive air Hg passive samplers (Mitchell et al. 2016) for better coverage in urban areas and around impacted reservoirs, and use these as surrogates for dry dep.

## Detailed Comments

p 25. The program is very ambitious.

Time lines after a decade not clear, but that would be hard.

Resource allocation to reservoir controls vs. mine clean up is not specified and should be.

Some of the language in the summary section doesn't match rule or summary in attachment 1 in places.

Not clear to me how the water quality objectives (fish MeHg levels) fit in this report – these are part of the TMDL, but are listed in the staff report as a “separate but related project in section S3. “The derivation of and scientific basis for mercury water quality objectives is provided in the Staff Report for Statewide Mercury Water Quality Objectives” – which we are not reviewing I think?

How will federally-owned reservoirs be managed? What about Bureau of Reclamation reservoirs?

Commend choice of fish for numeric water quality targets. “Staff proposes numeric targets that are equal to the mercury water quality objectives for COMM, WILD, and RARE beneficial uses because these targets will allow direct assessment of whether beneficial uses are being met.” – agree! P. 74

Appendix L – how to sample fish

### Good –

- avg. over whole lake, must include Hg-impacted areas if they exist
- Quantify at total Hg, EPA Method 7473 (thermal)
- 90% UCI of arithmetic mean – good new approach, conservative estimate of risk, smarter faster eval than “binomial” approach with grouped fish

### Less good –

- no frequency of sampling required; but frequency of ten years or less recommended
- Include stocked fish if present and important in creels, but not requirement about what fraction of the fish sampled, or if they should be evaluated separately. Recommend that they be reported separately even if included in determination of target attainment. p 3
- Wide allowable size range of fish.
- Min requirement of only 9 fish -too small for good stats, eval thru time, or eval of risk – at least 90% UCI captures some of the inherent variability in small sample size. May allow reservoirs with low fish Hg to avoid expensive sampling, while forcing borderline reservoirs to collect more samples. Why such minimal requirements when the fix can be very expensive??

90% UCI seems a big improvement on the “binomial” approach that required grouping all fish from one sampling date into a single data point for evaluation

**Chapter 3** – a very comprehensive look at CA reservoirs, and appropriate analysis of available data.

Analysis of individual fish and normalizing to standard size is great; also good to see relationship between avg and standardized fish as justification for avg fish target.

#### **Chapter 4.** Conceptual model

Agree that most CA reservoirs are old enough that they are steady state WRT reservoir construction impacts on methylation.

What fraction of CA reservoirs are stratified/anoxic bottoms? This data seems to be missing from the data included in the linkage analysis.

In general the references are outdated. Chapter 4 feels like it was taken from an older review, rather than a recent look at the literature. Some of the ideas on Hg complexation are incorrect as a result. Relationships between landscape patterns and watershed chemistry have been strengthened with recent work in the Bay-Delta and the Western Hg Synthesis that isn't cited.

Anoxic bottom waters and MeHg production - There are several papers on MeHg production in the anoxic hypolimnia of lakes. MeHg may be produced both in the water column, and efflux from sediments when the sediment-water interface is anaerobic. oxygenation of the water column blocks bottom water methylation and significantly reduced efflux from bottom sediments - including Onondaga Lake papers by Matthews 2013 [Matthews *et al.*, 2013]. See also [Watras *et al.*, 1995] Eckley [Eckley and Hintelmann, 2006; Eckley *et al.*, 2005], and the METAALICUS lake [Harris *et al.*, 2007]

Inorganic Hg in sediment p 90. - Sediment Hg is a predictor of sediment MeHg across ecosystems and should not be discounted (See new synthesis in Hsu-Kim 2017). See [Fleck *et al.*, 2016] – Western Hg synthesis. For the large set of lakes and reservoirs examined,

including CA systems, the THg -MeHg relationship was weak ( $r^2=0.25$ ) but significant across the landscape. The results of the linkage analysis should be compared quantitatively to the relationships in Fleck et al.

Hg bioavailability: discussion on page 91 says that Hg<sup>0</sup> may be more readily oxidized and available than HgS. I don't think that's supported by real world data. The Bloom 2003 sequential extraction study of different Hg forms was never linked to bioavailability to microbes for methylation, and the extractions don't represent how Hg behaves in the anoxic conditions of sediments. Agree overall that that cinnabar has low availability however.

Also the availability of Hg in atmospheric deposition depends on the path and timing to sites of methylation – reactions in the watershed can make Hg much less available (sorption to particles prior to transport for example). Atmospherically-deposited Hg may have the same availability as mine waste after moving thru watersheds – it could react for form sulfides. The science of Hg source availability is NOT settled.

The neutrally-charged sulfide hypothesis has been updated. What's really going on is the formation of HgS nanoparticles that pass filters and appear “dissolved.” Hg in the presences of just about any measurable concentration of bisulfide precipitates as HgS, however particles interact with DOM to form colloids that reduce the growth of particles. DOM helps keep HgS particles small and bioavailable to microbes for methylation [Andrew M. Graham et al., 2012; 2013; A. M. Graham et al., In review; Zhang et al., 2012]. The practice upshot is that Hg can be highly bioavailable in sulfidic settings if DOM is present to reduce the rate of HgS precipitation. See a discussion of the process in Aiken [Aiken et al., 2011] and in Hsu-Kim 2017. HgS can be “dissolved” in the presence of DOM as well (Ravichandran papers).

P 92 **Wetlands** – Foundational references for the importance of freshwater wetlands in MeHg production in watersheds are [Driscoll et al., 2007; Hurley et al., 1995; Mitchell et al., 2008; St. Louis et al., 1994; Yee et al., 1995]. Several recent papers on MeHg production in Bay Delta wetlands by Marvin-DiPasquale and Windham Meyers are important, missing citations.

**DOC** p 93. Two key points to be made in this section: DOC is a carrier for Hg and MeHg from watersheds to reservoirs; DOC may enhance methylation rates in reservoirs. These are the mechanisms by which MeHg is related to DOC in surface waters.

Some of the strongest refs for MeHg export on DOC are local ones:

[B. A. Bergamaschi et al., 2011; Brian A. Bergamaschi et al., 2012]

The mechanism of DOC dissolution of HgS is not it's weak acid character, but the strong binding of thiols in DOC with Hg [Aiken et al., 2011; Deonarine and Hsu-Kim, 2009]

The key message for reservoir management is that high DOC systems (and systems with high DOC in inflows) are much more likely to have high Hg and MeHg levels. Perhaps high DOC/high Hg watersheds should be ones for early attention for remediation. DOC was not one of the parameters included in the linkage study, but should be included in ongoing data collection for reservoirs.



Fig 4.2 doesn't distinguish between Hg-contaminated systems and others. Suggest marking them, or noting that the orange arrows are locally-contaminated systems. It's an incomplete graph of the very large literature, but it makes the point that wetlands and reservoirs can be sources of MeHg production.

P 94 MeHg loss. Photodemethylation is probably the major loss term in lotic surface waters. However, MeHg degradation in sediments causes larger mass losses of MeHg in reservoirs. The mechanism for this loss is still being sorted out. It may be microbial [Oremland *et al.*, 1995] or more likely abiotic [Jonsson *et al.*, 2016]

Biomass removal. Removal of aquatic plants is a potential mechanism for reduction of MeHg production [Windham-Myers *et al.*, 2009].

Sweden has tried many MeHg remediation techniques for lakes including intensive fishing, liming for pH [Lindqvist *et al.*, 1991]. It would be wise to review their real-life experience over several decades in evaluating options for CA. See [Bishop *et al.*, 2009; Hultberg *et al.*, 1995; Munthe *et al.*, 2007; Verta *et al.*, 2010]

P 99 Reservoirs. See a new summary of reservoir effects/literature in Hsu-Kim 2017 (ICMGP synthesis paper, posted on ICMGP website).

The Western Hg Synthesis [Fleck *et al.*, 2016; Willacker *et al.*, 2016] provides quantitative relationships between reservoir characteristics and MeHg risk that should be cited and used in CA reservoir management planning.

**Chapter 5. Linkage analysis.** Very good statistical analysis of available data. But the available data have substantial limitations. The linkage analysis did not include several parameters that may be strongly related to MeHg production and MeHg in water or fish, including the degree of stratification or anoxia, the organic content of sediments, growth of submerged aquatic vegetation (the last two enhance microbial activity and MeHg production), DOC, and critically the loading rate of Hg to reservoirs.

I assume that Hg loading wasn't used because data aren't available. But sediment Hg concentration may not be a good surrogate for loading. Relationship between sed Hg and fish Hg doesn't consider the idea that Hg in sediments becomes less available for methylation over time (faster than burial). This is why source reduction is so impt. I suspect the linkage analysis is underestimating the efficacy of source control in reducing fish MeHg; because the model is based on sediment Hg concentrations rather than Hg load.

The use of unfiltered (and often spatially and temporally scarce) water MeHg data may be problematic. Unfiltered MeHg would include MeHg on particles including phytoplankton. It isn't really the amount of MeHg available to enter the base of the food web but may represent MeHg in the base of the food web itself. I couldn't tell if MeHg data for the reservoirs included in the linkage analysis were averages that included hypolimnia or not – and this could make a big difference in the outcome of the analysis. Unfiltered MeHg concentrations can be driven by particulate concentrations – which can vary enormously with season and depth.

The chl<sub>a</sub>:MeHg ratio was available for only ~40 reservoirs. MeHg and chl<sub>a</sub> data may not have been taken at the same season, depth, or frequency in all lakes. The multiple regression model that includes the chl<sub>a</sub>:MeHg ratio uses a subset of only 26 reservoirs.

The linkage analysis only included data from Hg-impaired reservoirs. An analysis that included all reservoirs may have shown stronger relationships between total Hg, MeHg and MeHg in fish.

The analysis conducted made appropriate use of available data, but could go further in acknowledging the limitations imposed by the data. Model 1 includes only a small subset of all CA reservoirs. The use of unfiltered MeHg data reservoirs makes this model more uncertain than it might appear in the formal statistical analysis presented.

Goal of no detectable water column MeHg (at a 0.009 ng/L DL) is a great goal, but may be unachievable in some reservoirs.

Hg loading rates from mines – don't have a good measure of this – using number or density of mines in the watershed as a proxy? Is there any data to suggest this is a good proxy?

I don't see a measure of stratification or bottom water anoxia in the data set. – does dam height capture this? Is chl<sub>a</sub> related to anoxia?

Despite the linkage analysis, I remain skeptical of the conclusion that “mercury source control alone cannot achieve the sport fish target” - because of the limitations of the linkage analysis.

What does “goals lower than natural background” mean – are these sediment concentration or loading targets?

Recommendations for sampling/monitoring in reservoirs undergoing TMDL implementation:

- switch to filterable and particulate MeHg for the water column
- conduct more detailed temporal and spatial sampling of MeHg and chl<sub>a</sub> in the water column, especially in stratified lakes
- collect data on the volume and duration of anoxia.
- MeHg in sediments.

Sediment goal: Sediment Hg concentrations and assessment of background values: would these be more informative if normalized to sediment organic content? What is the avg organic content of reservoir sediments? Either target on p 125 – 2 or 20 ug/kg – are low values in the context of available data for the US (See Fleck 2016 for example). The Western Hg synthesis found an avg on 29 ug/kg for lakes, near the Water Boards suggested target. Suggest putting the CA data into context with the Fleck synthesis. Recommend collecting grain size and/or organic content for reservoir samples collected going forward, and incorporate Hg:LOI ratios into models going forward.

**Chapter 7** – Source remediation.

The report noted (p 183) that “it could take decades to centuries for industrial-era mercury in watershed soils to be depleted” – meaning that contaminated soils could be contributing to high Hg in fish for a very long time. It’s certainly true that reservoir fish in mining areas are impaired more than 100 years after the CA gold rush.

Clean up should focus on flow paths to reservoirs, to reduce continued erosion of contaminated sediments that leads directly to reservoir Hg loads. In soils disconnected from main flow paths in the watershed, Hg may become immobile to leaching and transport over time. Solute transport of Hg from soils in these areas may be effectively zero (Oswald, 2016?)

Did staff consider that concentration-based TMDL load allocations for suspended sediments (instead of mass loading to reservoirs) could be a problem for watersheds with large suspended solid loads, or a high fraction of upstream soils that are contaminated. I understand that a concentration-based allocation is easier to measure and enforce. Nevertheless...

In any case, implementation of Hg load allocations as “management practices” and not clean-up standards will need to be evaluated once data become available for more reservoirs after remediation. The only available study data listed is for one mine (Gambonini) and no mention is made of resulting Hg load reductions to a downstream reservoir.

P 185. Mine site density may or may not relate to reservoir Hg loads. While no other data relating to reservoir Hg loadings may be available, the Water Boards should recognize that mine site density may be a very weak predictor of Hg loads from mine sites and mine waste in stream channels.

P 186 Lake San Antonio, Lake Nacimiento comparison. Here’s an obvious case of mines in the watershed impacting Hg in fish. Yet the discussion centers around the fact that the reservoir w/o mines has fish levels of 0.27, which is somewhat above the sportfish target. The state of California CA will be lucky to reduce Hg in the most impaired reservoirs given the number of impaired reservoirs and available resources. I find it unlikely that CA will be able to focus management tools on reservoirs like Lake San Antonio where fish Hg is only slightly higher than the sport fish target. The discussion also includes other factors that might result in differences in fish MeHg... when the obvious remediation approach is mine clean up.

## **Chapter 9. Implementation Plan.**

P 262. I agree with this approach: “The Water Boards encourage a coordinated approach for fewer, focused tests rather than tests in all mercury-impaired reservoirs.” Testing in more than a few reservoirs would be very expensive and incurs more risk of unknown consequences.

Pilot management testing will require a high level of measurement and monitoring. This should be more explicitly built into this TMDL implementation plan, so that resources for

that monitoring can be built into planning and cost estimates. There is danger in implementing too many strategies in too many lakes w/o a hard look at results along the way.

A outside technical review panel should be involved in design of remediation plans, not just evaluation of results.

P 264. Pilot tests should explicitly include mine site (and stream channel) remediation, along with detailed monitoring of results.

Potential water chemistry pilot tests: Is the TMDL implementation program open to options other than those listed here? There are several other potential approaches, including:

- approaches to reduce loading, like sedimentation basins
- addition of clean sediment to lakes by eroding clean soils
- in situ Hg sorption technologies
- decrease in nutrient loading to increase light penetration (demethylation) and reduce organic load to sediments (decrease methylation)
- reduction in sulfate load in systems with WTP, industrial, or mining sources of sulfur

Per Table 9.1 there are 74 reservoirs in Phase I TMDL implementation. This 303(d) impaired doesn't include the reservoirs that might be designated impaired based on fish sampling in the last few years. How would those reservoirs be staged for implementation?

### **Section 9.1 Mine clean-up**

There is no mention of potential available funding for mine clean up that will have to be paid for by state or local governments. It's hard to judge the potential time frame of efficacy of even Tier 1 clean up without some estimate of resources and cost. But it seems that this undertaking could be hugely (billions) expensive and take decades. Have the number of Tier 1 mine sites (or reservoirs) been identified?

Appendix I, p 4. "...the Reservoir Mercury Control Program will not pose new economic costs or environmental impacts to address discharges from mercury and gold mines." ... because existing regulations already require clean-up. But mine remediation has just barely begun – there is a huge disconnect here. If the new TMDL implementation program does not force additional clean up through allocation of additional state, federal and local funds, the TMDL loading goal will never be met.

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Gerald W. Bowes, Ph.D.  
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July 24, 2017

Dear Dr. Bowes:

As requested I have reviewed The Scientific Basis of the Proposed Plan Amendment to Establish the Statewide Implementation Program for Mercury in Reservoirs and the associated supporting documents that were supplied to me via the Water Boards FTP site. As discussed in our earlier correspondence, my review focused on conclusions 3,4,5,6,7 and 8, but also touched on conclusions 1 and 2. These are the areas where I have the most scientific expertise.

My detailed review is given below. Please let me know if you need further information or if there are questions about my review.

Sincerely



Dr. Daniel (Dan) Jaffe  
Professor and Chair, Physical Sciences Division  
School of Science, Technology Engineering and Mathematics (UW-Bothell)  
Professor of Atmospheric Sciences (UW-Seattle)

## Summary and “big picture”

In general the staff report is a remarkable review of mercury biogeochemistry. While I do have significant comments, and a few concerns over the modeling component, I think the staff should be congratulated for producing such a high quality scientific report.

Mercury is a complex problem and there are many scientific uncertainties. I appreciate that the State has conducted a detailed and comprehensive review of the fish mercury problem and proposed some workable solutions. In general terms, I am in agreement with many of the conclusions that form the scientific basis for the mercury provisions, but have some significant comments/concerns on the atmospheric deposition modeling that was conducted to do source apportionment. This will be discussed in the section on conclusions 6, 7 and 8 (sources). Other “big picture” comments:

1. The program proposes to use “adaptive management” (including modest fertilization) and continued research on Hg to guide future policy decisions. This is a very important step as there is much we do not understand about the sources and biogeochemical cycling of mercury. I strongly recommend that the state reinvest a fraction of the implementation costs on research to improve the scientific basis of these actions.
2. The documents use a variety of terms involving “background” (e.g. “natural background”, “modern background”, “global background”, or just “background”). As near as I could tell, these were never defined and there is a great deal of ambiguity in the documents over their meaning. I recommend that the report include some over-arching definitions of all of these terms and stick to these definitions throughout the report.
3. The conclusions use the term “many reservoirs” several times, but without providing specific %. It would be helpful to provide a % for each conclusion in these statements.
4. The fact that the land area around many reservoirs (half) have naturally occurring mercury is an important and supported conclusion (pg 6-5 and conclusion #4). In these cases it is the presence of the reservoir (or its management) that enhances fish methylmercury. I suggest that conclusion #4 be restated to clarify this (see suggestion below).
5. While I did not review in detail conclusions beyond #8, I did review section 7.2.2 concerning future global sources contributing to deposition in Chapter 7.2.2. While it seems reasonable to expect that California emission sources will continue to decrease, I was surprised at the level of reduction assumed for global sources. On page 7-19, the report states “...anthropogenic sources outside of California incorporate a 50% reduction from the 2001 baseline...” This is a highly optimistic conclusion based on a 2008 AMAP study. It is not clear what time frame is relevant here. More recent and much more carefully done studies indicate a reduction in deposition over the continental US from global sources suggest that for the year 2050 global non-US anthropogenic emissions may decrease deposition by a few percent or as much as 10% by the year 2050. These same studies also suggest that global emissions could continue to increase as countries like India develop. There is much uncertainty around future global emissions, but a 50% reduction in deposition from global sources seems highly optimistic and inconsistent with the most recent published studies. See for example:

Corbitt et al. Global Source–Receptor Relationships for Mercury Deposition Under Present-Day and 2050 Emissions Scenarios. *Environ. Sci. Technol.*, 2011, 45 (24), pp 10477–10484. DOI: 10.1021/es202496y.

Giang and Selin, Benefits of mercury controls for the United States. Proc. Natl Acad. Sci. 113 (2), 286–291, 2016. [www.pnas.org/cgi/doi/10.1073/pnas.1514395113](http://www.pnas.org/cgi/doi/10.1073/pnas.1514395113).

### **Specific comments on conclusions 1-8**

#### **Conclusion 1:**

I concur with this conclusion.

#### **Conclusion 2:**

I would agree that the first two factors are clearly important in controlling fish methylmercury. The evidence for the third factor, water level fluctuations, is much weaker. On page 5-11, the report states that this factor is weakly, and negatively associated with aqueous methylmercury. Only by including this factor in the multiple linear regression model does it show up as “significant”. I would argue that this demonstrates an overall weak controlling influence and, as such, its inclusion in conclusion 2 is probably over-stated. In addition, due to uncertainties in the atmospheric deposition and modeling of deposition, I would argue that we do not currently know how important deposition is in directly controlling fish mercury concentrations.

#### **Conclusion 3:**

I agree with the general sense of conclusion 3, but there are problems with several parts. First, the term “primary” is problematic. Certainly sources of inorganic mercury are a necessary ingredient in mercury impairments. Are these “primary” or not? Judgement call, what do we mean. I would suggest wording such as “Inorganic sources of mercury, by themselves, do not determine mercury impairments in California reservoirs.” Next the term “amount of mercury” is too vague. Are you referring to the concentration in the reservoir or a flux into the system? Is this THg or something else.

#### **Conclusion 4:**

I find the wording here confusing. What is natural background? What % of reservoirs fall into this category? As worded, it sounds like the goal is to reduce fish mercury to levels that are lower than a “natural background”. Is this really the intent? The conclusion might be reworded to something like “Many reservoirs (%) have inorganic sources or fluxes in that are near background/natural levels (define). However the presence of the reservoir and/or reservoir management have resulted in increased mobilization of that mercury and increased the concentration of methylmercury in fish.”

#### **Conclusion 5:**

This is certainly true. I agree with this conclusion.

#### **Conclusion 6:**

This is a broad conclusion and covers the three primary sources. As such the statement is largely correct. However I have significant concerns on the atmospheric deposition modeling, discussed below.

#### **Conclusion 7:**

Suggest minor edit to “Reducing watershed mercury sources alone is not expected...” Might add an additional sentence “Source reductions combined with management actions are needed to reduce fish methylmercury in many reservoirs.”

### **Conclusion 8:**

This conclusion depends heavily on the atmospheric deposition modeling, which is problematic. See below.

### **Atmospheric deposition modeling:**

Modeling can provide a useful tool to estimate source-receptors relationships, transport processes and deposition. Models are also very useful to identify sensitivities to key processes. However models are not a panacea for all environmental analyses, their underlying assumptions must be stated and evaluated and the results must be evaluated with observations. Even in cases where the modeled result and observations agree, it is possible to get the right answer for the wrong reason. However in this case, the model appears to have little skill in reproducing the observations.

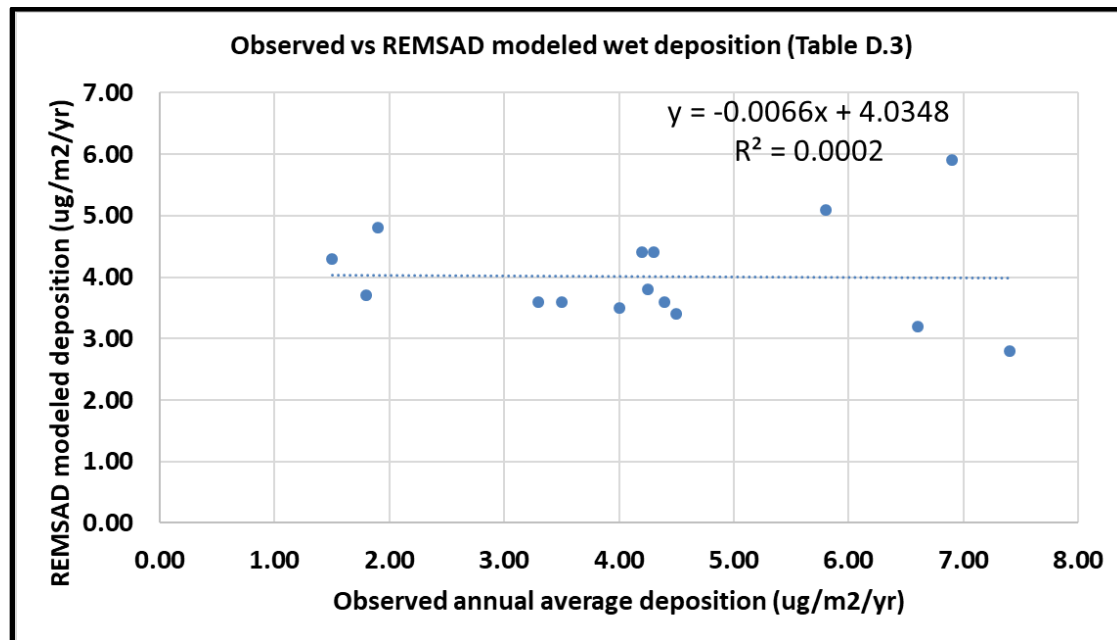
For this analysis, the REMSAD model used was used. Based on the citations, it appears that the most recent model evaluation took place in 2008 (Bullock et al 2008). However it is important to note that the Bullock analysis actually made no comparisons with observations (in contrast to what is implied in Appendix D (“the model was found to be reasonable.”). In fact the three models differed by up to a factor of 10 for some parameters. Dry deposition showed strong disagreements between the three models and this is particularly problematic since dry deposition is thought to be a large fraction of the deposition over California (Figure 6.17). So it is not clear what is meant by the statement in Appendix D “the model was found to be reasonable”.

It appears the model has not been updated since 2008 despite some significant progress in our understanding of mercury cycling. For example the REMSAD uses ozone and OH as the sole oxidants for elemental mercury, yet we now know that these oxidants are almost certainly not relevant and that halogens are probably the dominant oxidant. See:

1. Gratz, L. E., et al., Oxidation of mercury by bromine in the subtropical Pacific free troposphere, *Geophys. Res. Lett.*, 42, oi:10.1002/2015GL066645, 2015.
2. Shah, V., et al., Origin of oxidized mercury in the summertime free troposphere over the southeastern US, *Atmos. Chem. Phys.*, 16, 1511-1530, doi:10.5194/acp-16-1511-2016, 2016.
3. Horowitz, H.M., D.J. Jacob, Y. Zhang, T.S. Dibble, F. Slemr, H.M. Amos, J.A. Schmidt, E.S. Corbitt, E.A. Marais, and E.M. Sunderland, A new mechanism for atmospheric mercury redox chemistry: implications for the global mercury budget, *Atmos. Chem. Phys.*, 17, 6353-6371, 2017.

In addition, I was surprised at the lack of discussion on source profiles with respect to mercury speciation. It is known that industrial sources that emit mercury in the Hg(II) form will have much greater local deposition, compared to Hg(0). What is known about the California emissions and how well is this speciation understood? While the total mercury from these facilities is probably reasonably known (+/- 30%), the speciation will have much higher degree of uncertainty. What is the speciation, what is the uncertainty and how important is this?

Finally, in terms of model evaluation, Table D.3 provides a possible look at the model's ability to reasonably reproduce observations. This was not done for the report, so I made the graph myself. The graph below shows the annual wet deposition from observations in California vs the REMSAD modeled value. Two sites, which had multiple observed values, were averaged in this analysis. The observed values range from about 1.5 ug/m<sup>2</sup>/yr to 7.4 ug/m<sup>2</sup>/yr, whereas the modeled values range from 2.8 to 5.9. Some of the sites with the highest observed mercury wet deposition have the lowest modeled values. The graph below shows that the REMSAD model has essentially no skill at reproducing wet deposition fluxes in California.



However as mentioned above, dry deposition is even more important than wet for most of California. Unfortunately there are few or no observations of dry deposition in California. But given the large uncertainties and model disagreements in Bullock et al (2008) this is certainly a large uncertainty in the analysis.

So in summary, the largest uncertainties associated with the REMSAD source attribution modeling arise from:

1. Incorrect oxidation mechanism for Hg(0).
2. Unknown accuracy of emission inventories and speciation of emissions.
3. Model failure to reasonably reproduce observed wet deposition fluxes in California.
4. Inadequate data to evaluate model dry deposition.

A good summary of model uncertainties relevant to policy are given in:  
 Kwon, S.Y. & Selin, N.E. Curr Pollution Rep (2016) 2: 103. <https://doi.org/10.1007/s40726-016-0030-8>.

**Other comments:**

Pg. 5-3: I do not understand the last sentence in the first paragraph “Consequently, staff proposes a goal for reservoir...”.

Pg 5-3, next to last bullet point: Natural background is unclear here. A natural background would be much lower than a present day background....

Pg 5-9, second paragraph: the R value of 0.2 is very weak, despite the P value. Given the challenges with the modeling, what do we really infer from this?

Pg 5-20, next to last paragraph: This introduces several new variables (methyl mercury production, food web transfers) . I don't recall seeing these anywhere in the document up to this point (possibly in an appendix?) How are these defined and measured?

Pg 6-6, second bullet: I assume this is for soil or sediment?

Pg 6-10, recommendations: I assume there should be a "<" (less than) symbol in front of these values (e.g. <0.1 mg/kg).

Pg 6-11, second thru fourth bullets: There are large uncertainties here. I agree that these definitions are useful, but you should point out that these ranges overlap.

Pg 6-24, second bullet: I assume these are direct anthropogenic emissions and exclude re-emission. Suggest to clarify this.

Pg 6-27, third line from bottom: I did not see any comparisons between the model and observations in any part of the report. The comparison I showed above indicates very poor model performance for wet deposition.

Pg 6-28, second bullet: The discussion of "global background emissions and "re-emissions" is confusing. What are global background emissions? Why are re-emissions only considered for one year when it is the net accumulated deposition that causes these emissions?

Pg 7-16, first factor: The linkage analysis found a minor relationship between modeled deposition and fish methylmercury, but given the uncertainties in modeled deposition, I find this result rather inconclusive.

Pg 7-18, bottom: No timelines are given for these allocation. The 66% reduction factor for California sources seems reasonable.

Pg 7-19, top: The 50% reduction factor for global sources is highly optimistic and inconsistent with recent published studies.



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**University of Connecticut**  
**July 26, 2017**

## **Response to Assumptions, Findings, and Conclusions:**

Note: As the conclusions were answered mostly in order there is much more comment and discussion earlier in the document as many concerns and comments are provided in the first instance. Therefore, much of what is commented on in the earlier statements is pertinent to other sections of this document, and I have attempted to indicate this where applicable.

### **1. Many factors—not just the amount of inorganic mercury in water and sediment— influence methylmercury concentrations in reservoir fish.**

The statement is entirely true that the amount of total mercury (Hg) in a system does not often provide a good prediction of the concentration of methylmercury (MeHg) in fish. Chapter 4 provides a detailed overview of the many factors that can influence the concentrations of MeHg in water and sediment, and therefore in fish, and has accurately covered most of the important variables in sufficient detail to provide a suitable conceptual model of the factors that influence fish MeHg. Overall, I felt that the references seemed skewed to older publications and I am sure there are more recent studies that should be cited. There are some missing details and more information on some aspects would improve this discussion and the development of the conceptual model:

1. The report indicates that methylation in the epilimnion is important but the implication is that this methylation is still in the sediment. Recent studies have highlighted the potential for methylation in periphyton and in biofilms on surfaces, and within settling particles, and this should be discussed.
2. The role of reduced sulfur in binding Hg in sediments and influencing methylation was not really discussed. While this is less important in freshwaters than in saline systems it is worth some discussion.
3. There have been recent studies indicating the potential for the formation of colloidal Hg in the environment and that colloidal Hg can be available to methylating bacteria. Again, this may not be a big issue for reservoirs and I don't know if there are any studies examining this but it may be worth mentioning when discussing bioavailability to methylating organisms.
4. There is very little discussion in Chapter 4 of the potential importance of demethylation in sediments, and in the water column, which is both abiotic and biotic, and how this may impact net MeHg in fish, and the factors that may influence this demethylation should be discussed, even though there is likely no strong specific information about the controlling mechanisms for this process in the literature. There is some reference to demethylation and its potential impact on water MeHg in Ch 7 but it could also be mentioned here.
5. It is stated that the fish concentration correlates strongly with aqueous MeHg and Fig. 4.4 is given as an example. Again, while the data may not be for reservoirs, there are studies in the literature that indicate that the relationship to aqueous MeHg is not always that strong, especially for studies that compare across systems. Within one ecosystem the relationship may be strong but across many it may be weak. The assumption that aqueous MeHg is the key is that the partitioning into the particulate phase is constant across ecosystems and that may be reasonable for systems where most of the biomass is algae and where suspended solids levels are similar, but this may not be so across all the reservoirs. The concept of biodilution is discussed and this could be important in confounding the relationship between water column MeHg and fish MeHg.

6. Fig. 4.5 is given to support the idea that MeHg in fish correlates with MeHg in sediment. Again, for reservoirs of similar size and depth this may be so but there are likely to be other variables that impact this relationship in many reservoirs. Some discussion of the effect of physical conditions (size, depth etc) on this relationship would be useful.
  7. While there is discussion of the role of anoxia there appeared to be little discussion of the effect of eutrophication on the MeHg in fish in Chapter 4. Again, maybe this is not a big problem in CA reservoirs but some discussion of the link between eutrophication and fish MeHg in Chapter 4 is needed. In Appendix A, there is discussion of the potential impact of increasing nutrient levels as a mitigation strategy but the potential impact of too many nutrients needs to be highlighted. It is clear that the problem of nutrient limitation is more important than the opposite of excessive nutrients. There is some more discussion of the impact of nutrients in Ch 7. In discussing the impact of nutrient levels, most of the impacts are highlighted but there appears little mention of the potential for longer food chains in more oligotrophic systems which would lead to higher fish MeHg. Also, the sequestration of phosphorous (P) in sediments can be altered by anoxia or low oxygen conditions which could lead to its release from sediment. This should be discussed as its sequestration in sediments could change with changes in nutrients and ecosystem status.
  8. The dissolved MeHg/chlorophyll ratio also takes into account the likely impact of higher biomass on influencing dissolved organic carbon (DOC) levels and therefore the bioavailability of MeHg to the base of the food chain. Some more discussion of the complexity of the role of DOC may be useful – it is not always the case that higher DOC leads to lower bioaccumulation and the type of DOC plays a role. There are studies that suggest for example that Hg bound to DOC may be more available for methylation than Hg bound to other ligands. The complex role of DOC in Hg and MeHg cycling could be further highlighted in Ch 4, and discussed further in Ch 7. The role of DOC and higher plankton levels on light penetration and therefore photochemical demethylation and the likely impact of this is not mentioned in Ch 4, although it is discussed in Ch 7. While biotic methylation is not well understood there is increasing evidence that it can occur in the water column and maybe there is some manner in which this could be enhanced in some reservoirs over others. This could be worth mentioning.
  9. Also, there is little discussion of the potential for removal of Hg from the reservoirs by Hg reduction and evasion. As with MeHg photodemethylation, this is likely related to DOC levels and TSS, but there could be the potential for enhancing net reduction and evasion of Hg from the reservoir. There is essentially no discussion of this pathway and its potential importance in the cycling of Hg within the reservoirs. For the ocean and large lakes (e.g. Great lakes), evasion is the most important Hg sink and its importance has also been shown for smaller lakes, and could be more important for oligotrophic reservoirs.
  10. The implication in much of the discussion is that sulfate reducing bacteria (SRB) are the principal methylators in reservoirs. Given recent research about methylating genes and the role of other organisms, this may not be completely correct. Some discussion of the role of other organisms in methylation in CA reservoirs is needed.
  11. The correlation model in Appendix A appears to capture most of the important variables, and as indicated, the ratio of dissolved MeHg to chl-a includes the impacts of many of the other variables that can influence MeHg production, fate and transport, and bioaccumulation.
2. **The three most important factors that control fish methylmercury concentrations in California reservoirs are: the ratio of aqueous methylmercury concentration to chlorophyll-a concentration, aqueous total mercury concentration, and annual reservoir water level fluctuations.**

1. This conclusion is based on the evaluation of the available data for reservoirs which does not include all reservoirs and is therefore the best empirical evaluation of the controlling factors given the data. However, the variables that are important make scientific sense and are reasonable and indicate that it is the trophic dynamics that have more control over the fish MeHg levels than the relative differences in net production of MeHg across the reservoirs. This is likely the result of similarity in the conditions across the reservoirs and the fact they are mostly oligotrophic and therefore differences such as degree of anoxia in bottom waters in summer, sediment organic content and its redox chemistry, and other factors that are more important in other ecosystems are of less importance for CA reservoirs.
2. While the model includes four variables (two as a ratio) there are other variables of some importance that have been found in other systems that clearly do not have as much importance for the CA reservoirs because of their likely similarity in the reservoirs from where the data was available. However, the importance of these variables may become more apparent as more data is collected on other reservoirs. Many of these other variables are discussed in more detail in Ch 7.
3. The targets that are derived from the analysis are reasonable based on the data but it is not clear that these are easily attainable and so this is of course the major problem. As noted for the total Hg in sediment criteria the derived value is lower than background levels and therefore the criteria are set for a reservoir based on the background value in the region. That levels have to be at background makes this very difficult to achieve as there still may be a small "reservoir effect" in many locations which would likely make the reservoir exceed the criteria. One issue that should be mentioned is the potential impact of stimulating algal production - a suggested remedy - on the concentration of Hg in sediments. Increased algal biomass has the potential to lead to more deposition of organic material to the sediment and this may lead to a higher concentration of Hg being stored in the sediment over time, and therefore changing primary productivity may lead to the reservoir exceeding the criteria. There is some mention of these links in Ch 7. In many ecosystems there is a relationship between sediment Hg and organic content especially at low organic matter levels, which is likely representative of the reservoirs given that they are oligotrophic – the sediment organic content and its potential impact is not really discussed in the report. Was this one of the variables that was considered in Ch 5? Perhaps some consideration should be given to normalizing the reservoir sediment Hg to OC when comparing it to the content of the background Hg in the watershed as differences in OC likely will be important – for example a watershed with high forest coverage probably has higher Hg and OC levels than one that is not. Overall, throughout the report there is discussion of a potential controllable variable as if its impact is in isolation, but most of the factors that can be manipulated to influence MeHg concentrations could easily result in other changes that could lead to an increase in MeHg in the longer term. The overall timescale of these interactions will differ and this should be discussed and acknowledged in the report.
4. The dissolved MeHg level of 0.009 ng/L is the concluded level for protection given the specific fish concentration of 0.03 mg/kg. One issue is whether such low levels can be measured on a relatively routine basis and it is indicated that this should be possible. Indeed, such low levels are found in ocean waters and routinely measured by investigators in these waters although intercalibration studies show that there can be high variability in the reported levels at lower concentrations. Therefore, if this low level is to be used then there needs to be an excellent QA plan associated with the reservoir TMDL to ensure that the values reported are accurate and precise, either by having one accredited lab doing the analysis or having the labs participate in regular low level intercalibration studies. This issue needs to be discussed in the report as this is not a trivial undertaking.
5. The idea of trying to stimulate algal growth and biodilution is put forward and based on the information put forward from the literature and the current oligotrophic status of the

reservoirs this appears to have merit but it needs obviously to be carefully controlled. It may also however be that it is not just the major nutrients that are limiting in the reservoirs. While there has been less study in freshwaters, there is definitely evidence that trace metals (e.g. Fe, Mo) may be limiting productivity in lakes, and so if this is the case, then adding major nutrients could lead to a shift in the species composition rather than a stimulating of the existing algal species in the system. While this may not be a major issue, it should be considered. Iron limitation may be important if the systems have oxidic sediments. Another issue that is not really discussed is whether the change in primary productivity would leave to a shift in zooplankton and other secondary consumers, or how it may affect the relative amount of benthic to pelagic production. As noted in the report, benthic production is an important component of system productivity. The timescale over which studies that have been done to examine the impact of adding nutrients is not detailed in the report, as it may be that changes associated with changing nutrient dynamics could be slow.

6. Other things that could be manipulated that are not mentioned here but are touched on later in the report is whether it may be possible to alter the seasonality of the water level drawdown as this may lessen its impact. Also, in the systems that do have low oxygen bottom waters in summer, water column oxygenation may be useful. Some discussion of these may be warranted to indicate that they were considered but not found to be appropriate. While it may not be of use for the reservoirs, there has been success in the addition of nitrate to bottom waters in Onadaga Lake in NY. While this is likely a completely different system, some mention of this would be appropriate here – it is mentioned somewhat in Ch. 7.

**3. Inorganic mercury sources alone are not the primary driver of mercury impairments in California reservoirs. Multiple factors drive reservoir fish methylmercury levels: amount of mercury, methylmercury production, and bioaccumulation.**

This statement is valid and the reasons have been well outlined and any additional information needed is described in the comments above.

**4. Inorganic mercury levels in many reservoirs would need to be lower than natural background to achieve the TMDL targets<sup>1</sup> and mercury water quality objectives if no other factors are addressed.**

1. I am assuming this statement refers to Hg in sediments. This statement is the logical conclusion of the analysis in Ch 5 and elsewhere and is the basis for the conclusions for 2 and 3 above that other factors need to be addressed besides Hg inputs. The implications of this conclusion are that, for example, sediment levels should not be lower than the “modern” background at the reservoir locations, as discussed above. This does raise the concern that the criteria are not attainable and the obvious implication of this is that if levels have to be reduced below modern background concentrations then the organism that this level is designed to protect may have been exposed for a long period – of course, the reservoirs are not natural but it is likely that lakes in the region would behaved similarly historically. The fish level values that are determined for the protection of humans and wildlife will have uncertainty associated with their calculation and perhaps this needs to be further considered in evaluating how this statement is possible. There is likely a large error range in the exposure estimate. This may be discussed in the report somewhere but I might have missed it but perhaps some measure of the uncertainty in the wildlife and human estimates needs to be incorporated into the evaluation. As a simple example, most exposure estimates use a single value for human assimilation of MeHg from food, but recent studies suggest there is actually a much wider range in this value. Can the uncertainty be incorporated into the choice of the sediment levels associated with the various risks?

2. Manipulation of environmental conditions to try and achieve goals, and to reduce one effect by perturbing the system in another way, has always the potential for unknown adverse effects and there are many historical examples of this. The report states that the reservoirs would revert back to the pre-perturbed state if nutrient addition is stopped and claims evidence to support this but it is not clear over what timescale the experimental perturbation was enacted, and the recovery monitored. Further, over what timescale would the interventions be considered as it appears that there may need to continue such interventions indefinitely.
  
5. **Mercury sources are not evenly distributed across the State and no one source type is responsible for all reservoir impairments.**

This is a relatively obvious statement given that mining and other point source inputs are not evenly distributed around the state, and that external inputs such as atmospheric deposition do not dominate the inputs for many of the reservoirs.
  
6. **The most important anthropogenic sources to impaired reservoirs are historic mine sites and atmospheric deposition from global and local (California) industrial emissions.**

This statement is reasonable because the location of other potential point sources and other anthropogenic inputs (urban runoff (this is however primarily derived from atmospheric deposition), wastewater treatment plants effluent etc) are in locations relatively far removed from the location of most of the reservoirs in the state. However, this is not entirely true and so in a way the statement is too definite – maybe it would be better to add: “...to the majority of the impaired reservoirs...”. Also, modeling indicates that a small fraction of the Hg in atmospheric deposition in CA comes from anthropogenic emissions within the state, and further that a small fraction of the reservoirs are impacted by these CA-based anthropogenic emissions. Therefore, the inclusion of the local (CA) emissions in the statement seems to indicate that these sources may be more important than they are. Overall, regulation of emissions in CA would have a small impact on Hg inputs to the reservoirs. In total, the statement is supported by the presented information and analysis of the distributions of sources in the state and the locations of the reservoirs but could be altered to focus on the most important sources to the majority of the reservoirs.
  
7. **Reducing watershed mercury sources is not expected to result in substantial reductions in reservoir sediment mercury concentrations and fish methylmercury concentrations in many reservoirs.**
  1. This statement appears to contradict the Statement 6 above and some of the others statements. Above, it is concluded that historic mining sites are an important source. Is this statement referring to the natural and background sources in the watersheds or all sources in the watersheds? This needs to be clarified and better supported in the document. Perhaps this background information for this statement could be better presented – there is little detail in Ch. 6. Is there some subset of reservoirs where this could be true, based on reservoir size, watershed area, watershed Hg levels, water depth, trophic state etc as one could envision factors that could make this true for some locations – e.g. very low sedimentation rates, larger watershed and a small number of mining sites, naturally Hg-enriched soils in the watershed etc. If this statement is correct, then why is there a proposed outcome and an effort in some of the statements below to reduce inputs from historic sites. Surely this will always have an impact? Additionally, the timescale needs to be considered and this is not discussed. While I have no idea of the sedimentation rates, they could be less than a cm per year, and therefore with sediment mixing due to benthic organisms, it

would take many years for the sediment concentrations to change. There is no discussion of the timescale or the expected response time of the reservoirs to changes in watershed inputs. I would expect this response time to be many years to decades and perhaps this is the reason why this conclusion has been reached. While water bodies will respond reasonably rapidly to changes in atmospheric inputs the timescale of response to changes in watershed loading are much slower. This has been examined in a number of modeling papers and these should be detailed and discussed in the report.

2. There could be a further categorization of the reservoirs. As noted in the report in various places, the watershed/reservoir area ratio varies by many orders of magnitude so combining the data for large reservoirs with small watersheds with small reservoirs with large watersheds will lead to confusion in the driving factors as these would be different – atmospheric deposition in the first case and therefore changing watershed inputs would have little effect, while the opposite would be true in the latter case. Overall, the major differences in the sources to the different reservoirs, and the complexity of issues such as water transfer between systems, which is outlined in section 6.8, is not properly conveyed in this statement and the others, and a better effort is needed to do so.
  3. While many statements are put forward in a general way there appears to be little generality in terms of the actual reservoirs, their sources and it appears to me that in most cases each will have to be dealt with as an individual case with little ability to extrapolate from “case studies” as is proposed.
8. **Global industrial emissions are the predominant anthropogenic source to about 20 percent of mercury-impaired reservoirs.**  
This statement is entirely based on the computer modeling results and therefore is valid if there is confidence in the ability of the computer model to reflect reality. There is no other way with the current understanding and information available to evaluate this in another manner – mercury isotopes could help perhaps but this approach is still being developed and likely would not be sufficient to provide a conclusive answer. There are other computer models in the literature and it would be useful perhaps to compare the REMSAD results with other models if possible.
9. **There are a variety of mercury source control options and reservoir water chemistry and fisheries management practices that may be effective for reducing fish methylmercury concentrations.**  
This is true and the determination of which will be the most effective will require an evaluation for each reservoir as it is likely that each will have a unique set of inputs, and factors influencing in situ net methylation, that will need to be considered. It is not clear to me given the large difference in the reservoir characteristics, their locations, the size relative to the watershed etc that it will be easy to extrapolate results from one reservoir to the next, or even to identify without further study and sample collections which approach may be the best for a particular reservoir.
10. **A combination of source control actions and reservoir and fish management practices—versus source control alone—will be needed to achieve both timely and measurable fish methylmercury reductions in most of California’s mercury impaired reservoirs.**  
This appears a valid statement based on the information provided and discussion in the report.
11. **Actions to reduce fish methylmercury levels may need to vary for each reservoir because of the many combinations of different mercury sources (e.g., some are natural or global and therefore not regulated by state and federal agencies), competing factors that control methylmercury production, and reservoir operational constraints. Reservoir-**

**specific characteristics and operational requirements and mandates may not allow for all methylmercury management tools to be used in all reservoirs. Even so, there should be a possible solution to mercury impairment for every reservoir.**

This leads on from the previous statement however I am not sure that the final sentence is entirely valid. This is a hope rather than an expectation, I would conclude, as it is indicated that there could be a limit to which strategies could be invoked and therefore there is no *a priori* reason why success is guaranteed. Also, what is the timescale of expectation here.

**12. The TMDL loading capacity and allocations, combined with reservoir water chemistry and fisheries management pilot tests and implementation actions identified in the proposed program of implementation), are adequate to achieve the proposed mercury water quality objectives and TMDL numeric targets for methylmercury in reservoir fish.**

1. The load allocation approach seems confusing as there is allocation for external sources of inorganic Hg as well as an allocation for the *in situ* formation of MeHg. The relative impact of these loadings may not be adequately taken into account because of the reliance on concentrations rather than actual loads. For example, a small reservoir with a large watershed with mining contamination would have a very different source allocation (high inputs from the watershed of contaminated sediments compared to a small *in situ* net production of MeHg for a specific concentration because of reservoir size) than a large reservoir with a small watershed. Maybe these extremes don't exist but the reservoirs are not relatively similar in size and in relative watershed/lake area. Therefore, it is easy to imagine that the allocation approach may lead to the disproportionate allocation to one source over the other, and therefore the result would be incorrect assignment of the necessary source reduction. The information in Tables 3.2 & 5.1 indicate that the watershed/lake area ratio varies by many orders of magnitude, and the dam height by a factor of about 200, so there is the need to take these concerns into account when assigning inputs on the basis of concentrations in particulate load and MeHg concentration in water. This needs to be discussed in more detail in the report.
2. The allocation of a site to a particular region may be easily assessed but this is not clear on reading the document but it is of substantial importance given the differences in allocation for "mineralized" versus "enriched" areas in particular. I would have liked to have seen a better justification of the choice for these values. This was not adequately described in the document.
3. It was not clear to me how the atmospheric loading would be allocated – based on the reservoir surface area relative to the total area of CA, or on the watershed ratio? Whichever it is will have an important impact of the allocated importance of atmospheric inputs the large watershed to lake area ratio. While atmospheric inputs are not important for most of these reservoirs, the allocations should still be done in a scientifically defensible manner that is well articulated in the report.
4. Given the timescale over which the TMDLs will be implemented there may be the need to redefine the atmospheric loading allocations in the future.

**13. The allocations are adequate for both current and future mercury sources to the mercury-impaired reservoirs.**

This statement relies on the projections of future global Hg emissions being accurate as there are a number of reservoirs that have atmospheric deposition, predominantly from global sources, as their major input (Statement 8). So, this statement should be refined to indicate this. Also, given that one of the sources is *in situ* production of MeHg, this may change due to other factors besides inputs of Hg such as climate-related increases in temperature, rainfall etc so there is the potential for other climate changes to impact this "source". In addition, the timescale of the term "future" needs to be stated more clearly.

**14. The Reservoir Mercury TMDL incorporates an adequate margin of safety.**

This is not well described in the report and is difficult to assess. The choice of the upstream loading concentrations is based on the available information and appears to be conservative in terms of the current data. The atmospheric inputs are constrained by the modeling results but the distribution of inputs between CA and global sources is a model result and not based on any actual data, and therefore is as valid as the errors and uncertainties in the model predictions.

**15. The Mercury Reservoir Provisions requirements for inorganic mercury controls are adequate to reduce anthropogenic discharges of inorganic mercury to reservoirs.**

The requirements are consistent with the scientific report and therefore are reasonable if the concerns and caveats outlined above are considered and either shown not to be a concern or are incorporated into the revised versions of the documents.

**16. During the first phase of the implementation program for the impaired reservoirs, the Mercury Reservoir Provisions require reservoir water chemistry and fisheries management practices pilot tests. Implementing reservoir pilot tests to develop and evaluate and water chemistry and fisheries management practices in a phased approach is an adequate approach to reduce reservoir fish methylmercury levels. This phased approach includes State Water Board review of the Mercury Reservoir Provisions prior to full scale implementation of effective and feasible management practices.**

The approach appears reasonable, and the oversight is required, given that it is unlikely that there will be a consistent approach and set of actions for the reservoirs. While the report suggests in numerous places that there may be test sites that would allow application to other similar reservoirs, I think this is likely not to be the case as the variables that are proposed for manipulation are not independent and therefore the actual changes will depend on the location and exact characteristics of a particular reservoir and it is unlikely that extrapolation will be possible without actual testing in each case. So, it is likely that each reservoir will be a “special case”. If experimental manipulations or other measurements are done it would be worth making the measurements to assess the relative importance of external sources of MeHg to the reservoir.

**17. The upper 90th confidence limit of the mean is an appropriate statistical method to determine compliance with water quality objectives based on an annual average fish methylmercury concentration. In addition, it is appropriate to use consistent fish trophic levels and sizes, sample type and quantity, and sampling locations when determining compliance with water quality objectives.**

I am not a statistician but the 90<sup>th</sup> percentile appears a reasonable confidence limit and is protective in the majority. The first part of this conclusion is best evaluated by someone trained in statistical methods. However, the second part which relates to the use of consistent fish trophic levels and sizes in making allocations has definite merit, and I agree with this approach. The more detailed and consistent the approach to examining compliance based on fish concentration, the more valid will be the outcome of the determination of compliance.

**18. Biosentinel fish monitoring provides a means to evaluate relatively rapid changes to biotic methylmercury levels.**

Many papers, books and chapters on monitoring have endorsed the approach of using resident, young of the year fish for determining change due to a particular implementation even if the overall evaluation of compliance relies on the concentration in a larger, higher food chain species consumed by humans and wildlife.