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Delta Mercury Control Program Phase 1 Tidal Wetlands and Open Water Methylmercury Control and Characterization/Control Reports – Independent Scientific Review

A report to the Delta Science Program

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Delta Stewardship Council
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1. Executive Summary

This Independent Scientific Review (henceforth referred to as “Review”) considered two Control and Characterization Study Reports (2019 Control Studies and this Tidal Wetland/Open Water Report) to evaluate compliance with approved work plans and to assess the scientific validity of the results and conclusions of the work. The Review Panel (henceforth referred to as “Panel”) was also charged with providing an overall assessment of the studies to inform an Advisory Panel that will ultimately guide the implementation of the Mercury Control Program.

The Review Panel recognizes the significant progress that is made in understanding mercury dynamics in the Yolo Bypass/Delta through the development and testing of two different modeling efforts presented in the Report. These are complex models of complex systems, and it is apparent to the Panel that the work presented here represents a first step toward the development of models that may be used for critical scenario testing. That said, they are far from usable in a predictive way at this stage due to challenges in model validation as well as the quality of input data and definition of boundary conditions. Extensions to these efforts are encouraged through refinement of the representation of critical processes as well as the addition of a biological component to address methylmercury bioaccumulation and biomagnification.

The Open Water Technical Studies provided valuable input data and boundary conditions for the models, confirmed pre-existing knowledge about the mercury and methylmercury dynamics of this system, and provided new empirical data. The impact of some of these findings was weakened by high variability, flaws in design, incomplete measurements, and poor representation of the heterogeneity of the land cover, soil types, and mercury in the Yolo Bypass. Despite strong statements in support of some potential control measures (e.g. disking), the Panel cannot support these conclusions yet given the weaknesses identified. A careful re-examination of data for these studies is warranted. Similar statements apply to the Tidal Wetlands Study, where the Review makes recommendations concerning data quality and interpretation.

Overall, the empirical studies and the models point to the same conclusion: the overwhelmingly dominant source of mercury and methylmercury is upstream inputs. Only through reductions of upstream sources which both supply methylmercury directly, as well as inorganic mercury which may be methylated within the Yolo Bypass/Delta, will meaningful downstream reductions be achieved. Within the Yolo Bypass/Delta, flooded agricultural soils are the largest source of methylmercury, to the point where other sources are likely of marginal significance (albeit with very high uncertainty in some cases). Although loads may be reduced, it is unlikely that methylmercury TMDL targets for the agricultural areas of the Yolo Bypass can be met. Even with high uncertainty, the Panel can say with some confidence that reductions from other small sources within the system are not large enough to offset these loads.

Finally, the impacts of climate change (temperature, precipitation regime, snowmelt, drought frequency/intensity/duration) remains inadequately explored, particularly given the potentially profound changes that are forecasted for the Central Valley of California. These changes can only be truly addressed through experimental data and down-scaled climate models linked to landscape simulations. When these climate-driven changes along with potentially substantial future hydrologic diversions and changes in salinity are factored into well-parameterized models, there will be considerably more certainty about the future impacts of measures implemented to control methyl-mercury uptake by biota in the Delta.

2. Acknowledgments

The Independent Panel wishes to acknowledge the substantial effort made by all of the authors and contributors to the studies and models included in this report. The Panel is also grateful to Dylan Stern of the Delta Science Program for his coordination of the Panel activities, scheduling of meetings, accommodation of requests for information, and overall patience with our deliberations and debates.

3. List of Key Abbreviations

ADCP: Acoustic Doppler Current Profiler

BMP: Best Management Practice

D-MCM: Dynamic Mercury Cycling Model

DSM2-Hg: Delta Simulation Model, version 2, Mercury Model

Hg: Mercury

LA: Load Allocation

MeHg: Methylmercury

PMeHg: Particulate methylmercury SSC: Suspended Sediment Concentration

TAC: Technical Advisory Committee

THg: Total mercury

TMDL: Total Maximum Daily Load

uHg: Unfiltered total mercury

uMeHg: Unfiltered methylmercury

WLA: Waste Load Allocation

YBP: Yolo Bypass

4. Background, Intent and Scope

The Sacramento-San Joaquin Delta (Delta) is identified in the Clean Water Act Section 303(d) list of impaired water bodies due to harmful levels of mercury in some fish eaten by people and wildlife. In response, the Central Valley Regional Water Quality Control Board (Central Valley Water Board) developed a methylmercury Total Maximum Daily Load (TMDL) and associated Delta Mercury Control Program (Mercury Control Program) to control mercury and methylmercury in the Delta. The TMDL is the analysis of the methylmercury impairments, a review of the primary sources, a linkage between the sources and the impairments, and recommendations for mercury reductions to eliminate the impairment. The Mercury Control Program is the implementation program for the TMDL, and addresses controls for mercury as well as methylmercury. The TMDL and associated Mercury Control Program were adopted by the Central Valley Water Board as an amendment to the Water Quality Control Plan for the Sacramento River and San Joaquin River Basins (Basin Plan) in April 2010. The TMDL and Mercury Control Program received final approval from the US EPA on 20 October 2011.

(From the Independent Scientific Review Panel Charge)

4.1. Review Panel Charge and Approach

This report of this independent scientific review panel (Review Panel) is the review of the reports of tidal wetlands and open water characterization/control studies approved by the Regional Water Board. The Review Panel was asked to evaluate the completeness and scientific validity of the tidal wetlands and open water characterization/control study reports, determine whether the scientific methods used were consistent with those outlined in the workplans, and assess whether the conclusions were reasonably derived from the data and informed by the best possible scientific information. While the characterization/control study reports were the primary documents to be reviewed, the study workplans were also to be reviewed to ensure the stated scientific objectives were met in each study.

For this review, the Review Panel was provided with the following documents:

1. DWR - Tidal Wetlands

a. Mercury Imports and Exports of Four Tidal Wetlands in the Sacramento-San Joaquin Delta, Yolo Bypass, and Suisun Marsh for Delta Mercury Control Program Compliance April 3, 2020

b. Workplan for Methylmercury Import and Export Studies on Tidal Wetlands in the Sacramento-San Joaquin Delta and Yolo Bypass, December 20, 2013 “Tidal_Workplan_Wetland_121913_Final.pdf"

2. DWR - Open Water Studies and Model

a. The Final Report for the Open Water Control Study (August 31, 2020)

b. Open Water Workgroup Methylmercury Control Study Work Plan (December 20, 2013)

c. Technical Memorandum for the Methylmercury Control Study Workplan (December 20, 2013)

3. Supplementary Materials

Open Water

1. Memo summary Open Water original December 2013 work plan drafted by Jennie Fuller to inform the Review Panel #2 review of the Open Water Report "Memo Summary open water original December 2013 workplan"

2. First extension request approval from Central Valley Water Board, November 28, 2016, extending deadline from October 20, 2018 to December 31, 2019 "DWR 28 Nov2016"

3. Second extension request from DWR (April 24, 2019) to extend deadline from December 31, 2019 to June 30, 2020 "Open Water Extension Request 2019-04-24"

4. Second extension request referenced Final Sensitivity Analyses – part of second extension request "Final Sensitivity Analyses_042518"

5. Approval of second extension request from Central Valley Water Board (June 11, 2019) "Open Water extension request granted 190506"

6. Third extension DWR request (May 29, 2020) to extend deadline from June 30, 2020 to August 31, 2020 "Messer-Pulupa CVRWQCB MeHg Open Water Req for Ext FIN-SIG 2020-05-29"

7. Third extension Central Valley Water Board approval (June 16, 2020) "DWR Open Water Extension Approval Request"

Tidal Wetlands

8. Tidal Wetlands DWR second extension request (October 29, 2019) moving deadline from 31 December 2019 to 3 April 2020 "Tidal Wetlands Extension request April32020"

9. Central Valley Water Board approval of tidal wetland second extension request (November 27, 2019) "DWR Tidal Wetlands Extension Request Approval_112719"

10. Tidal Wetlands data from Final Report

The Review Panel Chair assigned two reviewers to each section of the Report. The reviews for the individual Reports/Chapters are included in Section 6 of this Report. Materials from the Open Water Report are divided among the technical studies, climate change impacts, and the two modelling efforts because of the structure of the Report. Detailed reviews were framed around the questions in the Charge to the independent Review Panel; however, it should be noted that, in many cases, the questions were not applicable to the Reports and/or were addressed in a more comprehensive way (e.g., Climate Change Impacts were synthesized in the Open Water Report as a separate chapter and were reviewed as such).

The Review Panel held an initial organizational teleconference and several mid-point teleconferences to present and discuss the reports. During the mid-point calls, overall assessments were presented. Information from individual report reviews and in-depth discussion from the review mid-point teleconference were incorporated into the Overall Review Panel Assessment. Individual sections of the overall Report were produced by the two Panel members and are included in Section 6 of this Report verbatim (with minor formatting corrections only) in order to preserve the independence of the reviews. This consequently results in some inconsistency in language and formatting. The Panel reviewed and commented on a draft of this final report prior to submission to the Delta Stewardship Council.

5. Overall Review Panel Assessment of Phase 1 Tidal Wetlands and Open Water Methylmercury Control and Characterization/Control Reports

5.1. General Feedback

The studies presented in the reports and associated technical appendices represented a substantial work effort both in terms of the research undertaken as well as the reporting thereof. Given documented challenges with execution of original workplans, revisions, and extensions, the Review Panel was impressed with the scope of work executed and the synthesis efforts in the form of both modeling efforts. Indeed, the development of models for the Yolo Bypass and for the Delta represents the first mechanistic synthesis of decades of mercury data collection and monitoring, and represents an important first step toward valid tools for scenario testing and prediction of future conditions. Feedback in this Review is intended to provide recommendations and improvements (where applicable) for the effective interpretation and application of knowledge generated through this process, as well as recommendations for extensions and future work.

The Panel review of the reports and associated appendices was complicated by a lack of coherent structure in presentation of the Report materials. Typically, such a report would comprise the main body of the report, with data and technicalities in appendices. As presented, the substance of much of the work presented was found in technical appendices, with much of the latter content not referred to in the main report, despite its importance. This confounded the review process, particularly considering the scope of the Charge to the Panel and limited time allocated to the Panel for the review. It is possible that some of this Review's critique of aspects of the report may be a result of the lack of coherence in presentation and lack of reference to specifics in the Technical Appendices that would have facilitated the review process.

One of the hallmarks of rigorous scientific reporting is reproducibility in collection, measurement, and presentation of data. Given the critical nature of some of the parameters reported to modeling efforts (e.g., diffusion rates, tidal water fluxes), Panel members committed effort to verify values, rates, and other measures presented in the Report. In some cases (discussed in the detailed reviews), the Panel was unable to reproduce critical values from the data provided, and/or generated outcomes that were different from those reported, and/or identified data quality issues that were not highlighted in the report. The Panel would like to emphasize that it does not conclude that the information presented, calculations, or conclusions drawn in the Report are necessarily erroneous. It is possible that the issue lies in the incomplete presentation of data, the lack of articulation of methods used, or assumptions made (but unreported). That said, the Panel encourages a thorough and critical assessment of the data, calculations, and interpretations where problems were identified in the detailed reviews, particularly for values and rates that then support critical parameters in the models. The Panel also encourages the open curation of all data collected and utilized in studies undertaken as part of the methylmercury TMDL effort so that third-party analyses may be undertaken as extensions to these studies or others. This is particularly relevant for data that is not mentioned in the formal reporting. Technical Appendix F contains invaluable data on the distribution of mercury and methylmercury from nearly 70 sites in the Bypass that were sampled by various groups (DWR and USGS) for this study, with other valuable ancillary data. The appendix provides maps of Hg, MeHg and %MeHg across the Delta, but no raw data were included. These data were used in the Yolo Bypass Model but were not formally analyzed in any other way. Despite being underutilized for this Report, these data have extraordinary value for future synthesis efforts and must be made available.

5.2. Specific Charge Questions

As part of its charge, the Review Panel was asked to consider a series of overarching questions to inform the Phase II Advisory Panel. The issues raised by these questions encompassed all of the studies, and as such are presented globally rather than on a per study basis. Where applicable, consideration is given to the first half of this review concerning the Control Studies submitted August 2019.

1) What additional information would be needed, if any, to adapt the studies' results for changes in climatic and hydrologic conditions in the Delta?

The first charge question concerning adaptation of results to changes in climatic and hydrologic conditions in the Delta (and the second question related to the individual studies) was addressed as a full separate chapter in the Open Water Report (Chapter 6; and reviewed in more detail in Section 6.2 of this Review) and incidentally elsewhere. The Review Panel considers this question to be of considerable importance. Future changes in fundamental controls on mercury biogeochemistry (e.g., temperature, vegetation type) and hydrology (evapotranspiration, precipitation frequency, duration, and intensity, snowmelt, seasonal flooding frequencies) are critically important to the question of the suitability and feasibility of load allocations. It is the Panel's opinion that there was insufficient consideration paid to adaptation to future changes in climate and hydrology in the current Report and as such, considerable additional information is required.

Temperature: Changes in water and sediment temperature will impact net methylmercury production via methylation and demethylation; likewise, water temperature changes also will affect Hg(0) volatilization fluxes which will impact the overall mass Hg balance for the system. None of the likely effects are mechanistically explored in the Report either experimentally or conceptually. Indirect effects of changes in temperature include the duration and amplitude of drought conditions, timing of snowmelt, and effects on vegetation, which are important factors governing load allocations but are not discussed nor evaluated. The most defensible means of assessing climate change impacts is through modeling, especially since system-level feedbacks truly cannot be considered in any other way.

Precipitation: The effect of climate change on precipitation, the intensity of storms, the extent of snowpack, and seasonal runoff are all linked through the expectation that in the future, there will be more precipitation as rain than presently and less as snow, and therefore, there will be a change in the timing and magnitude of delivery of water (and mercury) into the Delta region. The report makes no predictions of how these changes would affect methylmercury within the Delta and its supply.

Soil Wetting and Drying Cycles: Changes in both temperature and precipitation regimes will lead to a change in frequency in soil wetting and drying cycles, which will influence the oxidation of organic matter and reduced sulfur, and thus the release of both inorganic mercury and methylmercury. As well-documented mechanisms that regulate net methylmercury availability, this should be further considered to better constrain methylmercury production in the Yolo Bypass. Although not approached this way in the Report, an examination of wet vs. dry year differences in mercury biogeochemistry and loading would allow for some inferences to be made about these processes at a larger scale.

Changes in Particulate and Dissolved Mercury Transport: Climate change predictions indicate a high likelihood of more intense storms, more sediment transport, and a change in particle transport of Hg/MeHg in the system relative to historic conditions. The impact of more intense storms on the MeHg loading is difficult to predict, but the likely impact of these changes is more MeHg production and transport to the Delta. Additional work clearly needs to be done to develop more detailed and robust (including improved statistical model specificity where appropriate) predictions of the impact of changes in precipitation on MeHg transport delivery to, and mobilization in, the Delta. The role of dissolved organic carbon (DOC) quality and quantity is clearly important for Hg transport, for maintaining dissolved Hg in solution (protection from photodegradation), and its impact on methylation and is discussed in some detail in the report, but more mechanistic details on the relationships between Hg/MeHg and DOC are needed in in order to better understand future changes and resultant impacts on loads. Current modeling presented in the Report does not evaluate changes in DOC concentrations, nor its chemical character and will represent an important avenue for future investigation.

Other Climate Change Impacts: The impact of sea level rise on levee failure and resultant impact on MeHg levels is discussed in the Report in more detail than any other aspect of predicted climate change effects but is focused on the aspect of levee breaching and flooding and not the impact of salinity changes in the system. Changes in water and soil biogeochemistry as a result of sea water intrusion are also potential impacts that warrant future investigation.

The potential increase in wildfire frequency and extent are reasonably addressed in the report with increased release of Hg to the atmosphere and subsequent local deposition. However, the Panel believes that the effect of fires on the transport of Hg from the watershed is likely of much greater importance due to increased erosion and runoff from burned areas. This is not discussed in the Report but contributes to increased uncertainty in input estimates from the upper watershed.

Other Hydrological Changes Affecting Load Estimates and Allocations: Human alteration of Bay-Delta hydrology through hydrological “short-circuiting” has the potential to significantly alter load allocations and change biogeochemical conditions due to freshwater diversion. For example, the proposed “Delta Conveyance/Delta Tunnel” will place two very large water intakes in the north Delta, further decreasing natural freshwater flows. This would be a near-future disruption that would impact both hydrology and Hg cycling and would exert a cumulative effect on top of climate change. However, this is not discussed in the Report.

The Role of Modeling: Clearly the impacts on hydrology by climate and land-use change/water diversion are far beyond the scope of a model focused on mercury in the Delta. Climate change impacts must be tackled by combining climate model down-scaling with regional hydrological simulations, which is feasible using current knowledge, science, and existing modeling approaches. Simulations from these models could then define the input conditions for models such as those presented in the Report for the Yolo Bypass and the Delta. This type of sequential modeling would then assess if the potential impacts outlined above are likely subject to feedbacks that are not accurately described on an impact-by-impact basis, but rather for which a cumulative impact may be confounded and only reasonably discernible through an appropriate model framework. Although neither mercury model included in this Report is at the stage of development needed to test scenarios such as this (current limitations are discussed further in the Detailed Reviews), the Review Panel sees further mercury model development, and integration with regional simulations of climate/hydrology as the path forward.

2) How comprehensive is the current understanding of methylmercury sources and processes in the Delta and its tributaries? Is this extent of knowledge sufficient to identify scientifically robust mercury/methylmercury controls that will meet fish tissue objectives? What are the critical gaps in information that would prevent scientifically sound conclusions to be made regarding mercury/methylmercury controls?

As posed, this question goes far beyond the scope of the Panel Review of the Characterization and Control studies that were completed as part of this overall Review. Substantive scientific effort has been applied to this complex question by many agencies and researchers since the CalFed program, and the full synthesis of the disparate components comprising this knowledge has yet to be undertaken (and would be a worthwhile exercise). From a broad perspective, the major methylmercury *sources* in the Delta and its tributaries are known (e.g., upstream sources, Yolo Bypass seasonally-flooded soils) but are understood mechanistically to greater and lesser degrees. The extent of knowledge is insufficient to identify robust, actionable controls that will meet fish tissue objectives beyond the conceptual stage at this point.

It is clear from the work presented in the Characterization/Control studies that at the scale of the entire Delta, the Sacramento River is the dominant source of methylmercury (model estimates of >50%, largely consistent with prior empirical mass balance estimates). While controls associated with this source are beyond both the scope of the TMDL studies and the scope of this review, such controls are nonetheless clearly related to the remediation of historically contaminated sites in the watershed. The importance of this input of mercury and methylmercury to the Delta is well known and ultimately must be addressed if movement toward the load allocation is to be realized.

The Yolo Bypass seasonally-flooded vegetated lands have also been previously identified as important sources of methylmercury to the Delta, and the Characterization/Control studies in this report confirm (and to some degree refine) this knowledge. Some weaknesses in the diffusion and vegetation studies raise questions about the findings presented (see detailed reviews). However, even with this caveat, the production of methylmercury is largely related to the flooding of vegetated land, and results from these characterization studies suggest that pasture lands may be the largest source within the Bypass. Although the technical studies explored land management strategies for the reduction of methylmercury production, the findings are preliminary at best and their application largely unfeasible. The removal of vegetation (through cultivation or grazing) had a net benefit of reducing methylmercury production. However, modeled scenarios that simulate the complete removal of ALL vegetation only reduced net methylmercury production by ~50% from pasture land cover. Larger reductions were seen in other land cover classes (Rice, Wetlands) with smaller net contributions. Even with some land management practice changes, the likelihood of achieving these reductions is slim, and when combined with the requirement to manage private landholder practices, the likelihood is reduced further. The Report authors were cautious in recommending that discing and vegetation removal are feasible BMPs given the limited scope of the experimental work presented (independent of other limitations raised in this review) and the Review Panel concurs.

The models that were presented in this Report have made progress toward a system-wide model of both methylmercury and inorganic mercury sources and sinks; however, considerable work remains before these approaches can be used in a predictive way. The current understanding is lacking in the aspects that are outlined above in the report and many of these sources, which mostly relate to internal production of MeHg within the Delta, have not been well characterized or studied and should be a focus on any future research on the Delta. The current Delta model predicts that the Delta is essentially a conduit for transport of Hg and MeHg through the system, with some net removal to the sediment, and little internal formation of MeHg and release into the water column. This is likely a simplistic and inaccurate prediction of a more complex system where mixing processes enhance net MeHg formation and other substantial sources of MeHg production and flux into the water exist besides the bed sediment. This needs to be further studied. With the current model conclusion, the management strategy will be clearly focused on the external sources. This may result in the lack of successful management if there are important internal sources that have not been properly quantified and modeled.

As indicated in our prior review, the Review Panel has concerns that the TMDL exercise draws a relatively simplistic link between the source of mercury (or methylmercury), methylmercury concentrations in water, and mercury concentrations in fish tissue in the Delta. Both methylmercury that is discharged directly and inorganic mercury that may be methylated in receiving waters may ultimately be bioaccumulated, and there are complex factors that regulate bioavailability, bioaccumulation and biomagnification. More information is required about the relative sources and contributions of methylmercury and inorganic mercury that drives the production of methylmercury that is then bioaccumulated in biota, rather than presuming that all loads are created equal and contribute proportionally to fish tissue mercury concentrations.

As there is no bioaccumulation model yet developed for the Delta, the impact of such factors on the prediction of change in MeHg load with climate and hydrological changes cannot be predicted with the current information and model. The bioaccumulation model was part of the work plan initially, and the development of the model should be pursued. Additionally, if needed, more information should be collected on the levels of MeHg in biota to allow for model testing and validation. It is suspected that there is sufficient biota information available, but this may be lacking for the lower levels of the food chain. Further, the significant complications that are imposed on this understanding by future climate change, land-use change, and hydrological manipulation must be incorporated into prediction scenarios.

3) Do the characterization/control study reports provide adequate support to change the Delta mercury load and waste load allocations? If so, in what direction (e.g., increase or decrease) does the report support a change in waste load allocations?

The mass balance results (Open Water Technical Studies) and the Yolo Bypass and Delta modeling efforts all convey similar findings with respect to the importance of internal sources of methylmercury. The Yolo Bypass is the largest internal source of methylmercury, and both experiments and model scenarios suggest that the WLA objectives (nearly 75% overall reduction) are unrealistic. The production of methylmercury is largely related to the flooding of vegetated land, and results from these characterization studies suggest that pasture lands may be the largest source within the Bypass. Although the technical studies explored land management strategies for the reduction of methylmercury production, the findings are preliminary, limited in scope, and their application challenging. The removal of vegetation (through cultivation or grazing) had a net benefit, but as described above, is unlikely to be implemented in a way that would allow for the loads to approach those allocated in the TMDL.

The tidal wetlands study currently concludes that tidal wetlands are not significant sources of MeHg to the Delta. At face value, such a conclusion would suggest that the current allocation could be reduced. However, it is our opinion that the study's conclusions are not yet adequately supported. For example, the mass balances for methyl and total mercury to the Yolo Bypass and the Delta are not robustly determined and have considerable uncertainty. Further analysis of the water balance and periods of clear wetland flooding are still needed to better understand these systems.

4) Do the cumulative study results address the ability to control inorganic mercury and methylmercury sources to attain assigned load and waste load allocations?

The cumulative study results directly and indirectly address the ability to control inorganic mercury and methylmercury sources to attain load and waste load allocations. The prior review of Control Studies from this Panel (August 2019) concluded that the sources investigated (e.g., urban stormwater, municipal wastewater) were already (or would be with planned upgrades not related to the TMDL) significantly below WLAs. The very small contributions from these sources (relative to those presented in this report that their WLAs) could be reduced (with the caveats concerning data quality and uncertainty presented in the August 2019 Phase 1 review). That said, reductions in these WLAs would not make substantive impacts on the overall load allocations for the system.

As noted previously, the characterization studies considered in this review were not intended to address controls in support of assigned load allocations, and few recommendations indeed are made in this regard. The only controls discussed focused on land management practices in the Yolo Bypass (discussed above) and as indicated are unlikely to help attain WLAs. The tidal wetlands study concluded that control measures may not be needed due to the negligible net contributions of methylmercury, but it is our opinion that this conclusion is not yet adequately supported. Some analysis of relationships with other ancillary chemistry were completed in the study, but these findings are not clear enough to infer possible control measures, if indeed they are needed.

5) Does the evidence presented in the reports suggest that if mercury/methylmercury loads from the studied sources were reduced to the assigned allocation or beyond, would the reduction be sufficient to offset loads from other sources either within the Delta or upstream?

Despite flaws in the studies and reporting, the Review Panel is confident in stating that changes to WLAs associated with the small stormwater and wastewater sources (August 2019 Report) could in principle offset loads from other sources to a very minor degree; in the case of larger loads the offset would be well within the margin of uncertainty and therefore may have little effect on the overall mass balance. The evidence presented in the Characterization/Control Report combined with the Review Panel's familiarity with past studies and the scientific literature suggests that the load allocations to critical compartments such as the Yolo Bypass agricultural lands and wetlands are not likely to be met, and as such cannot be used to offset loads from other sources. The Review Panel concluded that there is insufficient evidence (or confidence in that evidence) to draw clear conclusions about the loads from other compartments investigated as part of this Report (e.g. Tidal Wetlands). Indeed, models identify other potential net methylmercury sources (e.g. Liberty Island) that are wholly uncharacterized. Upstream sources remain the majority input of inorganic mercury and methylmercury to the Delta. Control measures within the Delta cannot be expected to offset these external inputs without significant proactive measures to reduce the magnitude of the upstream sources, such as the remediation of upstream mercury mine sources. Only *the combination* of controls focused on the Yolo Bypass seasonally-flooded lands and remediation of upstream sources will lead to meaningful future reductions in surface water

6. Detailed Reviews of Characterization/Control Studies and Modeling

6.1. Open Water Report: Yolo Bypass Technical Studies

This chapter summarizes a number of extensive lab and field studies designed to help parameterize the Yolo Bypass model. It provides estimates of overall net MeHg production within the bypass during a flood year, as well as experimental estimates of MeHg flux from soils and vegetation during flooding.

The technical studies associated with the Open Water Report represent a significant work effort, and were largely designed to support the Yolo Bypass Model with empirical information that were identified as required, but not well constrained by available data. They also represent a substantial redirect from the original workplan which was to focus on the use of experimental ponds to evaluate BMPs for methylmercury. The Review Panel considered the Reported technical studies at face value and aligned with the scientific objectives of the revised workplans. The reviewers also agree with the overall conclusions of the Report that scaled up experimental testing of control BMPs remains an important task to be undertaken.

In the context of the Technical studies, many of the Review Panel Charge questions are not applicable. They are explicitly not designed nor intended to address Load Allocations (Q.1a) nor provide alternatives for improving load allocations (Q. 1b). Changing climatic conditions (Q.2d) were addressed collectively in the Open Water Report Chapter 6 (reviewed separately in this section). The review comments here are focused on Charge questions 2a (new or additional information), 2b (design of future control studies), 2c (identification of knowledge gaps), and 3 (commentary on scientific issues related to the study).

The Yolo Bypass Technical Studies comprise four main work packages:

- Yolo Bypass Mass Balance Study
- Sediment-water Flux Study
- Gust Chamber Erosion Study
- Vegetation Senescence Study

Each work package is discussed separately here in the context of the relevant Charge Questions that are specific to the technical review and framed differently from the overall charge questions described above. The Reviewers expressed several general concerns about this Chapter of the Open Water Report. As noted in the overall assessment, this Chapter in the Report failed to capture the depth of scientific information presented in the substantial related Appendices to the Chapter, making thorough scientific review challenging. There is considerable complementarity between the data presented here and prior work that is not quantitatively synthesized. As presented, the data were difficult to extract and use. Errors and omissions in data tables, and lack of full context/metadata limit the useability and value of these important studies, and in some cases prevented an independent assessment of study findings from raw data.

6.1.1. Yolo Bypass Mass Balance Study

The Mass Balance Study, a field study of Hg and MeHg inputs and outputs to the bypass during the 2017 flood season, was of high utility. This sampling program provided critical context for the other experimental work and confirmed that the YBP is source of MeHg to receiving waters during flood years. However, the magnitude of that source is highly uncertain. The study was quite limited in its sampling program, with only a few sample dates and grab samples rather than flow-weighted samples. There was also no sampling within the Bypass, which could have provided spatial information on sources of MeHg. Despite these shortcomings, it expands on previous studies by providing another season of data and the first information for the lower Bypass (the "Stairsteps").

Question 2a: Do the study results provide new or additional information about mercury or methylmercury sources, mass balances, or loads in the Delta or Yolo Bypass?

MeHg production during floods. We agree that the field data from 2017 support the finding of internal MeHg production in Yolo bypass during the 2017 flood season. As noted in Chapter 3, net MeHg production in Yolo bypass was also demonstrated by another study for the 2005-6 flood year. The comparison with the 2005/6 data was valuable, showing that inputs were very roughly the same for the two seasons, although exports appeared to be higher in 2005-6. However, because of differences in study design between the two studies, it's really not possible to quantitatively evaluate differences in net MeHg production between two flood years. Observations also support the idea that much of the MeHg production that occurs during flood season happens soon after initial wetting. The drying and rewetting effect on MeHg production has been observed in many other ecosystems. For the Yolo bypass, it appears that initial MeHg production may be in soils as they wet up; while MeHg production in decaying vegetation contributes more MeHg later in the flooding cycle. The study is based on a previous similar work by Foe et al. (2008) except that the current study importantly added dissolved (filtered) Hg and MeHg (Foe et al., 2008 focused on total/unfiltered samples.

Question 2b: Will the studies help design future control studies?

This study does not contribute to the design of future control studies; however a well-designed ongoing mass-balance sampling program is the most effective means by which to assess the impact of controls in the future.

Question 2c: What knowledge gaps must be filled to inform future control studies or management strategies?

The reviewer's agree with Report recommendations about adding other internal points of measurement in future mass balance sampling in order to both better spatially constrain areas of MeHg contributions from the YBP, as well as provide improved parameterization and validation of models. Higher frequency data during floods, at minimum capturing rising and falling limbs of major events, but preferably using flow-weighted sampling would allow for inference about processes of MeHg production and mobilization. Data presented in the Report hint at the value of this but fall short in utility. Very little data were collected for much of the study period and most of the Report is focused on a longer, but single, flooding event in 2017. There were some smaller events investigated, but sampling of these events generally was only a single time point. To base loading on a single sample is extremely weak in establishing a net sink or source role for the Yolo Bypass. In the Reviewer's' opinion interpretations of the smaller two events are simply not possible because of this. Similarly, the "First Flush" sampling for the 2017 flooding provides interesting and important hints but is relatively unconstrained. This is a relative term at best and generally TSS (and associated) total Hg concentrations peak in most systems ahead of peak flow. In this case it is unclear to me if "first flush" is actually more akin to "first sample", which would technically be different.

We recommend caution in the use of proxies, like TSS for total Hg, or DOC fluorescence for MeHg. These proxies only work if Hg inputs and MeHg production stay relatively constant over time. If used, the relationships between proxies and Hg or MeHg should be checked routinely, and direct Hg/MeHg measurements should be done routinely for some portion of continuous flow-weighted samples (perhaps 10%). .

A strategic and scientifically informed extension of the annual mass balance sampling will address gaps in knowledge about the landscape level contributions of MeHg from different management areas and land-use covers in the YBP. The difficulties in drawing connections between disparate years of mass balance sampling is made clear in this report. Annual sampling for mass balance is very straightforward, low-cost, and very high return in terms of understanding the interannual dynamics of the system, trends related to climate, and ultimately the efficacy of control measures. With no consistent mass-balances conducted before, there will be no means by which to differentiate the effects of control measures from background variability.

Need for risk assessment separate from MeHg fate and transport models. Importantly, net MeHg production and efflux from Yolo Bypass during floods is only one part of exposure, bioaccumulation and risk. MeHg accumulation within the Bypass was not examined and may be the major contributor to risk to biota utilizing the system. This has been demonstrated for other systems, e.g. tidal wetlands (Kopec et al. 2018) and rice fields (Ackerman and Eagles-Smith 2010).

Need to understand the distribution of Hg in soils across Yolo bypass. Identification of hot spots of Hg accumulation and net MeHg production within the Bypass is critically needed for BMP development (more below). This would include a map of soil Hg concentrations across the bypass, as well as field evaluation of MeHg production from different landuse and ag types during flood.

> *Question 3. Do you have comments on other scientific issues related to this study or mercury/methylmercury source type?*

High uncertainty. There is very high uncertainty surrounding the magnitude of net MeHg production and flux from Yolo during flood (given as 42 ± 42 g/d on p3-62). The variability 'isn't all that surprising given the complexity of the water flows in the system being monitored, and the very limited data collection (less than a dozen sampling points thru time, all grabs, and all on inputs and output structures). However, a better seasonal estimate of total MeHg flux will be needed for managers who will want to monitor the efficacy of any BMPs put in place to limit MeHg production and exposure.

Hydrological uncertainty especially for outflows (as one might assess from the large differences in modelled vs. assumed outlet flows, for instance), is high, even if it is somewhat arbitrarily given a relatively low uncertainty in the study. Given the size of the Yolo Bypass, it is surprising that there was no consideration of atmospheric inputs or possible groundwater inputs/outputs. It is possibly that the majority of the mass balance is dependent on surface flows in and out of the Bypass, but it would be good to know with some level of uncertainty that other possible inputs or outputs of water and mercury are indeed unimportant. This seems especially important in light of forcing a closing of the water budget based solely on outputs presumed to match inputs, particularly where SCHISM model estimates for outputs do not agree.

The mass balance assumes that differences in inputs vs outputs equate to a net production of MeHg in the Bypass. This can only reasonably be interpreted as a combination of MeHg production *and mobilization*. If much of what is being observed as an increasing load at the outlet were as much MeHg bound to particles in soils that are then mobilized, this would suggest a nuanced or possibly different approach to control, compared to attempting to reduce the actual production of MeHg (and then still its mobilization with flows through the Bypass).

This study assumes that "A net increase in filter-passing MeHg (fMeHg) loads is indicative of an increase in diffusion of MeHg from sediment porewater. Conversely, increasing particulate MeHg (pMeHg) loads suggests increased suspension of organic and inorganic particles enriched with MeHg." This may be reasonable, but it is an untested assertion. Given the large size of the Yolo Bypass, it would be reasonable to expect that this behavior is **not** conservative. This is an important consideration because it will drive future decisions about control measures. Although outside the scope of this particular study, it is important to point out the implications of the assumptions made.

The majority of MeHg (even unfiltered) samples do not have particularly high concentrations. The average unfiltered MeHg is 0.09 ng/l, which suggests some methylation capacity in the system, but not on the order that one might observe for other wetlands or newly flooded reservoir systems that have been studied around the world. Given the otherwise quite high concentrations of total Hg entering this system, one must presume that limitations on Hg bioavailability for methylation would be a worthwhile focus for understanding impacts in this system overall. We do believe there is quite a lot of literature in this region on this particular topic.

There does not appear to be much interpretation at all related to quite a number of ancillary chemical analyses done, including for potentially helpful things like boron and manganese. There are methods and some results given, but nearly no interpretation of how variability in some of these parameters might help for inference of certain processes. This seems to be a lost opportunity.

Attempts to correlate loadings with water flows are difficult to interpret as that these are collinear variables. A strong linear relationship in this case can simply be inferred as a relatively unchanging Hg concentration in relation to increases in flow. The point of these analyses appeared to be to infer the role of flooding more land and how this would therefore impact MeHg production. A relatively straightforward and direct approach would have been to look at satellite or other imagery through time to assess the actual extent of flooded land.

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6.1.2. Sediment-Water Flux Study

Soil cores from several sites in Yolo bypass were used for laboratory evaluation of efflux of total and MeHg from soils upon flooding. The goal was to provide estimates of flux for several land use types for use in the Yolo Bypass model. Soil Hg and MeHg concentration data were also used to evaluate differences among land-use types.

Question 2a: Do the study results provide new or additional information about mercury or methylmercury sources, mass balances, or loads in the Delta or Yolo Bypass?

The Report concludes that overall, seasonally flooded Yolo Bypass soils are generally a source of MeHg during flood events. We agree that the data support this conclusion. Porewater MeHg concentrations generally exceeded MeHg in overlying water of experimental cores and MeHg generally accumulated in overlying water over time.

We agree that these the results provide "insight to the chemical speciation of Hg and the biogeochemical conditions that control Hg cycling and transport in the Yolo Bypass." The flux measurements were carefully done, with associated biogeochemical data to help understand the control of flux. QA/QC for experimental data look good. Fluxes were appropriately estimated both from porewater profiles, and from the change in concentration in overlying water over time. This type of information can be used to extrapolate to sites with similar, measured biogeochemical characteristics. Unfortunately, the was little of no analysis of data in this way. Flux data was evaluated (qualitatively) with regard to sediment Hg and MeHg concentrations, and they were unsurprisingly related. But much other low-hanging data analysis was not done.

We **do not agree** that this study provides a quantitative evaluation of differences in Hg and MeHg concentration or flux among land use types within the Yolo bypass. Spatial coverage was quite limited, with only one site used to represent each land management type. There is not enough spatial distribution information to provide good estimates of concentrations or fluxes by land use type.

These data provide a start on understanding fluxes of Hg and MeHg from YBP soils. Unfortunately, the data **do not** directly support the conclusion that "This work clearly demonstrates that different land use types in the Yolo Bypass introduce different amounts of Hg to overlying water during a flood event, have different Hg methylation potential, and contain different Hg solid phase reservoir size." (p C-24). With a sample size of one for each land-use type it is impossible to separate site differences from land use or land management differences.

We believe that this study provides a broad range of MeHg flux values for flooded soils that are appropriate for use in the model.

However, the data should not be used to assign different values for specific land practices. The estimated range of "open water" MeHg flux values overlaps significantly with experimentally estimated fluxes from vegetated soils. Note also that while this study was designed to estimate MeHg fluxes soils in "open water" during flood, the cores used may have been vegetated, so the model separation of sediment/water flux and flux from decaying vegetation is somewhat artificial.

The report concludes that "Land use type is an important consideration in the Yolo Bypass with respect to Hg movement between sediments and overlying water, Hg methylation potential, and

Hg solid phase reservoir size." We agree, but that conclusion was probably already apparent without additional data collection.

Question 2b: Will the studies help design future control studies?

This work was not related to control studies *per se*, but data like this is of value as it can complement data on MeHg production in different land-uses/areas of the YBP.

Question 2c: What knowledge gaps must be filled to inform future control studies or management strategies?

Although a useful start, the limited spatial coverage of the core sampling limits the applicability of the findings. Much more spatially extensive and stratified sampling related to soil parameters needs to be undertaken before a true picture of sediment-water exchange of Hg under flooded conditions can be fully assessed. This would significantly focus control efforts since the data as presented cannot be used to generalize about land-use type across the YBP. The soil distribution data in this Appendix (and in Appendix F) are of significant value for understanding controls on Hg methylation, and should guide future efforts in this regard.

Associated biogeochemical data could be used to help understand controls on MeHg in Yolo soils. The data are there to start on this, but little analysis was presented. How was soil Hg related to soil MeHg across the study sites? How was %MeHg in soils related to soil saturation, or soil organic matter? Some of the data were provided in Appendix C1 (although much of the ancillary data noted in the methods section was not, or at least not in detail through time). It would be great to see more evaluation of these data, and it would be useful for BMP development for the Delta.

Simple analysis of the soil core data shows that MeHg is not strongly correlated with total Hg in soils, nor is the percent of Hg as MeHg in soils correlated strongly with antecedent moisture conditions (see Figure 1 below). What is (are) the main control(s) on differences in MeHg among soils? This is critically needed information for management of MeHg in the Yolo Bypass.

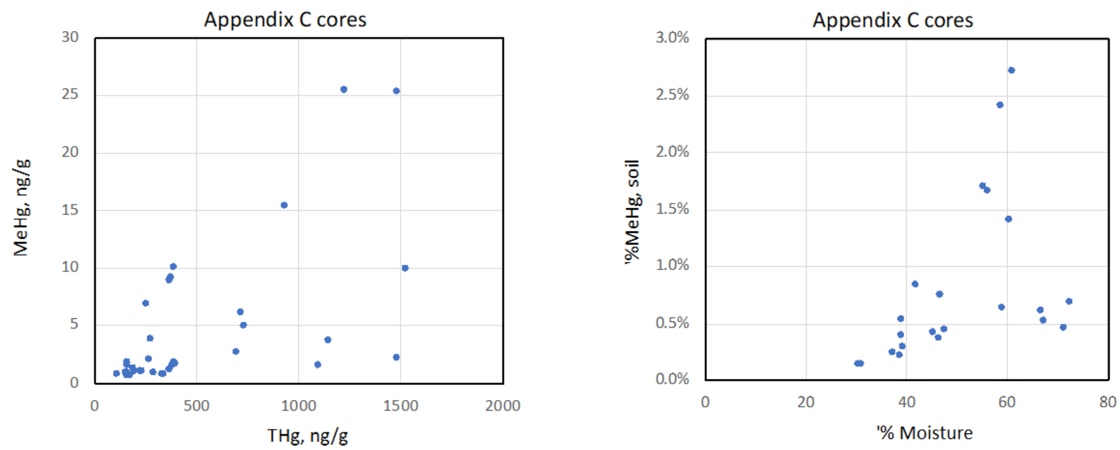

Figure 1: Data from Report Appendix C. %MeHg plotted against Total Hg (left) and %Soil Moisture (Right).

> *Question 3. Do you have comments on other scientific issues related to this study or mercury/methylmercury source type?*

The highly variable estimates of Hg and MeHg flux within and among sites provides a very rough estimate of the contribution of flooded soils to Yolo Bypass waters during floods. The lack of data on vegetation in the study cores does not allow the separation of sediment MeHg efflux from MeHg production in decaying vegetation. Although this study is labeled "open water sediment-water fluxes" the method section also notes that vegetation was included in core samples where present. As reported, average fMeHg flux across all the seasonally inundated study sites was roughly 20 +/- 18 ng/m^2/d. Compare this to estimates of MeHg contribution from decomposing vegetation (from the VegSens studies) that ranged from roughly 10 to 200 ng/m^2/d. The data produced by these two studies overlap, and in some ways measured the same things in different ways. The separation of MeHg production in sediment loaded with plant roots or dead plant material, from MeHg production in decaying plant material alone seems somewhat artificial.

The sample size in this core study was too small to say much about differences in flux among land use or land management types. <u>For this study, each land use or land management type was represented by soils from only one site;</u> not enough to say anything statistically about differences among land-use or management types. Although there were replicate cores from each site, this is <u>pseudo-replication</u> that does not capture differences among sites with the same land use. With a sample size of one, specific conditions to the sample site chosen (especially differences in soil total Hg) could easily be what controls MeHg production.

The soil distribution data in this Appendix (and in Appendix F) are of significant value for understanding controls on Hg methylation. For example, the study found that porewater MeHg was positively correlated with porewater organic matter content. The study also found relatively low soil/water partition coefficients for Hg in Bypass soils; which suggest high bioavailability of Hg for methylation. However, this was not discussed. Much more could have readily been done with these data in terms of understanding biogeochemical controls on MeHg accumulation in Bypass soils.

Variability in this study was high within and among sites (not surprising), sometime with different directional fluxes among cores taken from the same site, and in several cases not significantly different from zero. Here is where info on vegetation (or other differences) among the cores might have been helpful. No statistical analysis was presented. A preliminary analysis done as part of this report reveal statistically significant differences among the sites. For MeHg, irrigated pastureland had significantly higher flux than all other sites (see Least Square Means table below).

Table 1: Statistical Least Square Means comparison of MeHg Flux among land cover types. Levels not sharing letters in compact letter display are significantly different.

Level	Compact Letter Display	Least Square Means
Irrigated Pasture	A	53.3
Fallow	B	28.6
Undisked Seasonal Wetland	BC	25.9
Disked Seasonal Wetland	BC	25.7
Dry Pasture	CD	12.1
White Rice	DE	8.9
Mixed/Other Farmland	DEF	3.6
Wild Rice	DEF	3.0
Tidal Wetland	EF	-3.7
Open Water	F	-10.9

The sediment-water flux study is plagued by incomplete/unclear presentation of data. In Appendix C (Table C1-1), the components of the table aren't defined. What does 1,2,3, etc. in the column headers represent? Replicate cores? Sampling times? If replicate cores, please provide information on when these data were taken within the time course of the study (how many days after incubations were started). Were these measurements made in water or in soil? Lack of information about flux calculation approaches meant that the Reviewers were not able to explore these relationships at all, and when 'best guesses' were made about how the fluxes were calculated, different flux rates were found (sometimes with different directions than that reported). Missing metadata for data tables throughout this study will limit its usefulness to managers and other wanting to use these data. This is important information that should be extended spatially to better represent land-use types across the entire YBP; greater transparency in methodology and data will ensure coherence between this work and future studies.

6.1.3. Gust Chamber Erosion Study

The Gust Chamber Erosion study was intended to experimentally assess the erodibility of YBP soils from 10 locations under different land covers to provide critical shear stresses, suspended sediment concentrations and erosion rates for sediment entrainment parameterization in the Yolo Bypass Model. This is a USGS Open File report that we presume received internal peer review prior to release. Appendix D comprises the Open File report reproduced in full.

Question 2c: What knowledge gaps must be filled to inform future control studies or management strategies?

This study was undertaken purely to generate input data for the Yolo Bypass Model. There are no direct implications for control studies and no knowledge gaps related to the development of future control measures that could be explicitly addressed. No Hg or MeHg concentrations are linked to the soils that were subject to the experimental tests. Samples may have been co-located with samples taken for other experiments or data collection such as that in Appendix F; however, no links were made in the overall Report to this effect. The lack of any link between particulate bound Hg and MeHg concentrations and particle size fraction means that the results are incorporated into models in a relatively coarse way. These latter points are beyond the scope of the specific study.

Question 3. Do you have comments on other scientific issues related to this study or mercury/methylmercury source type?

The approach generated robust mechanistic data that supports the model effort. The limitations of the results are laid clear in the Report (only 10 locations each representing a land-use type) with only two cores per site. Future sensitivity analyses in the model would reveal whether greater certainty in this parameter would warrant further work. Appendix D/Open File Report contains maps of elevation, Loss-on-ignition (organic matter content) and grain size for YBP soils, which are extremely valuable as they represent important controls on hydrology and Hg behavior. An opportunity exists to couple these data with those in Appendix F to explore approaches to generate a more spatially explicit map of Hg and MeHg in soils in the YBP. Soil Hg distribution is a factor that is more strongly related to water flow patterns and Hg inputs than to land use *per se* and should be a variable in future extensions to experimental work such as that presented in this Report.

6.1.4. Vegetation Senescence Study

Question 2a: Do the study results provide new or additional information about mercury or methylmercury sources, mass balances, or loads in the Delta or Yolo Bypass?

Thirty years of studies of MeHg production in other types of flooded systems support that idea that vegetation is an important driver of MeHg production. Vegetation can enhance methylation within soils by increasing organic matter availability for microbial methylation (Windham-Myers et al. 2009). Live emergent plants in wetlands produce DOM in their root systems, while senescent or dead vegetation (e.g. for rice fields; see Tang et al. 2019) also contributes. MeHg production can also occur in decaying plant matter itself during floods (Cleckner et al. 1999).

The data reported from the several years of VegSens experiments generally confirm that vegetation is the dominant source of internal MeHg production during YBP bypass flooding. We agree that the data support a link between biomass (above and belowground) and MeHg production. The relative importance of MeHg flux from sediments vs. decaying vegetation is less clear. Certainly, the biomass of both above and below-ground organic matter contribute to microbial activity that fuels MeHg production.

Issues associated with experimental design confound the interpretation of the experimental results, which reduces their utility in exploring control measures and BMPs. There were only a small number of sample sites, presenting challenges in extrapolating lab manipulations to mimic field practices. Further, a lack of data for Hg and MeHg in soils or vegetation means that it is not possible to separate the effects of different Hg concentrations from treatment, limiting the strength of conclusions.

Question 2b: Will the studies help design future control studies?

The study results point in an important direction for further work on future control studies, however, **the data presented are not sufficient unto themselves to develop BMPs based around specific agricultural practices.** Specifically, the Report concludes that disking, and probably grazing, significantly reduce MeHg flux to flood water. We do not feel that the data in the report are strong enough to support conclusions with the level of certainty stated in the Report, nor justify implementation of those practices as BMPs at this time, although further evaluation is certainly warranted.

Question 2c: What knowledge gaps must be filled to inform future control studies or management strategies?

Issues around experimental design and data interpretation are elaborated further under Question 3. Knowledge gaps here are posed as a series of questions.

What is the source of Hg for methylation in decaying vegetation and in surface soils? The vegetation studies did not investigate the source of Hg for MeHg production in decaying vegetation. Does it come from Hg in flood water or underlying local soil? Chapter 3 lists this as a data gap (p. 3-68) and we agree. This has important ramifications for MeHg risk control. Was Hg accumulated in vegetation and then methylated? Was decaying organic matter fueling MeHg production in sediments? Had vegetation or soils accumulated MeHg prior to the experiments and just released it during decay? Analyses (and mass balance) of Hg and MeHg in vegetation, soils and water would have been very useful. An experiment examining changes in MeHg concentrations in bagged rye grass in some of the experiments was briefly described, but the experimental design of that work wasn't clear so it's hard to interpret (p. E-33). In VegSens2018, substantial MeHg production occurred in beakers with vegetation but no soil, but we don't know where the vegetation got its MeHg (was it produced or just released during the study?). If MeHg production in vegetation comes from soils, then the identification of soil Hg hot spots within YBP soils could provide a management approach. If Hg in flood water drives methylation, a focus for control should be upstream Hg inputs. Lab studies could help sort this out, with field measurements for up-scaling and verification. As noted by the study authors (p. 3-69), field studies are limited by cost, access issues and safety; while mesocosm studies (better proxies for the real world than lab studies) are limited by cost and the lack of availability of flood waters during non-flood seasons (which can go on for years). Nevertheless, development of effective BMPs for MeHg control requires field trials and sufficient monitoring. This type of information wasn't collected and could help with management of rice and pasture lands. Lack of total Hg data for the soils and vegetation, or any measure of MeHg in either soils or vegetation was an unfortunate oversight. Future studies should attempt to mass balance experimental microcosms.

What is the role of other vegetation types? Chapter 3 notes that vegetation decay studies focused on rye grass and that information for other types of vegetation is needed (p. 3-68) to generate BMPs. We agree. The vegetation data collected in this study was also entirely experimental and will need to be backed up by field trials for verification. The significant body of research on MeHg in rice fields and associated potential BMPs was not captured in recommendations of Chapter 3 or Technical Appendix E. Were these data used in modeling? Or proposals for BMPs going forward? For example, in rice fields, slow flows of water across the fields can lead to accumulation of MeHg in sediments; and to risk pathways that may not be captured in sediment flux studies (Windham-Myers et al., 2014a; 2014b, Fleck et al. 2014). This potential risk pathway was not evaluated here. Future studies of MeHg production in decaying vegetation should do full mass balance on study microcosms – measuring Hg and MeHg in soils and vegetation over time –not just filterable MeHg in overlying water (which was used as a proxy for flux).

> *Question 3. Do you have comments on other scientific issues related to this study or mercury/methylmercury source type?*

Generalizability of Findings: From and experimental design standpoint, the VegSens studies suffered from the same challenges as the sediment-water flux studies with limited spatial coverage of land-use types across the YBP. The VegSens study designs generally had good within-treatment replication. Variability was sometimes high, but that is typical of the heterogeneity of field collected materials, but also speaks to the potential concerns about applications of the findings more broadly across the YBP. It is clear that soil Hg concentrations vary considerably across the YBP. No data on Hg in soils or vegetation was collected for the VegSens studies. This means it's impossible to separate the effects of differing Hg concentrations among the sampling sites from the treatments, particularly in VegSens2018 where soils from different sites were used to represent different agricultural practices.

Lack of underlying biogeochemical data, and potential sulfate limitation. All VegSens studies were done with tap water rather than YBP flood waters, so the studies may not be representative of field conditions, especially if low sulfate concentrations limited microbial sulfate reduction. No ancillary biogeochemical data for water or soils were reported for these studies, so the impacts of these factors could not be controlled for. Further, the slow movement of flood waters over vegetated fields not only has the potential to deliver Hg and MeHg, but also refresh the pool of available nutrients, potentially supporting higher rates of MeHg production than reported here.

Flaws with experimental designs. In the 2016 studies, irrigated vs. non-irrigated pastured under aerated water found higher MeHg production in soil cores take from irrigated land. These sites had more biomass. However, because irrigated and non-irrigated cores came from different locations, the results could have been confounded by difference in Hg between sample sites, reducing confidence in the conclusions. For the VegSens2017 mesocosms means coolers instead of beakers. Sod was used instead of soil cores, increasing the realism of the studies and potential applicability to field conditions. Grazed and ungrazed sites were paired by fencing off areas adjacent to grazed sampling sites; although not documented, this hopefully meant that the soil Hg concentrations were similar. For this experiment, "disking" was one of the experimental treatments being tested, however it was merely simulated by mixing senescent vegetation into study soils, instead of using field-disked soils, reducing confidence in the conclusions

Instead of clarifying the interacting effects of agricultural practices and biomass, the VegSens2018 study design was perhaps the most confounded. VegSens 2018 was a beaker study designed to test the impact of biomass and agricultural practice on MeHg efflux, using defined additions to a single soil. The amount of biomass (and type) varied among treatments designed to mimic grazed, ungrazed or disked soils. For example, grazed treatments received half the biomass of ungrazed treatments, but also received manure. Disked treatments had both manure and biomass mixed into soils, but at lower concentrations that the grazed and ungrazed treatments. There was no way to evaluate the individual effects of manure, vegetation or mixing, dramatically reducing the utility of the experiment. Beakers with high, medium and low biomass across treatments could really not be scientifically compared because the amount and ratio of biomass between high, medium and low treatments was NOT the same for all treatments.

Study Conclusions and BMP Recommendations: Overall, these lab-based studies identified decaying vegetation as the dominant source of internal MeHg production during Yolo bypass flooding. Fresh vegetation (growing up after early rains in October, but before flood) appears to be a more important contributor to MeHg production that decay of older, senesced vegetation. Overall, MeHg production in flooded pasturelands is clearly a function of vegetation biomass. The estimates of MeHg flux from flooded soils were reasonably similar among the three major VegSens experiments (for similar treatments). The broad internal consistency of the results of the microcosm/mesocosm experiments indicates that there are underlying mechanisms that may be more fully explored in more carefully designed experiments and/or scaled-up field plot studies.

Despite definitive statements to this effect in the Report, it is our opinion that the study <u>does not</u> provide clear data to support disking as a BMP. The mesocosm mixing in the studies does not physically mimic field disking at the field scale. Further, the experimental design for comparing disking to other practices in VegSens 2018 was confounded by differences in biomass between mixed ("disked") and unmixed treatments. No measure of below ground biomass was made in any of studies. Moreover, the wholesale recommendation of removal of vegetation cover does not address any of the potential negative implications of the practice on soil nutrient levels, soil carbon storage, soil biodiversity, or higher trophic level impacts such as resident or migratory bird ecology. Statements about these results must be tempered to better reflect the uncertainties associated with the experimental designs and the extension of the results over a larger spatial scale, as would consideration of the implications of different vegetation types. Rice straw management has been undertaken for some time. What have the findings been?

Lacking information about the sources of Hg (or MeHg) in any of the studies presented, it is entirely possible that reductions in Hg inputs from upstream tributary sources may be an effective BMP in controlling MeHg production in decomposing vegetation along with the regulation of biomass.

In summary we agree with the Report that MeHg accumulation increases with mass of vegetation in experimental beakers (Hypothesis 3). We do not agree with the report that:

H1) Grazing reduces MeHg production. Although it's clear that MeHg production is related to vegetation biomass, the summed effect of grazing (e.g. reduced vegetation biomass with addition of manure to land) was not full mimicked by the study design which didn't include mixed vegetation and manure treatments. The study as designed really only looked at the effect of vegetation biomass. H2) Disking reduces MeHg production. Disking was only mimicked by mixing vegetation into the experimental soils. The mass of vegetation was different between the disked/grazed and ungrazed treatments and confounds the effect of mixing.

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6.2. Open Water Report: Climate Change Impacts

General Comments: This chapter is focused on the Question 2 of the non-modeling studies charge to the reviewers as it addresses how well the report identifies impacts of changing climatic conditions, outline next steps, and assess feasibility of future actions. The narrative below provides the details about the chapter and how the chapter does not provide a concrete and comprehensive examination of the question, but provides a series of simple assessments of the various potential climate impact factors, and does not consider their interaction and any synergism that climate may have because of the interaction of factors. The chapter does not provide any suggestions for control over climate impacts. Overall, the chapter is an inadequate response to the posed question.

While the work plan does not directly address the issue of climate change but focuses more generally on changes in the future to hydrology, this is an important aspect of the report and it has not been well addressed. It is also a specific question posed to the Scientific Review Panel. Thus, it is necessary to conclude that the report fails to address the scientific objectives of the work plan as there is no real attempt made to develop a rigorous response to the question of climate impacts. As noted below, there are impacts that can be assessed based on current knowledge that should have been included in the report, and further developed and quantified beyond the observations made by the Review Panel below which highlight specific impacts not mentioned in the report. Clearly, future environmental changes are fundamentally important to the question of the suitability and feasibility of the load allocations.

Some aspects of climate are addressed elsewhere in the Report, but overall the Report fails to address this question which is of importance to managers who are trying to predict future environmental changes and the implications for load allocations. Throughout this section there is discussion of how changes in a parameters will impact MeHg production, but there is no examination of direction, magnitude and certainty. Indeed, it may not know be possible to know the outcome for sure, but not all of the considerations are "unknown" as indicated throughout the Report.

Specific Comments: The effect of temperature is discussed in a very generic way, with the focus only on its impact on methylation, and there is no effort made to place its potential impact in the context of the study results. For example, the Delta model concludes that sediment methylation and subsequent inputs are small compared to other sources so that the likely impact of temperature would be small,, although it could impact methylation upstream of the Delta, but this is not part of this analysis. There is also no mention on how temperature could impact photodemethylation which is a small but important sink within the Delta. Further, how temperature could impact other factors, such as increasing the duration of dry conditions and the impact of this, is not evaluated. It should be possible to assess the impact of increasing temperature and climate on these processes. Also, the relationship between microbial activity and temperature is not linear but a complicated relationship.

Importantly, the most defensible means of assessing climate change impacts is through modelling, especially since feedbacks are not adequately represented in the simplistic "generalized impacts" that are discussed in Chapter 6. One would reasonably expect some assessment of the main climate change impacts within the modelling frameworks that are detailed in the reports.

The effect of climate on precipitation, the intensity of storms, the extent of the snowpack and the seasonal runoff are all linked through the expectation that in the future there will be more precipitation as rain, and less snowpack, and therefore there will be a change in the delivery of water into the Delta region. The report makes no predictions of how these changes would affect MeHg within the Delta and its supply. Climate change prediction is for more intense storms and likely more sediment transport as a result and given the Hg/MeHg relationships with suspended sediment load discussed in other chapters it is likely that more intense storms will lead to more transport of Hg/MeHg into the system. The impact of more intense storms on the MeHg loading to, and flux from, the Yolo Bypass is more difficult to predict but given the mass balance predictions in Fig. 3-42, the likely impact of these changes in climate is more MeHg production in the Yolo Bypass and subsequent transport to the Delta. This is not discussed in sufficient detail in Chapter 6 and needs further evaluation. More work clearly needs to be done to develop more detailed and robust predictions of the impact of changes in precipitation on MeHg transport to the Delta.

Some of the statements made are not evaluated fully. For example, in the Sacramento-San Joaquin system section, the authors state: "It is reasonable to assume that larger and more frequent storm driven stream flows will result in greater erosion of Hg contaminated sediments in the Sierras and Coastal mountain ranges, resulting in enhanced loads of Hg entering the Delta. If this does occur, then there will likely be a greater abundance of Hg available for methylation than in the current case,". This is an overly safe statement in that differences in Hg bioavailability are not considered. There is indeed an overall potential to be more Hg available for methylation, but it is also possible that most of the Hg coming off the Sierras and Coastal mountain ranges would likely less bioavailable overall.

The overall impact of the predicted precipitation changes is likely to be less of a spring flush and more runoff in winter which would also affect sediment transport and other aspect of the upstream hydrology, therefore affecting the Hg and MeHg input into the Delta from external sources. This is likely a greater impact than changes in storm intensity. Direct atmospheric deposition is likely not a large Hg source relative to tributary inputs and therefore changes in the timing and extent of precipitation will not substantially affect this.

Chapter 6 makes reference to the importance of DOC/POC in Hg/MeHg transport and how this may change with climate. Changes in DOC quality and quantity are clearly important factors for Hg transport, for maintaining dissolved Hg in solution, and its impact on methylation rate. This is discussed in some detail in the report and clearly more details on the relationships between Hg/MeHg and OC are needed in terms of both the inputs, and within the Yolo Bypass and the Delta. Relationships examining this should be constructed using currently available data.

The impact of drought is discussed more than other factors in the report with a focus on the impact of drought on the Yolo Bypass and the potential for the resultant changes in the system to affect MeHg production and transport from the system. There is a reasonable discussion of the effect of different scenarios on the net MeHg transport. However, it would seem that drought could have impacts both in the Delta and upstream with lower flow and soil and sediment drying. It is known that exposure of submerged sediment leads to overall oxidation of the sulfide in the sediment and that these areas tend to produce more MeHg during re-flooding, such as is found when new reservoirs are constructed. The impact of drying/wetting within the Delta system is not discussed, and needs to be evaluated both in terms of drought and sea level rise, which is discussed more below. The balance of drying effects and sea level rise should be discussed and evaluated. The effect of flooding of dry land (the so-called "reservoir effect") is discussed in terms of the impact of sea level rise on levee breaching and flooding of cropland and dry land, and this may be a much more important impact than that of drought on other parts of the Delta, but this should at placed in the context of upstream changes in hydrology and the relative combined implications of all of these changes in hydrology.

The Yolo Bypass is characterized by a variety of land cover that is dominated by agricultural lands. It would be expected that agricultural land would behave differently to changes in flooding frequency than other types of natural land cover. There's nothing in this section that considers land uses or land cover, which would have helped make the consideration of the effects of flooding more mechanistic.

As noted above, the impact of sea level rise on levee failure and the resultant impact on MeHg levels is discussed in the report in more detail than any other aspect of predicted climate change effects. This is a reasonable discussion but is focused on the aspect of levee breaching and flooding and not the impact of salinity changes in the system. Salinity itself may not be an important aspect, but saline waters contain more sulfate and therefore increased salinity could impact the degree of Hg methylation within the system and/or the degree to which Hg is sequestered in the system if there is additional removal of Hg by Hg-sulfide precipitation in sediments. The role of sulfide in complexation of Hg and influencing bioavailability needs to also be considered. Additionally, the amount of reduced sulfur in organic matter is a function of the degree of sulfate reduction and there are higher levels of reduced sulfur ligands in organic matter in more saline ecosystems, and this will affect Hg/MeHg speciation, fate, transport and bioavailability. None of these important controls on methylation are evaluated to any degree in the Report.

As discussed elsewhere, the model does not adequately evaluate changes in DOC concentrations and how this impacts Hg and MeHg cycling and the impacts of sea level rise and other factors on DOC concentration within the system are not mentioned. Increased salinity may lead to coagulation and precipitation of organic matter which is known to occur in estuarine mixing zones and this would definitely impact Hg cycling. The role of chloride in influencing Hg and MeHg speciation should also be considered although given the relatively high DOC within the system it is likely than changes in salinity would likely be less important than changes in DOC levels in terms of impacting MeHg speciation and bioaccumulation at the base of the food chain. If passive uptake of chloride complexes into methylating microbes is important in the region, then changes in salinity would affect the relative MeHg concentration.

The two impacts of wildfires are reasonably addressed in the Report. Firstly, increased release of Hg to the atmosphere during burning could increase regional deposition, although the extent of this is not evaluated - the potential for increased deposition is purported and this conclusion is supported by literature references but the issue is barely discussed. In terms of the effect of fires on the transport of Hg from the watershed, this is likely to increase with more erosion and runoff from burned areas. This is not discussed.

The Report concludes with a set of specific questions that is indicated need to be addressed in the future in terms of climate impact. It is apparent that many of these could be addressed to a greater degree than done in the report based on available information in the literature. As noted above, the Report is deficient in this regard, and more evaluation of the models and their results and predictions could be done based on existing literature, with the aim being to specifically answer the questions posed at the end of this chapter.

Finally, factors besides direct climate impacts are likely to have a large impact on Hg cycling and concentrations. One issue that was discussed during the Review process was the potential for regional changes as a result of climate adaptation in the future, and these are not addressed in Chapter

6. An example is the proposed “Delta Conveyance/Delta Tunnel”. This plan calls for a couple of new, very large water intakes in the north Delta (further decreasing natural freshwater flows) and conveyance through a 40 mile tunnel to existing reservoirs. This is because sea level rise, salt water encroachment, and seismic activity threaten the current intakes which are closer to the ocean and saline waters. This would be a very large near-future disruption that would impact hydrology and Hg cycling, but this is not discussed in the report.

A similar concern is related to the potential of levee and island failures, which are expected in the future. If expected, are there proposed adaptations specifically carried out to circumvent the impact of these events? It seems that 60-80% of levees failing, for example, is an alarming prospect with a large impact, if it does not consider adaptation (e.g., making the levees larger or moving them).

The impacts of climate change will vary widely across the state. Even temperature variability is not that easy to specify (e.g., the Report indicates “2 to 7 degrees statewide”, which is a large range). If the resources existed, regional climate down-scaling in the area to be more specific to the Delta region and upstream drainage basins would reduce uncertainty.

6.3. Open Water Report: Yolo Bypass Mercury Model

Do the results of the modeling studies fully achieve the scientific objectives of the workplan?

Based on the Workplan, the goals of the modeling studies are two-fold: (1) to serve as the starting point for hypothesis testing; and (2) provide a basic understanding of the important processes governing MeHg production and how open water and flood conveyance loads could be impacted under different proposed operational scenarios. Inherent in this latter goal, which the Workplan articulated using both sensitivity and scenario analyses to address, was using the models as management tools "to evaluate the potential effects of operational changes on Hg cycling and MeHg supply" and thus provide "CVRWQCB staff with a mechanism to evaluate future regulatory proposals and prioritize and focus future actions."

The Workplan delineated three objectives to satisfy the modeling goals. These include:

1 Provide working models for Hg and MeHg supply, transport, and fate in the open waters of the Delta and Yolo Bypass;

2 Apply the models to identify processes governing MeHg supply to the Delta and Yolo Bypass (YBP);

3 Apply the models to examine the potential impacts of proposed operational changes in water management and flood conveyance in the Delta and YBP on MeHg supply and compare to TMDL allocations.

Phase I modeling was intended to include initial development and application of models for the Delta and YBP, and identify modeling, knowledge or data gaps.

Goal 1 – Provide a working Hg and MeHg model to simulate supply, fate and transport in the Yolo Bypass: This goal was satisfied for the YBP through the application of the Dynamic Mercury Cycling Model (D-MCM; Harris et al., 2012a). D-MCM is a well-established Hg biogeochemical cycling model that has evolved substantially in its functional representation of Hg cycling since its original incarnation as the Mercury Cycling Model (MCM) as part of the Mercury in Temperate Lakes (MTL) project (Hudson et al., 1994). This evolution of MCM to D-MCM has notably included the ability of D-MCM to simulate lakes, wetland and estuarine/marine ecosystems with multiple cells (including both 2- and 3-dimensional spatial configurations) (Harris et al., 2003; Harris et al., 2012a; Harris et al., 2012b).

Goal 2 – Apply the YBP model to identify processes governing MeHg to the YBP and Delta: D-MCM configured to the YBP using a total of 47 cells. Depending upon the model endpoint selected, calibration was conducted for six or seven cells, all with limited water and sediment chemistry that precluded a rigorous calibration over space and time. This limitation results in little confidence regarding how well the model represents internal spatial dynamics. External input and outputs (defined by releases from the YBP at the Stairsteps) can be constructed using observational data and can be used to define whether the YBP is a net sink for total Hg and total MeHg. Extending the analysis much beyond that simple mass balance to elucidate key processes governing Hg and MeHg fluxes internal to the YBP and ultimately from YBP to the Delta is, however, problematic. The calibrated D-MCM in theory can be used to identify which internal processes help govern Hg and MeHg export from the YBP. However, this was not formally conducted during this phase. For example, the D-MCM ostensibly could be used to evaluate the likely change in relative and absolute magnitudes of internal *vs*. external sources of MeHg moving spatially downstream through the YBP from the various external input locations to the Stairsteps. At this juncture, the limited amount of calibration data coupled with problems such as unconstrained fluxes (e.g., no data on net sediment burial rates and Hg(0) volatilization) preclude a meaningful evaluation of the spatially and temporally variable mass balance dynamics necessary to identify Hg key cycling processes.

Goal 3 – Forecast sensitivity scenarios: The D-MCM model was used to conduct simulations for nine different management scenarios developed by the DWR and Regional Board staff. Each scenario considered the effect of a change in a single input. The utility of these scenarios is questionable given the apparent lack of constraint in the parameterization of certain key parameters. For example, reducing Hg(II) concentrations (and thus loadings) by 50% at the Fremont Weir resulted in only a < 5% in MeHg fluxes at the Stairsteps. Hg(II) loadings via the Fremont Weir constitute 43% of the total external inputs to the YBP. The authors acknowledge that the effect was less than expected. Likewise, a similar scenario related to reducing TSS (total suspended solids) by 50% at the Fremont Weir indicates a less than 5% decline in MeHg export at the Stairsteps as well. Similar results were obtained for the same scenarios for CCSB inputs. From a mass balance perspective, these results are troubling and may reflect issues with how the particle balance is implemented and maintained for the active surface sediment layer. The authors should discuss more fully which scenarios may be reasonable and why and which scenarios are indicative of model problems that should be resolved

1) How well does the model account for key processes, assumptions, and uncertainty? Are these documented adequately?

The Yolo Bypass (YBP) model comprises two components – hydrologic and Hg biogeochemical cycling – with each component addressed by a separate model. Modeling of Hg biogeochemical cycling was conducted using the Dynamic Mercury Cycling Model (D-MCM), which is a proprietary, mechanistic (dynamic) model developed for the Electric Power Research Institute (EPRI); hydrologic inputs and outputs were modeled using the segmented 2-D hydrodynamic model TUFLOW. TUFLOW was previously applied and calibrated to the YBP for the period 1997-2012. Such "soft-linking" of a biogeochemical mass model that is not structured to directly simulate hydrodynamics and hydrologic forcing with a separate, external hydrologic model is both appropriate and necessary in complex hydrologic settings such as the YBP. A previous example of soft-linking D-MCM with a separate and more highly resolved (spatially and temporally) hydrologic model is the application of D-MCM to model Hg biogeochemical cycling in the Gulf of Mexico (Harris et al. 2012a).

D-MCM is well-established as a tool for modeling Hg biogeochemical cycling across a range of freshwater and marine environments, including wetlands, lakes, estuarine systems, and fully marine waters. As such, it is well-suited to serve as the Hg modeling framework for the YBP.

D-MCM has algorithms designed to simulate all the major key processes thought to govern Hg biogeochemical cycling. Table 1 summarizes these key processes and indicates how the process was parameterized and a qualitative assessment of how well the process parameterization meets the objectives of the study.

<u>Model Configuration:</u> The D-MCM model is configured with 17 permanently wet cells and 30 cells classified as seasonally wet or land cells. Each cell is considered to have uniform

characteristics within the cell and the land cells vary based on land use type. The grid structure was based on GIS coverage (land use, sediment concentrations of Hg and MeHg, agricultural disking, hydroperiod duration and hydroperiod wet/dry cycling frequency). The authors state that disking was not considered as part of the model calibration; as a result, it is unclear why disking was included in the model or how it's parameterization can be justified.

The number of grid cells was established as a compromise between the spatial complexity limitations of D-MCM and the desire to adequately capture the spatial heterogeneity of the YBP environment. Moreover, the authors observe that Hg cycling data limitations preclude implementing a more complex grid. The paucity of Hg cycling data with respect to the number of cells included in the model (seven cells with observed data *vs*. 47 cells in the model) precludes a formal assessment of the adequacy of the D-MCM to represent the spatially variable dynamics in Hg cycling within the YBP and thus its adequacy to represent how different management scenarios imposed upon and/or within the YBP will affect Hg export dynamics to the Delta.

The D-MCM model was implemented with a two-dimensional cell structure, with flow vectors input from TUFLOW in both horizontal directions; vertical conditions in the water column were well-mixed. The range in water column depths is not clear, nor do the authors discuss why the assumption of vertical well-mixed conditions is justified. The assumption likely is appropriate but should be explicitly discussed.

<u>Particle Dynamics:</u> Four types of particles are considered in the YBP implementation of D-MCM: (1) organic matter other than vegetation; (2) vegetation solids; (3) fine inorganic solids; and (4) coarse inorganic solids. Processes that are important with respect to particle dynamics include: (1) mineralization/decomposition [organic matter including vegetation]; (2) settling [all particle types]; (3) resuspension [all particle types]; and (4) deep burial below the active surficial sediment layer (assumed 2 cm thick; this likely is reasonable based on modeling studies of other systems that require an active sediment layer that turns over rapidly to be consistent with observed short-term dynamics).

Resuspension was calculated external to D-MCM by TUFLOW using computed flow velocity shear stresses (presumably calculated at the sediment-water interface based on the vertical profile of horizontal velocities) coupled with measured critical shear stresses conducted as part of the Open Water study. Values were determined for surficial sediments collected in intact cores from 10 separate sites within YBP (GUST chamber erosion studies; Work and Schoellhamer, 2018) to determine erosion potential. Resuspension is a critical process influencing the movement of solids through YBP and the ability to use experimentally determined τ_c specific to different land-use types in the YBP is important feature of how the model has been implemented in reducing uncertainty.

The text discusses solids supply to the YBP related to vegetation die-offs. How those fluxes were determined is apparently based on other studies (inferred from Figure 4-6). Given the importance of vegetation *vis-à-vis* both solids loading and as a source of carbon supporting methylation, how the flux calculations were conducted should be discussed more explicitly. Decomposition of vegetation particles that have settled and been incorporated into the surficial sediments is assumed to occur more rapidly than other organic particles although it is unclear quantitatively what this means. The authors do state that the vegetation modeling is preliminary and could be refined if more modeling is conducted in the future. It would be useful to see gross particle budgets for the various particle pathways constructed for the YBP.

The D-MCM implementation for the YBP thus has an essentially unconstrained particle budget. External inputs are inferred (and potentially biased with large uncertainties). Internal production of vegetative organic matter is estimated based on changes in standing crop derived from other published studies but the uncertainty in these estimates is not presented. The interplay between gross particle settling, organic matter decomposition and resuspension defines how management-related changes in particle dynamics both transition through the YBP and (because organic matter decomposition rates influence methylation rates) impact net MeHg production and export from the YBP. Net burial (not discussed) is the product of the calculated particle mass balance with the model tracking net accumulation and erosion of the surface sediment layer. Organic matter decomposition rates are not known and thus net burial (and thus turnover rates of the active surficial sediment layer); this fact, coupled with the fact that no data are apparently available to characterize net sediment accumulation rates, results in an unconstrained particle budget. This uncertainty also means that the turnover rate for the active sediment layer – which has important implications on the time scale of response to different management scenarios – is essentially unconstrained.

It also is unclear whether the model allows for solids buried immediately below the 2 cm surficial horizon to be re-introduced into the 2 cm horizon (in order to maintain a fully 2 cm thick layer) during periods of net erosion.

Uncertainty: Uncertainty in the loadings of suspended solids, total Hg and total MeHg was **not explicitly addressed in the report.** Uncertainty in model parameterization was evaluated with the PEST++ optimization framework (Welter et al. 2015; White et al. *in press*). This approach led to a substantial reduction in the root mean square error (RMSE) – apparently a 51% reduction in the aggregated RMSE for all the calibration parameters although this (whether the uncertainty is aggregated) is not clearly stated in the text. The implementation of PEST++ is also useful from the perspective of parameter identifiability. Parameter identifiability refers to the degree to which a model parameter can be constrained to a unique value given the experimental data (Daly et al. 2018). Parameter identifiability also has implications for model forecast uncertainty – model predictions that rely upon influential parameters that are not well-constrained will be more uncertain. PEST++ was used to quantify identifiability for 147 D-MCM parameters. Results were presented for the least and most identifiable subsets of model parameters (Figure H2 and H3); while qualitatively interesting, however, the information lacks quantitative context relative to the resultant, inherent uncertainty in model predictions. Moreover, the reader can only infer as to the actual nature of parameters included in Figures H-2 and H-3, which identify the parameters according to their acronym but for which no acronym list is provided in any of the D-MCM related modeling documents.

Results for improvement in model predictions (forecast uncertainty) related to improvements in parameter uncertainty following the implementation of PEST++ are presented as examples for two different forecasts (annual average loadings between 1998-2012 for total Hg and total MeHg). While the example plots (Figure H-6 in Appendix H) show a striking improvement in forecast uncertainty for both scenarios, the analysis is only valid within the context of which parameters were actually allowed to vary in the uncertainty analysis or forecasts. Which parameters and their uncertainties included in the analysis and which parameters that were not included in (and thus considered fixed or known exactly) and their uncertainties were not defined in the text. In brief, while the tools to conduct a full uncertainty analysis that considers the effects of joint parameter uncertainty are conceptually available, such an exercise was not undertaken. Such an exercise should also consider that some parameters are inter-related, and thus how each of those parameters varies during the uncertainty exercise should account for that covariance.

Table 2. Overview of major processes included as part of application of the D-MCM to YBP. Qualitative review rankings regarding how well the model process is characterized for the YPB implementation of the D-MCM range from 1 (very poorly characterized relative to importance) to 5 (very well characterized relative to importance).

Model Process – *External Inputs*	**Comments**	**Ranking**
Atmospheric deposition	MDN data for Hg wet deposition from most proximal site. Tsai and Hoenecke (2001) for dry deposition. Uncertainty in these inputs not important given magnitude of external inputs.	4
Tributary inputs	Based on measured flow and inferred concentrations.	2/3
Other point source inputs	Not applicable – apparently no other point sources directly discharge into the YBP.	N/A
Model Process – *Physical Transport/Particle Dynamics*	**Comments**	**Ranking**
Advection	Advective transport simulated by TUFLOW	5
Particle settling	N/A	N/A
Diffusion	Based on measured pore water chemistry (Sediment-water Flux Study) to calibrate model pore water chemistry and fluxes.	3/4
Resuspension	Calculated from TUFLOW velocities to generate shear stress and particle-type specific resuspension rates based on experimentally derived critical shear stresses GUST Chamber Erosion Study.	4
Deep burial	No data presented. Has potentially large impact on Hg mass balance.	1
Volatilization	No data presented. Has potentially large impact on Hg mass balance, particularly in wetlands with emergent deposition.	1
Particle decomposition	Information of particle die-off for several different vegetation types derived from Vegetation Senescence Studies report. The vegetation modeling is acknowledged as preliminary and could be refined if more modeling is conducted in the future.	2/3
Model Process – *Hg Species Interconversion*	**Comments**	**Ranking**
Methylation	Methylation assumed to occur in an active surficial sediment layer comprising 2 cm thickness. Model ignores methylation associated with periphyton associated with submerged and emergent macrophytes – a process pathway known to be important in other wetland systems such as the Everglades (Cleckner et al., 1999)	2
Demethylation	Not discussed. Presumably based on literature values.	2/3
Photo-reduction of Hg(II)	Not discussed. Presumably based on literature values. Uncertainty in this process directly propagates to uncertainty in Hg volatilization.	2/3
Photodegradation of MeHg	Not discussed. Presumably based on literature values. This process will be influenced by DOC dynamics which presumably can be altered by certain management strategies.	1

Photo-oxidation of Hg(0)	No discussed. Presumably based on literature values. Uncertainty in this process directly propagates to uncertainty in Hg volatilization.	2/3

2) Are the assumptions used for missing or inadequate data appropriate?

<u>*Tributary Loadings of Suspended Solids:*</u> How to treat missing data was an important consideration for developing tributary loads of both suspended sediment (SSC) and total MeHg/Hg. For SSC, the problem of missing data is particularly important for Fremont Weir, which constitutes the largest hydrologic input to YBP during the wet season (and thus overall). For the model calibration period, a total of only 22 observations were available for two wet seasons (2006 and 2016/2017) to estimate SSC loadings out of a total of 16 wet seasons simulated. Because SSC concentrations will vary as a function of time and flow, linear regression was used to develop a relationship between contemporaneous values of SSC at Freemont lagged by one day (36 miles downstream in the Sacramento River) and Fremont Weir. The actual bivariate relationship between the two sets of observations is shown in Figure L-4 of Appendix L. Inspection of the plot shows that the regression relationship significance is likely inflated due to the presence of a highly influential observation; likewise, the slope may also be biased because of the leverage induced by this observation. This is also illustrated by the plot of observed vs. predicted SSC concentrations at Fremont Weir (Figure 4-5) also suggests that the fit may be greatly influenced by a single observation and that the slope used to drive the functional relationship between Freeport and Fremont SSC would likely be substantially different if a more statistically robust regression modeling approach was used. A cursory examination of the lack of fit in Figure 4-5 indicates that the regression model substantially overpredicts SSC for observed values < 75 mg/L and generally overpredicts at SSC > 75 mg/L. At a minimum, the regression modeling should be redone excluding the apparent outlier or using robust linear regression and then evaluating the effect on inferred SSC loadings at Fremont Weir. Also, it is not clear what is gained by developing an inferential model for Fremont Weir using Freeport data rather than developing a relationship between measured SSC and flows directly at Fremont Weir. This latter approach was used for the other tributaries and is conceptually appropriate, although the report is silent regarding the actual number of observations, basic fit statistics, and the uncertainty in loadings inherent in these estimates. In addition, a regression model that considers flow interactions may yield improved results.

<u>*Tributary Loadings of Total Hg and MeHg:*</u> Tributary loads of both unfiltered Hg and MeHg were inferred based on empirical relationships between either flow and the constituent or SSC and the constituent. In the latter case, the relationship between SSC and flow was then used to infer MeHg and Hg concentrations as a function of flow. The limited information presented in the report presents a concern regarding the robustness of the calculations. For example, the predictions for high flow loadings of unfiltered Hg from the Cache Creek Settling Basin are based on a regression relationship using only three points (Figure 4-8; see also Figure L-26). The comparison of observed with predicted MeHg loadings for the Cache Creek Settling Basin (Figure 4-9; see also Figure L-33(a)) suggests the regression relationship is biased toward substantially underpredicting MeHg fluxes for high loading events and overpredicting loadings for low loading events. As with the SSC modeling, robust regression coupled with uncertainty analysis of the resultant predicted fluxes should be conducted. It is also noted that Figure 4-9 includes an apparent outlier (observed load ~ 40 g/day; predicted load = 0 g/day) and the calculations should be checked both to ensure this value is correct and whether this value unduly influenced the underlying regression analysis.

Similar concerns apply to the total Hg loading boundary condition estimates. For example, based on information provided in Appendix L, we estimate the RMSE for predicted Hg concentrations at Fremont Weir (Figure L-10) to equal 3.9 ng/L, which appears to approximate 40% of the mean value for the observed data used to fit the model. The comparison of observed and predicted values for Hg at Fremont Weir also indicates that the model error is not evenly distributed as a function of predicted values, with higher predicted concentrations biased towards underprediction, and lower predicted concentrations biased towards overprediction. This would suggest that the boundary conditions for total Hg – at least at Fremont Weir – are underpredicting the flux of Hg to the YBP. That fact, coupled with the relatively large RMSE, argues for more analysis of the effects of uncertainty on the boundary conditions and resultant conclusions derived from the modeling.

The regression relationship between MeHg and TSS at Fremont Weir shown in Figure L-14, similar to L-4, shows strong leverage and thus over-inflation of fit significance due to a single observation. Figure L-15 suffers from spurious correlation and has no meaningful predictive value (see comment on Delta modeling). Prediction bias identified for unfiltered Hg at Fremont Weir also applies to unfiltered MeHg predictions at Fremont Weir (Figure L-16).

No significant relationships between unfiltered Hg and unfiltered MeHg and flow were observed both for Knight's Landing Ridge Cut and Putah Creek. As a result, loadings for TSS, uHg, and uMeHg for KLRC inflows to the YBP were assumed fixed at average values. Given the results from the regression modeling, this assumption is reasonable. The effect of the uncertainty in these fluxes should be explicitly stated in the text, although from a system perspective, the effects clearly will be relatively small (Figure 4-12).

The liner regression models for estimating TSS at Sacramento Weir used log-transformed TSS as the dependent variable. Back-transformation of predicted TSS concentrations to their original metric form will be biased as a result. To avoid this bias, the model should be fitted using a general linear model with a log link for TSS. Although this error is likely not that large compared to other sources of error in the analysis, it is still important. The same concerns apply to predicting uHg for Sacramento Weir, which in turn imposes a bias on uMeHg since uMeHg is calculated as a fixed fraction of uHg.

Wet-Dry Period Transition

To facilitate conducting the D-MCM modeling as a single simulation for the entire period of record (rather than simulating 16 wet seasons separately), dry season flows (June-September) were constructed each year to hydrologically link TUFLOW results for different wet seasons. Dry season flows were constructed by calculating the necessary summer flow for each model cell that would transition from the water volume at the end of a TUFLOW simulation in May to match the volume of water in October. In the absence of actual hydrologic data, this approach is reasonable.

> ***3) Are priorities for future data collection (or model development, if needed) to reduce model prediction uncertainty identified and appropriate?***

The authors separate the priorities for future data collection into three categories: (1) Knowledge gaps; (2) Modeling scope issues; and (3) Model development issues.

The knowledge gaps include areas of uncertainty that translate to model uncertainty. Four major areas were identified, including: (1) vegetation influences on MeHg production and cycling; the question of whether legacy or newly deposited Hg react differently as substrates supporting methylation; variations in Hg exchange (partitioning) dynamics as a function of different particle types; and (4) the influence of wetting/drying cycles. All of these knowledge gaps are important and appropriate. In addition, the relationship between changing vegetation dynamics within the YBP and resultant (if any) changes in DOC is important and should be evaluated. While Appendix L does discuss DOC dynamics in the YBP in relation to total flow to the YBP and site hydrologic type (canal or interior non-canal sites), the text in both the report and Appendix L are not explicitly clear regarding precisely how DOC dynamics are modeled.

Modeling scope gaps include: (1) incorporating biota; (2) further testing of management scenarios; (3) climate change; and (4) uncertainty analysis, including the continued use of PEST++ to help identify uncertainty. As the report states, inclusion of biota is important because fish and shellfish Hg concentrations are the ultimate endpoint of interest. Further testing of management scenarios is warranted at this juncture as a tool for helping to evaluate model deficiencies rather than helping to define management strategies. We agree that future modeling should consider climate change; it should of course be coupled with a delineation of the hypotheses to be tested to ensure that the appropriate studies are undertaken to better structure and parameterize D-MCM. The continued use of PEST++ is to be encouraged. Indeed, it seems likely that the existing work already done with PEST++ coupled with mass balance information should be more fully analyzed to better define model uncertainties and priorities for future data needs.

Model development gaps include: (1) development of a single integrated model that incorporates hydrodynamics, sediment transport, and Hg biogeochemical cycling in a highly spatially resolved 2-D framework; (2) better representation of vegetation dynamics; and (3) a supporting field program as a necessary prerequisite to expanding model capability. We agree wholeheartedly with model development gaps (2) and (3). However, given that the current implementation of the D-MCM within the YBP had calibration data for a maximum of 7 out of 47 cells, we do not see how developing an integrated model for the YBP is worthwhile.

4) How relevant and reliable are the field data used for estimating parameters, establishing initial conditions, and specifying boundary conditions?

<u>Parameter Estimation:</u> Field data including critical shear stresses and diffusion collected as part of the Technical Studies were used for parameter estimation, along with literature values from other studies.

<u>Initial Conditions:</u> Insufficient surface water chemistry data are available to establish initial conditions as a calibration constraint within the model domain. For example, the D-MCM simulation starts in October 1996; the first observed data for uHg(II) for all but one of six model cells used for calibration do not predate October 2003 (cell 23 includes two observations from late 1996/early1997 and seven observations for early to mid 1998).

<u>Boundary Conditions:</u> Data for specifying boundary conditions include atmospheric deposition inputs and tributary loadings. Point source inputs directly to the YBP were not specified in the model report; these expectedly are relevant for quantifying tributary loading dynamics in response to management-related mitigation scenarios. Atmospheric deposition inputs for total Hg include wet deposition weekly data obtained from the Mercury Deposition Network (MDN) site most proximal to the YBP (CA72 near San Jose). The period of record for this extends from January 2000 through December 27, 2006. Dry deposition inputs were assumed constant and were based on the mean value for the San Francisco Bay Estuary from August 1999 through November 2000 reported by Tsai and Hoenecke (2001). Although neither wet and dry deposition are directly available for the YBP, nor does the period of record encompass the simulation period, the effects of likely uncertainty in these inputs are expectedly minor compared to tributary loadings entering the

YBP (estimated to be < 3%). Atmospheric inputs of MeHg were not reported but are likely to be small relative to external inputs from tributary sources.

5) Were appropriate calibration, validation, and verification steps taken to ensure model performance and accuracy?

Calibration: Calibration of D-MCM and parameter estimation are intrinsically interrelated. Calibration of the D-MCM was implemented by first varying uncertain parameter coefficients iteratively to produce simulated output values across the model domain in space and time; these results were then checked for goodness of fit with observed values. Goodness of fit was evaluated by computing the root mean square error (RMSE) for the all observed-predicted observations for a given response variable. This iterative process was continued until the RMSE was either judged to be acceptable or minimized. For D-MCM implementation for YBP, calibration was first conducted manually until the overall fit between observed and predicted values was judged reasonable. Often model calibration for complex biogeochemical models such as D-MCM does not extend beyond this step. For the YBP, however, the manual D-MCM calibration was followed by a second formal process using the PEST++ software package to optimize model parameter estimation and thus calibration. The implementation of PEST++ appears to be have been conducted with guidance from one of the key practitioners using PEST++ (R. Hunt of the USGS) and appears to have been well-implemented. As part of the PEST ++ optimization process, parameters related to suspended sediments, Hg, and MeHg were varied simultaneously in order to reduce the overall error for these three components combined. Because the number of observations available and some observation values can have order-of-magnitude differences, variable weighting was applied to allow PEST ++ to obtain a more balanced match across the different types of observations. The use of PEST++ resulted in a substantial reduction "in the error between observations and model estimates by 51% relative to the initial manual calibration". The text does not explicitly indicate what "reduction" means. For example, does the reduction pertain to the percent reduction in RMSE averaged across all the calibration parameters?

D-MCM calibration was conducted for the period October 1996 through May 2012. Calibration results for the following surface water variables – suspended sediment, unfiltered Hg(II) and unfiltered MeHg – were presented in Appendix G for six and seven cells for SSC/uHg(II) and uMeHg respectively.

From a hydrologic perspective, the calibration of the TUFLOW model underlying D-MCM encompassed a wide range of hydrologic conditions extending from wet to critical (wet > above normal > normal > below normal > dry > critical). This range helps instill confidence in the hydrologic modeling. In contrast, the observed surface water data for various Hg species used to calibrate D-MCM were sparse and discontinuous. For example, the cell with perhaps the most observations (cell 42 with 30 observations for uMeHg) included sampling from just three different time windows: October 2003 – April 2004; November 2004 – June 2005; and October 2005 to June 2006. Hydrologic conditions during the entirety of the above years ranged from below normal to wet and thus did not include more severely restricted water conditions. Comparisons of the dynamic variability in observed concentrations for uHg(II) and uMeHg in cell 42 (perhaps the most important cell of all the calibration cells for ensuring the accuracy of predicted export fluxes from the YBP below the Stairsteps) were reasonably good but did not well represent more extreme events. It would have been useful for explicit information on the RMSE for these variables to have been presented both overall and for cell 42 specifically.

D-MCM calibration also included sediment Hg and MeHg data. According to the text, most of the available data for these two variables were available for 2015-2016 with no reference to data actually available from within the calibration period. The implications of this issue were not discussed. This arguably less important a calibration issue compared to the paucity of surface water chemistry data given that the turnover time of the 2-cm surficial active sediment lens may be on the order of a decade or so.

*Validation: B*ecause of the limited amount of observed data, all the observed data were used for model calibration. This resulted in no formal model validation in which the performance of the calibrated model is gauged against a set of observations withheld from model calibration.

Verification: No information on Hg(0) dynamics was presented. Although there are no observations to compare with predicted results, volatilization is presumably a major loss pathway, particularly in flooded areas with emergent vegetation. Predicted fluxes should be presented and discussed relative to fluxes observed for other wetland systems. Likewise, the uncertainty in these fluxes should be evaluated and discussed *vis-à-vis* the impacts on the Hg mass balance and turnover times for the YBP.

This same concern applies to sediment burial rates below the active sediment lens. It also is unclear whether the mathematical implementation of gross sedimentation, erosion, and burial for the active sediment layer maintains a fixed depth during periods of excess erosion and, if so, how the Hg/MeHg mass balance for the sediment layer is maintained.

The D-MCM modeling report does include comparisons of tributary loadings and export fluxes of suspended sediment for the D-MCM with estimates developed by Springborn et al. (2011) as a form of model verification. That comparison, which the authors state is not precise with respect to time, nonetheless shows good agreement for suspended sediment tributary loadings. The comparison for export fluxes shows D-MCM approximately 20% less compared to the Springborn et al. estimates. Tributary loadings for total Hg(II) and total MeHg were similar or somewhat lower depending the actual tributary for the D-MCM inputs *vis-à-vis* the Springborn et al. estimates. No comparison was made for total Hg(II) and total MeHg export fluxes from the YBP. Thus, although the D-MCM tributary inputs for these three variable are consistent with the Springborn et al. (2011) results, the comparisons cannot be extended to help verify the model outputs for Hg(II) and MeHg because of the lack of data presented.

6) Were any revisions in model structure, initial or boundary conditions, parameter values, or assumptions made during the calibration, validation, and verification steps appropriate?

Revisions to the D-MCM structure appear to include at least one revision related to the mass balance of the active surficial sediment layer. It is not clear whether the sediment layer is maintained at a fixed thickness when particle resuspension exceeds settling. How this mass balance is maintained has direct implications regarding turnover rates of sediment Hg driving the primary assumed locus of *in situ* methylation; it thus has direct implications regarding the rate of response of the YBP to management strategies that influence particle mass balance or Hg inputs.

Another assumption which needs to be evaluated is that senescing vegetation particles are quickly delivered to the sediment-water interface whereupon the particles are instantaneously mixed with the active sediment layer. In reality, these particles likely have lower particle densities and likely are resuspended much more easily than the other particles comprising the sediments. As the authors indicate, how vegetation modeling is addressed within D-MCM for the YBP needs to be evaluated more carefully.

SENSITIVITY ANALYSES

1) Are the sensitivities of the model outputs to inputs and parameter values clearly identified?

The YBP modeling report included a series of sensitivity scenarios. Four categories were related to loading dynamics (particle related; external loadings of inorganic Hg; external loadings of external MeHg; and internal loadings of MeHg) with the last category related to vegetation dynamics. The scenarios and how the variable of interest for each scenario was manipulated are clearly defined in Tables 4-3 and 4-4, respectively. Results for the scenario runs are presented in Table 4-6. This table shows a range of results for each scenario simulation. suggesting that multiple runs were conducted for a given scenario; if so, the text makes no references regarding how the different runs were conducted.

Based on a review of the Chapter 4 report and Appendices G and H, no otherwise formal analysis of the sensitivity of the model to individual parameter uncertainty was conducted. The report does present the results from using the PEST++ software to conduct uncertainty analyses for modeled export fluxes of unfiltered Hg and unfiltered MeHg (Figure 4-11). In these analyses, the uncertainty in the model predictions (forecasts) was estimated by perturbing a selected number of parameters based on the *post hoc* calibration uncertainty defined by PEST++. The actual parameters that were jointly perturbed and their values was not identified in the text. The text indicates that "the uncertainty analysis performed conveys the primary findings regarding which model forecasts had relatively higher uncertainty, and which were most improved during calibration." The suite of uncertainty analyses results that apparently underly and thus justify this statement were not presented.

2) Were the appropriate model inputs or parameters varied to conduct sensitivity analyses? Are there additional inputs or parameters you would recommend for sensitivity analysis investigation?

The model inputs that were varied to conduct the sensitivity scenarios are defined in Table 4-4. How the manipulations were implemented are straight-forward with the exception of the vegetation category simulation. This latter simulation involved manipulating (reducing) vegetation biomass loadings by 50%; it is not clear how this was actually implemented (presumably by changing biomass production rates shown in Figure 4-6).

The sensitivity scenarios essentially focus on the management related questions regarding how MeHg fluxes exported from the YBP will change if (1) external inputs of particles, Hg or MeHg are reduced by 50%; (2) if MeHg internal production declines by 50%; and (3) internal production of vegetation particles is reduced by 50%.

The effect of vegetation on MeHg production within the YBP appears to be confined to carbon cycling impacts on the active surficial sediment layer. It is not clear if the effects of the extent of areal changes in vegetation were included in the analysis as an approach towards reducing vegetation-associated particle fluxes. Changes in vegetation dynamics should have a concomitant effect of methylation more directly associated with the macrophyte community. As discussed previously, this is a conceptual issue that should be addressed.

In order to help guide future data collection efforts (and with the goal of improving model uncertainty), it would be useful to conduct sensitivity analyses coupled with parameter uncertainties defined by PEST++. This would ensure relevant changes in parameter values rather imposing a single value on all parameter changes are used, thus producing a more meaningful sensitivity analysis.

Additional parameters to consider for sensitivity analysis would include variables related to the sediment mass balance, Hg(0) production/reduction/volatilization, and MeHg photodegradation rates.

3) If a scenario analysis was conducted, do the model studies appropriately represent the defined scenarios?

These scenarios included a suite of five major categories of analyses which correspond identically to those agreed upon for the YBP by the Open Water Workgroup. The parameters modified are appropriate to each scenario with the possible exception of the vegetation category. As discussed in the response to Question 2, how the vegetation loadings were manipulated requires more clarification.

4) Are the scenarios examined and the model outputs adequate and appropriate to support decision making? Are there additional scenarios you would recommend for evaluation?

Each scenario considered the effect of a change in a single input. The utility of these scenarios is questionable given the apparent lack of constraint in the parameterization of certain key parameters. For example, reducing Hg(II) concentrations (and thus loadings) by 50% at the Fremont Weir resulted in only a < 5% in MeHg fluxes at the Stairsteps. Hg(II) loadings via the Fremont Weir constitute 43% of the total external inputs to the YBP. The authors acknowledge that the effect was less than expected. Likewise, a similar scenario related to reducing TSS (total suspended solids) by 50% at the Fremont Weir indicates a less than 5% decline in MeHg export at the Stairsteps as well. Similar results were obtained for the same scenarios for CCSB inputs. From a mass balance perspective, these results are troubling and may reflect issues with how the particle balance is implemented and maintained for the active surface sediment layer. The authors should discuss more fully which scenarios may be reasonable and why and which scenarios are indicative of model problems that should be resolved.

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6.4. Open water Report: Delta Mercury Modeling

Response to the Charge Questions

1. The results of the modeling study achieved the majority of the objectives of the revised and reduced work plan.
2. The major processes were included in the model but potentially important processes and sources were not, as detailed below. The discussion of uncertainty was not highlighted in the report and not adequately documented.
3. Priorities for future work were presented but additional suggestions are made below.
4. One significant lack was sufficient data on Hg, and especially MeHg concentrations, that are needed to further improve and refine the model. More data is required on tributary concentrations to allow for better relationships to be developed on Hg/MeHg and flow/sediment load for some sources, as detailed further below, as some estimation approaches do not provide a defensible relationship with flow. Additionally, some parts of the Delta are lacking in Hg/MeHg data. Important processes that could be important but are not in the model currently need to be investigated.
5. As A weakness of the calibration was its relied reliance on the existing data that was limited in some respects, this limited the ability to calibrate the model.
6. The development and inclusion of a sediment exchange portion into the model was a good development and likely improved the model accuracy and predictions. Some of the parameters in this model could be better constrained based on recent findings in the literature.
7. The sensitivity analysis was limited and also did not properly account for linked variables in the process of doing this analysis. Moreover, the magnitude of imposed sensitivity parameter reductions (10%) was much lower than the likely uncertainty in some model inputs, including tributary loadings, limiting their usefulness, as detailed further below. A more detailed analysis is definitely required both looking at more parameters in more detail and placing the uncertainty in boundary conditions in context.

2. Details on the Reviewers' Response to the Charge

Overall, the model provided and the work done is mostly in accordance with the substantially modified and reduced objectives of the work plan, as detailed in Technical Appendix A. Overall, the model development and its results somewhat addresses the first two objectives of the original work plan, but does not address the other objectives. The reviewers were concerned that the report does not present a summary mass balance of THg and MeHg for the Delta that specifically shows what are the major sources and sinks, and provides details of the specific process pathways for both sources and sinks for each Hg species coupled with a discussion of the uncertainty in those input/sink pathways. Given that, we conclude that the model report currently does a poor job of providing "a basic understanding of the important processes governing MeHg production."

The lack of completing all the objectives reduces the usefulness of the model in its current framework for making predictions about how to reduce MeHg levels in the Delta, or how changes would exactly propagate into the food chain. Development of a bioaccumulation model in the future is an obvious need to allow for the better prediction of the impacts of changes in MeHg loading on MeHg levels in biota. Recent studies in coastal environments (e.g. Chen et al., 2014; Buckman et al., 2021) have suggested that concentrations in forage fish are more closely linked to the water column MeHg concentration, rather than the sediment MeHg;THg; therefore, if this is true for the

Delta region, then there is a critical need to obtain accurate predictions of water column MeHg, and controlling processes, and this does not appear to be the case with the current version of the model, as discussed further below.

The model development used all the available Hg speciation data in formulating and testing the model so no data was available to test and validate the model predictions. More data should be obtained in the future to further test and refine the model. The lack of inclusion of various sources/processes in the model is depicted in Fig. 5-7 and while some of these are reasonable – e.g. WWTP inputs of MeHg are minor compared to other sources – there are some sources (urban runoff and wetland inputs) that need to be further evaluated and incorporated into future versions of the model, although these are characterized as minor sources in the work plan. It was not discussed what data these conclusions are based on. Also, as discussed below, other potential sources of MeHg need to be considered.

The main hypotheses that were to be have been tested with the model according to the original work plan were somewhat addressed given the modified scope and model development. The model results appear to confirm the first two and the final hypothesis, which have also been demonstrated by earlier mass balance studies, that tributary loads are the main source of MeHg to the Delta and outflow is the major sink, and that other external inputs (atmospheric, agricultural and open water sediments fluxes) are not important. The third hypothesis that these sources may be important in some locations is not well addressed in the report. However, there are specific aspects of the models and the lack of information on sources (and consideration of other potential MeHg sources) that make it impossible to validate these conclusions because of the model structure and the processes incorporated. Further refinement of the model will likely result in the conclusions being further modified if there was better process characterization, and the inclusion of missing potential sources in the model.

Fig. 5-10 shows the comparison of the model with data for the suspended sediment concentration (SSC). While some of the model results compare well, there is evidence that the peak concentrations are not well captured for some locations, and for others the low flow conditions are not well characterized. The same is true for Hg, as shown in Fig. 5-14, and discussed further below. It is clear from the data shown that there is a critical need to gather further Hg data during conditions of high flow. For MeHg (Fig. 5-15), the low flow conditions are also not well captured and there is not sufficient explanation of why this may be so. As the Delta is a sediment sink, it is not surprising that it is also a sink for Hg, but the conclusion that it is also a sink for MeHg is somewhat surprising, and may need to be further examined, as discussed further below.

Fig. 5-21 shows that there is substantial annual variability in the relative amount of MeHg inputs from the various tributary sources and it is not clear why this is and how such differences can be confirmed, either with existing data or with future studies. This was not adequately discussed in the report.

The differences in the relative importance of the sediment sink within the Delta is not well explained in the report and requires further investigation. Indeed, while the model suggests that the Delta is a sink for 50% of the sediment, only 25% of the Hg and 13% of the MeHg is removed. In addition, the uncertainty in input fluxes would indicate that the model is presently not capable of resolving whether the Delta is truly a net sink. Indeed, the conclusion that the Delta is a net sink for MeHg may simply be an artifact of errors in the model inputs and calibration. The differences in the strength of the dissolved-particulate partitioning for Hg and MeHg could account for their differences in their removal or the differences in removal may reflect differences in sources/sinks for Hg compared to MeHg in the Delta. This was not highlighted in the report.

While the model accounts for most of the processes that are known to affect the concentrations and exchange of all Hg forms, the model does not appear to take into account some processes that may be important and that have been a focus of more recent investigations in the literature. The ability of the model in terms of accurately predicting observations in terms of Hg and MeHg concentrations in the filtered and unfiltered fractions is likely the result of not incorporating all the relevant processes in the model. Before discussing this, it should be noted that the model does not always provide even a good prediction of the sediment load, demonstrated by the comparison to USGS data for the Middle River, as shown in Appendix J, Fig. J-14. There is little data presented for the model comparison in the appendix, but the data in Fig. J-14 shows that the model overpredicts the fluctuations in the sediment load (SSC), and if this is the case, then the difficulty in predicting the concentrations of Hg and MeHg is even harder. While Hg is strongly associated with particles, and the load estimate regressions show that the relationships are relatively well constrained (Fig. I-10), the same is not true for MeHg (Fig. I-11). It was somewhat disconcerting that the model relationships used to calculate the MeHg flux from the suspended sediment flux were not universal across the locations. Some of the relationships used are %MeHg vs. SSC are exponentially decreasing while others are linear for uMeHg vs. SSC. The rationale for doing this is not adequately presented. A detailed evaluation of the potential errors with this approach is outlined below in Section 3.1. It can be said, however, that the data is very limited for many of these relationships. and if the model is to be further developed and used, then the collection of additional data is obviously a priority.

The lack of inclusion of the wetland model was agreed to in the revision to the work plan but not having this as a part of the model is a large omission. While the model and data show that the Yola Bypass is a source of MeHg, the conclusion for the Delta is that it is a sink. If true, this may be due to the differences in sedimentation in the two areas, and there should be further discussion of the differences in the dynamics and Hg/MeHg cycling in the two systems. Again, the lack of inclusion of processes may account for this difference to some extent. This issue is dealt with further in this Report on Tidal Wetlands (section 6.5 as there are concerns about how the studies were done and that the conclusions that wetlands are not a source are not totally supported.

One important omission is the potential for methylation of Hg in other locations besides the open water sediment. It is known that methylation in the water column is the main process whereby MeHg is formed in large water bodies and more recent studies, such as Schartup et al. (2015), have shown that mixing zones, where tributary inputs mix with estuarine waters, lead to substantial methylation and that the sediments may not be the most important source of MeHg to dynamic water bodies. Also, the potential for methylation on biofilms/periphyton associated with vegetation, or associated with roots (as summarized recently in Branfireun et al. 2020), or the methylation within large particles in the water column (e.g. Ortiz et al., 2015), has not been considered in the model. Indeed, in the Yolo Bypass model (Figs. 3-39 & 3-42) and the data, it is shown that the vegetated areas are much larger sources of MeHg than the non-vegetated zones and, while it is not identified why, it is likely the fact that the vegetation promotes methylation. All these processes could lead to in situ formation of MeHg in the Delta and thus their inclusion would improve the Delta model.

The process of sediment resuspension and the potential transfer of Hg/MeHg is another area where there seems to be a lack of consideration of the recent literature. For example, Seelen et al. (2018)

suggested that bulk sediment measurements were not a good predictor for the flux of MeHg associated with resuspension and that the resuspended particles more closely resembled the suspended particulate than the bulk sediment. In addition, resuspension can input sediment plankton biomass into the water column and also result in inputs of nutrients, and these processes are not considered within the model framework. Many studies in dynamic ecosystems have shown that there is not a strong relationship between the suspended particulate MeHg concentration and that of the bulk sediment, and so this process does not appear to be well formulated in the model (Appendix I).

The collection of additional data is a priority also because the model appears to be relatively unconstrained. This is a consequence of a lack of data on two key loss pathways – evasion of Hg(0) (which is highly uncertain in wetlands); and net (permanent) burial of Hg below the active sediment layer engaged in methylation and resuspension.

The sensitivity analysis of the model was added as Appendix K. The sensitivity analysis was restricted to examining the impact of a small (10%) decrease in variable value based on concerns that higher parameter reductions would result in model stability problems. This concern about model instability and the need to constrain the parameter reductions to 10% is troubling given that, for example, the uncertainties in individual tributary inputs of SSC and THg are certainly much larger than 10%. More details are given in Section 3.4.

The analysis did not also link the impact of reduction in the value of one variable on closely related variables, for example, reducing the sediment load but not simultaneously reducing the Hg and MeHg load that would be associated with this, which makes the interpretation of the results more difficult. Given the model results that the major source of MeHg is external inputs from the tributaries, the results of the sensitivity analysis are consistent with the observation that the major impact on MeHg in the Delta is from the reduction in the major loads to the system. The lack of linking parameters in the sensitivity analysis is demonstrated by the fact that a reduction in sediment loading is shown to cause an increase to MeHg export from the Delta, which is due to the fact that with less sediment load, there is less removal of MeHg to the sediment within the Delta itself. Overall, the sensitivity analysis would appear to confirm the conclusion that the external load of MeHg from the major tributaries is the factor controlling the extent of MeHg export from the Delta. If this is truly the case, then the management option from the modeling study is clear that reducing upstream MeHg sources is key to reducing MeHg in the Delta. This conclusion, however, presupposes the accuracy of the boundary conditions loadings and the appropriateness of the resultant model parameterization based on calibrating the DSM2-Hg to those boundary conditions. As discussed earlier and detailed at the end of this section of the report, the loadings are based at least in part on tenuous or inappropriate regression relationships which in turn have direct impacts on model calibration. At this juncture, the sensitivity analyses provide value only in illustrating that the model results are affected by changing inputs.

Detailed Evaluation of the Model and Model Calibration

Concerns were raised about the *boundary condition computations* for the external inputs.

Unfiltered Hg: The Cosumnes relationship suffers from heavy leverage by a single SSC point that is nearly 5x greater in concentrations than the next higher observation. As evidenced by the high coefficient of determination, this results in fitting a bivariate linear regression model through two points. The Calaveras regression model has only three observations and is further hindered by the clustering of the points into two groups.

MeHg: The estimates for boundary inflow concentrations using the regression relationships for SAC Freeport and Cosumnes shown in Figure I-11 are highly problematic for two reasons. First is that the functional relationships are based on spurious correlation given that the relationships essentially regress the inverse of unfiltered Hg (multiplied by concentration of unfiltered MeHg) as a function of itself. It can be shown through simulation exercises (cf. Pollman and Axelrad, 2014) that the same bivariate relationship can be obtained for a data set with similar ranges in unfiltered Me and Hg and for which the relationship between both species is specified *a priori* to be uncorrelated (Figure 2). Thus, the derived regression models for both sites may have essentially no predictive utility (Figure 3). The Cosumnes relationship also suffers from heavy leverage imposed by a single uHg point that is > 3.5 times the concentrations of the remaining observations.

The second problem is that modeled unfiltered Hg concentrations rather than observed values are used to infer uMeHg concentrations, as only limited observational data are available for uHg. It would be more appropriate, however, to use a model derived as a function of SSC values directly since the inferred values of uMeHg ultimately are calculated as a function of strictly SSC.

The uMeHg regression relationship for Calaveras is based on only four points.

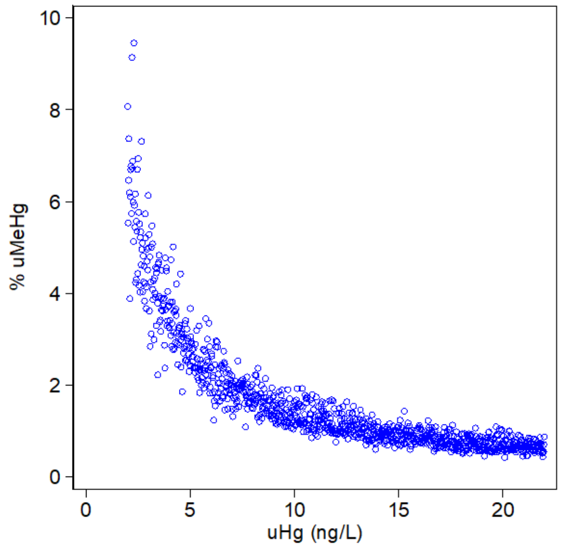

Figure 2. Plot of relationship between % unfiltered MeHg and unfiltered Hg. Relationship is based on assuming: (1) the range of both variables approximates that observed for SSC Freeport in Figure I-10; and (2) the relationship between unfiltered MeHg and unfiltered Hg is completely random. The apparent inverse regression relationship (which is highly significant: $r^2 = 0.92$; $p < 0.001$; N = 1,000) – and which mimics that shown in Figure I-10 – is spurious because the bivariate relationship includes unfiltered Hg as both an independent variable and a modifier of the dependent variable.

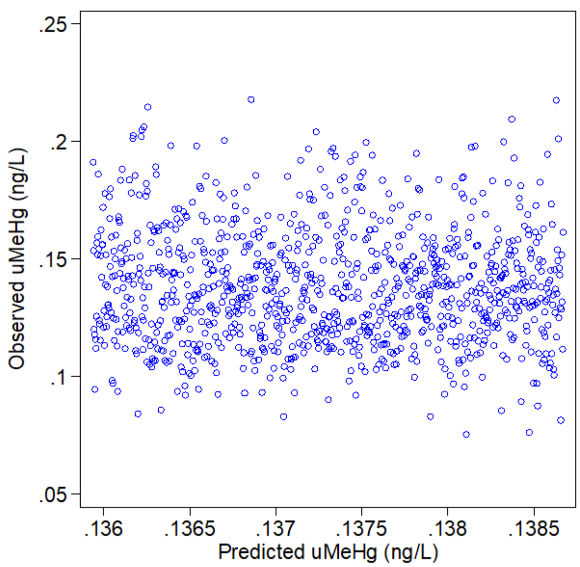

Figure 3. Comparison of observed MeHg concentrations used to construct with predicted values obtained from the fitted model. The figure illustrates that, despite the seeming excellent fit of aspects of the model to data(?), the model has <u>no</u> predictive ability because of the underlying spurious correlation. The statistical modeling for both uHg and uMeHg is presented with no evaluation of model robustness. Assuming the regression models are implemented correctly with proper model specificity (as is particularly a concern for uMeHg), it would be useful to see plots of the

distribution of residuals as a function of predicted values as well as an assessment of leverage/influential observation impacts. We recognize the modelers are trying to make the most out of very limited data to estimate meaningful boundary conditions or inputs. Unfortunately, the problems with limited data/leverage/influential observations (uHg and uMeHg) as well as inappropriate regression variables (uMeHg) suggest that considerable error is likely inherent in the boundary condition estimates. As a result, the boundary condition estimates should also include some estimate of error in predicted values. For example, the RMSE for the hypothetical example shown in Figure 1 is 0.077 ng/L compared to a mean value of 0.138. Errors of these magnitudes likely will have important implications regarding mass balance assessments and elucidation of sources and sinks. It should be noted that the authors do acknowledge in the final section of the report (Data/Knowledge Gaps and Next Steps) that boundary inflow loads are based on limited data that do not adequately characterize the highly variable nature of these inputs over short time intervals. Moreover, the authors also indicate that these boundary conditions strongly influence Hg concentrations in the Delta. This has important implications for the sensitivity analyses.

Modeled Processes

It appears that pH is modeled as a conservative constituent based on using pH inputs from 8 different regions delineated in Table I-4. A better approach in lieu of a thermodynamic complete approach would be to use alkalinity and then estimate alkalinity concentrations in different cells assuming alkalinity is conservative. This admittedly could be complicated by DOC effects which would need to be considered, as well pCO_2 dynamics. To the extent that pH varies between inputs, this could be a meaningful correction.

DOC is assumed conservative in the model. It is not clear how DOC is modeled. Since it is such an important variable in Hg cycling, including methylation and photodegradation of MeHg, and because management options may influence DOC dynamics, how DOC is modeled needs to be explicitly addressed.

Comparisons with Other Studies

Table 5-6 in the report shows input loadings from the Sacramento River, the San Joaquin River, and the YBP to the Delta estimated from the DSM2-Hg model in comparison to previous (observed) estimates developed as part of the CalFed (2008) report. The Delta model report indicates that the agreement for MeHg loadings is "similar" between the observed and modeled estimates but is silent on the THg and SSC loadings. As Figure 3shows (which is a bar chart constructed herein from the data presented in Table 5-6), the agreement for both SSC and THg individual component loadings is quite poor. Why the large discrepancies between modeled and predicted SSC and THg fluxes needs to be addressed. Moreover, these discrepancies have implications for conclusions drawn regarding major source inputs for both parameters. For example, the observed data for SSC loadings show the following ratios (SCR:SJR:YBP): 0.48:0.42:0.10, while the modeled ratios are: 0.17:0.71:0.12. Similar discrepancies are noted for THg loadings. These results call into question whether the Discussion section statement on page 5-33 that "[t]he Sacramento River was the largest estimated source of water, suspended sediments, uHg(II) and uMeHg to the Delta for the simulation period" is truly meaningful.

Sediment Data

A total of 201/424 raw observations for sediment MeHg and Hg concentrations respectively that were collected between 1999-2017 were used as input for initial conditions to the Delta model. The raw data were aggregated for co-located samples and restricted to include observations from the uppermost 5 cm only (which is useful with respect to minimizing biases on the effective concentration imposed by including sediments from below the surficial active exchange layer). This process resulted in a total of 130/210 observations of MeHg/Hg respectively as input to the model.

Figure 5-17 shows spatial plots of the observed and simulated results for sediment MeHg and Hg. While the authors note that further analysis of the data, as well as the collection of additional samples, is necessary to better establish spatio-temporal dynamics, Figure 5-17 may be confusing to the reader, particularly when the stated intention is to visually demonstrate that the patterns are 'comparable'. Even a cursory examination shows that there are many regions of disparity, and the stated challenges of merging data collected at different times across many years. This is a simulation to demonstrate some internal model coherence, and is not a true prediction (i.e. the model is calibrated with an empirical dataset, and then the model generates current or future patterns that can be tested).

Data Gaps and Sensitivity Analyses

Model Development: We wonder if the D-MCM and the DSM2-Hg models would predict the same results if both models were applied to the same test system with the same parameterization of the model coefficients. This seems like a relatively simple exercise to demonstrate that the two models are functionally equivalent.

Sensitivity Analyses: The sensitivity analyses were conducted as a series of 21 separate simulations where a single input parameter was reduced by 10% based on concerns that higher parameter reductions would result in model stability problems. This concern about model instability and the need to constrain the parameter reductions to 10% is troubling given that, for example, the uncertainties in individual tributary inputs of SSC and THg are certainly much larger than 10%.

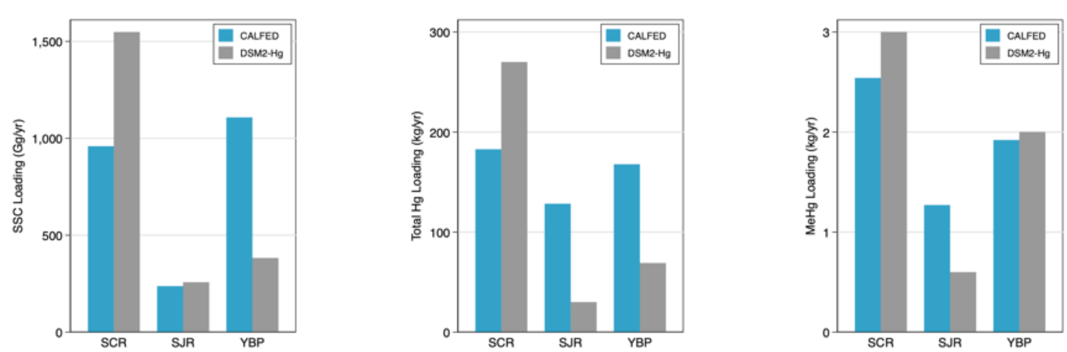

Figure 4. Comparison of estimated Delta input loadings from the Sacramento River (SCR), San Joaquin River (SJR) and Yolo Bypass (YBP) obtained from the CalFED (2008) report and the DSM2-Hg model. Left-hand panel: suspended sediment (SSC); middle panel: total Hg; right-hand panel: MeHg. Data from Table 5-6.

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6.5. Tidal Wetlands Study

The scientific objectives for the tidal wetlands study were largely well conceived and the workplan was carried out as closely to "as planned" as we have seen for any of the Reports, though this final report does still lack a rigorous analysis of the data and is missing some key data components from the data submission. The workplan sought to examine net tidal fluxes of total Hg and MeHg from 3-8 wetlands. The final report includes studies on four wetlands, one across each of four different years. The study leaders did a lot of difficult work and were successful with many things. Sometimes the setup of these types of studies is the most difficult part and though it is apparent from the workplans that a lot of consideration went into choosing sites, there are some potential issues related to the rigour with which the generated data is able to support the scientific objectives. We believe that some of the data are more believable as presented for some wetlands than for others. Importantly, we are struck that this study has no tangible link to the "open water" modelling report/study.

The total Hg and MeHg analyses are high quality and QA/QC measures are good. Though we may have chosen ourselves to focus on filtered samples and the particulates themselves (rather than filtered and unfiltered), we were happy to see the authors distinguish total and filtered samples, which is a major improvement from studies we examined in the first phase of control studies review.

This particular study was greatly needed because the current wetland allocations (including tidal wetlands) are based on work from one wetland, which is non-tidal, and unlikely to be representative of the significant coverage of tidal wetlands in the region. This Delta report focused on freshwater tidal wetlands, so it remains uncertain if tidal saline or brackish wetlands would be represented by any of the current science or this report for adequate allocations in relation to the salinity of the systems. There is some peer-reviewed literature available for total Hg and MeHg net tidal fluxes in tidal wetlands that are not freshwater, such as Bergamaschi et al. (2011) within the San Francisco Bay area, Mitchell et al. (2012) within the Chesapeake Bay region, and Turner et al. (2018) within the Penobscot River estuary. Of particular importance is that the main findings of this report are ***not in agreement with these published works,*** though some of this work does suggest that tidal wetlands may be only relatively small sources of MeHg. While this is possible, our elaboration below suggests that there are technical issues in this report that lead to uncertainty about the strength of the conclusions drawn.

Overall, the findings of this tidal wetlands report point to a conclusion that freshwater tidal wetlands in the system are negligible sources of MeHg to the Delta; or are small sinks of MeHg. However, we are concerned that some issues with representativeness and almost certainly, some hydrological measurement errors may have potentially biased this conclusion. ***As yet, we are unable to fully support the report's conclusions that tidal wetlands do not contribute MeHg to the Delta***.

There is a general lack of detail in analysis and presentation, as well as with the submission of data. We provide these points as examples:

- There is a fairly substantial gap between the raw data provided and the much more summary data presentation/graphics in the report. It was a challenge to review this study because the underlying data were not presented or evaluated. We feel there that significantly more data could be presented in Appendices, which would allow us to more substantively review the results. At a minimum, this should include time series of numerous variables measured continuously and should include calibration data relevant to the velocity indexing (not just the equations). Where possible, we did some analysis of raw data that highlights some potential problems, but we are unable to complete a full re-examination of the data within the limits of this charge.

- One example relevant to the point above is that data are not presented as time series for any of the 25-hour, more intensive sampling campaigns. We have examined some of the data ourselves (see point #4 Page 57) and find that patterns in concentrations do not support a role for these wetlands as MeHg sinks. The patterns are variable and concentrations at times are quite close to detection limits, but there are generally not higher concentrations of MeHg coming into the wetlands, compared to leaving.
- More analysis could have been done with the ancillary measured variables. In our own analysis, we found that turbidity tended to most often peak on the ebb tide, sometime more closely to slack low tide. An example time series is given below. Over longer time periods, it appears quite clear that the Yolo marsh at least is a source of particles. Figures 12 and 20 of the report show that this marsh is almost always a sink for particulate MeHg and total Hg, respectively. This seems in contrast to one another. We suggest that further analysis could be done to take advantage of suspended solids and turbidity data, examining relationships with particulate Hg and MeHg separately for ebb and flood tides.
- Though this fits with the workplan, there is only one year of data for each wetland, and little of no context of how water levels in each year fit into long-term inter-annual trends. Most of the wetlands were evaluated during a long drought. An opportunity to sample during major flooding in 2017 was missed.
- There is no context provided for water levels during the studies relative to the surface level of the marshes. Beyond Blacklock, which was described as flooded most of the time, were the marsh surfaces flooded at any time during the studies or was flow restricted to the channels? We think it would be important to have this information for context about Hg and MeHg fluxes relative to wetland flooding. An evaluation of MeHg concentrations or flux vs the extend of inundation (over time) would have been helpful. It is quite possible that measurements and/or surveying exist that could still enable this analysis, but this is not with the data product currently.
- It would be helpful to have more information about context of the conditions during the 25-hour sampling periods, especially as this relates to tidal heights.
- Additional information about the wetlands themselves would be helpful. How well do the wetland characteristics match up with the distribution of tidal wetlands in the Delta? It would have been helpful to have information about when the wetlands were created, the prior land use, type of soil, and type of vegetation, and importantly – the level of Hg contamination. From the particulate numbers, it does not appear that any of the marshes were in Hg contaminated areas. Though we understand that this may have been outside of the authors' control, we were struck somewhat that no wetlands were within the Central Delta, San Joaquin or Sacramento River areas.
- There remains a disconnect between this study and the "open water" study. For example, why does the Yolo mass balance show net MeHg production in Yolo during flood and se tidal wetlands did not? We can think of several reasons that were not explored, including a first flush effect during the 2017 Yolo flood, that might not have happened in any of these

wetlands. Alternatively, it is unclear from the presentation of data if any of the small wetlands tested here, outside of the Blacklock wetland, ever had water overtopping the marsh. This disconnect need to be understood to figure out BMPs for Hg in the Delta.

There are data analytical issues with the report and potentially some errors. Before the report's findings can be taken at face value, some consideration of the following is needed.

1. Possible hydrological calculation issues. We were struck by the very large water flow sum differences presented in Tables 4 and 7, which seem too large to us. ***We suspect that fluxes of water (and possibly Hg) were incorrectly calculated***. According to Tables 4 and 7 (same flow data in both tables) there is a median (n=10 x 25-hour tidal cycles) of 92,155,620 liters per tide more water entering the Blacklock wetland than flowing out from the wetland, at least where it is measured. This is 92,156 m^3 per tide, or an average of about 1 m^3 s^{-1}. For the other wetlands, these imbalances were 12,095 m^3 per tide for Yolo, 3,987 m^3 per tide for Westervelt and a net average outflow at the North Lindsay Slough of 1,637 m^3 per tide. These values do not match with our own examination of the spreadsheet data submitted, which is explained here for Blacklock. In an examination of 13 random 25-hour tidal cycles spread across the year for the Blacklock South Breach (one of two breaches measured for Blacklock), we calculated an average net inflow of 502 m^3 per 25-hour cycle. The average total flow (not associating negative vs. positive for flows in versus out) for each 25-hour cycle was 7,940 m^3. These numbers are two orders of magnitude lower than those given in Tables 4 and 7. Given the flow values are similar for the North Breach, there must be some error in the overall calculation of flows. This could lead to issues with the magnitude of the calculated fluxes, but we are not able to re-run all of the flux analyses to confirm.

2. Hydrological/flow velocity measurement bias. From the flow data provided, there are easily distinguishable directional biases in terms of net tidal fluxes of water (Figure 5 below). If one plots a time series of the tidal flows (an analysis not included in report), the range of values is biased toward the negative (into wetland) compared to the positive and this bias appears to worsen through time in most of the wetlands. This suggests that significant amounts of water are accumulating in the marsh through time, which is implausible without very substantial evapotranspiration (see next point). Though the flow differences in Tables 4 and 7 may be incorrect (see above), it is still likely that the trends are real. In Tables 4 and 7, the measurement bias varies depending on the wetland, with the worst performance from the Blacklock wetland, followed by the Yolo wetland, and then bias that may be within measurement uncertainty for the North Lindsay Slough and Westervelt wetlands. The measurement bias could be due to a number of reasons. As one potentially illustrative example, ADCPs will provide data in 2-3 dimensional directions, depending on the particular instrument. It would be good to know if at Blacklock in particular, the ADCP was shifted a bit in relation to the dominant flow direction and whether this shift was noticed and data were corrected. The hydrological imbalance in that site's flow data in particular appears nearly non-existent over the first stretch of time, but worsens considerably later on.

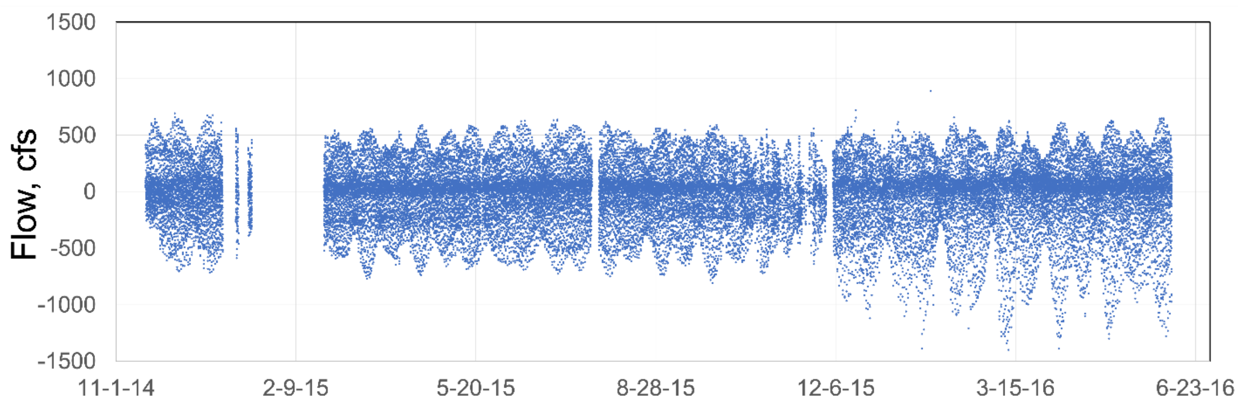

Figure 5. An example of worsening directional bias through time from the tidal flow record at the Blacklock North Breach. Notice that positive flows tend to stay consistently at a maximum of about 600 cfs through time, whereas the negative flows (into wetland) are initially similar but increase to nearly 1,500 cfs by the end of the record.

3. Water budget issues likely impact estimated sink/source function of wetlands for Hg and MeHg. In general, the evaluation of tidal marsh fluxes using the report's approach requires a close examination of, and hopefully near closure, of the water budget. This should/could be done at two times scales; 1) the "intensive" 25-hour tidal cycles; and 2) the longer ADCP deployment periods of approximately a year, at least where data integrity allows. As outlined above, the data presented in Tables 4 and 7 suggest a significant tidal flow imbalance in most wetlands. The large imbalances between water flowing into vs. out of the wetlands (note substantial differences for two of the studied wetlands – Blacklock and Yolo) will have an impact on whether the wetlands act as net sinks or sources for Hg and MeHg. In other words, the Hg and MeHg flux data should be and could be more appropriately interpreted in the context of the water budget. Problematically for example, the wetlands acting as the strongest net sinks for MeHg are also those with the strongest net imbalance of flows, such that much more water is coming into those wetlands than going out. Though evapotranspiration and precipitation were considered negligible fluxes in these systems, these data should be obtainable from nearby sources or should be estimable from reasonably available data, in order to demonstrate this. Additionally, no information was provided on the potential terrestrial inflows to the wetlands. Finally, specific conductance data could be used to help support or refute the water budget estimations.

4. An example from the Yolo wetland that illustrates the above point. Using the data provided, we plotted total Hg and MeHg concentrations and water depths over the tidal cycle at the Yolo wetland during one 25-hour sampling period in June 2014. This is shown below in Figure 6. It is clear that unfiltered total Hg and uMeHg especially are highest at slack low tide – so there is a source in marsh water. Pairing chemistry data with the ADCP flow data, we were able to calculate flow-weighted concentrations on ebb and flood tides. We found that average concentrations (both arithmetic averages and flow weighted averages) of nearly all constituents measured - uMeHg, PMeHg, and PTHg – as well as TOC and POC - were higher in ebb tide waters. So, the marsh is a source of all of these things on this tidal cycle. Our rough flux calculations then do match relatively closely with what is shown for this report on Figure 8 for MeHg. However, the report shows PMeHg efflux from the marsh for this event in Figure 9, but net influx for the monthly loads in Figure 12. How does that possibly come about? We suggest that this is most likely driven by issues with tidal water flux calculations.

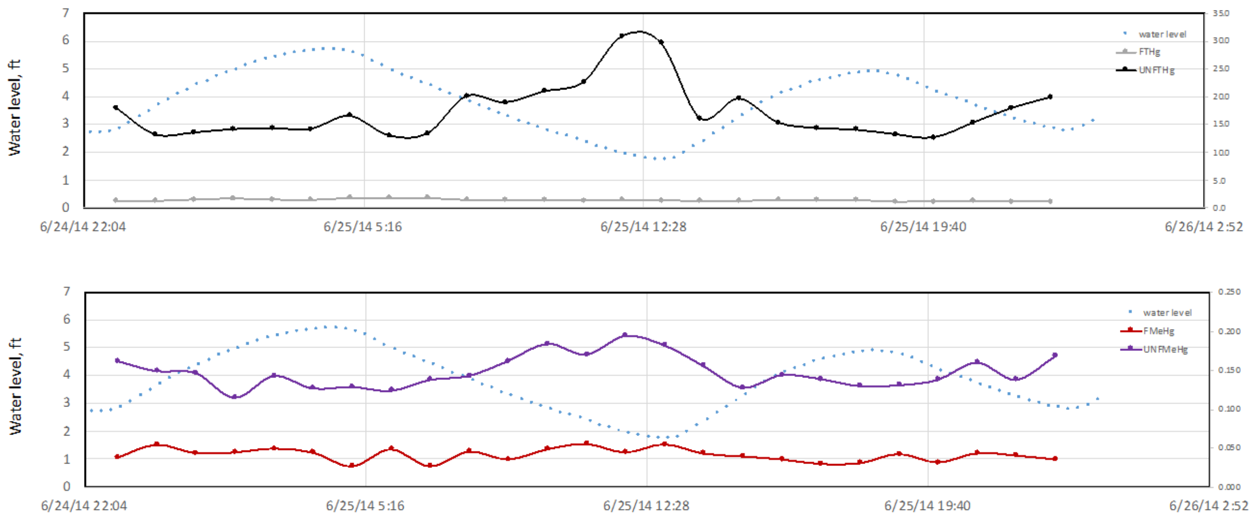

Figure 6. Time series of Hg/MeHg concentrations with tidal water levels at the Yolo wetland in June 2014.

5. Lack of uncertainty analysis. The hydrological discrepancies noted above point to a need to directly assess errors and uncertainties in the loading calculations, something that was not part of the study. Though not explicitly part of the objectives or even workplan, it is scientifically deficient not to include some sort of uncertainty analysis as part of a study such as this. Measurements and analytical work that can lead to loading uncertainties in tidal wetland studies include multiple aspects of the index discharge calibrations, including errors in the index velocity measurement, the channel average velocity measurements, stage-area relationships, and the index rating curve, in addition to sampling, precision and accuracy uncertainties for chemical constituents. We point the report authors to Ganju et al. (2005), which is a helpful reference for examining errors and uncertainty for mass fluxes in tidal wetland systems. This type of context is very much needed for this report. With the report and data as provided, we are unable to make an assessment of uncertainty ourselves. We would therefore encourage the retrieval of further data on the project, including some of the raw data and calculated files. Alternatively, the study authors could be contracted to examine these issues further.

6. A further examination of the data specific for times when some flooding of the wetlands' surfaces would have occurred. Like the open water report, the tidal wetlands report may have also suffered from taking place partly during a long stretch of drought. Additionally, though the setup of measurements in channelized areas of tidal wetlands is necessary for decent empirical measurements and sampling, we are somewhat concerned that the chosen sites may not have been measured often when the wetlands were actually flooded, but with tidal flows staying largely channelized (with the exception of Blacklock, which was nearly always flooded to our knowledge). Water staying in the channels (and therefore mixing less with the majority of the wetland area) may be exacerbated by less water in the region overall during drought. If most flows are sustained only within channels, this would also bias low the net outputs of MeHg from these systems if the rest of the wetland systems themselves are not substantially flooded/interacting with tidal flows. We performed only a cursory review of the DOC data, but the finding that the wetlands are not sources of DOC supports a hypothesis that most of the marsh areas do not interact substantially with tidal flows. In our opinion, it would be a worthwhile endeavor for the study authors to revisit their data and focus on measured tidal cycles when substantial flooding of the tidal wetlands was apparent. This could be compared to tidal cycles when it is clear that tidal flows remained channelized. Such an exercise would more clearly delineate the role of the wetlands and would potentially be helpful with some inference about climate change impacts.

It is our opinion that carrying out a more substantial analysis of the data and focusing on the above suggestions would likely help to improve the loading allocations. Improvements to the analyses this way would also make this and produce a robust dataset invaluable for future modelling efforts; something pointed out by the study authors and that we agree with in principle. An alternative way of estimating or assessing loading allocations could simply involve simply include more MeHg measurements at more appropriate times and places. This could include sampling, including drainage areas across a breadth range of tidal wetland area coverage. These additional measurements which would help to resolve differences between this study and observations of the YBP mass balance in the Open Water report.

With regards to mercury/methylmercury source characterization and control, how well does this report identify impacts of changing climatic conditions, outline next steps, and assess feasibility of future actions?

a) Do the study results provide new or additional information about mercury or methylmercury sources, mass balances, or loads in the Delta or Yolo Bypass?

Yes, though the biases implicit currently in the loading numbers (as described above) need to be dealt with. With this improvement, this would possibly be the most comprehensive study on wetland MeHg net tidal loading that we know of. Currently, however, the main findings are somewhat contrary to findings from other studies that generally find tidal wetlands to be sources of MeHg and are seemingly in contrast to the Yolo Bypass mass balance findings in the Open Water report.

b) Will the studies help design future control studies?

Given the study design and approved workplan, making more than an inferential assessment of controls was explicitly not part of this work. Given the paucity of data about MeHg production and release from tidal wetlands, this more fundamental loading assessment was needed before even being able to think about or carrying out control studies. There is not all that much information that can be leveraged into particularly predictable control outcomes. Further analysis of the dataset in relation to some of the breadth of site characteristics that are given in the report, may be useful in this regard. At a minimum, once the data are assessed further for hydrological biases, the dataset would be useful in modelling studies or for scaling in situ work in these same wetlands for alternative assessments of loading.

c) What knowledge gaps must be filled to inform future control studies or management strategies?

The certainty with which tidal wetlands do or do not net export MeHg is not yet much more than before this study. As explained previously, going through some of the data and reassessing biases and uncertainties would help to fill some knowledge gaps, potentially quite substantially. There is some breadth in wetland characteristics across the 4 wetlands studied, but possibly not enough to infer much about control studies or management options. The large amount of data collected at these study wetlands, however, may help future research at these sites that focuses more specifically on aspects of control and management.

There already exist a number of mechanistic studies on mercury methylation in the region. Our suggestion for additional work would be to focus on improved spatial and temporal sampling. Sampling across times when it is clear that marshes are actually flooding would be very important. This would require a flexible sampling program and significant attention to weather and flows. Additionally, understanding the importance of loading from wetlands on a larger, landscape scale could be examined by sampling drainage areas across a breadth of tidal wetland area coverage. In this regard, examining areas across a breadth of Hg contamination levels would also be helpful.

Finally, it is unclear whether some wetland design features that are common to wetland creation have been considered in relation to MeHg production and transfer. Design and/or control features of created or restored wetlands sometimes include the following:

- Extent and depth of tidal flooding of wetland sediments
- Tidal channel vs. wetland area ratios
- Tidal channel interface perimeter with wetland
- In the case of wetlands such as Yolo, impacts from large water inputs (such as when the bypass is used to divert flows). There are a lack of studies of flooded agricultural soils in particular (flooded non-wetlands) that may be impacted by the diversions into the bypass. This is an important link with the Open Water study, that we think is needed to better understand Hg loading in the region.

d) How could changes in climate and other conditions in the Delta impact the relevance of study findings?

The study workplan was explicit in pointing out that assessing climate change impacts would not be feasible within the report. In our opinion, one could foresee issues from climate change that could affect MeHg production and movement in tidal wetlands via processes such as:

- Temperature – likely to impact MeHg production rates by stimulating microbes
- Temperature and hydrological dynamics – likely to affect DOC production rates through impacts on heterotrophic metabolism. This could increase MeHg transport through DOM complexes, but may also decrease demethylation by reducing UV light penetration.
- Plant community – this could impact soil aeration, which could impact MeHg production and DOC complexes. It could also physically affect flows in the wetlands via impacts on roughness.

In any future assessment to better understand how changes in climate or other conditions could impact study findings such as in the tidal wetland report, comprehensive modelling efforts are needed. Even the potential impacts above could be subject to feedbacks that are not accurately described on an impact-by-impact basis, but rather for which a cumulative impact may be complicated and only reasonably discernible through modelling.

e) Does the final report suggest any appropriate controls, and if so, are they feasible?

The final report is cautious to suggest controls. According to the workplan, there was no intention of a full control measures evaluation, which was thus not part of the study design. If further analyses of existing data or further studies conclude that indeed tidal wetlands are not important sources of MeHg to the Delta, then control measures would not be needed. Such a conclusion is not yet supported.

Do you have comments on other scientific issues related to this study or mercury/methyl-mercury source type?

We would like to point out that despite not yet having fully supported conclusions, we were impressed with the amount of work done by the study authors and we note that they stuck closely to their workplan. However, the data analysis was not deeply interpreted and significant corrections for hydrologic measurements are still needed. We encourage support further analysis of the data as we have outlined.

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