

Appendix S. Responses to the External Peer Review

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S.1 Marc W. Beutel (MWB)

Thank you for this opportunity to review the draft proposed rule for Mercury Water Quality Objectives. I would like to commend Water Board staff for developing a comprehensive and detailed proposal to protect the State's human and environmental resources from the threat of mercury pollution. My comments are presented below. Since my background is in environmental and civil engineering, I have focused my comments on addressing Concerns 5-8.

Conclusion 5 – Water column target of 12 ng/L total mercury is appropriately protective

COMMENT MWB 1

In reviewing the narrative in (6.11) Issue K in the draft staff report, I agree with the need for a consistent and simple method to develop effluent limitations for mercury and to draft permits. The recommended Option 1 in Section 6.11.3 of the draft staff report, with its focus on a water column target for total mercury (Figure 6-2), seems like the most appropriate approach. This contrast with Option 2 (Figure 6-3), in which effluent limitation is based on site-specific fish mercury content. I agree that the barriers to implementing Option 2 on a wide scale, which include on-going collection and evaluation of site-specific fish tissue data, are significant.

RESPONSE TO MWB 1

The reviewer's agreement with the approach is noted.

COMMENT MWB 2

One question I have regarding Option 2 [(6.12, Issue L)] and Figure 6-3 is the rationale for using ≥ 4 ng/L as an effluent threshold for potentially accepting an effluent limitation. Where did this value come from and why was it used? Was the 4 ng/L from a 0.2 mg/kg fish tissue concentration translated to a water column target using the USEPA mean lake/river bioaccumulation values as detailed in Appendix I (top of p. I-3)? And what happens in the flow chart if the effluent has a measurable total mercury concentration < 4 ng/L?

RESPONSE TO MWB 2

Yes, the value of 4 ng/L in Figure 6-3 (now Section 6.12) was used as an example. The value used here could be 4 or 12 ng/L or another value, depending on the effluent limitation that was chosen from the three options presented in the next section. Text has been added to clarify.

Existing text:

“Alternatively, if there is no fish tissue data then the dischargers could opt out of the fish collection obligation by agreeing to use a water column target to determine if they will be issued effluent limitations (same as option 1).

New text has been added:

“That water column target may be based on the effluent limitation ultimately chosen. The water column target could be 12 ng/L (from Option 1, Section 6.13), a value based on facility type (Table 6-1, Option 2, Section 6.13), 4 ng/L (from Option 3, Section 6.13) or another value based on the effluent limitation ultimately chosen. In Figure 6-3, the value of 4 ng/L is shown as an example.”

COMMENT MWB 3

The logic that since ionic mercury can be transformed to methylmercury in receiving waters, total mercury should be the focus on the water column target, is sound. The rationale for making the water column target the same as the effluent limitation is also clearly described in the draft staff report. As detailed in Section 6.12.3 (p. 117), based on State and USEPA guidelines dilution credits are not appropriate for bioaccumulating compounds like mercury. Since mercury bioaccumulation is a relatively long-term process within an ecosystem, an annual average also is the appropriate time scale on which to assess any effluent limitation. That said, for an annual average to be meaningful, a suitable minimum number of samples need to be collected annually. The minimum quarterly monitoring for larger dischargers detailed in the draft amendment is appropriate.

RESPONSE TO MWB 3

The reviewer’s agreement with the approach is noted. However, staff revised the Provisions to give the Water Boards the discretion to allow dilution. The existing California permitting policy for the relevant dischargers (the SIP), discourages using dilution for persistent bioaccumulative pollutants, such as mercury, but there may be cases with low background levels of mercury where dilution is appropriate.

COMMENT MWB 4

In my opinion, there is a disconnect in the presentation of the calculation of the proposed effluent limitation of 12 ng/L in Section 6.12.3 of the draft staff report and the calculations presented in Appendix I Section 1. In Section 6.12.3, the text states that the water column target was “calculated by using California bioaccumulation factors and translators based on data from river and streams only.” But in Section 1 of Appendix 1 the primary calculations (i.e., Tables I-1, I-2 and I-3) are based on USEPA national values for bioaccumulation factors and translators. The USEPA-based value for rivers was 11.5 ng/L, apparently rounded up to 12 ng/L in Table I-3 (The rounding up of values in Table I-3 seems inappropriate; consider presenting data with 2-3 significant figures, as was done in Tables I-5 and I-6.)

RESPONSE TO MWB 4

Table I-3 (now Table I-4) was edited to match the number of significant figures used by U.S. EPA and the California bioaccumulation factor (BAF), and to include the data for the California BAF, similar to table I-5 and I-6. The U.S. EPA final BAF and the California BAFs were presented with 2 significant figures, while the water quality objectives have only one significant figure. The level of precision implied by 3

significant figures is not supported by the large uncertainty in the BAFs and the uncertainty in the water quality objective. The final effluent limitations were rounded to the nearest whole number for practicality (one or two significant figures).

COMMENT MWB 5

There is then a discussion in Section 3 of Appendix 1 of the California bioaccumulation factors and translators, and acknowledgement that the California data was not of “high quality” and “provided limited [spatial] representation of the state as a whole.” Then the text includes a supporting calculation using California bioaccumulation factors and translator values for rivers (12.1 ng/L). It would be more appropriate for the narrative in section 6.12.3 to say that the water column target was estimated using the USEPA national bioaccumulation factors and translator, and that an additional calculation with California values, which apparently is not an especially rigorous data set, yielded a similar value.

RESPONSE TO MWB 5

The text (now Section 6.13.3) was updated as suggested, to better reflect the calculations. The following text was revised: “The water column target of 12 ng/L (total mercury) was calculated by using the U.S. EPA bioaccumulation factor from rivers and streams only, as shown in Appendix I. Most of the discharges from wastewater and industrial facilities flow into rivers (Appendix N). An equivalent threshold of 12 ng/L was derived using the California bioaccumulation factor. The California bioaccumulation factor was derived from data from rivers (Appendix I).”

COMMENT MWB 6

This raises the additional question of why 12 ng/L was the final water column target. Why not 11.5 ng/L or 11.8 ng/L, the average of the USEPA and California-based calculations.

RESPONSE TO MWB 6

Since the bioaccumulation factors were expressed with only two significant figures it is not appropriate to be expressing the water column target with three significant figures. The difference between 11.5, 11.8, and 12 ng/L is not significant, especially given the uncertainty in the bioaccumulation factors.

COMMENT MWB 7

Also, Tables I-1, I-2 and I-3 would be more effective if they were formatted like Tables I-5 and I-6, which included a presentation of both USEPA and California values.

RESPONSE TO MWB 7

These tables were adjusted as suggested (now Tables I-1, I-3 and I-4).

COMMENT MWB 8

Note that the California translator for MeHg_{dissolved}/MeHg_{total} was never numerically presented, even though it was used for the calculations presented in the last column of Table I-6.

RESPONSE TO MWB 8

The California translator is now shown in Table I-3.

COMMENT MWB 9

That said, the narrative presentation in Step 1 on p. A-10 of the draft amendment is nicely presented. But in the interest of transparency, the text in Step 1 on p. A-10 of the draft amendment should make clear that these bioaccumulation factors are “river-based” bioaccumulation factors.

RESPONSE TO MWB 9

This section of the amendment, which is now referred to as the “Provisions”, was reorganized and now there is a clear distinction between the water column values for “flowing water bodies (generally rivers, creeks and streams)” and values for other waters. .

COMMENT MWB 10

In both Appendix I (p. I-10) and the last paragraph of Section 6.12.3 Option 1 of the draft staff report, the documents state that the water column target of 12 ng/L, calculated on the basis of the sport fish objective (0.2 mg/kg in trophic level 4 fish, 150-55 mm), is also protective of wildlife, “or very close” to being so. Since this is a significant outcome, the report would benefit from an actual numerical calculation and presentation to support these claims. This could be presented in Appendix I.

RESPONSE TO MWB 10

The data to support such calculation is not available. This passage (now Section 6.13.3 Option 1) was revised as follows to explain this better:

“The wildlife objectives are consistent with meeting the one meal per week objective in trophic level 4 fish or very close. Data are not available to make this determination in a very exact manner, but see Section 6.1 through Section 6.6 of Appendix K for estimations. The wildlife objectives would not require a different limitation for wastewater and industrial discharges (unless a TMDL indicates otherwise).”

COMMENT MWB 11

After reviewing the SIP and the draft amendment text, it is not clear to me what total mercury effluent limitation concentration is called for in the event that there is measurable mercury in a discharge and total mercury in the receiving water is above 12 ng/L. It appears that the effluent limitation simply defaults to 12 ng/L. Is this the case? Is this approach

adequately protective of environmental quality? Or is this such a low probability scenario that it is not a concern, or perhaps other regulations or guidelines apply. Please clarify this issue.

RESPONSE TO MWB 11

Yes the effluent limitation is 12 ng/L in many cases, this had been revised somewhat to clarify and account for situations where 12 may not be protective (see the Provisions: Appendix A). The bioaccumulation factors used (described in Appendix I) suggest this water column target is consistent with meeting the sport fish water quality objectives in rivers, and therefore is protective of environmental quality for rivers. We clarified the requirements for slower moving waters (waters other than rivers or streams), waters where the tribal/ subsistence fishing water quality objectives apply and other exceptions may apply. These situations (where 12 ng/L may not be protective) are discussed in the next few comments (to Comment MBW- 12 through Comment MBW- 17). Additionally, if there is an exceedance of the water quality objectives, then a TMDL is required, and that TMDL may result in more protective requirements.

Conclusion 6 - Water column target for slower moving waters

COMMENT MWB 12

I agree that a more protective water column target is warranted for discharges to waters that are slower flowing than rivers. As detailed in Appendix I (i.e., USEPA bioaccumulation factors for rivers versus lakes) and as generally acknowledged by environmental scientist working on mercury cycling, lakes, reservoirs, wetlands and estuaries are expected to have higher potential to methylate mercury than rivers and streams. But it seems to me that the draft Mercury Water Quality Objectives do not adequately apply a more protective water column target in these cases. While I concur with the general approach and rationale used to develop the water column concentration of 12 ng/L, I am not convinced that it is appropriate to apply this standard to wastewater and industrial dischargers that discharge to waterbodies close to or designated as non-river in character (i.e., near or into lakes, reservoirs, wetlands or estuaries). As noted in Attachment 2 (p. A2-8) of the request for scientific peer review, the rationale for using a river-based water column target for all discharges are twofold: we only have bioaccumulation factors for California river/streams, and most treatment facilities in the State discharge to rivers/streams (greater than 90%). These themes are echoed in Section 6.12.3 Option 1 of the draft staff report. But it seems to me that the California bioaccumulation factors and translators, as noted above, were based on limited data, and in fact the USEPA bioaccumulation factors and translators were more appropriate to use. This raises the question: if we have both river and lake bioaccumulation factors and translators from the USEPA, why not apply both? If lakes, reservoirs, and estuaries are acknowledged as ecosystems with higher methylation potential relative to rivers, and if we have compelling metrics from the USEPA national dataset to calculate a water column target for non-river systems, then why not propose a water column target for non-river dischargers so as to better protect these more vulnerable systems from mercury bioaccumulation? Since we

do not have solid bioaccumulation factor and translator values for estuaries, perhaps estuaries should default to the lake water column target.

RESPONSE TO MWB 12

This requirement in the Mercury Provisions was changed to incorporate the reviewer suggestions. There are now three categories of water body types, each with a different water column concentration. For rivers and streams, the water column concentration remains 12 ng/L. Lakes and reservoirs were put in a different category. There are very few dischargers to such waters in the state, and we expect most of those will be included in a TMDL soon. In the meantime, if any of the permits for discharges to reservoirs come up for renewal we have included a case-by-case procedure for the permit writer to derive appropriate concentration for lakes/reservoirs.

For a third category of water bodies: “slow moving waters” (which could be estuaries or bays), a more stringent water column concentration (4 ng/L) has now been included. This water column concentration was based on the U.S. EPA national bioaccumulation factor (derived from combined data for lakes and rivers). For waters other than lakes, reservoirs, rivers and streams, it is somewhat difficult to determine the appropriate water column concentration, based solely on the water body name. Therefore, we have given the permit writer the discretion to determine if the water body is not a “flowing” water body similar to a river, but a “slow moving” water body. The staff report (now Section 6.13.3 Option 1) was revised accordingly and includes examples.

Additionally, subsequent to the scientific peer review, an unpublished study on bioaccumulation factors for California bays was located (Stephenson et al. 2009). This information has been added to Appendix I. The resulting water column concentration for all bays (2 ng/L) based on data in Stephenson et al. is not very different than using the U.S. EPA national bioaccumulation factor lakes and rivers combined (4 ng/L). Given that the results for bays were not much different from the U.S. EPA national data, and because these data were not included in the scientific peer review (and the study was not peer reviewed on its own), the bay study was not specifically used to alter the requirements in the Provisions. The bay study (Stephenson et al. 2009) provides additional supporting data.

COMMENT MWB 13

The argument that there are not very many treatment systems that discharge to non-river environments, or that it is difficult to distinguish between treatment systems that discharge to river-like systems and non-river like systems, as argued in Appendix I (p. I-9), do not seem compelling to me. If a treatment system discharges to an ecosystem known to be a more potent transformer of mercury into methylmercury, should it not need to meet a more stringent water column target? In addition, based on Table N-3a of Appendix N, the characteristics of treatment facility receiving waters appear to be fairly well defined. As detailed in Section III.A.2.d.3 of the draft amendment, the permitting authority may calculate alternative water column targets for non-river discharges, or may require non-river dischargers to develop site-specific bioaccumulation factor and translator values for their unique receiving

water system. But it seems prudent to first have ecosystem specific (rivers/streams and lakes/estuaries) targets of an appropriate magnitude. Under the proposed scheme, rather than protecting non-river systems from the start, there is need for some additional finding and action to implement a potentially more stringent water column target other than 12 ng/L. My concern is that the default 12 ng/L value will have an inertia that could impede implementation of more protective effluent limitations for wastewater and industrial facilities discharging to non-river environments.

RESPONSE TO MWB 13

This requirement was changed as the reviewer suggested. See comment MWB 12.

COMMENT MWB 14

I have one additional question related to site-specific bioaccumulation factors. Currently the draft amendment states that the “permitting authority may require a study” to develop bioaccumulation factors. I am curious about the Water Board’s perspective on whether the development of site specific bioaccumulation factors should be undertaken only if required by the permitting authority, or if dischargers should be given the explicit option in the amendment to develop site specific bioaccumulation factors if they want to

RESPONSE TO MWB 14

Agree -the dischargers should have the option to do a site-specific bioaccumulation study, although the permitting authority must review and approve the study. The Provisions were intended to allow that option. The staff report was edited to make this clearer (Section 6.13.3, option 1).

Conclusion 7 - Water column target for subsistence fishing

COMMENT MWB 15

I agree that a more protective water column target is warranted for discharges to waters that impact subsistence fishers. The draft staff report details a number of studies, some of which are recognized as limited in scope, which detail higher fish consumption rates by subsistence fishers. Presuming that other peer reviewers with expertise in public health toxicology affirm that appropriateness of the higher consumption rates for subsistence fishers, then it is appropriate to have a water column target that is more stringent than the 12 ng/L river-based target estimated using sports fish (0.2 mg/kg). The calculation methods presented in Appendix I are appropriate and scientifically sound. But there are some acknowledged weaknesses of the method, including limited bioaccumulation metrics specific to California and specific to estuaries.

RESPONSE TO MWB 15

The reviewer's agreement with the approach is noted. The concerns for protection of estuaries are addressed in Response MWB 12.

COMMENT MWB 16

Keeping in mind the uncertainties in bioaccumulation metrics discussed above, and with the anticipated low water column targets likely to be calculated with the use of subsistence fish tissue levels (e.g., 0.05 mg/kg for subsistence fishing and 0.04 mg/kg for Native American subsistence fishing), it seems appropriate to also allow dischargers to develop site-specific bioaccumulation factors when discharging to waters impact subsistence fishers. Thus, I recommend that the Water Board consider adding text to item d.4 (p. A-14) of the draft amendment, similar to that in item 3.ii (p. A-13), that allows for two potential outcomes for dischargers impacting subsistence fishing: the recalculation of a modified water column target or the develop site-specific bioaccumulation factors by the discharger.

RESPONSE TO MWB 16

Agree. The text in section IV.D.2 has been clarified by rearranging. This reorganization was also done to address Comment MWB 12.

Yes, the dischargers subject to the subsistence water quality objectives are allowed to develop site-specific bioaccumulation factors. Also, the dischargers subject to the subsistence water quality objectives could be given the small disadvantaged community exception or the insignificant discharger exception if the discharge meets the criteria.

COMMENT MWB 17

An additional question arises for both of these options: given the uncertainties in fish consumption patterns of subsistence fishers, which is a key driver of the target fish tissue limit, should dischargers be permitted to develop site-specific fish consumption metrics? Should this section of the amendment include an explicit acknowledgment of this issue and note that dischargers could be required, or dischargers could choose themselves, to develop site-specific fish consumption metrics?

RESPONSE TO MWB 17

The subsistence fishing water quality objective has been modified, so that the objective can be implemented in a site-specific manner. In Section 6.5 of the Staff Report, Option 6 is now recommended, which is the narrative water quality objective. Previously, a numeric water quality objective was recommended. With a narrative subsistence fishing water quality objective, dischargers could potentially fund or perform a fish consumption study to support a site-specific water quality objective, but the Regional Water Board must find such a study acceptable before it would be implemented in permits.

A narrative water quality objective has the advantage of allowing permit-specific implementation. A site-specific fish consumption rate could be used to implement the water quality objective or a provided default fish consumption rate (142 g/ day) could be used to implement the water quality objective. A permit writer could consider relative loading from the discharge compared to other sources. A permit writer could also consider other site-specific factors, such as if there are no trophic level 4 fish, requirements would not need to be as stringent as in a water with trophic level 4 fish

(This does not apply for the Sport Fish Water Quality Objective since one of the two prey fish objectives would still need to be achieved). Finally, an additional advantage of the narrative water quality objective is that these site-specific considerations could be taken into account without the lengthy regulatory process of adopting a site-specific water quality objective. On the other hand, a site-specific water quality objective must be adopted by the Regional Water Board through a regulatory process called a Basin Plan Amendment, which includes public input. This is a process similar to the process that the Provisions is undergoing. See also Comment MBS 8 and EVW 14.

Conclusion 8 – Sediment controls and transport of mercury into waters

COMMENT MWB 18

The focus on sediment and erosion control in the Storm Water Discharges section of the draft amendment, with a particular emphasis on control measures in areas where soils are naturally rich in mercury or have a history of mining activity, is appropriate.

RESPONSE TO MWB 18

The reviewer’s agreement with the approach is noted.

COMMENT MWB 19

The focus on wetland restoration projects is also a commendable component of the draft amendment, since important wetland restoration efforts in the State will spatially overlap with mercury-impacted regions, such as the South San Francisco Bay for example.

RESPONSE TO MWB 19

The reviewer’s agreement with the proposal is noted.

COMMENT MWB 20

One issue to keep in mind is that some BMPs, such as anaerobic components of structural BMPs used to enhance microbial denitrification, may have the potential to promote methylation of trapped mercury. Can or should this issue be acknowledged as part of the draft staff report or in the context of BMP implementation in the draft amendment?

RESPONSE TO MWB 20

Yes, BMPs for microbial denitrification could be required by other Water Boards programs and it is possible that BMPs could be anoxic and methylate mercury. Text was added to Section 4.4.6 “Conversion to Methylmercury as a Source” at the end of the first paragraph:

“ Additionally, structural Best Management Practices used to enhance microbial denitrification, such as treatment wetlands, can have anaerobic zones and are rich in organic matter both, factors that promote mercury methylation. Also, storm water catch basins can become anaerobic. Therefore, while these Best Management Practices serve important function in controlling nutrients and

possibly other pollutants, these Best Management Practices may also inadvertently incorporate conditions that promote mercury methylation.”

COMMENT MWB 21

The proposed 300 ng/L total mercury numeric action level for industrial dischargers is far below the current level of 1,400 ng/L, which is appropriate. But I am not convinced by the rationales for the new numeric action level, as detailed in Section 6.10.3 of the draft staff report and Appendix P Section 4.1. A key rationale, as stated in the draft staff report (p. 102), is that the numeric action levels are “technology based” and that “it is not clear that a lower threshold would be achievable with currently available storm water treatment methods.” Presumably the treatment methods alluded to in this statement encompass other non- structural BMPs such as good storage and handling practices. What is the basis for this statement? Is there documented studies that find that achieving industrial storm water levels below 300 ng/L is technically infeasible?

RESPONSE TO MWB 21

No documentation was found on the effectiveness of storm water treatment for removing mercury that could be included in the Staff Report. An example when a lower numeric action level may not be achievable with current technology is if the mercury is from atmospheric deposition. We also included information on typical concentrations of mercury in rain from atmospheric deposition in Appendix P: Averages were around 3-13 ng/L, storm events had mercury levels up to 70 ng/l and few samples were above 200 ng/L (See also Comments MWB 22-23). If the mercury is from atmospheric deposition and not from the industrial facility, treatment methods such as good storage and handling practices will not be sufficient to control mercury.

COMMENT MWB 22

A second rationale was the potential cost of water quality analyses. The 300 ng/L action level seems to have been selected partly because it is comfortably above the detection limit of 200 ng/L for USEPA method 245.1, which cost \$18 to \$35. This compares to USEPA method 1631E (quantitation limit of 0.5 ng/L), which can costs over \$115. No cost was stated for the method 245.7 with the intermediate quantitation limit of 5 ng/L. Presuming method 245.7 is on the order of \$75, use of this method would increase sampling costs by around \$60 per sample. Noting that industrial dischargers are required to sample around once per year (Appendix P Section 4.1), this additional cost does not seem to me like a significant enough financial burden to dictate the numeric action level for mercury for industrial dischargers.

RESPONSE TO MWB 22

In general, sampling mercury with the most up to date methods is much more expensive and complicated than monitoring for other constituents, such as copper.

More information was gathered on the costs of method 245.7 and added to Appendix P. The cost for method 245.7 is more difficult to estimate since few labs perform this test. Cost estimates also ranged widely. So the cost may be roughly similar

to method 1631, because the clean hands technique is still required. The clean hands technique could increase the cost to roughly \$250 for method 1631. In some instances, much of the cost of these mercury sampling methods can be attributed to the travel expense for persons qualified to perform the clean hands sampling. Since few labs perform 245.7 (possibly because there is little difference in cost compared to 1631, but 245.7 is much less sensitive), method 245.7 is a less feasible solution.

While normally industrial storm water dischargers (those enrolled in the general permit) may only be sampling one per year, a discharger with an exceedance of the Numeric Action Level may need to sample multiple times, and the cost for 1631 (or 245.7) could be over 1000\$ after three or four samples. These dischargers range from small businesses to large scale industrial operations. For the small businesses dischargers this is likely a significant cost, and it does not include the cost of the actions required to reduce the mercury in the discharge.

COMMENT MWB 23

A third apparent rationale for not having a low action level for total mercury was that pollutants will be diluted by storm water. But, as detailed above for wastewater and industrial dischargers, mitigating effects of dilution are not appropriate for bioaccumulating substances like mercury. While acknowledging that numeric action levels “are not meant to be water quality standards, objectives or criteria,” Water Board staff should consider using a more scientifically-based method for developing a new numeric action level for industrial storm water dischargers for total mercury. The draft staff report notes that the original 1,400 ng/L numeric action level was based on outdated aquatic life criterion for mercury. Is there an updated metric related to mercury’s environmental impact that could be used to inform development of a numeric action level for industrial storm water dischargers?

RESPONSE TO MWB 23

Comments are understood. However, there is not clear more scientifically-based threshold to use for the numeric action level. Section 3.11 of the Staff report reviews other U.S. EPA water quality criteria for freshwater. That section lists a chronic U.S. EPA aquatic life criterion of 770 ng/L, which is higher than our proposed numeric action level. Also, there is another aquatic life criterion of 12 ng/L (U.S. EPA 1985a, U.S.EPA 1986), which is equivalent to our water quality based threshold for wastewater treatment plants. To achieve this threshold (12 ng/L) requires a wastewater treatment facility.

Storm water discharges are sporadic, and the discharge is diluted by other storm water while the discharge is occurring. This is different than a continuous discharge (such as wastewater) that is constantly occurring. Hence, for storm water discharges, the numeric action level that is applied to a specific discharge is not water quality based, unlike wastewater discharges, and consideration of dilution is incorporated in a different manner for storm water.

S.2 Mark B. Sandheinrich (MS-UW)

Review of Draft for Scientific Peer Review: Amendment to the Water Quality Control Plan for Inland Surface Waters, Enclosed Bays, and Estuaries of California—Mercury Water Quality Objectives and Program of Implementation

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This review of the document “Draft for Scientific Peer Review: Amendment to the Water Quality Control Plan for Inland Surface Waters, Enclosed Bays, and Estuaries of California—Mercury Water Quality Objectives and Program of Implementation focuses on the Draft Staff Report including the Draft Amendment Language (Appendix A), Review of Effects on Wildlife (Appendix J), Wildlife Targets (Appendix K), and the Derivation of Trophic Level Ratios (Appendix L). My review is limited to evaluating the proposed water quality objectives relative to the protection of wildlife and I attempted to address those areas discussed on pages A2-6 and A2-7 of Attachment 2 from the June 16, 2016 memorandum signed by Karen Larsen: “Request for Scientific Peer Review of the Draft Proposed Rule for Mercury Water Quality Objectives.”

Review of data collected on various wildlife species including food intake rates, reference doses, and diet compositions from previous published reports.

COMMENT MS-UW 1

The Draft Staff Report and USFWS (2003) based the water quality objectives on endangered and threatened freshwater piscivorous wildlife that occur in California as well as a select group of species that were included by regional water boards in the development of site-specific objectives. Food intake rates, reference doses (discussed below) and diet compositions were determined from extensive peer-reviewed literature and published reports from the USFWS and USEPA and used commonly accepted scientific practices.

RESPONSE TO MS-UW 1

The reviewer’s agreement with the approach is noted.

Review of uncertainty factors used to calculate the Reference Dose (RfD)

As stated in USFWS (2003) the RfD may be determined for a given taxonomic group by adjusting the test dose (TD) through the application of uncertainty factors (UFs) to incorporate variability in toxicological sensitivity among species (UFA), to extrapolate from subchronic studies to account for chronic exposure (UFS) and to account for spacing in concentrations of test doses (UFL).

For the mammalian RfD, USFWS (2003) evaluated TD, RfD and UFs from the Great Lakes Initiative (GLI) Technical Support Document for Wildlife (USEPA 1995) and the Mercury Study Report to Congress (MSRC; USEPA 1997). Though both USEPA (1995) and USEPA (1997) used different test doses from toxicity studies on mink (Wobeser 1976 a, b) and different uncertainty factors, the calculated RfDs were similar from the two reports (0.016 and 0.018 mg Hg/kg body weight/day). These RfDs were 10% to 33% of the test dose (no observed adverse effects concentration). Appendix K. (Wildlife Targets) of the Draft Staff Report for Peer Review: Provisions (hereafter referred to as the Draft Report) used a mammalian RfD of 0.018 mg Hg/kg body weight/day.

The avian RfD of 0.021 mg Hg/kg body weight/day in the Draft Report was also from USFWS (2003) and was based on a test dose of 0.064 mg/kg body weight/day from a study of mallard ducks (Heinz 1979) and uncertainty factors from the MSRC. The RfD is approximately 33% of the test dose.

COMMENT MS-UW 2

Though dated, the studies by Wobeser (1976 a,b) and Heinz (1979) likely represent the best available peer-reviewed studies that evaluated dietary concentrations of methylmercury on mammals and birds. Other studies, including those in which avian eggs were injected with methylmercury, may provide information on toxic concentrations of methylmercury but may not be germane because of different routes of exposure, toxicokinetics and toxicodynamics. USFWS (2003) reviewed and cited a number of other laboratory and field studies that supported the acceptance of the test dose determined from the studies of Wobeser (1976) and Heinz (1979). I am unaware of more recent studies that would contradict that conclusion.

RESPONSE TO MS-UW 2

The reviewer's agreement with the approach is noted.

Though different test doses and UFs were used by GLI and MSRC in determining the RfD for mammals, the final RfD values were similar. Because the Water Quality Objectives for wildlife in the Draft Report are based on the protective wildlife targets for the most sensitive species (Draft Report Table K-3) and avian species were the most sensitive species in each trophic level category, the UFs and subsequent RfD for the avian species deserve additional scrutiny. The avian test dose (0.078 mg/kg body weight/day) used by GLI and MSRC was the same. However, the cumulative UFs used by GLI ($UFA \times UFS \times UFL = 6$) and MSRC ($UFA \times UFS \times UFL = 3$) differed two-fold. USFWS (2003) concluded that the UFs presented in the MSRC were more appropriate for determining the avian reference dose than those from the GLI. Based on the TD from Heniz (1979) and UFs from MSRC, they calculated an RfD of 0.021 mg/kg-bw/day. This is the RfD used in the Draft Report. However, USFWS (2003) also stated (page 21) "because several of the bird species considered in this effort are not obligate

piscivores, the argument presented in the MSRC for using a UFA of 1 may not be appropriate for these species.” “An alternative avian RfD of 0.007 mg/kg-bw/day was also presented for the three clapper rail subspecies and the snowy plover.”

COMMENT MS-UW 3

Using the alternative RfD of 0.007 mg/kg-bw/day from USFWS (2003), I recalculated the Wildlife Values for the 3 species of rails and western snowy plover (Table 1).

Table 1. Alternative Wildlife Values (mg/kg in diet). Species body weight and FIR are from table K-1 of the Draft Report.

Species	RfD (mg/kg/day)	Body weight (kg)	FIR (kg/day)	Wildlife Value (mg/kg in diet)
California Ridgeway’s rail	0.007	0.346	0.172	0.014
Light-footed Ridgeway’s rail	0.007	0.271	0.142	0.013
Yuma Ridgeway’s rail	0.007	0.271	0.142	0.013
Western snowy plover	0.007	0.041	0.033	0.009

Using the alternative Wildlife Values in Table 1 and the same methods as presented in the Draft Report, I then recalculated the protective wildlife targets in various trophic levels for these same species (Table 2).

Table 2. Protective wildlife targets in various trophic levels.

Species	TL2	TL2/3 < 50 mm	TL3 <150 mm	TL3 150-500 mm
California Ridgeway’s rail	0.012			0.07
Light-footed Ridgeway’s rail	0.007			0.04
Yuma Ridgeway’s rail	0.003			0.017
Western snowy plover	0.036			

The proposed water quality objectives now can be evaluated relative to protective targets for various trophic levels in which these four species feed and based on the alternative RfD presented in USFWS (2003).

Target for Wildlife That Prey of TL3 Fish, 0-500 mm. Yuma Ridgeway’s rail remains as the most sensitive species in this category. However, using the same food chain multiplier of 4 (page K- 16 of Draft Report) and the protective target from Table 2 above, 0.017 mg/kg x 4 = 0.068 mg/kg in TL4 fish. Consequently, a water quality objective of 0.2 mg/kg in TL4 may not maintain 0.017 mg/kg in TL3 fish 0-500 mm based on the alternative RfD.

Target for Wildlife That Prey on TL2 Fish. Dividing the TL3 150-500 mm target (0.08 mg/kg) by the national food chain multiplier of 5.7 results in a corresponding TL2 values of 0.014. This is greater than the Table 2 recalculated targets for the California Ridgeway's rail and Light-footed Ridgeway's rail. Consequently a water quality objective of 0.2 mg/kg in TL 4 may not meet TL2 targets under this scenario.

Using the alternative RfDs presented in USFWS (2003) indicates that the water quality objective of 0.2 mg/kg in TL4 fish may not be protective of all species. The Draft Report Appendix K (pages K-26 and K-27) makes a logical argument why the alternative RfDs were not used and acknowledges points of uncertainty that suggest a less stringent or more stringent objective. In particular, the acknowledgement and discussion of the limitations and sources of uncertainty in the calculations is a strength of the Draft Report and supports the readers' assumption that best professional judgement was used in selecting UFs to calculate RfDs.

RESPONSE TO MS-UW 3

The reviewer's support of the logical argument and the discussion on uncertainties is noted. Staff appreciates the rigor with which the reviewer analyzed this issue.

Review of Trophic Level Ratios

COMMENT MS-UW 4

The Draft Report used food chain multipliers (FCM) from USFWS (2003; page 5) and/or trophic level ratios (TLR) to translate between methylmercury concentrations in different sizes of fish in different trophic levels. The FCMs and TLRs were either obtained from USEPA national data (if taken from USFWS (2003)) or from California site-specific data or California state-wide data (Appendix L). These FCMs and TLRs were used in deriving protective targets for individual species that consumed fish from multiple trophic levels. The most sensitive targets, in turn, were used to develop water quality objectives for all wildlife. In addition, the FCMs were used to calculate expected concentrations in TL 2 and TL 3 fish if the limiting methylmercury concentration is 0.2 mg/kg in TL 4 fish.

The FCMs in USFWS (2003) were calculated from Bioaccumulation Factors (BAFs) from draft National BAFs presented in the EPA's methylmercury criterion document (U. S. EPA. 2001. Water quality criterion for the protection of human health: methylmercury. EPA-823-R-01-001. Office of Science and Technology, Office of Water, U.S. Environmental Protection Agency Washington, DC.) and are presented here for discussion (Table 3).

Table 3. Draft BAFs for methylmercury empirically derived from field data collected across the United States and reported in the open literature. Based on BAFs calculated from lotic and lentic systems.

	BAF Trophic	BAF Trophic	BAF Trophic
5 th Percentile	18,000	74,000	250,000
Draft national values (approx geometric mean, 50 th percentile)	120,000	680,000	2,700,000
95 th Percentile	770,000	6,200,000	28,000,000

USFWS (2003) and subsequently the Draft Report used the approximate geometric mean BAF for each trophic level to calculate the food chain multipliers. The FCM for any trophic level is the ratio of the BAF for that trophic level to the BAF for the trophic level below.

$$\text{FCM } 4/3 = 2,700,000/680,000 = 4$$

$$\text{FCM } 3/2 = 680,000/120,000 = 5.7$$

From Table 1, and as the EPA acknowledges in the criterion document, it is evident that the range for BAFs for each trophic level varies by at least 10 (TL 2) to approximately 100 fold (TL 3 and TL 4). Moreover, the criterion document states “EPA fully recognizes that the approach taken to derive mercury BAFs collapses a very complicated non-linear process, which is affected by numerous physical, chemical, and biological factors, into a rather simplistic linear process.

EPA also recognizes that uncertainty exists in applying a National BAF universally to all water bodies of the United States. Therefore, in the revised 2000 Human Health Methodology (EPA, 2000) we encourage and provide guidance for States, Territories, Authorized Tribes, and other stakeholders to derive site-specific field-measured BAFs when possible. In addition, should stakeholders believe some other type of model may better predict mercury bioaccumulation on a site-specific basis they are encouraged to use one, provided it is scientifically justifiable and clearly documented with sufficient data” (page A-18 of USEPA (2001)).

Using the 5th and 95th percentiles for the BAFs instead of the geometric mean BAFs to calculate the FCM results in lower and upper bounds of the range of the FCM for any trophic level.

For FCM 4/3 the lower and upper bound of the range is at least

$$250,000/74,000 = 3.4$$

and $28,000,000/6,200,000 = 4.5$.

In turn, the lower and upper bounds of the range of FCM 3/2 is at least

$74,000/18,000 = 4.1$

and $6,200,000/770,000 = 8$

Although, the range of the calculated food chain multipliers is not as great as that of the BAFs from which they are derived, based on the EPA's admission of the limitation of the draft national BAFs and the importance of the FCMs to establishing the water quality objectives, the use of empirically derived national BAFs may or may not be appropriate. At the very least, the Draft Report should address the uncertainty associated with using these values and also address why FCMs or TLRs derived specifically from California water bodies were not used to calculate expected methylmercury concentrations in TL3 and TL2 fish if TL4 fish were limited to 0.2 mg Hg/kg.

RESPONSE TO MS-UW 4

Agree- these uncertainties should be acknowledged and these uncertainties were acknowledged in appendix K. In Appendix K there is a section on uncertainties (Section 9) and the second paragraphs says:

“The food chain multiplier and trophic level ratios are estimates that add to the uncertainty in these calculations. Some are site-specific while some were derived from national data. These values may not accurately represent all of California’s waters, but a more accurate alternative is not available.”

New text was added:

“More specially, FCMs could not be calculated, since sufficient data were not available for fish < 150 mm or TL2 organisms. California’s statewide monitoring program has collected a great deal of data on large TL4 and TL3 fish, but much less data on fish <150 mm or TL2 organisms. While there was a large data set for large TL4 and TL3 fish, the data that could be used to derive the TLRs provided poor geographic representation of California (see Appendix L). Since the TLRs were limited and a California FCM was not possible to calculate, values from various California projects, as well as targets derived from national values are all included in Table K-3 to provide an idea of the uncertainty in these values. However, this will not capture all of the uncertainty. If minimum and maximum values for the FCMs and TLRs were used the variation in the targets would be larger. The actual amount of mercury in fish in various waters will vary by the food chain in a particular water body and other waterbody specific factors. The variation in mercury concentrations in prey fish vs. sport fish in a particular water body is exemplified in the recent USGS grebe study (Ackerman et al. 2015, Figure 5, see also Section 7.1 of this Appendix). Only average FCM and TLR values were

used in this analysis to provide estimates for the whole state. These estimates may be either over protective or under protective for a particular water body.”

Next text was also added (at the very end of Section 4.1 of Appendix K) just after the introduction and descriptions of the TLRs and FCMs

“While California TLRs were derived for this analysis, California specific FCMs could not be calculated, since sufficient data were not available on fish < 150 mm or TL2 organisms. The FCMs are only used for a few species where a California TLR could not be used, including: river otter, southern sea otter, California Ridgeway’s rail and light-footed Ridgeway’s rail. Additionally, when possible, targets from site-specific projects and from site-specific data were included in Table K-3, such as for river otter. A range of values from various California projects, as well as targets derived from national values are included in Table K-3, to show some of the uncertainty in these values. However, this does not include all the uncertainty in these targets (see section 9).”

The description of the trophic level ratios (TLR) in Appendix L already discussed that the TLRs were from data based on a limited geographic representation of California.

Review of the resulting three proposed water quality objectives to ensure protection of wildlife

COMMENT MS-UW 5

Appendix J is a very good and concise review of the relevant literature on the effects of methylmercury on fish and wildlife and provides a summary of suggested dietary methylmercury thresholds in wildlife (Table J-1) and fish (Table J-2).

RESPONSE TO MS-UW 5

The reviewer’s agreement with the approach is noted.

COMMENT MS-UW 6

Appendix K (pages K-17 to K-19) compares the target values to a study of mercury in grebe blood relative to mercury prey and sport fish (Ackerman et al 2015 a,b). A concentration of 1 mg /kg mercury in grebe blood correlates to approximately 0.2 mg/kg in sport fish and 0.048 mg/kg in prey fish 10-123 mm and represents the boundary between low and moderate risk in loons (Evers et al. 2004). The target of 0.05 mg/kg for fish 50-150 mm is less than the suggested benchmarks for loons (Depew et al 2012b) and, based on food chain multipliers and dietary composition, is equivalent to the LOAEL (lowest observed adverse effects level) in white ibis (altered behavior; Frederick and Jayasena 2010), a species with the lowest mercury threshold reported in the literature.

RESPONSE TO MS-UW 6

Draft Staff Report: Part 2 of the Water Quality Control Plan for Inland Surface Waters, Enclosed Bays, and Estuaries of California – Tribal and Subsistence Fishing Beneficial Uses and Mercury Provisions

The reviewer's agreement with several conclusions is noted. Additionally, while the target of 0.05 mg/kg for fish 50-150 mm is equivalent to the lowest observed adverse effects level in white ibis (0.05 mg/kg, altered behavior; Frederick and Jayasena 2010), when considering the diet of the ibis, the target should be more protective of the ibis. Appendix K (section 7.3) describes how the target of 0.05 mg/kg for fish 50-150 mm is roughly equivalent to a *no observed adverse effects* level in white ibis, based on food chain multipliers and dietary composition (since ibis mainly prey on organisms with a lower trophic level status than fish 50-150 mm).

COMMENT MS-UW 7

Based on the assumptions in developing the RfDs for individual species (i.e., acceptance of UFs) and the use of FCMs based on nationwide rather than state-specific data, the proposed water quality objectives (0.2 mg Hg/kg in sport fish; 0.05 mg Hg/kg in prey fish 50 to 150 mm; 0.03 mg Hg/kg in prey fish < 50 mm consumed by the California least tern) may reasonably be expected to be protective of most species of piscivorous wildlife.

RESPONSE TO MS-UW 7

The reviewer's agreement with the approach is noted.

COMMENT MS-UW 8

Moreover, the Draft Staff Report recognizes that altered reproduction in birds is one of the more frequently observed effects of sublethal methylmercury exposure and that mercury concentrations in prey fish vary seasonally (Ackerman et al. 2015 a,b). Consequently, the Prey Fish Water Quality Objective and Prey Fish Water Quality Objective for California Least Tern defines the time period annually when the objective applies based on the avian breeding cycle.

RESPONSE TO MS-UW 8

The reviewer's agreement with the approach is noted.

COMMENT MS-UW 9

The lack of available data precludes evaluating the water quality objectives relative to insectivorous wildlife that consume the terrestrial stages of aquatic insects and may be exposed to relatively high concentrations of methylmercury. The Staff Report cites an unpublished study by Robinson et al. (2011) that documented concentrations of methylmercury (1.66 ppm) in the blood of riparian song sparrows downstream of New Almaden. These concentrations were similar to those that were associated with a 25% to 30% reduction in nest success of Carolina Wrens along two mercury-contaminated rivers in Virginia (Jackson et al. 2011). Additional studies will be required to determine the relation between mercury concentrations in prey fish and sport fish and those of aquatic insects that inhabit the same water bodies.

RESPONSE TO MS-UW 9

Agree. Text has been added to Appendix K (Section 9) stating that:

“The lack of available data precludes evaluating exposure to insectivorous wildlife that consume the terrestrial stages of aquatic insects and may be exposed to relatively high concentrations of methylmercury. High concentrations of methylmercury (1.66 ppm) have been measured in the blood of riparian song sparrows downstream of New Almaden, site of a large mercury mine (Robinson et al. 2011, Section K.10.2). These concentrations were similar to those that were associated with a 25% to 30% reduction in nest success of Carolina Wrens along two mercury-contaminated rivers in Virginia (Jackson et al.2011). Additional studies will be required to determine the relationship between mercury concentrations in prey fish and sport fish and those of aquatic insects that inhabit the same water bodies.”

S.3 Michael Bliss Singer (MBS)

Review of ‘Amendment to the Water Quality Control Plan for Inland Surface Waters, Enclosed Bays, and Estuaries of California—Mercury Water Quality Objectives and Program of Implementation’

Michael Singer, University of St Andrews (UK) and University of California Santa Barbara, 9/19/2016

Summary

In a regulatory framework, the current standards for protection of water quality and fish from Hg contamination have been found to be inadequate given the scale of the pollution problem in California and the risks to humans and wildlife. Thus, an Amendment has been proposed to strengthen the regulation of Hg contamination and its monitoring. Below I evaluate and comment specifically on items 5-8 (Attachment 2) regarding the scientific soundness of the proposed rules. Note: I combine my discussion for the related topics of 5-7, given they are interrelated. Subsequently I provide more detailed and considered comments on broader aspects of the proposed rule as stipulated in a and b in Attachment 2. Specifically, I provide contextual information and even some suggestions for how particular options may be improved and where potentially important data and/or theoretical gaps exist. I mostly limit my discussion to matters that are within my main areas of expertise, but do provide a professional (non-expert) opinion in some cases. My comments are keyed to specific sections/issues listed in the Staff Report. Overall, I am generally supportive of the science that lies behind the amendment. I believe the report represents deep consideration of the relevant issues in light of the contamination risks to wildlife and humans across the state.

Addressing the Science

5. A water column concentration of 12 ng/L total mercury in rivers is generally consistent with meeting the Sport Fish Water Quality Objective (0.2 mg/kg methylmercury in fish tissue) in rivers based on bioaccumulation factors...

6. Consideration of a more protective water column concentration than 12 ng/L total mercury is warranted as the effluent limitation for municipal wastewater and industrial discharges to waters that are slower flowing than rivers and streams, since these waters are likely to experience higher rates of mercury methylation and bioaccumulation.

7. A more protective water column concentration than 12 ng/L total mercury is warranted as

the effluent limitation for municipal wastewater and industrial discharges to waters where more stringent water quality objectives apply for subsistence fishing or tribal subsistence fishing.

COMMENT MBS 1

[Section 6.1 (Issue A)] I support Option 2 (the fish tissue objective). Water concentrations of total and methylmercury tend to be low and are often diluted over large areas by the mixing of highly concentrated water with the disproportionately common low concentration water. Given the apparently low risk of Hg contamination by skin contact and/or by drinking water with low levels of MeHg (or even total Hg), I agree with a more modern standard of measuring MeHg in fish tissues. However, fish sampling is obviously a destructive procedure, so the monitoring efforts of such concentrations over large areas may be complicated. Nevertheless, this is the most logical standard to implement since it is the primary pathway of contamination to humans and to wildlife in areas that are not designated impaired under the Clean Water Act.

RESPONSE TO MBS1

The reviewer's agreement with the approach is noted.

COMMENT MBS 2

In the latter cases, the local TDML standards (including those for sediment) should still supersede this objective.

RESPONSE TO MBS 2

The reviewer's agreement with the approach is noted.

COMMENT MBS 3

[Section 6.1 (Issue A)] I see the complication of not being able to regulate industrial/mining discharges, but perhaps a hybrid of Options 2 and 3 is possible, where Option 3 (only for discharged water, rather than the water column of the receiving water course) can hold. If not, selection of Option 2 will obviate regulation (at the state level) of Hg-laden discharges, except in cases that are (or become) classified as 'impaired'. If the designated concentration for Option 3 were elevated for discharges only (as I have conceived of it here), this may alleviate the concerns of industry about the infeasibility of implementation.

RESPONSE TO MBS 3

Agree. This suggested “hybrid” is in a way achieved in the recommended proposal. While the water quality objectives are in fish tissue, permitting requirements are expressed as water column concentrations.

COMMENT MBS 4

[Section 6.1 (Issue A)] I don’t totally agree with the supposition in Option 4 that anoxic sediments are the primary sources MeHg production. This has not been shown at the landscape scale and may instead reflect biases in sampling (of lowland wetlands, estuaries, etc). We DO know that a drop in oxygen levels is required to activate anaerobic bacteria, but this can also occur within sediments that not classified as anoxic, but are instead subjected to anoxic or even suboxic conditions temporarily (Briggs et al., 2015; Singer et al., 2016). These flood pulses have been suggested by others to induce MeHg production in hyporheic zones along streams (Bradley et al., 2012; Hinkle et al., 2014).

RESPONSE TO MBS 4

This sentence was deleted: “Moreover, the primary setting for methylation of mercury is thought to be anoxic sediments.”

COMMENT MBS 5

While it may be true that atmospheric deposition of Hg is a primary source in other regions, this is not likely to be the case in most of the California water bodies (see above). This statement needs to be clarified.

RESPONSE TO MBS 5

Agree, we changed “many” to “some” in the passage: “However, sediments are not a major source of mercury for all water bodies. There are several other potential sources including atmospheric deposition, which is likely the largest source of mercury in some water bodies.” The comment that atmospheric deposition is unlikely to be important in *mining impacted areas* is noted and reflected in the section on sources (4.4.3, see also comment MBS 19 & 20). However, as was noted in section 4.4.3 of the staff report:

“Mercury deposition from atmospheric emissions is thought to be the major source of mercury in some Southern California lakes and reservoirs (U.S. EPA 2012, Tetra Tech 2008).”

COMMENT MBS 6

It is encouraging that the Water Board is working on a separate set of Hg objectives for sediment that would sit alongside the fish tissue objectives recommended (if adopted). Presumably the same parallel objective approach could be adopted for wastewater and industrial discharges to ensure this pollution source is regulated, but at a feasible/appropriate level?

RESPONSE TO MBS 6

The proposal at hand does include a means to control mercury from wastewater and industrial discharges (discussed in Sections 6.12 and 6.13 of the Staff Report).

COMMENT MBS 7

[Section 6.1 (Issue A)] Option 5 is not viable. In environmental science/management, we need quantitative standards to ensure regulation is consistently applied and achieves its objectives.

RESPONSE TO MBS 7

Agree.

COMMENT MBS 8

[Section 6.5 (Issue E)] I support the establishment of quantitative (numeric) guidelines for MeHg in fish tissue for subsistence and tribal subsistence use. However, given the large variations MeHg concentrations for different sites, I actually support Option 5, which would enable further study to determine more precisely what standards are required for different sites. The option would also be less controversial to water dischargers, which might limit legal challenges. Overall, without some clear metric, it would be impossible to evaluate and/or enforce water quality standards. This is not my area of expertise, but a strong case is made that quantitative standards will ultimately be necessary, so it seems that now is the time to create them.

RESPONSE TO MBS 8

Agree that option 5 has the advantages the reviewer lists. The recommendation will be changed to option 6, which is the narrative water quality objective. This option will incorporate site-specific considerations as the commenter suggests. The advantages and disadvantages of all options will be considered more during the public comment period. See Comment MWB 17 for more advantages of the narrative water quality objective (also Comment EVW 14 is related).

COMMENT MBS 9

[Section 6.11 (Issue K)] I support Option 1. Option 2 is not logical because fish can be contaminated by Hg from various sources, so this limitation would be draconian, in that it assumes only the discharge from wastewater treatment or industry is responsible. Clearly there are legacy sources of Hg contamination in food webs of California that are not associated with these activities. In spite of the challenges in quantifying bioaccumulation factors, I still think it is preferable to use a water column concentration in the effluent. This makes monitoring and regulation more feasible.

RESPONSE TO MBS 9

The reviewer's agreement with the approach is noted.

COMMENT MBS 10

[Section 6.12 (Issue L)] I am not very familiar with the analysis of bioaccumulation factors (BAFs), but it is clearly an analysis of limited utility because it is based on so few studies, incomplete science, and the variability in the resulting metric is so high (note the log scale on Figure I-1 in the Water Column Appendix). I cannot reasonably evaluate what would constitute an appropriate water column limit for effluent, nor can I imagine that anyone can. However, another environmental scientist is probably in a much better position to evaluate this than I am. I do recognize that the limit must be higher than the current average water column concentration of 4.7 ng/L for California waters (as mentioned in the draft report). Another consideration is how and when the monitoring should proceed. The issue mentions quarterly sampling, but this may not fit with the timings of maximum concentration and/or maximum discharge. Perhaps the schedule of sampling for a particular discharge should be designed on an adaptive basis that could be determined from past discharge records of each company? Again, this is clearly not my area of expertise.

RESPONSE TO MBS 10

In line with the reviewer's suggestion, the actual monitoring frequency for a facility is determined based on the discharge volume, other facility specific variables, and the federal regulations (the Nation Pollutant Discharge Elimination System). For a typical wastewater treatment plants, often monthly samples are required. If the discharge is intermittent, sampling will only be conducting during the discharge. In the draft Provisions, a minimum frequency is set forth.

In regards to timing, the effluent limitations are derived using conservative assumptions about the variability of the discharge, so that timing the sampling to attempt to measure maximum concentrations or maximum discharges is not necessary. For example the maximum background concentration (not average concentration) is considered when assigning effluent limitations.

In regards to effluent limitations being higher than the average ambient concentration- this is not a general principle that must be followed. If waters are impaired (in other words, if pollutant levels exceed water quality objectives) then the effluent limitations that will be necessary to restore those waters may well be below the average ambient concentration. Also averages can be skewed by very high

concentrations, and many of the data are from waters where mercury concentrations are elevated (since TMDLs prompt more monitoring). New text with a reference to the figure on the spatial distribution was added to the Staff Report (Section 4.5.1):

“Many of the data were from areas with elevated mercury such as San Francisco Bay. See Figure N-4, in Appendix N, for the spatial distribution of samples.”

Since mercury is often bound to sediment, the use of sediment controls will effectively reduce the transport of mercury into waters, for discharges that can contain large amounts of sediment.

COMMENT MBS 11

[Section 6.8 (Issue H)] I generally agree with the recommendation for Option 2. However, I have concerns that may require more thought and revision before this option can be adopted in the Amendment. I am particularly concerned about the specific (and singular) emphasis on erosion control. I acknowledge that sediment-adsorbed Hg is the dominant source of Hg contamination of water bodies and food webs, that former mines are important contributors, and that erosion of mine tailing can move significant quantities of Hg-laden sediment to downstream locations (Singer et al., 2013). Thus, limiting erosion of Hg-laden sediment from Hg and gold mines (especially abandoned ones) is potentially important. However, this focus on future erosion does not acknowledge that most of the landscape downstream from large and/or important Hg-contributing mines (e.g., within Yuba R, Cache Cr basins, etc) is ALREADY contaminated with Hg-laden sediment over broad areas and to deep depths (e.g., (Bouse et al., 2010; Donovan et al., 2016a, b; Donovan et al., 2013; Marvin-DiPasquale et al., 2009; Singer et al., 2013)), so controlling erosion from these mines (which in and of itself may be infeasible in many locations) may only have a minor contribution in limiting further contamination to these water courses. In fact, the infrequent flooding regime that inundates previously contaminated sediments for long periods, may thus enable in situ MeHg production (in the absence of further erosion), which could drain back into rivers and become available to food webs. We have documented widespread contamination of sediment throughout the Sacramento Valley, so even though it might be helpful to control the erosion of sediment from abandoned mines, the non-point Hg source problem may be of greater concern. I would like to see this risk reflected in the language on Issue H (and others). This would put less blame/focus on owners of abandoned mines (including government agencies) and treat the problem as a legacy of former mining gone amok. This nonpoint upland contamination source is not explicitly included in Issue I.

RESPONSE TO MBS 11

Agree that the flooding of contaminated sediment is a source of methylmercury to biota. However, is not clear how this source of methylmercury could be controlled. The Provisions include “logical first order controls” as the reviewer phrased it in later comment, which is erosion control for areas enriched in mercury. Text was added to section 4.4.6 to acknowledge this source of methylmercury:

“Another potentially large source of methylated mercury is the landscape downstream from historic mining areas that are contaminated with mercury laden sediment. This sediment has become part of the landscape, covers large areas to deep depths (e.g., (Bouse et al., 2010; Donovan et al., 2016a, b; Donovan et al., 2013; Singer et al., 2013)). When occasionally flooded, methylmercury is produced, which could drain back into rivers and become available to food webs.”

That section (now Section 6.9) was reorganized to talk about historic mines first and mines tailings that are integrated into the landscape. Also new text was added at the end of Section 6.9.2:

“Another challenging aspect to the historic mining legacy is that much of the landscape downstream from mercury mines is already contaminated with mercury laden sediment over broad areas and to deep depths. These are not recognizable mine sites, rather the sediment has become part of the landscape. This type of mercury is very difficult to address and may be a more important source of methylmercury than the original mine sites. In some cases these sources could be addressed through the Clean Water Act 401 certification and wetland program and the nonpoint source program (Section 6.10).”

New text was added to Section 6.10.2:

“Also the inundation of mercury contaminated sediments from occasional flooding of land can produce methylmercury. A great deal of mercury contaminated sediment has already left mine sites and become part of the landscape as a result of historic mining. The methylation of the mercury in these contaminates sediments during occasional flooding is not a feasibly controllable process at this time. ”

Regarding sediment controls to reduce the transport of mercury into water and the comment that these sediments from mercury contaminated areas are only minor contributions- when viewed individually, each discharge is only a minor contributor. That can be said for every individual discharge that can carry mercury or methylmercury, whether it is a discharge from a mine, storm water, or wastewater. This does not mean some level of control for each discharge is inappropriate.

COMMENT MBS 12

[Section 6.9 (Issue I)] In this section, the terminology is a bit challenging to interpret. Nonpoint Hg sources include riparian zones as listed, but the discussion seems to be focused only on the lowland environment (e.g., emphasizing permanent wetlands and agricultural lands). To my mind, this is too narrowly focused and ignores the potential production and delivery to the food web of MeHg in nonpoint source areas that are only seasonally wet. I

generally support Option 2 here, but with a few caveats. The language here is focused on total Hg concentrations in sediment as an indicator of MeHg risk to water bodies (and the food web). There is not necessarily a direct link, even if this is logical to first order. Lower concentrations of total Hg (below 1 ppm), but well above background, may still provide important sources of MeHg to aquatic ecosystems. There should be acknowledgement here that we need to link the hydrologic (flooding) regime to the risk of MeHg production, since even highly contaminated dry sediments won't contribute Hg to food webs. The two risks are inundation that decreases oxygen levels in contaminated sediments and enables microbial methylation, and the erosion of Hg-laden sediment for delivery to downstream areas where methylation is likely (higher risk of inundation).

RESPONSE TO MBS 12

Agree that dry mercury contaminated sediments will not contribute methylmercury to food webs. Since the Water Boards regulate contaminants in water or “discharges”, the Provisions should not affect dry sediments. The Provisions includes requirements to keep sediments out of downstream waters, including areas that are likely to be inundated (riparian zones, wetlands). This type of requirement seems consistent with the comment. Yes, another issue is that areas that may be inundated are already full of contaminated sediment. It is not obvious how the methylmercury that results from flooding uplands could be controlled and the reviewer does not make a suggestion. The Provisions include “logical first order controls” as the reviewer phrased it. TMDLs or clean up orders will likely be needed to develop additional controls in highly contaminated areas.

Nonpoint source discharges are not confined to the “lowland environments”, that section stated that nonpoint sources included forests. Public forest land comprises much of the land at higher elevation in the Sierra Nevada Mountains. We added “open land” and “grazing land”: and this text was clarified (second sentence of Section 6.9.1):

“The Nonpoint Source Policy aims to minimize nonpoint source pollution from land use activities in agriculture, grazing, urban development, forestry, recreational boating and marinas, hydromodification and wetlands. This can include lands with historic mine tailings and other open land.

COMMENT MBS 13

Second, the spatial distribution of total Hg is not well established for most areas. We don't know the vertical distribution of Hg contamination in areas downstream of former mines, nor do we know how far this contamination extends laterally away from river courses (but is still susceptible to inundation during large floods). These aspects represent an important data gap that the Alpers study is unlikely to fill at the level of detail required to understand the nonpoint source risk of MeHg contamination.

RESPONSE TO MBS 13

Agree- this is a data gap. TMDLs or clean up orders will likely be needed to develop additional controls in highly contaminated areas.

a. In reading the Draft Staff Report and proposed rule, are there any additional scientific findings, assumptions, or conclusions that are part of the scientific basis of the proposed rule not described above?

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

COMMENT MBS 14

Hg poses a long-term problem for the State of California. In addition to low-level global inputs of Hg through atmospheric deposition (pollution source: global anthropogenic emissions), California has a long history of Hg mining and its use for (most dramatically) industrial-scale gold mining. These historical processes and activities, combined with industrial activities and the subsequent redistribution of Hg attached to sediment and dissolved in water, have created pollution over landscapes and regions. Due to its historical legacy of gold and Hg mining (and to a lesser extent the industrial legacy in the estuary as well), the largest region of Hg pollution in California waterways is in the northern half of the state (e.g., San Francisco Bay region including the Bay-Delta estuary and many of its contributing streams). Some of these areas have already been designated as ‘impaired waters’ under the Clean Water Act, yet others are less well-regulated or monitored. It is likely that the problem of Hg contamination will persist well into the future because the Hg pollution in waters and sediments is so widespread that clean-up efforts are challenging if not intractable. Most of the historic Hg attached to sediment and in waters is in an inorganic form, and therefore not particularly dangerous to biota because it cannot be incorporated into tissues and the bloodstream. However, at many locations throughout the landscape methylmercury (MeHg), the toxic form of Hg that affects biota (including humans) may be produced by methylating bacteria in conditions of low oxygen. Unfortunately, the Hg pollution of California waters has indeed led to the production of MeHg and the subsequent contamination of food webs that depend on these waters, and the problem is compounded with higher trophic organisms such as fish. This raises a major challenge in California because fish form the basis of the diet of many forms of wildlife (waterfowl along the Pacific Flyway and migratory anadromids). Fish that are potentially contaminated with MeHg are also an important component of the diet of many California residents, and especially that of subsistence communities including tribes that have depended on this food source (and associated waters) for their entire cultural history.

RESPONSE TO MBS 14

Agree. Comment noted.

Below I provide specific responses/impressions to aspects of the Staff Report (again keyed to the relevant sections of the report).

COMMENT MBS 15

Section 4.1 There is evidence that iron-reducing bacteria (FeRB) may also play an important role in methylating Hg some systems (Alpers et al., 2014; Gilmour et al., 2013), so it may be that conventional assumptions about where and how Hg is methylated are outdated. In particular, since sulfate-reducing bacteria (SRB) have been primarily implicated in MeHg production and sulfate is limiting in most of large basins (e.g., apart from wetlands), it is often assumed that wetlands comprise the only important loci for methylation. However, it is possible that FeRB play an important role, especially in locations where iron is in high supply compared to sulfate (i.e., upland locations that are not permanently inundated).

RESPONSE TO MBS 15

This information was added to section 4.1 of the Staff Report (now part of the second paragraph):

“There is evidence that iron-reducing bacteria may also play an important role in methylating Hg in some systems (Alpers et al., 2014; Gilmour et al., 2013), not only sulfate-reducing bacteria. The formation of methylmercury is a complex, far from fully understood, biogeochemical process driven by factors that control the activity of methylating bacteria, such as the availability of metabolic electron donors and acceptors, and the availability of aqueous phase mercury complexes (Jonsson et al. 2012).”

COMMENT MBS 16

Section 4.2 There is evidence that biofilms and algae also play an important role in providing MeHg at the base of food webs (Tsui et al., 2012). This is indicated elsewhere but missing here.

RESPONSE TO MBS 16

The Staff Report acknowledged that phytoplankton is a critical step in the pathway of methylmercury bioaccumulation. The information provided was added to Section 4.2 (second paragraph).

“Also, biofilms and algae play an important role in providing methylmercury at the base of food webs (Tsui et al., 2012). Zooplankton consumes phytoplankton, and then small fish and invertebrates consume zooplankton and algae.”

COMMENT MBS 17

Section 4.4 I suggest that there is too much emphasis on wetlands and reservoirs as the primary sources of MeHg production. It may not be the case, which really opens up a much larger regulatory question. Just because fish MeHg is higher in these environments (which is not universally the case—see below), it does not follow that all or most MeHg production occurs in wetlands. Resident fish in permanent wetlands have longer exposure times to MeHg locally produced. However, the rates of MeHg production may not be higher (especially after accounting for *in situ* demethylation).

RESPONSE TO MBS 17

The emphasis on the reservoirs and wetlands as the sources of methylations was reduced from other revisions to the Staff Report (from comments above). These revision added text on additional methylation sources. Also for wetlands, in the very beginning of section of 4.4.7, the text acknowledges that wetlands may be a sink for methylmercury, instead of a source of methylmercury.

COMMENT MBS 18

It is unreasonable to assume, in heavily Hg-contaminated environments of California (gold mining regions), that atmospheric deposition of Hg plays an important role in delivering MeHg to the food web. Recent work has shown that the isotopic signature of MeHg in food webs of Coast Ranges, Yolo Bypass, and Yuba/Feather Rivers, for example, is similar to that of the Hg stored in sediments deposited during the historical mining period (Donovan et al., 2016a, b; Gehrke et al., 2011).

RESPONSE TO MBS 18

Agree that atmospheric deposition is less important in gold mining regions, but in some water bodies it is thought to be the main source in California, as we note in Section 4.4.3. After the line:

“Mercury deposition from atmospheric emissions is thought to be the major source of mercury in some Southern California lakes and reservoirs (U.S. EPA 2012, Tetra Tech 2008).”

New text was added:

“However, in heavily mercury contaminated environments of California (gold mining regions), atmospheric deposition of mercury is unlikely to plays an important role in delivering methylmercury to the food web. Recent work has shown that the isotopic signature of methylmercury in food webs of Coast Ranges, Yolo Bypass, and Yuba/Feather Rivers, for example, is similar to that of the mercury stored in sediments deposited during the historical mining period (Donovan et al., 2016a, b; Gehrke et al., 2011). See also Table N-11, on the estimated mercury loadings from the Sacramento-San Joaquin Delta TMDL (Delta) and the San Francisco Bay TMDL.”

COMMENT MBS 19

Also, what is the evidence that Hg from wet deposition is ‘more readily methylated’, particularly in the California setting? This seems like speculation and is perhaps based on an outdated notion (citations from 2002 and 2003), especially when applied at the landscape scale.

RESPONSE TO MBS 19

That text has been revised with updated references to provide better evidence (this section was on the issue of bioavailability of different sources.)

“Related, there is a limited ability to predict how an ecosystem may respond to changes in the various sources of mercury (Hsu-Kim et al. 2013). Evidence suggests some forms or sources of mercury/methylmercury are more likely to enter the food chain. The inputs of methylmercury from terrestrial and atmospheric sources have been found to bioaccumulate to a substantially greater extent than methylmercury formed *in situ* in sediment (Jonsson et al. 2012, Jonsson et al. 2014).”

Again, while the mining legacy is important in many areas in California, in other areas, particularly in some reservoirs in Southern California, atmospheric mercury is thought to be an important source (see comments MSB 5 and MSB 18). (That passage made no assertion specifically for *wet* or *dry* deposition.)

COMMENT MBS 20

[Section 4.4.8] I’m also unconvinced of the relevance of the statement supported by the Fleck reference. I don’t understand how this establishes the importance of a wet deposition MeHg source to food webs.

RESPONSE TO MBS 20

This text was not about mercury atmospheric deposition. This section is on *bioavailability* of different types of mercury. The example in the Fleck reference is about bioavailability in the aquatic environment “...preliminarily results with isotopically labeled mercury indicate that the mercury that is taken up into food webs comes from mercury that is *dissolved in the water column*, rather than the mercury associated with the bottom sediments in a water body (Fleck et al. 2014)”

COMMENT MBS 21

Another important potential impact of climate change is increasing frequency and duration of inundation, which may enable higher net MeHg production in areas that are seasonally dry, but which contain high Hg inventories over multiple meters of depth (Singer et

al., 2016). We now have good evidence that such areas may be important loci of MeHg production and uptake into food webs (Donovan et al., 2016a, b).

RESPONSE TO MBS 21

New text added to section 4.4.10, second paragraph:

“Related to the storms, is the increasing frequency and duration of inundation of areas that contain high mercury inventories over multiple meters of depth from the historic mining legacy (Singer et al., 2016). This increase in flooding will enable higher methylmercury production in these mercury contaminated areas. Such areas may be important locations of methylmercury production and uptake into food webs (Donovan et al., 2016a, b).”

COMMENT MBS 22

Section 4.5 It seems that this monitoring effort is probably unnecessary. Efforts could be better targeted on sampling loci that we might expect to be disproportionately contributing to MeHg loads. In other words, we continue to operate sampling over broad spatial scales, yet mixing of highly concentrated water with water of low concentrations will tend to systematically dilute the signal and the timing of sampling is of particular importance. Similarly, the location within the water column should prioritize locations where benthic organisms, etc. might take up MeHg (at the base of the food web).

RESPONSE TO MBS 22

This comment is assumed to apply to monitoring mercury in the water column. The primary goal of the Water Boards ambient monitoring is to assess compliance with water quality objectives. If the objectives in the mercury Provisions are adopted, the focus of mercury ambient monitoring will move more towards fish tissue monitoring only, and away from monitoring mercury in the water column. Monitoring mercury in the water column may still be done for special studies or TMDLs. Monitoring mercury in the water column must also be done by dischargers for compliance with effluent limitations.

COMMENT MBS 23

Also, there appear to be major geographic biases in sampling efforts, where particularly contaminated streams are not being consistently sampled for water and/or fish (e.g., Yuba R, Cache Cr). See example from Fig. 8 in (Singer et al., 2016) below, where forage fish MeHg concentrations in the Yuba and Feather Rivers equate to an average of 0.083 mg/kg wet weight, higher than most values shown in Figs. 4-8 and 4-9:

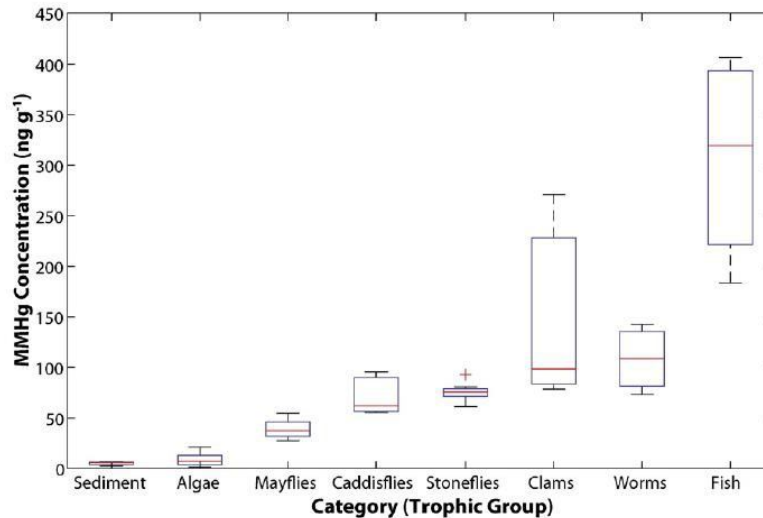


Fig. 8. Boxplots of MMHg (dry weight) in biota separated by organismal group for all samples in the Yuba-Feather system. Sampling locations are shown in Fig. 1. Sampling details provided in Donovan et al. (2016).

Other fish data from Cache Cr exhibit even higher MeHg concentrations. By contrast, the average MeHg concentrations for prey fish we analyzed from Yolo Bypass (a lowland wetland site expected to have much higher MeHg contamination) were 0.05 mg/kg. Note: the proposed MeHg limits for prey fish are 0.05 mg/kg for 50-150mm and 0.03 mg/kg for <50mm fish. Given that these fish provide a likely food source for higher trophic organisms, we may be missing important upstream sampling/monitoring locations that could better guide management and water quality control efforts. Given the migratory habits of many fish species, upland river sites represent an important data gap for understanding the regional picture of MeHg contamination, whether or not upstream reservoirs are providing a downstream MeHg supply.

RESPONSE TO MBS 23

This comment seems to be based on the data presented in Figures 4-8 and 4-9. These figures show data from prey fish not sport fish. Most of the data on prey fish to date is from special studies not from the Water Board's statewide monitoring program. So yes, the data is from only a few geographical areas. The Water Board's statewide monitoring program has just begun planning for sampling of prey fish. If the proposed water quality objectives are adopted that would provide additional justification for the statewide monitoring program to sample prey fish throughout the state.

The data in Figures 4-8 and 4-9 was taken from the Water Boards public database: the California Environmental Data Exchange Network (ceden.org). That data was fed into the data base by researchers who conducted the special studies. The data in Signer et al. 2016 was not in that database. The mercury projects staff at the Water Board encourages mercury researchers to add mercury data from their research to the public database, so it is accessible by all scientists. The Water Boards are working towards better connections of our databases with other state water quality databases and

national databases. The suggestions for monitoring designs will be shared with the statewide monitoring program.

COMMENT MBS 24

Figures 4-3 and 4-4 seem to contradict the notion that fish of 150-500mm are the most relevant to regulate for MeHg. The all sizes category on these plots is consistently higher (for both trophic levels 3 and 4). Was this designed because that is the size threshold allowed for fishing or what is typically eaten? If so, this was not made clear.

RESPONSE TO MBS 24

Yes that was not clear in the figures, and the figures were clarified. The figures show mercury concentrations in fish. There is an issue that the size of the fish is not reported in the database in many cases. Many of the fish in the “all sizes” category may well be from fish that were 150-500 mm, but it is unknown since the length of the fish was not reported in the database. If the mercury water quality objectives are adopted with the specified fish lengths, those specified lengths will guide future monitoring efforts. Additionally staff working on the mercury projects keep emphasizing the importance of reporting the length of fish in the database. The figure was clarified by adding text to the legend “ ‘All sizes’ includes many data points for which the length was not reported.”

COMMENT MBS 25

Section 4.5.5 This section is very incomplete. There are numerous studies documenting total Hg across various parts of the SF Bay Region (including contributing watersheds). Why is the information not included here? Some relevant papers include, but not an exhaustive list: (Bouse et al., 2010; Domagalski, 2001; Domagalski et al., 2004; Donovan et al., 2016a, b; Donovan et al., 2013; Singer et al., 2013). Several of these papers clearly documented that the threshold for background total Hg in various parts of the basin is ~0.08 ppm (similar to the results presented for Cache Cr). Furthermore, these studies document that concentrations an order of magnitude higher are common in many locations (including river floodplains, bypasses, and Bay-Delta bottom sediments) with some loci that are 2 or more orders of magnitude higher in total Hg. For example, our group has documented concentrations of 3-10 ppm in Yuba River sediments and up to ~200 ppm in sediments draining Hg mines in the Cache Cr basin.

RESPONSE TO MBS 25

Thank you for this additional supporting information. This was added to the report (Section 4.5.5). The Staff Report is not meant to be an exhaustive list.

“Additionally several studies in the San Francisco Bay region suggest that the threshold for background mercury (total mercury) in various parts of the basin is about 0.08 mg/kg (Bouse et al., 2010; Domagalski, 2001; Domagalski et al., 2004;

Donovan et al., 2016a, b; Donovan et al., 2013; Singer et al., 2013), similar to the findings for Cache Creek. Furthermore, these studies document mercury concentrations that are an order of magnitude higher or more in many locations (including river floodplains, bypasses, and Bay-Delta bottom sediments)."

COMMENT MBS 26

Section 4.8 I'm not convinced about the research on selenium and Hg. The interactions may be well understood in laboratory conditions and there may be negative correlations between Se and MeHg concentrations, but that does not clarify the process by which Se modulates methylation processes. Perhaps I'm just not familiar with the relevant literature on this, but I am not convinced by the references provided. Quite frankly, I'm not sure why this whole section is included in this draft report. It seems out of place because the evidence is not convincing that Se amendments would provide any benefit (and could potentially be harmful, as indicated) to ameliorate MeHg production/uptake. It is also not followed up in the development of objectives.

RESPONSE TO MBS 26

Agree. This section is included because we have received other comments suggesting that we included selenium in the development of the water quality objectives or suggestions that the Water Boards dose contaminated reservoirs with selenium. Since, as you point out, the benefits of using selenium is not clear, selenium was not included in the development of the objectives. Section 4.8.2 states: "Overall, the state of the science on selenium–mercury interaction is not close to a point at which it could be incorporated into regulatory limits for mercury."

S.4 Edwin van Wijngaarden (EVW)

Peer review of draft proposed rule for Mercury Water Quality Objectives and Program of Implementation

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and Community Health
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Thank you for the opportunity to review the draft proposed rule for Mercury Water Quality Objectives. I have a Ph.D. in Epidemiology and am a Fellow of the American College of Epidemiology. I have extensive experience in managing and conducting epidemiologic studies

and have published 95 peer-reviewed manuscripts with a focus on neurobehavioral outcomes and environmental and occupational health. In the past decade, my primary research efforts have focused on the influence of environmental exposures (in particular mercury and lead) on cognitive outcomes in children and adults. Because of my expertise in Public Health Toxicology, I will comment on the following three conclusions of the draft proposed rule:

1. The proposed Sport Fish Water Quality Objective was derived using sound scientific information and methods;
2. The California Tribes Fish Use Study (Shilling et al. 2014) contains a sound data set to use to establish a default water quality objective to protect tribes;
3. The consumption rate of 4 to 5 meals per week (142 grams per day) is a sound basis from which to derive a subsistence fishing water quality objective that would be applied to the highest trophic level fish.

The basis for my comments are sections of the draft staff report (dated June 2016) and supplementary appendices that are relevant to the three conclusions above (as identified in the request for scientific peer review, Attachment 2), the Shilling 2014 report, the San Francisco Bay Seafood Consumption 2000 report, the US EPA 2002 report estimating fish consumption in the United States and the related 2000 methods report, and literature pertaining to the health effects of mercury. References cited in this review are provided at the end of the document.

The proposed Sport Fish Water Quality Objective was derived using sound scientific information and methods

The Sport Fish Water Quality Objective for mercury is intended to protect the beneficial uses of commercial and sport fishing, wildlife habitat, and marine habitat. The Sport Fish Water Quality Objective is expressed as follows: the average methylmercury concentrations shall not exceed 0.2 milligrams per kilogram (mg/kg) fish tissue within a calendar year. This fish tissue concentration (FTC) is the methylmercury water quality objective. The objective must be applied to TL3 or TL4 fish, whichever trophic level is the highest existing level in the water body.

The objective for human health was derived using U.S. EPA's equation for calculating the fish tissue criterion (US EPA 2001):

$$FTC = BW \cdot (RfD - RSC) / FI \quad (\text{see page H-1})$$

where FTC is as defined above, BW = human body weight, RfD = the reference dose for methylmercury established by EPA (as described in Rice et al. 2003 and Dourson et al. 2001), RSC = the relative source distribution to account for store bought marine fish and other sources, and FI = fish intake. The FTC is affected by uncertainties in all these parameters, but RSC and

especially BW do not appear to greatly impact the water quality objective, especially since the objective will be rounded to one digit (Tables H- 2A and H-2B). Therefore, my comments here will focus on the two remaining parameters of the equation: the RfD and the FI estimate.

COMMENT EVW 1

As mentioned in Appendix H, the RfD was derived from a study of maternal-child dyads in Faroe Islands reporting on the adverse association between prenatal methylmercury exposure (as measured in cord blood) and child developmental outcomes (Grandjean et al. 1997). As noted elsewhere (e.g. Dourson et al. 2001; Grandjean et al. 2001; Weihe et al. 1996; Jacobson et al. 2015), the primary source of mercury exposure in this study population was through the traditional consumption of whale meat, not fish, and co-exposure to other contaminants such as polychlorinated bi-phenyls (PCBs) are of concern. It would be helpful if the staff report could discuss the generalizability of the findings from this study for the purpose of the proposed Sport Fish Water Quality Objective.

RESPONSE TO EVW 1

A paragraph on this topic has been added to the staff report in Section 4.7 (now the 4th paragraph):

“In the Faroe Islands, the primary source of mercury exposure in the study population was through the traditional consumption of whale meat, not fish, and co-exposure to other contaminants such as polychlorinated bi-phenyls (PCBs) are of concern. However, in California PCBs also contaminate fish tissue at levels that limit advised consumption (Davis et al. 2010, Davis et al. 2012). One hypothesis as to why adverse effects of mercury were not found in the Seychelles, but adverse effects were found in the Faroe Islands is that there are other neuroprotective nutrients in seafood, such as selenium and iodine, long chain polyunsaturated fatty acids, (Oken 2012, Meyers 2009). Freshwater fish do not have these nutrients in the same amounts as marine fish (Haldimann et al. 2005, Steffens 1997), and many Californians are exposed to mercury by consuming freshwater fish. While many people in the Faroe Islands and the Seychelles ate fish several times a week, in the Faroe Islands most of the methylmercury exposure was from infrequent (twice a month) consumption of pilot whale meat (Dourson 2001). Recreational fishers in California may also have infrequent high methylmercury exposure from weekend fishing trips, along with a steady methylmercury exposure from regularly purchased commercial fish. There are other theories as to why the two studies found conflicting results, such as study design (Oken et al. 2008, Debes et al. 2006). Ultimately, mercury is a known neurotoxin and the Faroes Island study provides data to support a reference dose.”

COMMENT EVW 2

Furthermore, since the derivation of the US EPA's RfD several additional studies have been published reporting on the association between prenatal methylmercury exposure and child development. There appears to be substantial uncertainty regarding the consequences of maternal consumption of fish with naturally-acquired MeHg contamination. For example, several studies in the Faroe Islands (Grandjean et al. 1997), New Zealand (Crump et al. 1998), United States (Sagiv et al. 2012) and Arctic Quebec (Jacobson et al. 2015) have reported adverse associations with cognition and behavior, but other studies in the Republic of Seychelles (van Wijngaarden et al. 2013; Strain et al. 2015), United States (Oken et al. 2016), the United Kingdom (Daniels et al. 2004), and Spain (Llop et al. 2012) have found no consistent evidence of adverse consequences of prenatal methylmercury exposure from fish consumption on children's development. It is likely that differences in study design, co-exposure to nutrients and contaminants, and genetic factors partially account for the inconsistencies in study findings which consequently may result in different RfD values (van Wijngaarden et al. 2006). RfDs vary by regulatory body and are often higher than US EPA's value; for example, it is four times higher in Alaska (<https://dec.alaska.gov/water/wqsar/wqs/pdfs/FishConsumption.pdf>) and the provisional tolerable intake is two times greater in Canada (http://www.hc-sc.gc.ca/fn-an/pubs/mercur/merc_fish_poisson-eng.php). Given the FTC equation, the water quality objective will increase or decrease as the RfD increases or decreases, respectively. While the lower US EPA RfD will result in a more protective FTC, the draft report could acknowledge the uncertainty and variability in determining the RfD and how this would influence the water quality objective.

RESPONSE TO EVW 2

The Staff Report does contain very brief paragraph (Section 4.7) on the conflicting evidence considered when U.S. EPA derived the reference dose –that while adverse effects were seen in the Faroe Islands, no effects were found in the Seychelles. The following sentence will be added to section 4.7: “While other studies in the Seychelles (van Wijngaarden et al. 2013; Strain et al. 2015), United States (Oken et al. 2016), the United Kingdom (Daniels et al. 2004), and Spain (Llop et al. 2012) have found no consistent evidence of adverse consequences of prenatal methylmercury exposure from fish consumption on children’s development.” The staff report also includes additional references that indicate adverse effects of mercury.

The references the reviewer provided on Canada and Alaska concern the development of fish consumption advisories, not water quality criteria. In the Alaska reference it states “The RfD was 2.5 times greater than EPA[’s] to account for health benefits of eating fish” (slide 8). The Alaska reference also correctly states that fish consumption advisories are not equivalent to water quality criteria, and that water quality criteria “do not account for health benefits of eating fish”. Therefore these references are not entirely relevant to the mercury Provisions, but to fish advisories. In California, fish consumption advisories are developed by another agency, the Office of Environmental Health Hazard Assessment, and the advisories are developed considering the beneficial effects of consuming fish (see Appendix E, Section 4).

COMMENT EVW 3

The RfD was derived based on data demonstrating adverse associations with prenatal methylmercury exposure. However, exposure occurs both prenatally and postnatally and throughout the life course. The health effects of postnatal methylmercury exposure are uncertain (Karagas et al. 2012), with no clear impact on cardiovascular disease and hypertension (e.g. Mozzafarian et al. 2011, 2012), and limited evidence of adverse associations with neurodevelopment and cognition in children (e.g. Myers et al. 2009; Boucher et al. 2016) and older adults (e.g. Weil et al. 2005; Yokoo et al. 2003). Use of evidence pertaining to risks in pregnant women and women of childbearing age results in a lower RfD and thus a more protective water quality objective. The draft report does not appear to distinguish between prenatal exposure (from fish consumption during pregnancy) and postnatal exposure (in either children or adults), and chronic vs. developmental risk. The U.S. EPA 2000 guidance document distinguishes between chronic human health risks and developmental health risks when discussing the default parameters but the water quality objective draft report is not clear on this point. Therefore, it may be informative to discuss the demographics of fish consumers targeted in the objective types (i.e. sport fish, tribal subsistence, subsistence) and the proportion of the target population that may be at the highest risk.

RESPONSE TO EVW 3

The text of the Staff Report was clarified as to how the reference dose was derived. In the section on “Methylmercury Effects on Human Health” (Section 4.7 of the Staff Report) after the sentence, “Toxicity to the developing nervous system of the fetus is considered the most critical endpoint” New text was added to clarify “ The water quality objectives were derived from a the U.S. EPA reference dose, which was based on protecting the developing fetus.” There was already mention in this same section of the Staff Report about possible effects on cardiovascular health. Nonetheless, U.S. EPA considers that the reference dose for the entire population, not only for women of child bearing age (U.S. EPA 2001,Rice 2003).

Additionally, another California agency, the Office of Environmental Health hazard Assessment is responsible for communicating to the public the risk of consuming mercury contaminated fish to the public and which segment of the population might be at the greatest risk of mercury toxicity.

COMMENT EVW 4

The San Francisco Bay Seafood Consumption study (hereafter called “SFEI 2000”) was considered to be one of the highest-quality studies of fish consumption in California done to date. This study provided the FI estimate of 32 grams per day which has already been used a various regulatory settings. The primary goal of the study was to collect quantitative data to

characterize exposures to contaminants in fish and shellfish caught in the Bay among the general fishing population of San Francisco Bay. The study included on-site personal interviews of 1,331 participants (77% response rate which is adequate) who were fishing at piers, beaches and banks, or private or party boats. Interviews were conducted over a 12-month period (summer of 1998 – summer of 1999), and asked about four-week recall of fish consumption. The recruitment approach was reasonable given the lack of a comprehensive list of anglers and the need to conduct in-person interviews to increase participation and understanding of the questions. Fish consumption rates were adjusted for avidity (i.e. how frequently anglers go fishing) in an effort to reduce bias; avidity-adjusted rates are lower than unadjusted rates. The magnitude and direction of any other biases in the fish consumption rate would be unknown. The SFEI 2000 report includes a comprehensive discussion of the study's strengths and limitations.

RESPONSE TO EVW 4

The reviewer's agreement with the approach is noted.

COMMENT EVW 5

As discussed in Appendix G of the draft report, short-term recall such as a four-week period may result in a skewed distribution as shown in Table 5 of the SFEI 2000 report, with a mean of 6.3 grams per day but a median of 0 grams per day. The SFEI 2000 report considers the 12-month recall to be less reliable because longer recall periods are more difficult for respondents to answer accurately. The rate of 32 grams per day is the 95th percentile in Table 5 and represents the rate among all consumers of Bay fish. The 95th percentile of the per-angler consumption rate in Table 6 is lower (24 grams per day) and represents consumption among all survey respondents including anglers that do not eat fish. For the purpose of the water quality objective, utilizing the results from Table 5 results in a more stringent FTC as it assumes that all anglers will eat the fish caught. (As noted in the report, it is also more conservative than utilizing the EPA default consumption rate of 17.5 grams per day apparently based on the 90th percentile of the fish intake data obtained in a national survey.) In all, the study's methods and design appear to be scientifically sound.

RESPONSE TO EVW 5

The reviewer's agreement with the approach is noted.

COMMENT EVW 6

Since the time of the SFEI 2000 report, health advisories regarding fish intake have been promulgated which may have affected fish consumption rates (e.g. Oken et al. 2003, Rehm et al. 2016). The impact of temporal trends in fish consumption, if any, on the water quality objective should be discussed, as should be the generalizability of the SFEI 2000 study to other angler communities in California.

RESPONSE TO EVW 6

Recent fish consumption studies will always be valuable, and the Water Boards are obligated to review water quality standards on a regular basis.

While public awareness of contaminants in fish and advisories may reduce fish consumption rates, the Water Boards are not mandated to revise water quality objectives to reflect artificially suppressed fish consumption rates. When agencies set environmental standards using a fish consumption rate based upon a suppressed consumption level, they may set in motion a downward spiral whereby the resulting standards permit further contamination of the fish. The mission of the Water Boards, set forth by the Porter-Cologne Water Quality Control Act, is to protect past, present, and probable future beneficial uses (in this instance the beneficial use is fish consumption). Therefore, if fish consumption rates are lower in the future, the Water Boards would need to carefully consider all information before altering the level of protection.

Rather than trying to estimate how representative the SFEI 2000 study may be, Appendix G provides data from other fish consumption studies from California for comparison to the SFEI study. Also Section 4.9 of the Staff Report summarizes these data, and Section 6.2 discusses why the data from the SFEI 2000 study was used as the fish intake parameter for California as opposed to another value.

COMMENT EVW 7

To compare methylmercury concentrations in fish tissue to the FTC, fish mercury samples are collected within a calendar year and subsequently combined into one value. The rationale for summarizing values over a longer period of time is that potential adverse consequences of methylmercury exposure are believed to be chronic in nature, and methylmercury exposure in fish are believed to not fluctuate strongly across seasons. Secondly, combining multiple values into one result is a statistically more precise estimate of concentration. This rationale sounds reasonable, although it may be necessary to add more references to support the statements about the chronic nature of toxic effects and lack of seasonal fluctuations. If there is empirical fish tissue data available (even if the sample size is small) to provide additional support for the latter assumption, it would be good to present those.

RESPONSE TO EVW 7

Agree that more data would be helpful. However, the Water Boards do not have data that can be used to compare the mercury levels in fish in different seasons. The statewide monitoring program generally captures a group of about ten fish on one day and then the water body is not sampled again for several years. Also, the sample locations, fish sizes and years all vary. For example, for Lake Berryessa, a water body with one of our largest data sets, there is data available from only one sampling event in the summer and five sampling events in winter, from the past 30 years. Data from one summer is hardly representative of the seasons. In another example Clear Lake, the largest natural freshwater lake in California, useful data are available from just three sampling events: one for May, September and October in various years. There is

additional older data, but, it should not be used to answer this question, since there is no accompanying data on the length of the fish. The mercury levels in fish are related to the size of the fish, so size is a confounding factor in determining if mercury levels vary by season. Overall, with the small number of fish sampling events, it would be hard to attribute differences in fish mercury levels to the season, when a number of factors could have been the cause.

Staff also consulted a California researcher to attempt to find such data in the peer reviewed literature. That researcher didn't know of such data, but stated that the seasonal fluctuations of mercury concentrations in fish are unlikely to be statically significant in larger sport fish. The Staff Report includes the references that were originally found on the stability of mercury level in fish, in Section H.4.

The California Tribes Fish Use Study (Shilling et al. 2014) contains a sound data set to use to establish a default water quality objective to protect tribes

COMMENT EVW 8

To derive the Tribal Subsistence Fishing Water Quality Objective, the draft report incorporates the fish intake estimates reported in the California Tribes Fish Study report (Shilling et al. 2014) into the FTC equation shown above. In this study, participants were recruited and interviewed across California in tribal offices or at tribal or inter-tribal events from May, 2013 to June, 2014. A strength of the study is its community-based participatory research (CBPR) approach, i.e., tribes identified the need to collect tribe- specific information about fish use, and questionnaires and field methods were developed in collaboration with tribes. Despite the CBPR approach, only 24 of 147 tribes (federally- and state-recognized except for one) participated in the project (16%). A variety of reasons for non-participation were provided, but there was no in-depth discussion of how this may have impacted the generalizability of the findings, both in terms of geographic representativeness of the participating tribes (although figures were provided) and whether factors related to tribal non-participation may be correlated with actual fish consumption. An additional uncertainty about the generalizability of the data is that participants were recruited using non-random sampling methods. While obtaining a random sample is difficult in epidemiologic surveys, volunteers may be non-representative of the target populations (i.e. participating tribes) which may result in biased fish intake estimates if factors that are related to volunteering are also related to fish consumption. It is believed that incidentally a random sample of each tribe was obtained, but no data were provided to support this statement. More discussion of participation bias, at the tribal level as well as the individual level (e.g. some tribes are only represented in the study by one participant), would provide a better understanding of any uncertainty associated with the fish intake data. This appears to be potentially important because Figures 2, 7 and 8, for example,

show that the number of types of aquatic organisms and the number of places as fish sources increase with an increasing number of participants interviewed.

RESPONSE TO EVW 8

Agree- Including more discussion on bias could improve the report. However, the Water Boards are not the authors of that report. The Staff Report acknowledges that the study only surveyed a portion of the tribes in California. This was repeated in the discussion on the water quality objective for tribal subsistence and subsistence fishing, in Section 6.5.

“The survey includes 40 California tribes, while there are more than 100 federally recognized tribes in California and many others (see Section 4.10).”

Discussion on the generalizability of the data to all tribes would be fairly speculative and difficult to determine. Discussion on the biases / uncertainties from the study has been added included in the Staff Report. See also Comment EVW 11.

COMMENT EVW 9

In addition to collecting information about traditional fish use, thirty-day recall of fish intake was collected for contemporary use which allows for direct comparison with estimates obtained in the other surveys used in the draft report for estimating the FTC. The coding of narrative responses is not described in detail in terms of both methodology (e.g. groupings established *a priori*?) and findings. As in previous studies, the 95th percentile was emphasized as a value that would protect most users. The 99th percentile was also reported though inherently this does not protect all users (only the maximum value would do so), which seems to be the intended use of this value. The mean use rate was not reported because this is not being used in regulatory policies; however, by presenting the mean and median, amongst others, a better understanding of the distribution of the data would have been achieved. Given the lack of information about this distribution, it would have been especially helpful to report the sample size (i.e. the number of respondents) upon which the data in Table 6 of the Shilling report are based, because those data (142 grams per day) are the basis for the tribal subsistence water quality objective and upper percentiles may be sensitive to small sample size.

RESPONSE TO EVW 9

Agree- Including the mean fish consumption rate would aid in understanding the distribution of the data better. However, the Water Boards are not the authors of that report.

The study author is correct that the 95th percentile is a value that would protect most users. When a 95th or a 99th percentile is used for population estimates, the goal is not literally to exclude 1 to 5 percent of the population. These estimates are often used because of the difficulty of accurately calculating a 100th percentile (a maximum value) from a limited subsample. Therefore, high end estimates are generally used (e.g. 95th, 99th percentiles) to protect the whole population.

Yes, sample size should have been reported in the tables with the 95th percentile (Table 6), but the sample size was reported earlier in the report. This information is also reported in the summary of fish consumption studies (Appendix G) included in the Staff Report.

COMMENT EVW 10

Though traditional fish consumption is not a primary variable, it would be helpful to clarify the frequencies reported (page 14 of Shilling et al. 2014) as it appears that there are missing categories (e.g. 2-3 times/month and 4-6 times/week).

RESPONSE TO EVW 10

Agree- However, the Water Boards are not the authors of that report.

COMMENT EVW 11

The research described in the Shilling report does a commendable job of addressing the study goals. However, unlike the SFEI 2000 report, its discussion and conclusion section does not provide a comprehensive discussion of the extent to which the fish consumption estimates could have been influenced by various study limitations. The draft staff water quality objective report would benefit from including such a discussion to provide a sense of uncertainty in the fish intake estimate used.

RESPONSE TO EVW 11

Agree. This study provided information beyond our expectations and the authors are to be commended for that. New text was added to the end of Section 4.9 of the Staff Report (second to last paragraph) about this study to describe some of the uncertainties/biases. This new text follows the discussion on the uncertainty in estimates used for recreational fishing, and the difficulties in deriving a rate for subsistence fishers in general:

“To derive a numeric water quality objective for the Tribal Subsistence Fishing (T-SUB) beneficial use, however, the California Tribes Fish-Use study (Tribes Fish Use study) provides a significant summary of statewide fish consumption by California tribes (Shilling et al. 2014). While the Tribes Fish Use study includes data from 40 tribes throughout the state, the study cannot be assumed to represent every tribe, since there are many other tribes in California. There are 109 tribes that are recognized by the federal government and 72 more communities are petitioning for recognition (California Environmental Protection Agency 2009). This study was somewhat unique in that study participants were volunteers, which may result in biased fish intake estimates. One obvious source of bias could be that people who eat large amounts are more motivated to participate in the study. However, the study authors list reasons why some tribe members would not participate, including resistance to governmental intrusion,

and knowledge of past failure of government to act to protect tribal interests (Shilling et al. 2014). These may be more significant for a person for whom fish use is very important (and frequently eats fish), resulting in underrepresentation of those who eat large amounts of fish. The effects of various sources of bias are complex and difficult to predict. Nevertheless, the rate of 142 g/day for contemporary fish consumption for California tribes found by Shilling matches the US. EPA recommended subsistence rate of 142 g/day (U.S. EPA 2002).

COMMENT EVW 12

As discussed above (see 1.), use of a calendar year averaging period seems reasonable but could be better supported with additional references and/or data if available.

RESPONSE TO EVW 12

See response to Comment EVW 7.

The consumption rate of 4 to 5 meals per week (142 grams per day) is a sound basis from which to derive a subsistence fishing water quality objective that would be applied to the highest trophic level fish.

COMMENT EVW 13

To derive the Subsistence Fishing Water Quality Objective, the draft report incorporates the fish intake value of 142 grams per day as recommended by U.S. EPA (2000); it appears that this value is based on analysis of the 1994-1996 Continuing Survey of Food Intakes by Individuals (CSFII) and uses the 99th percentile of freshwater/estuarine uncooked fish consumption. When the 1998 CSFII data are included, the value 99th percentile value is similar at 143 grams per day (see U.S. EPA 2002, page 5-6). The CSFII was an annual survey conducted by the United States Department of Agriculture obtained survey estimates of food consumption from nationally-representative samples of non-institutionalized U.S. individuals, using an approach to sampling design and use of survey weights that is similar to other federal government surveys (e.g. National Health and Nutrition Examination Survey). CSFII response rates varied from 75.9% in 1996 to 81.7% in 1998 which are acceptable, and non-response was accounted for in survey weights. Average daily fish consumption data were collected for two non- consecutive 24-hr days, which is a different scale than the 30-day period used in the studies discussed above and may have resulted in lower precision of the estimated daily average consumption. However, the CSFII survey methodology appears to be scientifically sound and should have resulted in reasonable estimates of fish intake *at the time the surveys were conducted* (also emphasized on the USDA website: <https://www.ars.usda.gov/northeast-area/beltsville-md/beltsville-human-nutrition-research-center/food-surveys-research-group/docs/past-surveys/>). It should be noted that subsequent trends in fish consumption rates in response to health advisories regarding fish intake (e.g. Oken et al. 2003, Rehm et al. 2016)

may have impacted the extent to which the CSFII fish consumption estimates are representative of current fish intake in the general adult population and subsistence anglers.

RESPONSE TO EVW 13

See response to Comment EVW 6.

COMMENT EVW 14

Because it is difficult to define and identify subsistence fishing population, the 99th percentile of uncooked freshwater fish consumption estimate in the CSFII survey was used as a somewhat arbitrary cut point (the 95th percentile is 50 grams per day). This percentile is different from U.S. EPA's recommendation to use the CSFII 90th percentile for general adult population and sport fishers, from the 95th percentile in SFEI 2000 report for sport anglers, and from the 95th percentile of the SFEI 2014 study for tribal subsistence fishers. Nevertheless, the value of 142 grams per day used for the subsistence fishing water quality objective is the same as that derived for Tribal Subsistence Fishing in Schilling et al. 2014 (see above) which gives confidence that this is a reasonable estimate to use for human health protection of subsistence fishing populations and it provides consistency across beneficial use types.

RESPONSE TO EVW 14

The reviewer's agreement with the approach is noted, as well as the reviewer's concerns on the difficulty of defining and identifying subsistence fishing populations. This requirement has been modified in manner that matches some of the reviewer's (and other reviewers') concerns. A different approach is now recommended to better address the variability and uncertainty in establishing one subsistence fish consumption rate. In Section 6.5 of the Staff Report, Option 6 is now recommended, which is the narrative water quality objective. Previously, a numeric water quality objective was recommended. A narrative water quality objective has the advantage of allowing permit specific implementation. A site-specific fish consumption rate could be used to implement the water quality objective or the provided default fish consumption rate (142 g/ day) could be used to implement the water quality objective. See also Comment MWB 17 for other advantages, and MBS 8.

COMMENT EVW 15

As stated above, use of a calendar year averaging period appears reasonable but could be better supported with references and/or data if available.

RESPONSE TO EVW 15

See response to Comment EVW 7.

Staff thanks all reviewers for their comments.

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