STATE OF TENNESSEE
DEPARTMENT OF ENVIRONMENT AND CONSERVATION
Division of Remediation, Oak Ridge Office
761 Emory Valley Road
Oak Ridge, Tennessee 37830

November 22, 2024

Mr. Roger Petrie
Oak Ridge Office of Environmental Management
U.S. Department of Energy
Post Office Box 2001
Oak Ridge, Tennessee 37831

Dear Mr. Petrie

TDEC Comment Letter for Bear Creek Valley Mercury Sources Remedial Site Evaluation for the U.S. Department of Energy Oak Ridge Site Oak Ridge, Tennessee (DOE/OR/01-2977&D1)

The Tennessee Department of Environment and Conservation (TDEC), Division of Remediation-Oak Ridge Office, received the above referenced submittal on September 6, 2024. The document has been reviewed pursuant to the Federal Facility Agreement for the Oak Ridge Reservation. The following paragraph and comments are relevant to the review of this document.

TDEC does **not** believe this Remedial Site Evaluation (RSE) meets the requirements of the Mercury Management Approach "*to evaluate mercury methylation in Bear Creek*" as outlined in the Environmental Management Disposal Facility (EMDF) Record of Decision (ROD) (DOE/OR/01-2794&D2) and does **not** accept the conclusion that no further action is needed to address methylmercury in the Bear Creek watershed.

General Comments

1. The Mercury Management Approach section of the EMDF ROD states, "Unless the conclusion in the RSE accepted by all parties is for no further action, the RSE shall lead to other milestones for removal or remedial actions, including developing the substantive equivalent to developing load allocations and waste load allocations under 40 CFR 130.7(c)(2) and 130.2(g)(h) and (i)." TDEC does not believe this RSE meets the requirements of the Mercury Management Approach as outlined in the EMDF ROD and does not accept the conclusion that no further action is needed. Please explain how mercury methylation will be evaluated in accordance with the goal as defined by the EMDF ROD considering the following issues:

a. The goal of the RSE and defined in the EMDF ROD was to "*evaluate mercury methylation in Bear Creek*" which includes but is not limited to looking for sources of methylmercury. For example, the first sentence of Section 1.1 (p. 1-1) in the D1 RSE states the RSE objective is to *evaluate potential sources of mercury and methylmercury* in the BCV watershed in accordance with the EMDF ROD. However, the EMDF ROD (p. 2-64, Paragraph #4) states DOE shall conduct a RSE to *evaluate mercury methylation* in Bear Creek and conduct pilot or treatability studies as needed. The italicized words highlight an important difference between the objectives of this RSE and the EMDF ROD.
b. The U.S. Department of Energy's (DOE) response to TDEC Comment 2 on the *Bear Creek Valley Mercury Sources Remedial Site Evaluation Sampling and Analysis Plan, Oak Ridge, Tennessee* (DOE/OR/01-2958&D1) states an objective of the RSE is to obtain data from hydrologic settings that may contribute to mercury methylation. It also states, "parameters will be collected that may contribute to a better understanding of mercury methylation…." The methylation objective is also included as a data quality objective (DQO) in Section 3.2 (p. 11). However, the report presents no information produced by the RSE regarding the mechanisms for mercury methylation.
c. If a conclusion of this RSE is that the concentration of mercury in fish tissue has decreased below the maximum allowable 0.3 µg/g criterion, then at a minimum the environmental factors that have contributed to the decrease in methylmercury must be evaluated to verify the levels of methylmercury do not cause future exceedances. Identifying the mechanisms of mercury methylation should then precede an evaluation of potential technologies that can be used to either disrupt or lessen the effectiveness of the methylation in the creek and ensure long-term protectiveness. For example, given DOE's findings that mercury methylation potential varies by organic matter type, evaluating the potential impacts on methylation from different organic matter types may help inform future remedial actions.
d. Based on the RSE's lack of information about methylation, it is not yet clear what additional actions might be warranted, nor is it clear how to develop the substantive equivalent of developing load allocations and waste load allocations.

2. Please include a discussion of efforts conducted or planned for identifying mechanisms of mercury methylation and/or ways in which the methylation of mercury in Bear Creek could be reduced.
3. According to the U.S. Environmental Agency (EPA) guidance, to properly evaluate trends in bioaccumulation and make decisions regarding risks associated with fish consumption, the size of the fish sampled should be both consumable and consistent with past data collection efforts. DOE should not use data from fish that are abnormally small for decision making, unless additional effort is made to account for the small size such as allowing proper time for population recovery and/or modeling of

bioaccumulation. In 2021 when the mercury strategy was developed and placed in the EMDF ROD, the mercury levels in fish tissue were trending steeply downward and projected to continue downward. Instead, the data has been variable as shown in Figure 4.10:

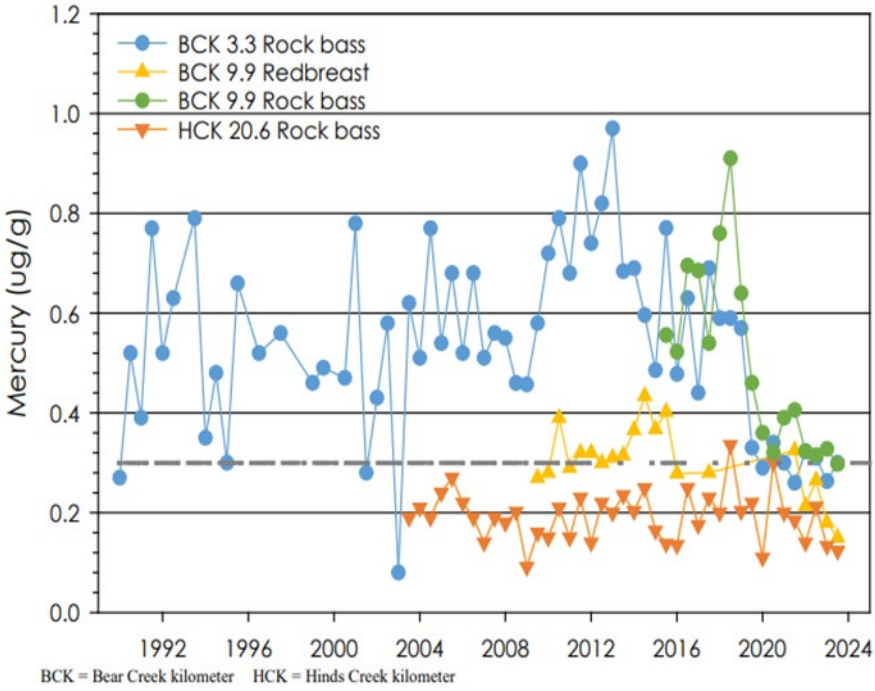

Figure 4.10. Average concentrations of mercury in Bear Creek fish.

It is very possible that, if not for the small fish sizes, average fish tissue concentrations would be significantly above 0.3 µg/g, as acknowledged in Section 5.1 in the discussion of the most recent sampling. Please include some discussion showing how the size of the fish sampled as part of this RSE effort compares to the size of the fish sampled in previous data collection efforts and provide an explanation for how the size of the fish will be accounted for while determining trends in tissue concentrations. DOE should consider non-lethal sampling of fish tissue to preserve an adequate population.

4. Given both that the last paragraph of **Section 5.1** of the RSE states that mercury contamination is "widespread", and the mercury speciation studies indicate organically-bound mercury dominates much of the Bear Creek Valley watershed, especially in soils, addressing areas with low-level mercury sources may be necessary to effect a change on fish tissue in the watershed. For example, even though Bear Creek mercury is orders of magnitude lower than the remediation goal of 400ppm for Lower East Fork Poplar Creek (LEFPC) (as referenced for comparison in presentations made at multiple project team meetings), LEFPC is characterized by mercury fractions that are less readily available to absorb and bioaccumulate than Bear Creek. These differences suggest that

actions targeting lower-level sources may be necessary to address methylmercury cycling in the food chain and restore Bear Creek to meet the recreational use designation, per the goal stated in the EMDF ROD Mercury Management Approach.

5. Data presented in this RSE report (including site description in Section 1.2.1) support the hypothesis that shallow groundwater/known contaminant plumes in the subsurface are potential sources of mercury and/or methylation impacting Bear Creek that should be investigated. TDEC requested that shallow groundwater be considered as a source of mercury, methylmercury, or mercury methylation in DQO meetings and in written comments on the related Mercury RSE Sampling and Analysis Plan (SAP) (Appendix A). While typical hyporheic zone/pushpoint sampling may not be appropriate given conditions in Bear Creek (as discussed in the Bear Creek Valley (BCV) Mercury Sources RSE SAP response to comments), TDEC is again recommending that shallow groundwater be investigated as a potential source that could be addressed to positively impact Bear Creek water quality.
6. TDEC recommends additional cooperation with Oak Ridge National Laboratory's Environmental Sciences Division to evaluate innovative strategies to mitigate Hg impacts and support the restoration of Bear Creek.
7. DOE should not use the conclusions of this RSE as a basis for decision-making, and there is not a consensus on future actions related to the water treatment and discharge at EMDF. One sampling event that is atypical due to small fish sizes is not a trend, and the variability of concentrations dictates that a longer period of observation is necessary. The data needed to support the removal of Bear Creek from the 303(d) list would have to be much more than what has been presented in this RSE. In order to maintain compliance with the fish tissue standard, if achieved, the RSE would also need to explain the mechanisms of methylation.

Specific Comments

1) **Section 1.2.1.2, Page 1-7, second paragraph** – Revise the wording in the final sentence of this paragraph that characterizes mercury concentrations in fish tissue as *occasionally* exceeding the ambient water quality criteria (AWQC). Multiple types of fish have exhibited methylmercury levels in tissue regularly exceeding the AWQC level for the past couple decades.
2) **Section 1.2.2, Page 1-8, last paragraph** – This section describes the issues with mercury at the Storage Yard-200 (SY-200) area and states, "free mercury was observed in some of the borings…." This text does not align with the conclusions of the report that the RSE did not identify a source of mercury warranting active remediation. Please add an explanation for why the report did not identify the SY-200 area as a potential source of mercury to Bear Creek.
3) **Section 2.1.3, Page 2-7** – Please include additional discussion on the width of the floodplain around Bear Creek, the lateral extent of floodplain samples, and whether any

biased samples were collected from identified depositional areas. Please explain why the floodplain soils further from the creek were excluded from consideration as a potential source of mercury to surface water.

4) **Section 2.1.3, Page 2-7, second paragraph** – The first sentence states soils samples were collected from "either" side of Bear Creek, should this say that samples were collected from "each" side of Bear Creek? Please provide additional details regarding sample volumes and how samples were collected and composited.
5) **Section 3.3, Table 3.9 and Figures 3.11 through 3.22** – Several locations where sequential extraction results are presented in Table 3.9 show a cumulative percentage significantly greater than 100%. For example, floodplain soil collected from the Hind's Creek Transect Reference (HCTREF) location has a cumulative percent that adds up to 128%. The graphs in this section normalize the data from each location to 100%. This makes it seem as though the HCREF location had significantly less organically bound mercury (F3) when in fact the percent reported in the table for F3 at HCTREF was 62.1%, which is within the range of F3 percentages measured at the BCV transects. Please verify the data reported is accurate and, if so, please explain what errors in the mercury fraction calculations could result in a sum that exceeds the total amount of mercury.
6) **Section 4.1** – Spring SS-5 is identified as a source of significant Hg flux, data in Oak Ridge Environmental Information System indicate that Spring SS-4 exhibits flux of Hg and MeHg comparable to SS-5, and Spring SS-6 also sees a steady flux of Hg. Given the multiple groundwater/surface water connections in Bear Creek, these data suggest that groundwater may be a source of mercury and methylmercury into Bear Creek. Please explain how the Hg fluxes from groundwater along Bear Creek will be evaluated.
7) **Section 4.1, Page 4-2** – The text here states "higher flow rates cause increased sediment transport, and with the strong particle retention of mercury, its mass transport can be greatly increased during high flow events". Provided the recognition that high flow events can greatly increase the short-term particle transport and mercury discharge to the stream, please explain how the current sampling strategy is sufficient when the time elapsed since last significant rain event prior to sampling has averaged from between 3 and 24 days as presented in **Table 1** of the *Bear Creek Special Studies Report 2021*.
8) **Section 4.1, Page 4-7, fourth paragraph** – The text here emphasizes the lack of dissolved mercury in the surface water "an average of 27% (+/- 5%) of mercury in surface water was dissolved, with the remaining 73% being associated with filterable solids". This dominance of particle-bound mercury indicates that the majority of mercury in the system is available to filter feeding organisms in the food web supporting the trophic transfer of mercury to larger organisms and the cycling of methylmercury in the system. Please provide an explanation for how the cycling and transfer of methylmercury was or will be evaluated.
9) **Section 4.1, Page 4-7** – The text in multiple areas on this page relates mercury in Bear Creek to the S-3 Ponds including the last paragraph of the section that refers to "the known source associated with the S-3 Ponds area." Please provide an explanation for

how the conclusion of the report was that no sources of mercury were identified in the context of a known source in the S-3 Ponds Area.

10) **Section 4.2, Section 5.1, Figure 4.10** – It is stated here "fish fillet concentrations were approaching and dropping below the EPA-recommended fish-based AWQC of 0.3 µg/g". As per the guidance document EPA-823-R-01-001 referenced in the EMDF ROD, this EPA-recommended value of 0.3 µg/g is a not-to-exceed criterion and is therefore inappropriate to be used to compare to average fish tissue concentrations. While recent fish tissue data from Bear Creek indicate tissue concentrations of mercury/methylmercury have decreased and are hovering around the EPA criterion *on average*, these data sets continue to include samples with concentrations above the EPA tissue criterion. Also, as indicated in this RSE, populations of rock bass and redbreast sunfish were significantly impacted/overharvested as a result of the intensive sampling effort in 2021 and are not representative of normal conditions. Therefore, these fish tissue data are likely not appropriate for supporting delisting of Bear Creek from the CWA 303(d) list, per the EMDF ROD Mercury Management Approach. Please explain how the size of the fish will be normalized to allow for accurate comparisons to previous sampling events and trends. Please include details on maximum fish tissue concentrations and how many fish tissue samples exceeded the AWQC criterion. Please add distribution boxes to Figure 4.10 similar to Figure 2.19 in the 2024 Remediation Effectiveness Report (DOE/OR/01-2960).
11) **Section 3.4, page 3-41, Table 3.10** - It is noted there is a distinct difference in partitioning of the forms of mercury in the sequential extraction data between Bear Creek and Lower East Fork Poplar Creek, and that this partitioning may be a key factor in the unsolved mystery of why the conversion of mercury into methylmercury is much more efficient in the Bear Creek watershed than in East Fork Poplar Creek. This should form the basis of future research opportunities.
12) **Section 3.2.2, page 3-13: Table 3.7 p. 3-19** - Given the earlier general comments and the difference noted in specific comment #11, DOE should consider the idea of removal of the relatively high concentration creek bank soils that have been identified in the general vicinity upstream:

> "Mercury concentrations in the sampled BCV soils and channel sediment are comparatively low. (Note the laboratory reported mercury and methylmercury concentrations in soil and sediment in units of µg/kg, equivalent to parts per billion [ppb]). The maximum measured mercury concentration in floodplain soil was 3500 µg/kg at BCT12A, upstream of the NT-3 confluence with Bear Creek. The maximum measured mercury concentration in creek bank soil was 7100 µg/kg at BCT13, located near the former HCDA entrance. The maximum measured mercury concentration in channel sediment was 530 µg/kg at BCK12A, upstream of the confluence of NT-3 with Bear Creek and downstream of the HCDA."

Because of the unique nature of the mercury partitioning in Bear Creek and the efficiency of methylation in the watershed, DOE should consider that the levels for triggering removal of source material may require a lower threshold than in East Fork Poplar Creek.

13) **Section 5.1, page 5.2** – The last sentence of this section concludes there is not a source of mercury that would warrant active remediation, but as identified in comments above, multiple statements throughout the report seem contradictory to this conclusion including known sources near the upper reaches of the creek and statements that mercury is being derived from the "widespread" contamination across the creek bank and floodplain soils. Please provide additional supporting detail on how the conclusion of the RSE is for no remedial action, and explain what further plans DOE has to study mercury flux to the creek, investigate the methylation occurring the creek ecosystem, and evaluate potential remedial actions for source removal, stabilization, etc.

Review of this document meets the review cycle protocol of 90 days. Questions or comments concerning the contents of this letter should be directed to David Carlone at the above address or by phone at (865) 839-3362.

Sincerely

Randy Young

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