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Confederated Tribes and Bands
of the Yakama Nation

Established by the
Treaty of June 9, 1855

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Subject: Review of the *Data Summary Report for the Remedial Investigation of the Hanford Site Releases to the Columbia River, Hanford Site, Washington* (WCH-398)

Dear Mr. McCormick, Mr. Faulk, and Ms. Hedges:

The Yakama Nation ERWM Program appreciates the opportunity to have reviewed the *Data Summary Report for the Remedial Investigation of the Hanford Site Releases to the Columbia River* (WCH-398), and offers the attached comments regarding data quality and other issues. Although no formal comment period was offered, we recommend that Tri-Parties consider how the issues raised here by the Yakama Nation might be addressed in the upcoming Columbia River Remedial Investigation and River Corridor Risk Assessments.

The Yakama Nation previously submitted comments on the Columbia River Remedial Investigation Work Plan (DOE/RL-2008-11) and the associated Sampling and Analysis Plans (DOE/RL-2008-11 Appendix A, WCH-286). Overall, the Data Summary Report (DSR) indicates that the Yakama Nation's previous comments and outstanding issues have not been addressed.

Review of the data presented in the DSR raises several questions regarding data quality and usability. Based on sampling and analytical deficiencies we have identified, the data presented in the DSR do not appear to be suitably representative, or of sufficient quality to properly assess the risks posed by the Hanford Site and Hanford-associated activities to the Columbia River. It appears from this DSR that the characterization of the river is inadequate, that the background sample locations are inappropriate such that associated data from these "reference" areas are not comparable to site data, and that issues related to data quality raise questions as to data usability.

We recommend that existing data be re-evaluated, and that additional data be collected and incorporated to ensure that contamination associated with the Hanford Site will be accounted for in the Remedial Investigation. Without reliable and sufficient information, how will cleanup decisions be made?

Sincerely,

Russell Jim
ER/WM Projects Manager

Cc:

RHW Committee
Phil Rigdon
Administrative Record

Attachment

Yakama Nation Comments on the *Data Summary Report for the Remedial Investigation of the Hanford Site Releases to the Columbia River, Hanford Site, Washington* (WCH-398)

The Yakama Nation has reviewed the January 2011 *Data Summary Report for the Remedial Investigation of the Hanford Site Releases to the Columbia River, Rev. 0* (WCH-398), and offers the following comments regarding data quality and other issues.

The Yakama Nation previously submitted comments on the *Columbia River Remedial Investigation Work Plan* (DOE/RL-2008-11) and the associated Sampling and Analysis Plans (DOE/RL-2008-11 Appendix A, WCH-286). Overall, the Data Summary Report (DSR) indicates that the Yakama Nation's previous comments have not been addressed. Many previous comments and outstanding issues that were not suitably addressed in the final revision of the Work Plan, data collection efforts, or subsequent DSR are reiterated in this memorandum.

Review of the data presented in the DSR raises several questions regarding data quality and usability. In particular: 1) there are inconsistencies with primary and duplicate uranium analyses; 2) contaminant concentrations in "reference" areas are frequently higher than those measured in the Hanford Reach and downriver, indicating unsuitable reference site selection; 3) many analytical results for onsite samples have very high detection limits, resulting in measurements of little value since screening levels and expected concentrations are lower than the detection limits; 4) sample coverage is inadequate for the comparatively large study area; and 5) many samples were collected in proximate groups, further limiting the sample distribution and design to accurately represent the area of interest.

Based on sampling and analytical deficiencies identified in this memorandum, the data presented in the DSR do not appear to be suitably representative or of sufficient quality to properly assess the risks posed by the Hanford Site and Hanford-associated activities to the Columbia River. We recommend that existing data be re-evaluated and that additional data be collected and incorporated to ensure that contamination associated with the Hanford Site will be accounted for in the Remedial Investigation.

HOW DOE ADDRESSED PREVIOUS YAKAMA NATION COMMENTS ON COLUMBIA RIVER SAMPLING DESIGN

The Yakama Nation ERWM Program submitted extensive comments on the Work Plan (DOE/RL-2008-11) released by DOE in 2008. A summary of the comments that applied to the data collection elements of the Work Plan is provided below followed by a discussion of how these comments were or were not addressed during the sampling and analysis effort (based on information presented in the DSR). Because the DSR is intended only to present the data collected under the guidance of the Work Plan, comments intended to address other issues such as remedial responses and future risk assessments are not addressed in detail here.

Comments submitted by the Yakama Nation regarding the Work Plan included the following main points:

- The Work Plan failed to follow EPA guidance for the Remedial Investigation and Feasibility Study process.
- The Work Plan did not address data gaps in the overall understanding of the site, planned data collection did not attempt to further refine the nature and extent of contamination, and deposition from air emissions was not considered when sampling was planned.
- Pathways for contaminant migration were not considered during planning of sampling and analysis; as a result, many questions regarding where and when contamination from the 100, 200 and 300 Areas will reach the Columbia River could not be adequately answered by the data collected.
- Insufficient biological characterization was planned.
- Background and reference locations that were selected for comparison with the Hanford Site were not suitable for that purpose.
- The list of analytes selected for data evaluation was not complete, and did not include high-risk radionuclides, such as iodine-129, and organic compounds, such as carbon tetrachloride.
- Data collected under the Work Plan are not sufficient to perform comprehensive ecological or human health risk assessments for the River component of the site.

Review of the DSR indicates that these comments were not adequately addressed in the final revision of the Work Plan, and thus not implemented in the subsequent sampling and analysis activities. The data collection effort as presented in the DSR included only some of the known or suspected sources of contamination or affected media. With the exception of pore water, the program selected and then sampled media that met specific physical or spatial characteristics with only marginal consideration for the potential for contamination. Previous comments related to sample collection and analysis that were not addressed are discussed below.

DOE is deviating from CERCLA by not planning to issue a ROD for the Columbia River. The Yakama Nation commented previously that no Record of Decision is scheduled for the Columbia River component of the site, and that the Work Plan appeared to be geared toward superficial studies that would justify "no further action" in this portion of the Hanford Site. The DSR implicitly acknowledges that this may have been one of the primary objectives of the data collection effort stating that "In general, the findings of the remedial investigation are consistent with the current site understanding." Cleanup decisions, including whether to issue a ROD for the river portion of the Hanford Site, cannot be made until adequate characterization data are evaluated and potential risks are assessed.

The nature and extent of contamination has not been fully characterized. Sample collection in the Hanford Reach was limited to depths greater than 6 feet and did not include the near shore aquatic zone that was previously characterized in the River Corridor Baseline Risk Assessment. The Yakama Nation previously stated that the assessment of the Columbia River must include the entire corridor including the near shore and riparian aquatic zones. Existing data from the

riparian and near shore areas must also be included in the Columbia River Remedial Investigation; otherwise the nature and extent of contamination in the river cannot be evaluated holistically.

Identifying the furthest reaches of Hanford-related contamination -- the full nature and extent -- was not addressed by the sampling performed and data presented in the DSR. Most samples were collected from the Hanford Reach of the Columbia River, the upriver area, and in some cases Lake Wallula. The only sampling performed downstream of Lake Wallula was collection of two sediment cores from the Bonneville Pool. Only 20 sediment cores were collected from the Columbia River (14 shallow, 6 deep) between the Priest Rapids Pool and the Bonneville Pool. The sampling design also appears to deliberately avoid potentially contaminated sites (for example reactor *intake* structures were sampled for sediment, but *outlets* were not, even though the latter transported contaminated cooling water to the river; see DSR Appendix L Figures L-7 and L-10).

The Yakama Nation also identified deficiencies in the biological characterization that was proposed in the Work Plan, and pointed out that the resulting data would not support the development of a complete Baseline Risk Assessment. Previous comments repeatedly noted that DOE should consider the total extent of contamination across all media types, including plants (particularly those that are culturally or ecologically significant), animals, and a wide range of fish. The final Work Plan and resulting data included tissue from only six species of fish: bass, carp, sturgeon, sucker, walleye and whitefish. Yakama Nation comments regarding expanding the list of analytes were not addressed by DOE.

Background sample locations are not appropriate. The background or upriver area sampled for comparison to contaminated site samples was identified as the area from River Mile 440 downstream to River Mile 338, where the Vernita Bridge crosses the river channel. The Yakama Nation previously commented in detail regarding the proposed background sampling locations identified in the Work Plan as unsuitable for that purpose. The concentrations of chemicals and radionuclides from soils and sediments in this area are already greater than statewide background concentrations and EPA preliminary remediation goals. We also had noted that invertebrate samples from the upriver area had higher concentrations of several metals, including uranium, than those observed in biota collected in the 100-B/C Area (Yakama Nation, 2008). More recent evaluation of DOE data from the Vernita Bridge and upriver also found fault with using the area for reference or background measurements, as stated in a Yakama Nation comment letter to DOE on the River Corridor Baseline Risk Assessment (Yakama Nation, 2011):

> *...average ratios of "Reference Area" doses for terrestrial pathways (180 millirem per year) to the doses predicted for local and broad area pathways in the River Corridor Baseline Risk Assessment's Tribal scenario was about 4.3. In both cases this was due, in large part, to high americium-241 levels measured in soil and fish....To use "Reference Area" doses that are dominated by man-made radionuclides of Hanford origin gives an incorrect picture of relative contamination in the 100 and 300 Areas since it makes it appear in many cases that the latter areas have been remediated below some reference or background level. This is like comparing two populations with excess cancers but designating one of them as a reference, unexposed group. This approach is incorrect.*

The problems associated with the selected reference area were not addressed in the final Work Plan or the data collected and presented in the DSR, and there appears to be substantial evidence supporting the hypothesis that the reference area is contaminated. Island soil, shallow sediment, and sediment core samples collected upriver of Priest Rapids dam provided maximum concentrations or notably higher average values for most metals than samples collected downriver. Because an appropriate reference site should "match the Superfund site in all aspects except contamination" (USEPA, 1994), consistently higher values for contaminant metals and radionuclides measured in the reference area indicate that either the area selected does not adequately match the Hanford Site characteristics, or has been contaminated and retained more residual contamination than locations sampled within the Hanford Site. We propose that alternate background sites or methods be identified for comparison to Hanford Site data.

REVIEW OF DSR DATA QUALITY AND DATA USES

DOE provided a data quality review in a separate report, *Data Quality Assessment Report for the Remedial Investigation of Hanford Site Releases to the Columbia River, Hanford Site, Washington* (WCH-381), which was comprehensive but presents a somewhat mixed picture of the data quality (WCH, 2010). The quality assessment consisted of two main components. First, the data were reviewed internally to compare the performance of the 10 quality metrics listed below against the Quality Assurance Program Plan criteria. Second, in addition to the internal review of these 10 metrics, approximately 5% of the samples of each media type were fully validated by a third party. The conclusions of the external validation were not substantially different than those of the internal reviews. The basic metrics were:

- Holding times
- Method detection limits
- Surrogate recoveries
- Method blank results
- Laboratory control sample (LCS) results
- Matrix spike/matrix spike duplicate results
- Tracer results (for radionuclides)
- Laboratory duplicate results
- Field Quality Control sample results (e.g., field blanks and duplicates, equipment blanks, and trip blanks).

Overall, the DOE reviews found few major quality assurance (QA) problems, and the data associated with those problems were properly rejected or qualified. However, the reviews identified other QA problems that were associated with almost all of the data sets. These problems included the following: Many analyses that exceeded holding times and had high detection limits; poor recoveries of spikes, tracers, and surrogates; contamination in sample blanks; and poor replication, both in laboratory and field duplicates. While none of these problems individually are sufficient to reject the data, and the results were flagged appropriately, they raise an overall concern about the care that was taken in the analyses.

A few water and sediment samples were split and analyzed by both DOE and either EPA or Ecology. Similar to the results of the internal analyses of duplicate samples, there were some considerable differences in the results between the split samples. The report stated that for the majority of the split sample analyses either one or the other of the split results were non-detects, but provided no further explanation.

Some of the radionuclides and metals analyzed were detected in both of the split samples, and these results could be compared. While the concentrations for most the substances detected in both split samples agreed fairly well, there were some substantial differences. Differences in the concentrations of the radionuclides between the split samples were attributed to different analytical techniques, but it still raises the question of which data are more representative. These differences were not consistent, but were observed frequently enough to question the true concentrations of the contaminants. The ranges of the differences were generally on the order of as much as a factor of 10. For example the activity of thorium-230 in one surface water sample was reported as 0.123 pCi/L in one split sample and as 0.0136 pCi/L in the other split. Similarly, the activity of uranium-238 in a surface water sample was reported as 0.281 pCi/L in one split sample and as 0.089 in the other split. Greater differences were noted at lower concentrations, as might be expected for results measured near the detection limit where precision tends to decrease. The differences between the concentrations in the split samples appear to be similar to those observed in the internal laboratory and field duplicates.

Overall, DOE's QA review indicated that the majority of the data were analyzed correctly, but there are substantial reasons to question the precision, and possibly the accuracy of any specific sample result. Our review of the data identified a number of potential problems or irregularities that bring into question the suitability of this data set for the stated purposes, and the general usability of the data overall. These potential problems and irregularities include the following and are discussed in more detail below:

- Measured ratios of radioactivity attributable to different uranium isotopes do not follow expected relationships.
- Duplicate analyses are treated the same as primary analyses, which biases the data set.
- Duplicate samples were analyzed by different methods with significantly different detection limits from primary samples resulting in poor agreement between analyses.
- Duplicate analyses were apparently only performed for two radionuclides, and data validation information is not available for these results.
- Unusually high concentrations or higher average concentrations were measured in reference (upriver) areas and do not match trends for Hanford Reach samples.
- Sample area coverage was geographically limited and, in some cases, confounded identification of contamination from the Hanford Site versus other sources.
- Many analytes had detection limits that were too high to provide useful results, e.g. to compare to screening levels.

Uranium concentrations do not have plausible isotopic signatures. Uranium at the Hanford Site should have isotopic activity ratios similar to those of natural uranium. Uranium-234 and uranium-238 should each account for approximately 49 percent of the total radioactivity, and uranium-235 should account for the remaining 2 percent. Review of the data provided in the "DSRResults.accd" database that accompanied the DSR identified several irregularities. First, the three uranium isotopes were not all detected in all samples. Table 1 presents examples of the rates of detection. Uranium-235 was not detected in approximately 80% of the surface soil samples where uranium-238 was detected, and not detected in over 90% of the sediment samples where uranium-238 was detected. Based on the reported activities of uranium-238, most samples should have had activities of uranium-235 in the range of 0.03 to 0.1 pCi/g. In the relatively few samples in which uranium-235 was detected, the activities were within the expected range, indicating that the detection limit was not the primary source for poor detection of uranium-235. In addition, since U-234/U-238 ratios are generally in the expected range, a low activity of U-235 could not have resulted from depleted uranium conditions.

Table 1. Summary Statistics for Uranium Isotopes from Data Summary Report

	Soil Sample Detections	Percent of Total Measurements	Sediment Sample Detections	Percent of Total Measurements
Total U-233/234 Detections	79		498	
Total U-238 Detections	79		489	
Total U-235 Detections	16		30	
U-238 Detections without U-235 Detections	63	79.7%	459	93.9%
U-238 Detections without U-233/234 Detections	0	0%	13	3%
U-238 Detections within 5% of Expected Value	54	68.4%	255	52.1%
U-235 Detections within 2% of Expected Value	14	87.5%	24	80.0%

Similarly, comparisons of the U-238 activities to the total uranium activity in the same sample demonstrated that approximately 50-70% of the soil and sediment samples were within 5% of the expected ratio, meaning nearly half of the data presented in the DSR failed to meet this basic quality assurance metric. Where detected, the ratios of U-235 activity to total uranium activity in the soil and sediment samples showed similar discrepancies, but not as extensively as for U-238 (with at least 80% of the U-235 activities being within 2% of the expected value). An additional evaluation of the uranium data quality might have been possible by comparing measurements of total uranium radioactivity against the chemical measurements of total uranium. However, the procedure used for the chemical measurement of uranium had a high detection limit. All of the results for the measurement of uranium based on chemical methods, not radioactivity, were below the detection limits so that comparisons between the different analytical approaches for total uranium could not be made.

Duplicate sample analyses and uses are flawed. The results reported for duplicate measurements raise concern. Radioactivity measurements for U-235 and U-238 regularly show that the isotopes are not detected in the duplicate analysis, even when they are detected in the primary analysis. Generally, the non-detections are associated with duplicate analyses that were performed by a different analytical method than the primary analysis, a highly irregular approach. This issue is most pronounced for U-238, where there are many duplicate samples; however, it also appears in the U-235 results.

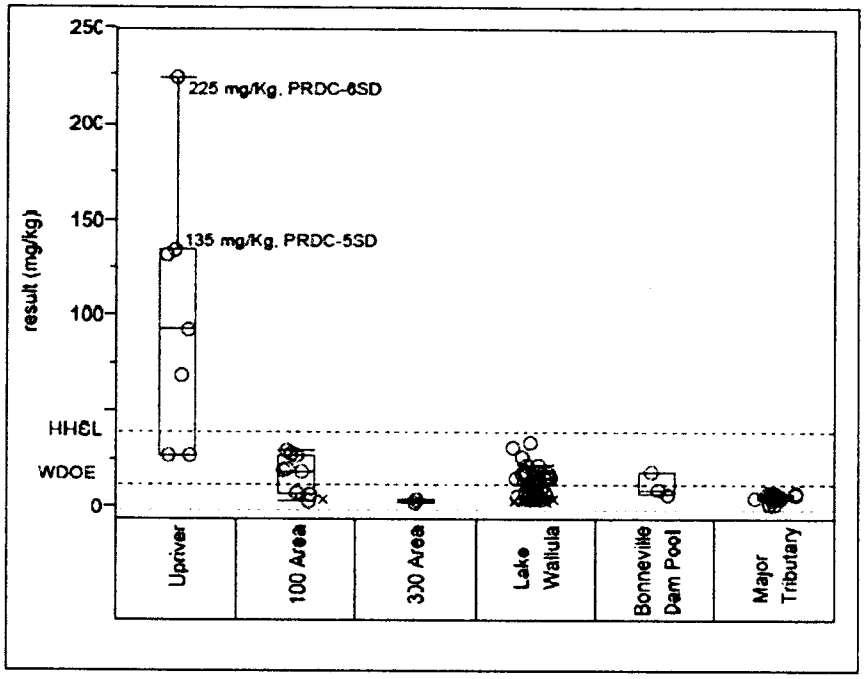
Section 4.0 *Analytical Results and Data Interpretation* of the DSR stated that "duplicate samples were also included in the data sets, and were treated as independent samples for the purposes of this evaluation" (p. 4-2). This treatment of duplicate data is biased and does not provide information representative of the site, and should not be used in future data evaluations or interpretations. This caution is particularly true given the near-systematic pattern of non-detections in the duplicate samples. These duplicate sample results, apparently based on less sensitive analytical procedures, should be qualified and used only when no other original measurement is available.

Finally, in the course of examining duplicate sample data for uranium, it was determined that no other duplicate analyses were included for *any* radionuclides other than U-235 and U-238. To achieve data quality objectives, all analytes should be analyzed in duplicate at an appropriate frequency.

Selected "reference" areas are not appropriate to compare to site data. As discussed in previous comments, a number of analytes, including americium-241 and uranium, were found to have higher average concentrations in media collected from the upriver area (upstream of Priest Rapids dam) than media collected in the Hanford Reach and downstream areas. Because the upriver area is used as the "reference" area and is intended to represent media unaffected by Hanford releases, these data are problematic. The upriver sediment samples were finer-grained than most of those collected downstream, which may account for some differences. In addition, these samples show patterns of contamination that are similar to the chemical signature of mining-related wastes known to have been released to the Columbia River upstream of the site (Majewski et al., 2003). Such discrepancies between the reference and downstream samples may seriously hinder their usefulness in determining releases from Hanford.

The boxplots provided in the DSR Appendix A also suggest that the limited number of samples collected in the upriver "reference" areas gives undue weight to single high measurements, resulting in higher average concentrations upstream than are observed in the Hanford Reach where significantly more data and greater variability are observed (see Figure 1). These high point measurements may again be related to grain-size differences or to offsite contamination; however, they are not representative of natural site conditions to which Hanford site samples should be compared.

Figure 1. Boxplot from the DSR Demonstrating Limited and Elevated Reference Values



Sample coverage was too selective and incomplete. While the total number of samples collected seems substantial, the area covered is large. In addition, samples were often collected proximate to each other rather than obtaining greater site coverage. As a result, the overall representativeness of the data is limited. For sediments, this issue is exacerbated by sediment sampling that was biased toward near shore samples, and many samples were collected from near or within marinas. For example, evaluation of selected data showed that high concentration of metals in the down-river Lake Wallula samples were almost all associated with marinas or boat launches, which are generally known to be local sources of such contamination. This confounds identification of the contaminant source.

Detection limits were too high. Many contaminants were not detected in water samples and fish tissue samples because of high detection limits (see Figure 2). In many cases, the detection limits are well above concentrations that would be expected in these media, as well as above the screening levels used to evaluate the results in the report. The data also show that detection limits varied greatly between samples. At least some of these results indicate that useful detection limits could have been achieved for all samples. It is a serious limitation that more sensitive analytical methods were not employed for these analyses, particularly since the concentration ranges could have been anticipated from previous sampling, and the sensitivities needed to reach the potential risk screening levels used by DOE in the DSR (Appendix C) were known.

Figure 2. Boxplots from the DSR Demonstrating High Detection Limits

Figure A-88. Boxplot of Dissolved Elemental Uranium in Surface Water.

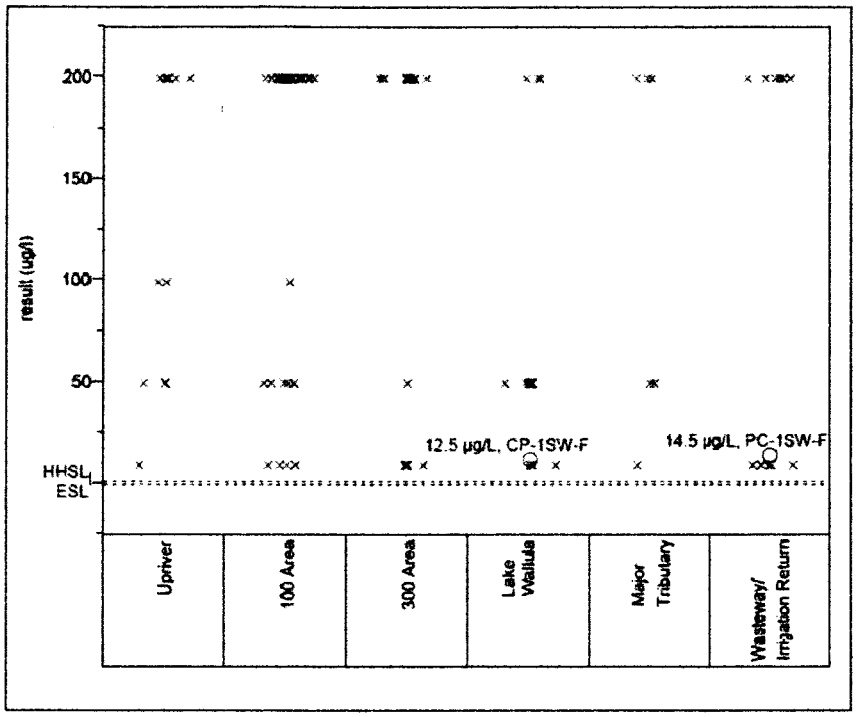
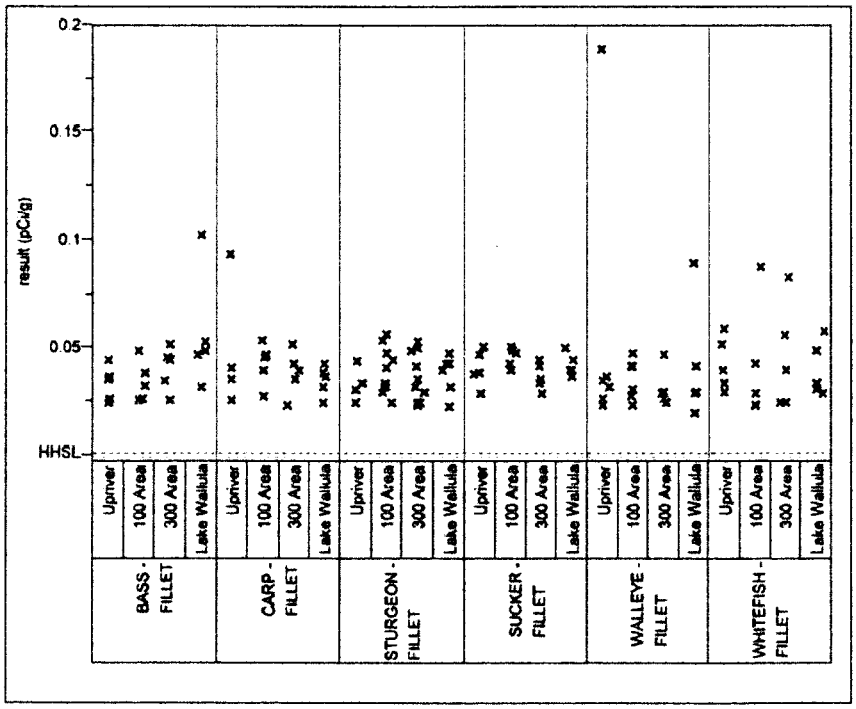


Figure B-33. Boxplot of Cesium-137 in Fish Fillet.



In the above boxplots, "x" symbols correspond to non-detects while "o" correspond to detections.

DOE could also provide additional verification of the utility of these data by comparing them to similar data collected for the River Corridor Remedial Investigation. Samples of the same media collected from spatially proximate locations could have comparable concentrations. The concentrations of substances in pore water collected from the same groundwater discharge area, for example, should be consistent with those data collected from the near shore as part of the Remedial Investigation and those collected farther off-shore during the river sampling.

The evaluations of the data presented in the report are of questionable value since the data are neither complete nor comprehensive. The concentrations of selected contaminants in the sediments were correlated to the percent fine material in the same sample; the relationship with the fine fraction is often useful for explaining the difference in concentrations among sediment samples. The report determined that few of the measured contaminants had a consistent relationship with sediment texture. This conclusion was based on simplistic analysis of the complete data set from the entire study area. Thus, samples from the "reference" area were not distinguished from samples in Lake Wallula, the Hanford Reach, or other areas of the Columbia River. However, closer scrutiny of the data and segregation of the samples into their respective river reaches, or in some cases smaller spatial units, yielded clear and consistent information. For example, inspection of spatially-segregated data identified a range of low concentrations of many of the metals consistent with a natural background, as well as a component in the upriver samples that may be related to mining-related source transport from Lake Roosevelt, upstream of the site. These more detailed analyses that respect the spatial relationship between the sample and analytical results can be used to isolate and identify potential inputs from the Hanford Site. Regrettably, it appears that such analyses were beyond the scope of what was accomplished, and the more limited analyses that were ultimately performed resulted in misleading and inaccurate conclusions that were presented in the DSR.

REFERENCES

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Yakama Nation ERWM Program. 2011. *Letter from Russell Jim to Matthew McCormick (DOE), Dennis Faulk (EPA), and Jane Hedges (Ecology), dated February 28, 2011: Comments on DOE/RL-2007-21 Volume II Draft C: River Corridor Baseline Risk Assessment, Human Health Risk Assessment, December 2010, Attachment 2 – Supplemental comments on RCBRA Draft A.* Confederated Tribes and Bands of the Yakama Nation, Environmental Restoration Waste Management Program.