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NSY PORTSMOUTH
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LETTER REGARDING U S NAVY RESPONSES TO U S EPA REGION I COMMENTS ON RADIOLOGICAL SAMPLING OF WATER, SEDIMENT AND BIOTA NSY PORTSMOUTH ME
1/9/2001
PORTSMOUTH NAVAL SHIPYARD

NAVSHIPYD PTSMH-5216/9 (REV 8-80)

DEPARTMENT OF THE NAVY
PORTSMOUTH NAVAL SHIPYARD
PORTSMOUTH, NH 03804-5000

IN REPLY REFER TO:
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Meghan F. Cassidy
Remedial Project Manager
U.S. Environmental Protection Agency
Region I
JFK Federal Building
Boston, MA 02203-0001

Dear Ms. Cassidy,

Your letter of October 2, 2000 forwarded comments on the draft Results of Radiological Sampling of Water, Sediment, and Biota for Portsmouth Naval Shipyard.

Enclosure (1) contains the responses to your comments. Please let us know within 30 days whether you have additional questions, or that the responses provided are acceptable.

Should you have any questions regarding this response, please feel free to contact me at (207) 438-1283.

J. A. BRANN
Director, Radiation Health

Enclosure: 1. Navy Responses to EPA Comments Regarding the Draft Version of the Results of Radiological Sampling of Water, Sediment, and Biota

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Enclosure (1)

Navy Responses to EPA Comments Regarding the Draft Version of the Results of Radiological Sampling of Water, Sediment, and Biota

1. Section 3.2: The procedure for determining if radium-226 is present from natural sources or is artificially enhanced appears flawed. The assumptions used are not valid, especially for water and biota. Fortunately, this does not appear to be an issue based upon the results (see further comments), but EPA cannot endorse or accept this "comparison" to determine if radium-226 is natural or enhanced.

First, in water and biota, the concentrations of radionuclides, especially those from different chains, cannot be compared. It is typically incorrect to compare concentrations in water for nuclides in the same chain (e.g., U-238 is typically lower than U-234). There are too many factors associated with uranium and radium concentrations, such as solubility and biota uptake rates that invalidate this comparison.

For soil/sediment, the comparison is not as bad, but it is still invalid unless numerous assumptions can be verified. First, it must be assumed that uranium-235 and uranium-238 exist in natural ratios (i.e., U-234 activity is 21.7 times U-235 activity). Unfortunately, even if this is true, it does not necessarily mean that radium-226 is present at that same concentration as U-234, thus making the U-235/Ra-226 comparison invalid. There are regions of the U.S. where glacial activity and other factors have caused nuclides to be present out of equilibrium. Furthermore, analytical uncertainties should be included in these types of comparisons. Finally, sealed containers are not necessarily radon tight containers. Some plastics allow diffusion of radon gas at greater rates than others. The method described does not specify that the container should be filled. It is sometimes accepted practice to assume that the radon daughters will be trapped in the crystal lattice structure and not concentrate on the top. In practice, this is not always the case; however, the resulting equilibrium should be approximate.

Response: This comparison was intended to provide perspective for the level of radium-226 found in one sediment sample from Meade Pond. This comparison protocol was included in the sampling procedure that was reviewed by the EPA (included as Appendix A to the current report). No such comparisons were

needed or made for water or biota samples (other statistical tests for water samples were specified in the approved procedure but were not needed due to radium-226 not being detected).

While the level of radium-226 and the equilibrium status of the decay chain may be subject to the factors and uncertainties discussed by the EPA, the comparison should still be useful as an estimate of the likelihood that radium-226 activity is due to natural background; i.e., as discussed in Section 3.2, if calculated radium-226 activity is substantially lower than measured bismuth-214, that would suggest (but may not prove, due to the uncertainties) that the radium-226 activity is technologically enhanced.

PNS notes that, although some uncertainties are associated with the measurements, the corrected radium-226 and bismuth-214 activities were in fact almost identical. When the activity values are comparable to natural background levels, and this radionuclide comparison matches, this information provides assurance that the detected radium-226 is probably due to nature.

2. Section 3.5 discusses other naturally-occurring radionuclides. It would be useful to see the results of chain nuclides during gamma spec analyses for agreement comparison and equilibrium. Only some were listed in the report, although others were probably identified in the analyses. The Tl-208 activities do not agree with the expected activities based on the Pb-212 and Bi-212 results and the Thorium series decay scheme. The nuclides of interest were limited. What nuclides were included in the gamma spec analysis library that could have been identified?

Response: Per the approved sampling procedure, all radionuclides identified during gamma analyses were included in Appendices D (PNS groundwater samples) and F (Meade Pond surface water, sediment, and biota samples). Isotopes not listed in Appendices D and F were not identified in any of the water, sediment, or biota samples.

The analysis library used included the radionuclides of interest per the sampling plan as well as a number of non-naturally occurring radionuclides associated with weapons testing or reactor operations (cesium-137, manganese-54, cobalt-58, etc.) and commonly observed naturally occurring radionuclides (e.g., potassium-40 and lead-212). An additional appendix will be included in the report listing the radionuclides included in the analysis library.

3. Section 4.1 states that Cs-137 was detected in four wells. These are identified in Appendix D as FW-1, FW-5, B184-MW3, and DW-10B. Based on the TAG map in Appendix I, three of these wells are in close proximity to each other. Discuss how this effects the interpretation.

Response: Although the locations of these wells appear to be close to one another on the map, in reality the wells are approximately a quarter mile from each other and are approximately evenly dispersed over the central portion of the Shipyard. In addition, they are in proximity to several other wells that did not show detectable cesium-137. The one characteristic these four wells have in common is proximity to large areas of pavement/concrete that may enhance incorporation of surface runoff (i.e., an expected accumulation point for fallout-related cesium-137). PNS notes that the concentrations observed were at the detection limit and, as discussed in the Historical Radiological Assessment (HRA; reference (8) in the report) previously forwarded to NAREL, cesium-137 is not a radionuclide of concern with respect to Shipyard operations.

4. Section 4.2 Indications that radium-226 is natural are determined (with the caution above) using mathematical comparisons and typical ranges for the nuclide. However, cesium-137 detections are simply explained as "at levels consistent with fallout from weapons testing." Reference is not given to the "typical levels" from fallout, nor are any background results determined at nearby bodies of water to determine if similar conditions exist. It seems rather inconsistent that the Navy goes through all of the assumptions and corrections to attempt to "verify" that radium-226 is naturally occurring, even when it is in the middle of the "typical" range cited, but simply state that cesium-137 is from fallout without references or other background data to confirm the statement. Additional information should be provided.

Response: As is discussed in reference (8) to the report, cesium-137 is not a radionuclide of concern from Shipyard operations. Radium-226 is, and hence additional discussion was warranted. Cesium-137 is often detected in environmental samples taken in the vicinity of the Shipyard, most notably in harbor sediment samples.

Consistent with the sampling procedure, no background samples from nearby bodies of water appear warranted. The report will be modified to include a reference to NCRP Report 50 for typical levels of cesium-137 in the environment.

5. Section 5.1 The results for the gross alpha/beta and tritium should be reported as calculated rather than only the MDC. The MDC could also be reported with the results.

Response: The gross alpha, gross beta, and tritium tables will be modified to include both the calculated activity and the MDC values.

6. Section 5.2 How can the one gross alpha detection be attributed to uranium/thorium when no analysis of these nuclides was performed?

Response: The Shipyard is not equipped to identify alpha emitters directly, either by radiochemical methods or by spectroscopic methods. However, the likely source of this one positive result could be predicted based on factors such as historical information, the fact that alpha-emitters other than radium-226 are not radionuclides of concern, the likelihood of naturally-occurring radioactivity, and the activity levels of the sample.

The only groundwater sample with detectable alpha activity came from Well SI-1DB, deep-bored into bedrock with a screened formation (sampling zone) from 130 to 150 feet below ground level. Groundwater from this depth is less likely to collect surface contamination from Shipyard activities than would a shallower well with a screened formation nearer the surface. However, no alpha activity was observed in any well other than Well SI-1DB, indicating that the detectable alpha radioactivity probably did not originate with surface contamination of an alpha emitter associated with Shipyard activities.

Additionally, Well SI-1DB is located next to Upper Meade Pond in an undisturbed portion of the central island far from any industrial waste sites. Basically: (a) only cesium-137 and naturally-occurring radioactivity were found in the ponds; (b) reference (8) documents no operations or releases that could account for a man-made alpha source in this well; and (c) the screened interval was in bedrock, where elevated levels of naturally-occurring alpha-emitting radionuclides are not uncommon. Hence, there does not appear to be any plausible basis to suspect this alpha result was due to Shipyard operations.

To verify the presumption that the alpha might be due to natural radioactivity from the deep rock formation, gamma spectroscopy was performed on the residue for the sample in question. That analysis found the gamma emitting radionuclides actinium-228, lead-212, bismuth-212, and

thallium-208, all part of the natural thorium decay chain. A note will be added to the final report to document this.

*7. Section 5.3 EPA does not agree with the statement that potassium-40 is the cause for elevated gross beta results. Statistical review of these data indicate that 60% of the sample results do not correlate well based on a standard normal variable (Z-score) test, sign test (probability of 34 of 40 K-40 [*0.9] being less than gross beta is about 4.2E-6), and other tests.*

The graph at the end of these comments also shows that gross beta results are elevated over K-40 contribution. If the K-40 contribution were the sole source of betas, the K-40 squares would be on the gross beta line. Since almost all squares are to the right of the gross beta line, this indicates that the gross beta results are elevated above the K-40 contribution. Based upon these data, please discuss how more accurate results could be obtained. One possibility is with the filtering of samples discussed below in comments on Appendix H.

Response: The Shipyard is not contending that potassium-40 is the sole source of beta activity in the samples. The point is that much of the beta activity is the result of potassium-40, given the correlation between high potassium-40 levels and high beta counts. Based on the nature of the subsurface into which the wells are drilled, and the fact that several radionuclides in the natural uranium/thorium chains are beta emitters, the observed total beta results appear normal, and no information exists to indicate that further sampling or data analysis are necessary.

8. Section 9.0 The statement "This demonstrates that radiological controls at PNS have been effective in preventing significant amounts of Naval Nuclear Propulsion Program radioactivity from entering the environment." is rather broad. There should be some type of disclaimer, such as "based on the results of this study, indications are that radiological controls at PNS have been effective in preventing radioactivity from entering local groundwater or Upper and Lower Meade Pond." The same is true for the last paragraph of this section.

Response: This recommendation will be incorporated into the report.

9. Appendix A Section 3.Sample Collection, e. indicates that biota and sediment samples will be collected directly into the sample containers and drained indicating that the units for

both media will be pCi/g-wet. Typically sediment results are compared on a pCi/g-dry basis. Confirm how the results were actually reported and clarify how the data was compared.

Response: The samples were drained and placed in the containers wet. Sediment samples show the consistency of thick mud. All Naval Nuclear Propulsion Program (NNPP) data have been reported in this manner since the inception of the Program. This allows for consistency in comparing past NNPP data.

Testing at Mare Island Naval Shipyard during the base closure process indicated that the difference in analysis sensitivity between wet and dry samples was acceptably small. The only benefit from the labor-intensive effort to dry samples would be to achieve a slightly lower minimum detectable activity (which could be done more easily with larger samples or longer count times, were it needed), and to improve analysis repeatability when detectable results are obtained. Since detectable results indicating the presence of potentially Shipyard-related radionuclides are rare and at low levels, as identified in the subject report and in reference (8) to that report, existing analysis protocols remain acceptable.

A note will be added to the applicable tables to clarify that the results are pCi/g-wet.

10. Appendix D Radium-226 MDC values (all greater than 100 pCi/L) are high. Based on an effective dose conversion factor of 3.58E-7 Sv/Bq and an intake rate of 1 L per day (half of the SDWA assumption), 100 pCi/L would equate to an effective dose equivalent of approximately 50 mrem/yr. These MDC values certainly don't support conclusions such as "This demonstrates that radiological controls at PNS have been effective in preventing significant amounts of Naval Nuclear Propulsion Program radioactivity from entering the environment."

In EPA's comments on the Groundwater Monitoring Plan for Radionuclides, we cautioned that gamma spectrometry may give high results due to the interference of the 186 keV gamma peak from decay of U-235, and also that the MDA may be too high to detect Ra-226 in most if not all samples. At that time, we indicated that using the Pb-214 and Rn-222 daughters of Ra-226 should not be done for water samples unless sufficient decay time was allowed for Rn-222 to decay to equilibrium levels with Ra-226 and that this also may not provide detection limits sufficiently low to quantify Ra-226 at desired levels.

The reporting units for the sediment samples were activity per gram. Based on the sample collection procedure in Appendix A.3.e., it is assumed that the results are reported on a pCi/g-wet basis. This should be clarified in the report and will affect the ability to compare between samples since the water content of each sample is not known and will be affected by the type sediment collected.

Response: The sampling procedure (Appendix A to the report) contains specific criteria for the analytical procedures used. No specific minimum detectable concentration (MDC) was specified for radium-226; rather, the data were reported at the sensitivity achieved based on targeting other radionuclides. Given the history of operations involving radium described in reference (8) to the report, no radium-226 was expected due to any Shipyard-related activities. Hence, PNS is not aware of any reason to require expensive additional analysis procedures to prove that ground water is within drinking water standards for all radionuclides. (PNS also notes that all the Shipyard wells sampled were specifically installed as part of on-going CERCLA investigations. As discussed in reference (8) to the report, there are no potable water wells within the Shipyard.)

The statement regarding NNPP controls is appropriate. Radium is not generated by NNPP work. A low MDC was achieved for the NNPP radionuclide of interest (cobalt-60). No non-natural radionuclides (other than cesium-137, attributable to past atmospheric weapons testing) were observed in any sample, at the analysis sensitivities achieved. PNS knows of no information that would indicate a need for further analyses at lower MDCs.

Please see the response to Comment 9 above for a discussion of the wet/dry issue regarding sediment sample results.

11. Appendix F There is a poor correlation between Tl-208 and Pb-212. Pb-212, part of the thorium chain, beta decays to Bi-212. Bi-212 then either beta decays to Po-212 (64% of the time) or alpha decays to Tl-208 (36% of the time). Therefore, the Tl-208 activity should be 36% of the Pb-212 activity. The results in Appendix F show Tl-208 concentrations slightly greater than Pb-212 concentrations. If adjustment to Tl-208 concentrations were made for the "appearance" of equilibrium with Pb-212, the report must clearly state this, although making that correction is unnecessary.

Response: Upon further review, PNS found that the Canberra Spectran computer software did make a decay correction during data reduction which was unnecessary and inappropriate since,

per procedure, the samples were allowed to reach equilibrium. When this factor is removed, the values show reasonable agreement with the expected ratio between thallium-208 and lead-212. The table will be corrected to reflect the proper (actual) values and a footnote added noting what the ratio should be.

12. Appendix H Gross alpha MDC values are too high (several greater than 1000 pCi/L). If this is due to TDS, the question becomes whether the samples had significant entrained particulates that became soluble upon acidification. EPA's comments on the Groundwater Monitoring Plan noted that the degree of suspended solids in the water, particularly the surface water, could necessitate filtering of the samples and analysis of both the filter and the filtrate to determine the radioactivity in the samples.

Response: The samples were taken by a contractor qualified in low-flow sampling per procedures approved by the EPA (i.e., the Shipyard's normal ground water sampling contractor). The concern with respect to entrained particles was discussed with the contractor prior to sampling and arrangements made for filtering if necessary. During sampling, the contractor identified no samples requiring filtering.

In hindsight, it is possible that some of the samples may have had improved alpha MDCs had they been filtered. Section 5.1 discusses the high alpha MDCs obtained and the elevated amounts of dissolved solids in the samples. The amounts of dissolved solids were not identified until after the samples had been acidified and evaporated.