INSTITUTE FOR ENERGY AND ENVIRONMENTAL RESEARCH

6935 Laurel Avenue, Suite 201
Takoma Park, MD 20912

Phone: (301) 270-5500
FAX: (301) 270-3029
e-mail: ieer@ieer.org
http://www.ieer.org

Additional Comments on the Pantex Plant Radiological Investigation Report

Brice Smith, Ph.D. and Arjun Makhijani Ph.D.
Institute for Energy and Environmental Research, Takoma Park, Maryland

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The following are supplemental comments prepared by the Institute for Energy and Environmental Research on the January 2004 Pantex Plant Radiological Investigation Report, henceforth referred to as the RI report. We have prepared this analysis for STAND (Sustainability in Technologies, Agriculture and Nature's Diversity) pursuant to a Technical Assistance Grant made to STAND by the U.S. Environmental Protection Agency. When our original comments were prepared in June 2004 we did not have a copy of the CD containing *Appendix D: Final Radiological Data Sets*. This was provided to us by Camille Hueni, the Remedial Project Manager, Region 6 Superfund Division at the U.S. Environmental Protection Agency (EPA). These comments address issues relating to the information contained in this appendix.

Main Findings and Recommendations:

Our examination of the Final Radiological Data Sets used in the RI Report to characterize the Pantex site has not allayed our concerns regarding the adequacy of the sample collection or data analysis procedures as raised in our revised comments of June 9th in relation to the determination of background.[1] We are pleased that in the July 6, 2004 additional comments from the EPA to BWXT Pantex that our recommendation for a complete review of the laboratory's Quality Assurance/Quality Control program has been incorporated.[2] Our analysis of the Final Radiological Data Sets for the soil and groundwater measurements has shown the same inconsistent and physically unreasonable uranium isotopic ratios as was found in the background samples. Thus we continue to recommend that the QA/QC program for all data samples be scrutinized and that the RI Report be redone using new samples that are analyzed in laboratories recently certified by the Environmental Measurements Laboratory for the appropriate isotopes of uranium, plutonium, thorium, and tritium.

In addition, the sampling for tritium in the ground and surface water must be done with a lower limit of detection than currently reflected in the data. As recommended in our June 9th revised comments, the background for tritium in ground and surface water should be determined from sampling techniques with a minimum detection limit of less than 5 picocuries per litre. The concerns we have discussed in regards to the uranium and tritium measurements raise questions as to the non-detection of plutonium in 75% of soil samples and 88% of ground and surface water

[1] Our original comments on the RI Report were presented on June 7, 2004 in a STAND meeting in Panhandle, Texas. A revised version of our comments was sent electronically to Camille Hueni at EPA on June 9, 2004.
[2] EPA 2004b

samples taken from the Pantex site. We continue to recommend that a suitable background for plutonium be determined from measurements with a lower limit of detection less than 0.001 to 0.01 pCi/gm given that releases to the environment cannot be ruled out from a historical analysis of Pantex operations.

Finally, we recommend that BWXT Pantex re-evaluate and seek external review for their Quality Assurance/Quality Control program used in the selection process of the laboratories to analyze the soil and water samples. We also recommend that they similarly re-evaluate and seek external review for their program to ensure an adequate examination and review of the resulting measurements in order to prevent, at a minimum, the use of data which violates basic physical laws and elementary principles of radiochemistry such as that presented in the Final Radiological Data Sets from the RI Report.

Determination of "Detected Result"

In the Final Radiological Data Sets, as in the background data sets, measurements were only considered if the analyzing laboratory "qualified" the result as being detected.[3] The procedure for the laboratory's decision to exclude these data points is not clearly specified in the text of the RI Report. There are a total of 364 soil samples, 50 ground water samples, and 2 surface water samples that are marked as non-detects even though their comparison to the reported detection limit determined by the standard procedure would have resulted in their being ruled detections. At least one measurement from each of the Site Relevant Contaminants (SRCs) (uranium-234, uranium-235, uranium-238, thorium-232, plutonium-239, and tritium) were excluded in this way. In a number of cases these measurements were above the "Decision Level" as measured by the counting error but below the "Detection Limit" as reported by the laboratory. The rationale for the exclusion of these data points must be more fully discussed both in the RI Report and in the response of BWXT Pantex to the request for all QA/QC information regarding the laboratories performing the measurements. The issue of "qualified" data is particularly important within the context of the serious flaws uncovered in the measurements of uranium in which the reported isotopic ratios are inconsistent with the basic principles of uranium radiochemistry and the measurements of total uranium are inconsistent with the reported activity of U-238 and physically impossible.

Thorium-232

The characterization of Th-232 contamination in water was insufficient in the Final Radiological Data Sets to use in characterization of the Pantex site. As noted in the July 6th additional comments from the EPA, no thorium measurements are reported for the groundwater in either the Ogallala or perched aquifers.[4] For comparison, a total of 962 ground water samples were analyzed for U-234 and 978 were analyzed for U-238. In addition, there were only 12 surface water samples analyzed for thorium contamination. This is in comparison to 541 surface water samples analyzed for U-234 and 540 samples analyzed for U-238. Considering the link between the source of thorium oxides and uranium oxides in the dismantlement of weapons and the potential for thorium contaminated materials to have been burned at the burning grounds as discussed in our earlier comments, it is important that a more thorough characterization of the water (both surface water and groundwater) for this SRC be conducted before any conclusions regarding the contamination of the Pantex site can be made.

Total Uranium

The measurement of total uranium in the soil from the Final Radiological Data Sets shows the same anomalies in regards to the measurements U-238 activity as found in the background data sets. Figure 1 below shows the results for the measurements of total uranium that also had measurements of U-238 activity reported as detected.

[3] RI Report p. C-4 and I-9
[4] EPA 2004b

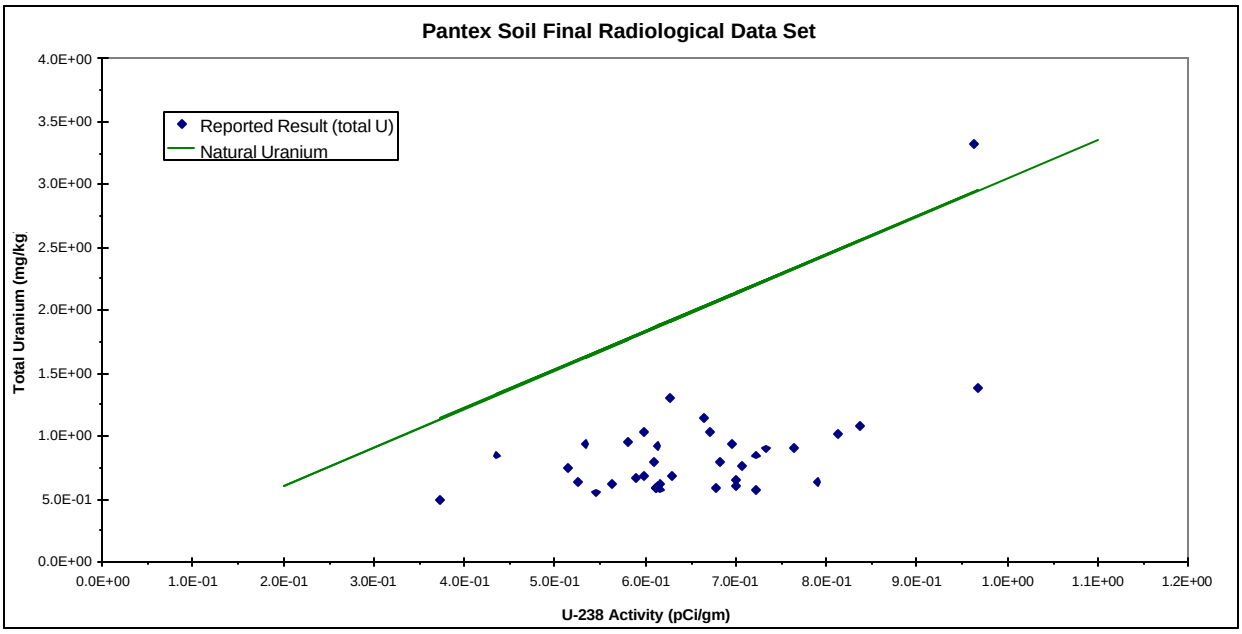


Figure 1: Graph of the measured values of total uranium and U-238 activity in soil at the Pantex site. The solid line indicates the trend that should be expected if it is assumed that essentially 100% of the mass of the uranium is due to U-238 as is the case in natural or depleted uranium.

In natural or depleted uranium nearly 100% of the uranium mass is attributable to U-238 and thus both forms of uranium should follow the solid line in Figure 1. With the exception of a single data point, all of the reported measurements show a U-238 activity in excess of that expected for natural or depleted uranium which is not physically possible. In addition, the data show no apparent strong correlation between U-238 activity and total uranium measured which further indicates that the measurements are not reliable and should not be used as part of a characterization of the Pantex site. This conclusion is further supported by the fact that more than 75% of the 1,375 soil samples analyzed for total uranium were recorded as being non-detections while just 0.26% of the 1,910 soil samples taken for U-238 were recorded as non-detections.

As with Th-232, the sampling for total uranium in the groundwater and surface water was inadequate to make a determination concerning the contamination of the Pantex site. While there were a large number of measurements taken for the activity of specific isotopes of uranium in the groundwater and surface water, there were only two measurements of total uranium in the groundwater (one from the Ogallala and one from the perched aquifer) and there were no measurements taken for the surface water. Given the concerns relating to the accuracy of the uranium measurements discussed here and in our revised comments on June 9th, additional measurements by a qualified laboratory should be made for the groundwater and surface water as part of preparing a new version of the RI Report in line with our recommendations.

Uranium-235

It is our conclusion that the measurements of U-235 in soil as presented in the Final Radiological Data Sets are not reliable and should not be used in order to characterize the Pantex site. Of the 912 samples analyzed for this isotope, nearly 47% were reported as non-detections. Figure 2 shows those that were reported as detected as a function of reported U-238 activity.

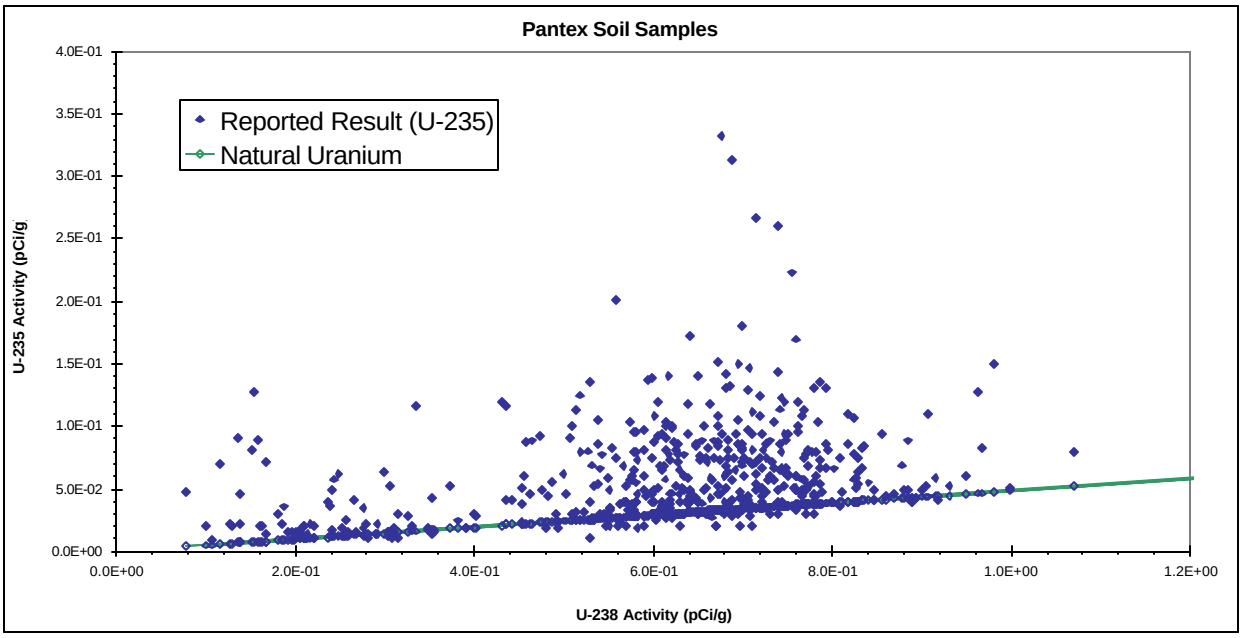


Figure 2: U-235 activity as a function of U-238 activity for soil samples in the Final Radiological Data Set that list both as detected. The solid line indicates the expected trend for natural uranium. The region above the line would be for enriched uranium while the region below the line would be for depleted uranium.

As with the data used to determine background, the isotopic ratios of U-235 to U-238 in the soil measurements are not consistent with natural or depleted uranium, and appear to show no clear correlation between the two isotopes. Since samples of depleted uranium should lie below the solid line in Figure 2, the data make even less sense in the context of Pantex operations. If these data were to be believed, it would indicate the presence of predominantly enriched uranium on site. The average ratio of U-235 to U-238 from these data is found to be 0.11 ± 0.09. The average is more than 2.2 times the expected ratio for natural uranium. However, the large standard deviation (80% of the average) indicates the significant spread in the data and makes it yet more difficult to say anything meaningful from this collection of measurements. It is our expert opinion that the soil data for U-235 as presented in the Final Radiological Data Sets is not physically reasonable, and that it is consistent with our previous conclusions regarding a likely problem with the QA/QC program at the laboratories performing the analysis.

For the groundwater and surface water, the number of samples analyzed for U-235 was far smaller than for soil or for measurements of other uranium isotopes in water. There were only a total of 30 ground water samples analyzed for this isotope. This is compared to 962 ground water samples analyzed for U-234 and 978 analyzed for U-238. The results for the ground water samples where both U-235 and U-238 were reported as detected are shown in Figure 3.

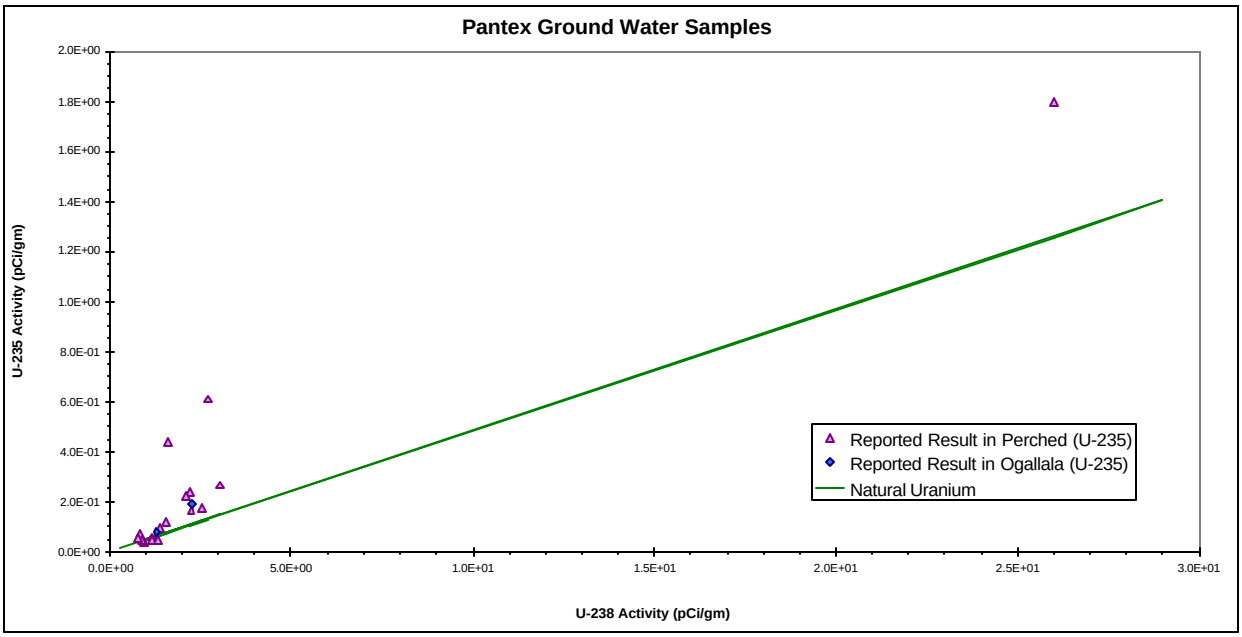
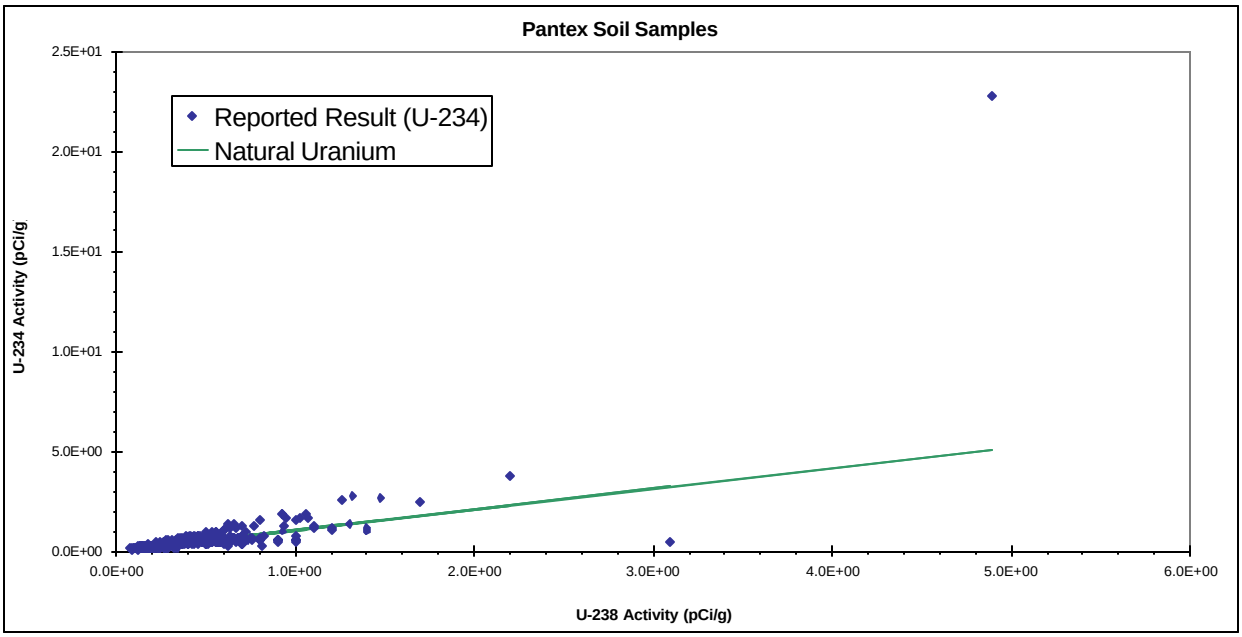


Figure 3: U-235 activity as a function of U-238 activity for the ground water samples in the Final Radiological Data Set that list both as detected. The triangles represent samples taken from the perched aquifer and the diamonds represent samples from the Ogallala aquifer. The solid line indicates the expected trend for natural uranium. The region above the line would be for enriched uranium while the region below the line would be for depleted uranium.

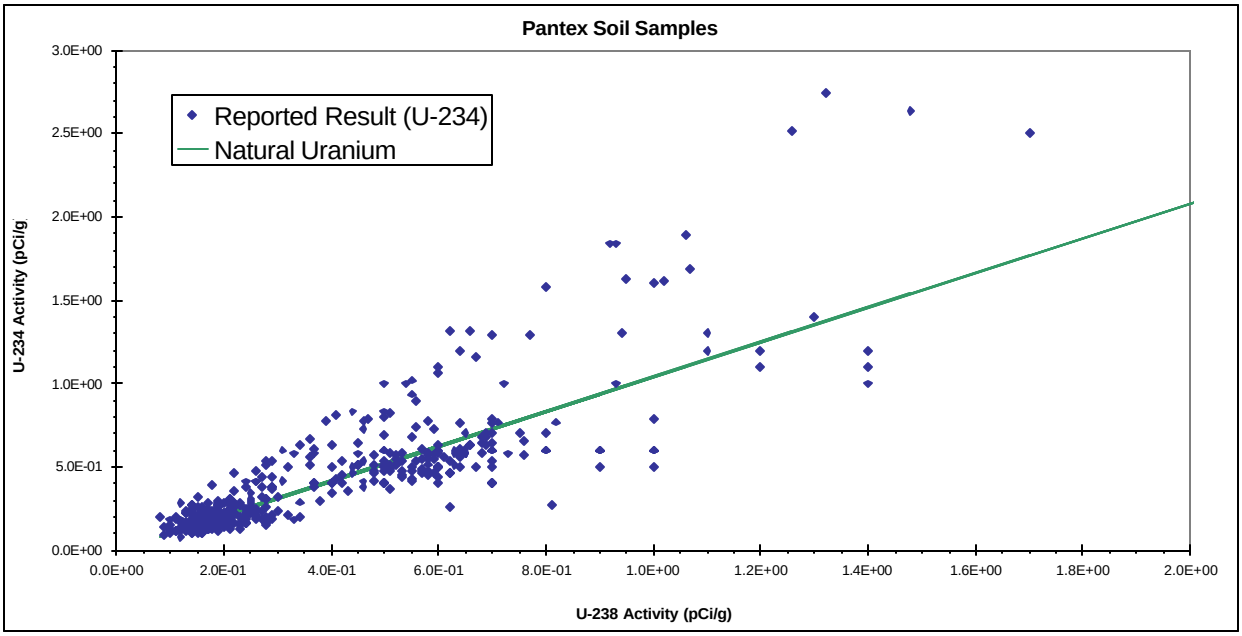
From the data in Figure 3, indications of the same type of anomalous ratio can still be seen as was found in the soil and in the samples analyzed to determine background. The majority of the samples that deviate significantly from natural uranium are above the line which would be appropriate for enriched uranium, but the scatter does not show any clear trend. For the surface water, there were only 4 samples taken and 3 were designated as non-detections. The indications of continued faulty laboratory analysis and/or data collection procedures, and the limited number of samples analyzed for U-235 support our recommendation for a new sampling of the Pantex site and analysis by a recently certified laboratory.

Uranium-234

Except for a single sample, the measurements of the U-234 activity in soil are broadly consistent with the presence of natural uranium. Figures 4a and 4b below show the results for U-234 activity as a function of the reported U-238 activity in the Final Radiological Data Sets for the locations at which both isotopes were reported as detected.



(a)



(b)

Figure 4: U-234 activity as a function of U-238 activity for the set of soil samples in the Final Radiological Data Set that list both isotopes as "detected" (a), as well as for the region of U-238 activities less than 2 pCi/gm to allow greater detail (b). The solid lines indicate the expected trend for natural uranium. The region above the lines would be for enriched uranium while the region below the line would be for depleted uranium.

For uranium in secular equilibrium the ratio of U-234 activity to U-238 activity should be close to one. For the measurements given in the Final Radiological Data Sets, the average ratio is found to be 1.03 ± 0.33. The only serious question surrounding this ratio is connected to the measurements of U-235 discussed above. In our opinion, the U-235 data are not reliable as it stands, particularly given the similarly inconsistent results for the isotopic ratios from the samples taken in order to determine background. However, in light of the indication of possible enriched uranium in the U-235 soil samples analyzed we note that there are 22 samples that have a ratio of U-234 to U-238

activity that is more than 3 standard deviations above the average which might indicate the presence of some level of enriched uranium as well. Only 2 of these 22 samples (Sample ID 20011107D01204 from the Building 12 Sump and Sample ID PTX07-AE-3D08-0-1 from SWMU 57) have U-235 results reported as detected, but both of these samples show ratios that are more consistent with enriched uranium than natural or depleted uranium.

	Ratio U-234/U-238	Ratio U-235/U-238
20011107D01204	2.06 (1.0)	0.454 (0.0484)
PTX07-AE-3D08-0-1	2.04 (1.0)	0.0755 (0.0484)

The numbers in parenthesis are the ratios expected for natural uranium. SWMU 57, also known as Landfill 6, was one of the landfills that was not found in the location that it originally thought to be (i.e. next to Building 12-95).[5] This uncertainty in the process history raises further concerns over these measurements. A re-sampling of the Pantex site with analysis carried out by a properly certified laboratory with reliable measurements for all uranium isotopes at each sample location is necessary to clarify the actual nature of the contamination present.

In relation to the groundwater and surface water samples, we concur with the EPA's comment from June 7th that a reference for the U-234 to U-238 ratio of roughly 2.2 in the ground water at the Pantex site claimed in the RI Report should be supplied, particularly in light of questions regarding the possible detection of enriched uranium in the soil at the site.[6] It has been argued that the surface water shows a U-234 to U-238 ratio closer to two than one because of the large amount of ground water that has been released to the surface onsite. The average ratio found for surface water was 1.8 ± 0.6 as compared to 1.9 ± 0.4 for groundwater from the information in the Final Radiological Data Sets. In light of this explanation from BWXT Pantex, it seems unusual that the percentage of samples that were reported as non-detections in the surface water was nearly 30 times greater than for the ground water (12.2% vs 0.42%). Again, these results further call into question the reliability of the data and support our recommendation that the soil and water sampling be redone before a new draft RI Report is issued.

Pu-239

In our June 9th revised comments, we concluded in relation to the question of plutonium contamination at the Pantex site that

> Discharges of plutonium on to the site cannot be ruled out as sources of contamination of ditches and playa sediment. For instance, the 1961 plutonium dispersal event may have resulted in plutonium contamination being discharged on to the site via the laundry or the shower drain. Further, the primary high explosives were in contact with plutonium. We recommend a careful, properly validated review and analysis of possible plutonium contamination be undertaken as part of a validated sampling plan, with the analysis done by a laboratory certified for plutonium analysis by the Environmental Measurements Laboratory. Fallout background for the site should be established and detection limits should be kept well below this level. The comparison of background levels should be made to surrounding offsite areas where there is high confidence that no contamination from Pantex operations exists. The comparison of Pantex to other DOE sites in very different locations relative to the Nevada Test Site is not a meaningful comparison for background fallout levels.

In light of these considerations, we note that the detection limits achieved for plutonium in the analysis of soil samples presented in the Final Radiological Data Set (0.02 pCi/gm) was on average 20 times higher than the lower detection limits of 0.001 pCi/gm commonly achievable with alpha spectroscopy as cited by the EPA in its Interim Final *Risk Assessment Guidance for Superfund Volume I*.[7] The detection limit for water was on average twice that commonly achievable with alpha spectroscopy. The higher detection levels from the laboratories conducting the analysis of Pantex samples calls into question their findings that 75% of the soil samples were non-detections, while 88% of the groundwater and surface water samples were reported as non-detections.

The repeated use of glassware on older samples and other difficulties associated with achieving a lower detection limit were cited in the RI Report as a reason to consider an even higher detection limit (0.05 pCi/gm).[8] In areas of the Pantex site where the potential for contamination with plutonium cannot be conclusively ruled out (this includes such areas as the playas and ditches) measurement techniques capable of achieving the lower detection should be

[5] RI Report p. 5-43
[6] EPA 2004a and RI Report p. C-13
[7] EPA 1989 p. 10-18 to 10-19
[8] p. 5-42 to 5-43 and Appendix I

used on a sufficient number of split samples during the execution of the re-sampling we recommend to provide confidence in the adequacy of the site characterization resulting from the use of a higher detection limit. The higher detection limit used to hold down costs should not be greater than 0.01 pCi/gm, which is one order of magnitude greater than that typically achievable for plutonium analysis.

Tritium

In the soil data presented in the Final Radiological Data Sets for tritium, there are 59 samples that are listed as "R" in the "Detected Result (Y/N)" column. These 59 samples are spread across Zones 4 and 12, the Independent Sites, and SWMU 82. The status of this data and the reason for its exclusion needs to be clarified in the Appendix as well as in the text of the RI Report. Clarification of this data is particularly important given the fact that the average value reported for those samples with an "R" was nearly 1,250 pCi/gm which is more than 320 times the Preliminary Remediation Goal (PRG) for tritium in soil.[9] The average detection limit for these "R" samples was more than 435 pCi/gm using the procedure set forth for data that the laboratory "qualified" as being detected, which is more than 110 times the soil PRG of 3.8 pCi/gm.

As with the plutonium analysis, the detection limits for tritium in water were on average too high to accurately characterize any potential impacts to the ground or surface water from Pantex operations. The average detection limit for the ground water samples as reported in the Final Radiological Data Set was 137 pCi/L, while the detection limit for the surface water was 165 pCi/L. The detection limit for the surface water was calculated with a correction to two data points that report clearly faulty counting errors that are 1000 times too large. Sample IDs 19950322A01009 and 19950321A00988 report values equivalent to a detection limit of 153.45 pCi/mL = 153,450 pCi/L. Comparing this to the other detection levels and the fact that pCi/ml and pCi/L were both used as units for reporting the surface water results it is most likely that this was supposed to be 153.45 pCi/L. These mistakes in the data collection further highlight the need for a thorough review of all future draft reports.

For comparison to the Pantex detection limits, we note that the typical background levels of tritium in lakes, rivers, and potable water were on the order of a few tens of picocuries per liter.[10] Thus, the use of a detection limit several times higher is not appropriate to properly characterize a site. This concern is further highlighted by the lack of a determination of background for tritium at the Pantex site, the observation of tritium at a level of 1.2 million pCi/L in water near the drip spigots on Building 12-64, and the known release from the "Cell 1 Incident" in May 1989.[11] The use of such a high detection limit calls into question the determination that 92% of the ground water and 90% of the surface water samples were non-detections as reported in the Final Radiological Data Sets. As per our previous comments, the background for tritium should be established using techniques with a lower detection limit of 5 pCi/L and then a re-sampling of both the ground and surface water should be undertaken to adequately investigate the potential impact of Pantex operations on the surrounding water supplies.

[9] RI Report p. 5-36
[10] Eisenbud and Gesell 1997 p. 182
[11] RI Report p. 2-24 and L-2 to L-3

References

Eisenbud and Gesell 1997	Merril Eisenbud and Thomas Gesell, Environmental Radioactivity From Natural, Industrial, and Military Sources Fourth Edition, Academic Press, San Diego (1997)
EPA 1989	U.S. Environmental Protection Agency Office of Emergency and Remedial Response, "Risk Assessment Guidance for Superfund Volume I: Human Health Evaluation Manual (Part A)", Washington, D.C., December 1989 (EPA/540/1-89/002)
EPA 2004a	Camille D. Hueni, Letter to Mr. Jerry S. Johnson "Re:Comments - Final Pantex Plant Radiological Investigation Report for the U.S. Department of Energy/National Nuclear Security Administration, Pantex Plant, Amarillo, Texas, January 2004", June 7, 2004
EPA 2004b	Camille D. Hueni, Letter to Mr. Jerry S. Johnson "Re: Additional Comments - Final Pantex Plant Radiological Investigation Report for the U.S. Department of Energy (DOE)/National Nuclear Security Administration, Pantex Plant, Amarillo, Texas, January 2004", July 6, 2004
RI Report	BWXT Pantex, L.L.C., "Final Pantex Plant Radiological Investigation Report", Amarillo, Texas, January 2004

Acronyms

SWMU Solid Waste Management Unit
SRC Site Relevant Contaminant
PRG Preliminary Remediation Goal