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Ms. Chris Borello
President
Concerned Citizens of Lake Township
P. O. Box 123
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Subject: Examination of the data and associated information on the radionuclide problem in the Industrial Excess Landfill Superfund site.

Dear Ms. Borello:

Thanks for faxing me the letter from Dr. William Muno, Director, Superfund Division of the EPA Region 5. I carefully read that letter and I would like to discuss couple of major points that I disagree on the content of the letter. To set the record clear, 0.28 pCi/L and 0.21 pCi/L of plutonium in the environmental samples are very high. To put these numbers in perspective, the total global fallout (from 1952 to 1980's) of Pu in Houston, Texas was estimated to be 0.20 pCi/cm². If this entire amount were dumped into a lake or ocean waters, in a depth of 200m, the expected Pu concentration would be 0.01 pCi/L. This is based on the following assumptions: a) All the Pu is mixed uniformly; b) There is no removal of Pu from the water column. This will be the total Pu concentration (particulate + dissolved + colloidal). Herewith I give the concentrations of Pu in several natural water samples and the pertinent references:

Lake Ontario ~0.0003 pCi/L (0.3 fCi/L) – Farmer et al. (1973)-

Great Lakes (1973 to 1976): ~0.0005 pCi/L – Alberts and Wahlgren (1981)-
Environmental Science and Technology 15, 94-98.

Hudson River - ~0.0004 pCi/L – Simpson et al. (1980) – In: Transuranic elements in the Environment. W.C. Hanson (ed.), DOE/TIC-22800, 684-690.

Narragansett Bay, RI – 0.0007 pCi/L (0.7 fCi/L) – Santschi et al. – 1980 – Pu in coastal marine environments. Earth and Planet Science Letters 51, 248-265.

Savannah River – 0.0001 pCi/L (Olsen et al. 1989- Environmental Science Technology 23, 1475-1481.

Northwest Atlantic Ocean (380 m) – 0.0006 pCi/L (Cochran et al. 1987, Earth and Planetary Science Letters 84, 135-152)

Greenland and Barents Sea surface water – 0.00035 pCi/L (Holm et al. 1986 – Earth and Planetary Science Letters 79, 27-32.

As you can easily see, the Pu concentration in the groundwater samples in the IEL superfund site is about 1000 times higher than other natural water systems. These natural water systems, such as Lakes, Rivers and marine waters have received their Pu from atmospheric fallout (either direct fallout or subsequently leached from continents). Now, what makes some parties to say this 0.28 pCi/L is low and background level is something I do not understand. If there is any presence of Pu in groundwater, then, it is most likely derived from one or more of the local sources.

Regarding the drinking water limit by EPA for the gross alpha, nobody expects Pu isotope to contribute significantly to that level. Indeed, the major players for the gross alpha are ^{238}U , ^{234}U , ^{226}Ra , and ^{222}Rn . Most other nuclides are quite insignificant. The importance of Pu in the ground water comes because of its high toxicity and its source to the groundwater. As I had mentioned in one of my earlier correspondences, most likely this Pu is derived from a local source, as opposed to the global fallout. If this Pu is derived from a local source, then, obviously there must be much higher levels of Pu in the soil above the water table. In my opinion, the truth must be found by a systematic investigation on this and adjoining aquifer systems.

In order to address the major issue on the quality of the data, we need the following information: a) How much water sample was used; b) Methodology of sample collection; c) What spike was used to assay Pu isotopes; d) What was the alpha detector background in the regions of interest and how long the sample was counted; e) What was the chemical efficiency; f) blank levels with the reagents. I will explain each one of these in detail below.

a) Size of the water sample: Normally, we use several hundred to thousand liters of water samples in the field. We do employ various techniques to preconcentrate the water sample in the field. These are routine methods and the environmental radiochemists have been using these techniques over the past 3-4 decades. For example, during our Arctic Ocean expedition in 1998 and 2000 (sponsored by the National Science Foundation), we filtered close to 100,000 liters of water samples at various depths for radionuclide analysis. For the ground water study, I would imagine to use at least 200-liter samples (for example, see the Pu work on the groundwater samples from the Nevada Test Site, Kersting et al. 1999- Nature, January 7th, 1999 issue). If an agency wants to prove that there is no Pu in this ground water site in order to fulfill the requirement to do some radiation measurements, then, they can collect very small volume of sample and get it measured in some place and they probably will get results what they want to hear. If we are really interested in finding 'the truth' on this matter, then, the sample collection, handling, and analytical procedures must follow the methods that are peer-reviewed, and accepted by the scientific community.

b) Methodology of Sample collection: Many of the radionuclides of concern belong to a class called 'particle-reactive.' What this term means is that these nuclides have a very strong affinity to particle surfaces, including the walls of the container and the filter paper used. These nuclides include all plutonium isotopes, thorium and lead isotopes, among

others. Uranium and radium are less particle-reactive. If one is really interested in measuring the Pu concentration in water sample, then, the sample must be acidified immediately after collection. This will prevent Pu adsorbing onto the wall container. We have conducted several experiments to investigate the affinity of particle-reactive radionuclides onto low-and high-density polyethylene containers (e.g., Baskaran et al. 1992- *Geochimica et Cosmochimica Acta* 56, 3375-3388). To know the truth (if there is any Pu in the ground water samples or not) prior to any filtration, the samples must be acidified immediately after collection and the total Pu (particulate + dissolved + colloidal) must be determined. It is pertinent to point out that in the Pu study on the ground water samples collected from Nevada Test Site, Kersting et al. (1999) found that >99% of the Pu were associated with colloidal and particulate fractions and only <1% of the total Pu was found as dissolved phase. Kersting et al study also indicated that Pu is not immobile in groundwater system and it has moved about 1.3 km in about 29 years (~0.4 ft/day, assuming a constant velocity).

c) What spike was used to determine Pu concentration: Normally, this is not an issue, as most of the labs use Pu-242 obtained from National Institute of Standards and Technology and the Pu-242/Pu-239,240 ratios are certified. However, we need to make sure what spike was used and the level of Pu-239,240 present in that spike.

d) Detector background and counting time: Since most of the analysis was done by alpha spectroscopy, it is very critical that we know the background of the alpha detector that is used for the work. For example, we always use detectors with extremely low background, as low as 0.006 counts per hour in each of the regions of interest for Pu isotopes. Since the errors on the final concentration value of the sample depends on all these factors, high background in the detectors will lead to higher propagated error on the activity. Another most important factor is the counting time in the alpha spectrometer. It appears that the commercial labs (including NAREL counted for 30 hours and depending on the sample size, this counting time may be inadequate. When we were investigating if any of the dumped nuclear reactors in the marginal seas of Russia started leaking Pu into the water, we counted some of the samples for about 2 weeks to get better precision. At least 100 counts in each of the peaks of Pu-239,240 and Pu-242 will lead to an error of about 30% (2-sigma error, using NIST Pu-242 standard). Instead of doing too many samples, it may be worthy to select a few samples and count them for long time and get high precision numbers. The goal should be that these data should be of the quality to be publishable in a peer-reviewed, international professional journal.

e) Chemical Efficiency factor: One should be able to obtain at least 40% chemical yield on these samples. Since Pu radiochemistry is fairly straight forward, this should not be a problem. However, one need to watch out for this factor, as poor chemical efficiency will result in higher uncertainty on the data.

f) Blank Level of the Reagents: Invariably, our blank levels are below the detection limit and thus, the blank level was never an issue for us. However, if glassware/Teflonwares are not cleaned properly (in boiling nitric acid, etc), residual Pu can cross-contaminate the sample and data will become highly unreliable.

I am not privy to the information pertaining to the issues (a) through (f) raised above. However, the uncertainty associated with the Pu concentration reported for this site is ridiculously high. Our University or any other academic institution where there is any active environmental radioactivity research is going on can improve the precision by 100 to 1000 times. I think that is where the ultimate answer lies for this puzzle.

On another related issue, you had mentioned about measuring Pu on a gravel-soil sample by acid-leaching technique. Since all the Pu on a gravel or soil is absorbed (as opposed to lattice-bound), normally we carry out acid leaching technique to remove the adsorbed Pu. If we use a fairly strong acid (such as one molar hydrochloric acid or nitric acid or stronger concentration), we will leach a major portion of the absorbed Pu in the first leach itself. In the second leach, most of the Pu will be leached out and there won't be any Pu left for subsequent leaching. Thus, it would be simply a waste of time to leach a soil or gravel third time for any Pu determination, as we won't have any leftover Pu. If some one is trying to prove that there is no Pu on the third or fourth or fifth leach, in my opinion, they are wasting their time and they are not supposed to get any Pu.

As my summary, 'truth finding mission' involves a systematic scientific research to determine if there is any Pu in any of the groundwater samples. The data that will be generated should be publishable in a professional, peer-reviewed journal. The data thus obtained should be reproducible by any environmental radiochemist in academia around the world.

I hope this assessment helps you to move forward with this issue. Please feel free to contact if you have any additional questions. I can also be reached by e-mail: Baskaran@chem.wayne.edu

With best regards,

Mark Baskaran

References:

Baskaran, M., P. H. Santschi, G. Benoit and B. D. Honeyman. 1992. Scavenging of thorium by colloids in seawater of the Gulf of Mexico. *Geochimica et Cosmochimica Acta* 56, 3375-3388.

Kersting, A. B., D. W. Efurud, D. L. Finnegan, D. J. Rokop, D. K. Smith, and J. L. Thompson (1999) Migration of plutonium in groundwater at the Nevada Test Site. *Nature* 397, 56-59.