

Review of Evidence for Anthropogenic Radioactivity at the  
Industrial Excess Landfill, Uniontown, Ohio

**Michael E. Ketterer, Ph.D., Consulting Analytical Chemist**

**3828 Steves Blvd., Flagstaff, AZ 86004**

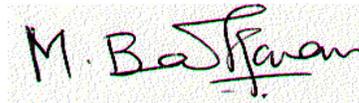
**[Michael.Ketterer@nau.edu](mailto:Michael.Ketterer@nau.edu)**

A handwritten signature in black ink that reads "Michael E. Ketterer". The signature is written in a cursive style with a long horizontal flourish at the end.

**Mark Baskaran, Ph.D.**

**35042 Lana Lane, Sterling Heights, MI 48202**

**[baskaran@wayne.edu](mailto:baskaran@wayne.edu)**

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**Final Report**

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## Summary

This report considers the following two questions concerning the Industrial Excess Landfill (IEL): 1) What evidence exists for the presence of “anthropogenic radioactivity” in the IEL, its surface soils, and the groundwater in the vicinity of the site? and 2) Have the previous environmental sampling and analysis efforts, conducted by USEPA and Ohio EPA, been appropriately designed and conducted so that best available methodology, maximally protective of the environment and human health, has been used to detect any potential anthropogenic radioactivity?

**Historical Background of the IEL.** The Industrial Excess Landfill (IEL) located on Cleveland Avenue in Uniontown, Ohio, about 16 km southeast of Akron, is designated as a CERCLA (Superfund) site. This site was originally known as the Summit Sand and Gravel pit and was used as an open borrow pit to mine sand and gravel. When the sand and gravel mining reached the water table, the mining ended. Subsequently, this site was converted to use as a licensed landfill. Over a period of 14 years (1966-1980), according to the Ohio EPA's submittal under the hazardous ranking system (3/8/84), the IEL contains “> 780,000 tons of hazardous substances”, which included at least 1 million gallons of liquids according to other EPA reports. In the initial phase, the landfill was licensed for fly ash disposal. During this period, the landfill was an open pit and would have served as a local recharge area for the shallow groundwater system and during this process, the recharge could have acted as a flushing mechanism to move contaminants through the landfill debris and down into the groundwater system. Other potentially hazardous contaminants that were dumped into the landfill include solid and semi-solid latex, flammable and non-flammable solvents, septic tank clean-outs, organic matter capable of generating ethane, and other garbage. The local health department documented up to 11,000 gallons being dumped per day by 1/72 records. Some of the buried waste in the eastern portion of the site was placed directly on top of more permeable sandy sediments and is reported to have direct connection to the uppermost aquifer (where many test wells were recently sealed from further testing). This site was placed on the US EPA's National

Priorities List (NPL) in October 1984, without any consistent monitoring data of groundwater in and around the site. Under the Superfund (CERCLA) program, the NPL is a list of the nation's top priority hazardous waste sites eligible for investigation and cleanup. The initial investigation, referred to as the Remedial Investigation and Feasibility Study (RI/FS), took place at IEL between 1985 and 1989. Subsequently, a Record of Decision (ROD) was issued for a pump and treat system, a cap and an expanded gas system. The ROD was issued approximately 22 years after the initial placement of waste into the landfill. The IEL site was capped with permeable soils and vegetated ca. 1980.

The IEL Superfund site was never licensed to receive radioactive waste and anecdotal evidence provided early in the 1980s from local citizens indicated that radiation had been trucked into the landfill in large quantities by tankers. While dumping of fly ash will result in elevated levels of  $^{238}\text{U}$ - and  $^{232}\text{Th}$ -series radionuclide, other anthropogenic radionuclides including transuranics (such as plutonium, Pu) are also alleged to have been dumped into the landfill. In addition to the notarized statements given by the residents - in what may be an unprecedented acknowledgement - the former owner of IEL, Charles Kittinger, came forward after a 30 + year silence, and provided Federal Court testimony in 2001 indicating that plutonium-238 was buried at IEL in approximately the late 1960's in large stainless steel "eggs" by the US Government. The US Justice Department investigation into Kittinger's statements did appear to reveal via magnetic resistivity testing that indeed egg-shaped "anomalies" were identified in close approximation to where Kittinger had flagged. Still, no further action was taken, with the Government claiming Kittinger's testimony lacked validity.

To address the concerns of local residents and state and federal officials, two rounds of initial groundwater testing were conducted by USEPA in August and December of 1990, albeit after the 1989 ROD issuance. USEPA still refused to conduct core sampling as was widely requested. By late fall, USEPA's NAREL declared the first round to be "invalid", claiming among other things, that plastic jars were erroneously used instead of glass for tritium analysis. EPA changed laboratories for the

December 1990 round. In August of 1991, the public was notified that elevated levels of tritium had been found in water samples collected from private wells that exceeded the federal drinking water limits. When serious aspersions were once again cast by EPA against this second lab that had reported the high tritium at IEL, former US EPA Administrator William Reilly, in response to public outcry, asked Thomas Grumbly, then president of Clean Sites, Inc., to look into the two successive rounds of analytical results that had been discredited by USEPA. As a result, Grumbly recommended that all the data from the second round be released. At that point, the public learned that not only were high levels of tritium reported, but there were numerous indications of man-made isotopes, including various forms of plutonium, including  $^{241}\text{Pu}$  and  $^{236}\text{Pu}$ . Reportedly, the Ohio EPA at this time was duly concerned and began conducting on a limited basis testing of a few offsite private wells. The Ohio State Health Department reported a sample collected from a shallow private drinking water well in March of 1991 with a reading of total Beta at 280 pico curies per liter. The Agency for Toxic Substances and Disease Registry (ATSDR) reported that the mean tritium levels in a suite of samples from the monitoring wells collected by Ohio EPA (OEPA) was found to be approximately 2,000 pCi/L (ATSDR, 1994). A sample drawn from a different shallow well in June 1991 reported over a million pCi/L tritium\* which is about 50 times higher than the Fed limits (SAB Report, p. 15). The well did not show high tritium activity before (in March) or after (in October) and only during June 1991 high activity was reported. It appears that there were possible extenuating hydrogeologic conditions during June 1991 and that the sample was collected at/near a sod farm when millions of gallons groundwater were pumped up during that period, could be a factor for the appearance of high level of tritium. This reading, if correct, could not plausibly due to background radiation. Rather than continuing to resample the wells more frequently in question, US EPA came into the site in the fall of 1991 and conducted "exploratory boreholes" at various locations at IEL. Under obvious pressure from Ohio EPA, USEPA-Region 5 agreed to drill a borehole down to the interface of the bedrock in January 1992, as specified by Ohio EPA, after the state had received aerial photographs of a suspect disposal area in the northeast corner that apparently had been neglected during the RI/FS. Once again,

initial data from these various boreholes suggested possible evidence of both <sup>238</sup>Pu and <sup>239</sup>Pu. The reader is referred to further discussion of the January 1992 ninety-two foot borehole.\*

In May of 1992, US EPA commenced groundwater sampling for radiation, conducting quarterly rounds that ended in March of 1993. (NOTE: No additional comprehensive radioactivity investigations of IEL has ever conducted by USEPA after March 1993). After discrediting the two previous commercial labs' IEL data, USEPA chose its own privately owned lab, S. Cohen and Associates, for the rounds conducted between May 1992 and March 1993. Technical experts hired by the citizens group, CCLT, via the "TAG" (technical assistance grant) program mandated under CERCLA by Congress, registered very serious written concerns in 1993 about the "field filtering" process conducted on the IEL samples during this time period. These experts questioned whether the actual levels of radiation were being grossly underestimated by this practice of removing the particles in the field prior to samples being sent for lab analysis. CCLT's TAG experts expressing grave concern that particulate removal could lead to inadequate risk assessment and a seriously flawed cleanup plan.

Because of widespread concern held not only by citizens and their experts, but by local, state and federal officials, former USEPA Administrator Mr. Reilly also adopted Mr. Grumbly's recommendation that the EPA Science Advisory Board (SAB) be utilized. However, instead of investigating the public's concerns about what went on between EPA and the labs, particularly regarding a non-standard testing method ordered for IEL by EPA that caused spewing and loss of suspended solids and colloidal material in the IEL samples, USEPA changed the charge of the panel to a more generic focus. The revised focus of the SAB was to determine whether the USEPA Finished Drinking Water Method 900 was appropriate not only at Uniontown IEL, but all other CERCLA sites in the nation suspected to contain radiation. Because the SAB's bottom line in the panel's report concluded that testing methods were adequate for sites expected to contain "common" radionuclides\* citizens and experts remained

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\*: Nuclides that are derived from crustal material, not from anthropogenic activity

deeply concerned, since it<sup>i</sup> was obvious that the synthetic radioisotopes believed to be buried at IEL were not considered to be "common." Therefore, the community group and elected officials pressed for an independent investigation of the issues by the USEPA National Ombudsman, Robert Martin. Soon after Mr. Martin's own preliminary investigation into IEL raised additional concerns that the site's waste had not been properly characterized, Martin left USEPA over a serious disagreement concerning his losing his independence, after being transferred to the USEPA Inspector General's office. The new Ombudsman replacing Martin, Paul McKechnie, picked up the IEL case in 2003 and hired an outside radiation consultant from Manitoba, Canada, Dr. Mel Gascoyne, for the putative purpose of reviewing the evidence for any anthropogenic radioactivity at IEL. While Dr. Gascoyne does list his concerns regarding the testing methods being sufficiently lacking in several areas to properly discern man-made radiation, Dr. Gascoyne appeared to ultimately provide to EPA the bottom line the agency seemed to desire, that the sampling conducted at IEL was adequate to support the contention that the site is "in compliance" with the Gross Alpha and Gross Beta Finished Drinking Water Standards. (NOTE: The authors of this report take serious exception to this application of this method being used for the detection of anthropogenic radiation, given that it is meant for treated water systems not expected to contain synthetic radioisotopes. The reader is referred to further sections of this report for more discussion).

The CCLT group continues to seek the Department of Energy and the Department of Defense's intervention into the IEL case, believing that these agencies are the most appropriate entities to handle what is buried at IEL in order to properly protect the health and welfare of the community and the aquifer in the long term. CCLT is strongly opposed to the Monitored Natural Attenuation remedy, namely, the continued flushing of the site into the area's groundwater through dilution. USEPA has unilaterally adopted the Monitored Natural Attenuation remedy after both the pump and treat systems and protective cap from the 1989 ROD were done away with by the IEL potentially responsible parties (PRP's) with Region 5's blessings in 1999 and 2002. CCLT and other area citizens are not only concerned about radiation

exposure to the public exposure via groundwater, but remain deeply concerned about migrating landfill soil vapors, given the high porous sand and gravel surrounding IEL in close proximity to thousands of homes. Radon in soil gas was detected as high as 3,640 pico curies in samples collected from the intermediate depth of perimeter landfill gaseous monitoring wells while EPA failed to sample for tritium vapors as discussed in the SAB report as a possible way of detecting the presence of other radionuclides with IEL.

**Objectives of this Report.** Based upon this site history and the uncertain framework of available information regarding the presence of anthropogenic radioactivity at IEL, the authors have been hired by Concerned Citizens of Lake Township (CCLT) to develop a technical report addressing the following two questions:

- 1) What evidence exists for the presence of “anthropogenic radioactivity” in the IEL, its surface soils, and the groundwater in the vicinity of the site?
- 2) Have the previous environmental sampling and analysis efforts, conducted by USEPA and Ohio EPA, been appropriately designed and conducted so that best available scientific methodology, maximally protective of the environment and human health, has been used to detect any potential anthropogenic radioactivity?

In consideration of these objectives, this study will consider a series of documents provided by CCLT, and the authors will also bring into consideration any other information from the scientific literature deemed relevant and reliable. These documents are described in Table 1.

**Question 1. What evidence exists for the presence of “anthropogenic radioactivity” in the IEL, its surface soils, and the groundwater in the vicinity of the site?** In this review, “anthropogenic radioactivity” is considered as any non-naturally occurring radionuclides, clearly present from an IEL-related anthropogenic source, or naturally occurring radioactive material (NORM) that is present in the landfill

environs at enhanced concentrations/activities due to IEL-related processes. This report will focus mainly upon the following substances: I) plutonium (in the isotopic forms  $^{238}\text{Pu}$ ,  $^{239}\text{Pu}$ ,  $^{240}\text{Pu}$ ), which is not naturally occurring except as minimal amounts of  $^{239}\text{Pu}$  in uranium ores resulting from spontaneous fission of uranium (Curtis, 1999); II) fission products, which are synthetic, non-naturally occurring radionuclides resulting from bomb or reactor fission of uranium or plutonium; III) any uranium isotopes present in ratios inconsistent with those found in Nature; and IV) other radionuclides such as  $^3\text{H}$  (tritium) which are mainly of synthetic, anthropogenic origin when found in the Earth's surface environment.

When considering the possible presence of plutonium, fission products (e.g.,  $^{99}\text{Tc}$ ) and other radionuclides ( $^3\text{H}$ ) in the landfill environ, it must be recognized that above-ground testing of nuclear weapons has artificially introduced and/or elevated the background levels of these isotopes at the Earth's surface (Beck and Bennett, 2002; Bennett, 2002). This review recognizes the existence of these other sources of synthetic radionuclides that are clearly not IEL-related, and hence considers their presence to be an expected part of the global background situation. Of direct concern herein, however, is any "anthropogenic radioactivity" that is ascribable, with preponderance of evidence, to an IEL-related cause.

**Tritium ( $^3\text{H}$ ).** Tritium is a radioactive, beta-emitting form of hydrogen that has a half life of 12.3 years. It is constantly being produced in the stratosphere by cosmic rays, but was also released in large thermonuclear tests conducted mainly between 1952 and 1962 in the Northern Hemisphere. From 1952 until recently, thermonuclear tritium dominated in precipitation and surface waters; however, the bomb fallout tritium has been steadily decaying, and cosmogenic tritium is once again becoming the main fraction. Theodorsson (1999) reports that the mean annual tritium concentration in Northern Hemisphere precipitation has fallen from a maximum of 1000-2000 TU in 1962 to about 10 TU in 1996. One TU (referred to as "tritium unit") corresponds to 0.118 Bq/L (Bq/L = Becquerel per liter), 3.2 pCi/L (pCi/L = pico curies per liter), or one  $^3\text{H}$  atom per  $10^{18}$   $^1\text{H}$  atoms. Earlier papers (e.g. Egboka *et al.*, 1983) also discuss the temporal changes of tritium activity in surface waters

and precipitation.

Tritium in the environment is usually found as “tritiated” water molecules (HTO), and the environmental/hydrologic behavior of HTO is essentially identical to that of stable water (Li-Xing *et al.*, 1995). In addition to cosmogenic and thermonuclear tritium, the presence of other anthropogenic environmental tritium sources has been documented in specific settings. Betti *et al.* (2004) summarize marine sources of tritium in the time interval 1985-1999, and show that nuclear fuel reprocessing stations at Sellafield (UK) and Cape de la Hague (France) are important contributors to the oceanic inventory. Thompson *et al.* (2006) documented the presence of tritium in Nevada Test Site vicinity vadose-zone waters from specific underground tests conducted in the 1970’s. Baeza *et al.* (2006, in press) reported the presence of above-background tritium from a nuclear power plant in the surface waters of the River Tagus in Spain. Beals and Hayes (1995) found elevated levels of tritium in surface waters at the Savannah River Site. Tritium has also been used as a tracer in biological and chemical laboratory research, and as an agent in luminous paints (Argonne National Laboratory, Human Health Fact Sheet, 2005). USEPA has developed a maximum contaminant level (MCL) for tritium of 20,000 pCi/L.

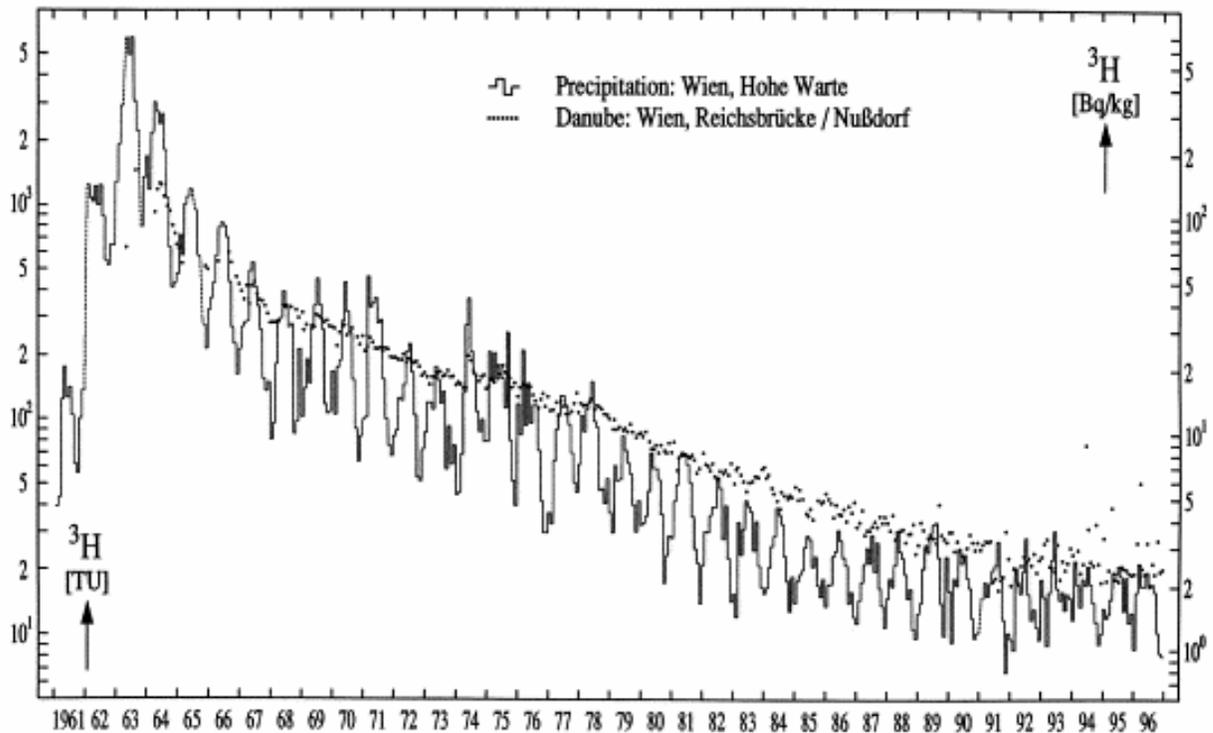
The presence of enhanced levels (i.e., above thermonuclear and cosmogenic) tritium in a local setting can generally be detected by comparing the activity to the expected curve for tritium activity vs. time in surface waters. An example of this type of a curve is shown below (from Theodorsson, 1999) for precipitation in Vienna and water samples from the Danube River:

There is existing evidence from groundwater samples collected in the IEL vicinity in 1992 that tritium is present at “enhanced” levels. These levels are trend outliers from Theodorsson’s tritium activity-time curve for surface waters and precipitation, and hence some other specific source is inferred. The samples in question were analyzed by S. Cohen Laboratories in 1992 and were reported as follows: Sample IE-7-GW-1D, 9/1/1992,  $470 \pm 180$  pCi/L ( $17.3 \pm 6.6$  Bq/L); Sample IE-7-GW-7I, 8/24/1992,  $400 \pm 180$  pCi/L ( $14.8 \pm 6.6$  Bq/L); Sample IE-7-GW-6S, 8/31/1992,  $400 \pm 180$  pCi/L ( $14.8 \pm 6.6$  Bq/L); and Sample IE-7-GW-7S, 8/26/1992,  $490 \pm 180$  pCi/L ( $18.1 \pm 6.6$  Bq/L). These samples are plotted in comparison to Theodorsson’s curve as

shown below:

It is seen that the IEL vicinity samples exceed the anticipated concentrations of tritium in precipitation and surface waters by approximately one order of magnitude. *Although the detected levels are well below EPA's MCL of 20,000 pCi/L, and in the absence of any other clear explanation for the elevation, the gross elevation of tritium in these samples can be taken as strong evidence of a local source of tritium at the IEL.*

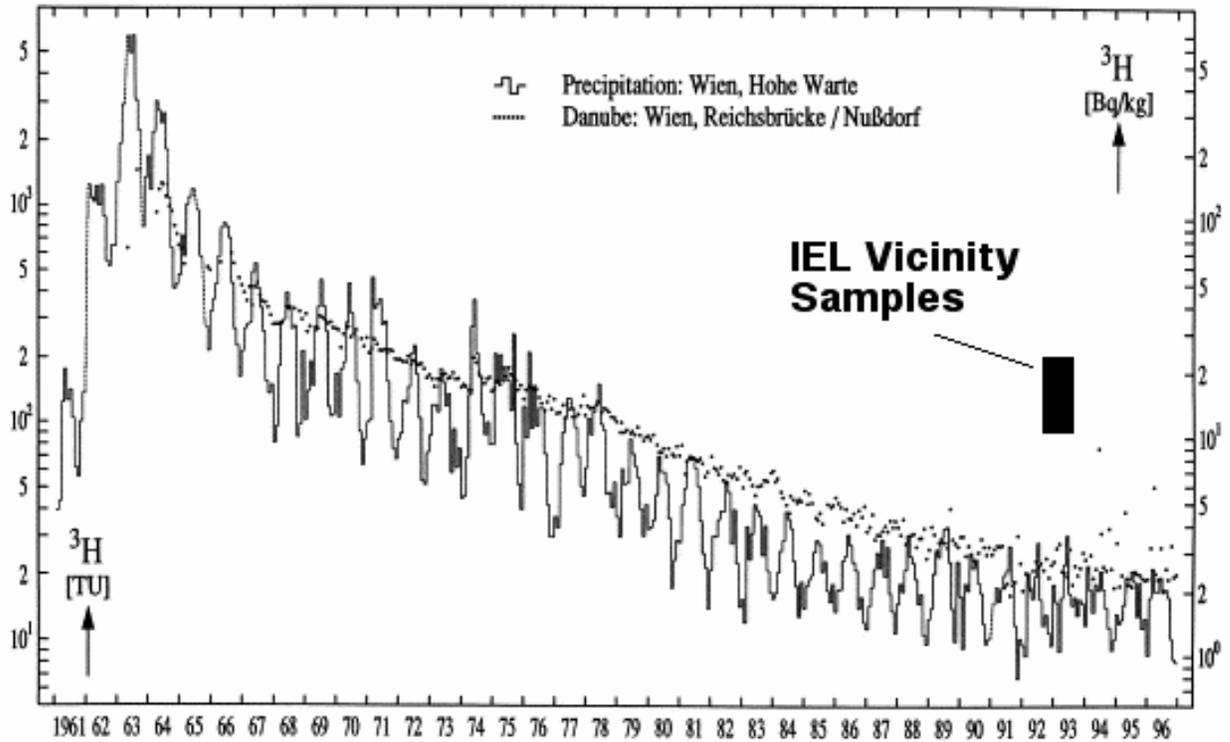
Rather than being one set of tests in isolation, the USEPA results from S. Cohen Laboratories are consistent with even higher tritium activity reported during limited split sampling conducted by the Ohio EPA (Betz Lab/ TMAEberline) in several other samples. One offsite monitoring well located in a residential area, MW 27s, indicated  $6665 \pm 2636$  pCi/L;  $246 \pm 97$  Bq/L. This data point would plot well above the black box sketched in the curve shown above, and resembles the levels of environmental



The precipitation and surface water data terminate in 1996, but contemporary levels are similar or slightly lower.

tritium found at the 1962-1964 peak of the thermonuclear tests. USEPA in 2004

nevertheless allowed this well to be sealed with the claim it had been "clean for greater than ten years". Although USEPA has maintained there has never been any evidence of radiation contamination above background, it must be noted that the USEPA Inspector General's own report on IEL does not seem to readily accept this



contention. In its report, appendix B, under the heading, Details on Testing Sensitivity," paragraph 1, the I.G. reveals that, ..."*Over the last 15 years, tritium in rainwater and snow melt (naturally occurring) has only been 40-60 pCi/L. Consequently, to determine whether tritium is naturally occurring vs. man-made, the analysis would need to be sensitive enough to detect 40 pCi/L (or less) of tritium.*"

In spite of this clear awareness within the agency about what would be considered background for tritium, there has been steadfast denials regarding the significance of the several examples found of tritium at IEL deemed valid, well above the 40 -60 acknowledged by the IG. It is the opinion of the authors of this report that the non-thermonuclear/cosmogony levels of tritium indicate the probable presence of a tritium-containing industrial, medical or governmental waste material within the IEL,

and that this situation warrants additional prompt and proper investigations by government agencies.

**Technetium-99 ( $^{99}\text{Tc}$ ).**  $^{99}\text{Tc}$  is a beta-emitting radionuclide with a half-life of 212,000 years that is present at extremely low levels in natural uranium ores (Curtis, 1999). This isotope is associated with the dispersion of fission products from nuclear tests and reactors. Technetium has no stable isotopes; its chemical properties and behavior strongly resemble that of rhenium (Re). In natural waters,  $^{99}\text{Tc}$  is readily soluble and geochemically mobile in the form of pertechnetate ( $\text{TcO}_4^-$ ), chemically analogous to perrhenate ( $\text{ReO}_4^-$ ). The solubility/mobility properties of  $^{99}\text{Tc}$  in the surface environment have long been recognized, and are discussed by Brookins (1986).

Data on the expected weapons-testing derived levels of  $^{99}\text{Tc}$  in natural waters are sparse. It is nevertheless possible to project that these  $^{99}\text{Tc}$  activities are extremely low, based upon results reported in seawater by Dahlgard *et al.* (2004). In a study of  $^{99}\text{Tc}$  and other anthropogenic radionuclides in North Atlantic surface seawater samples collected in 2000 near Greenland, Dahlgard *et al.* (2004) found levels ranging from 0.000016 – 0.000175 Bq/L (0.0004-0.005 pCi/L). The entirety of even these levels cannot be ascribed to weapons testing, since the  $^{99}\text{Tc}$  budget in the North Atlantic is influenced by nuclear fuel reprocessing. McCubbin *et al.* (2002) found much higher  $^{99}\text{Tc}$  levels ( $\sim 0.1$  Bq/L) in surface waters of the Irish Sea, proximal to Sellafield and la Hague. Beals and Hayes (1995) found  $^{99}\text{Tc}$  levels of  $\sim 0.1$  Bq/L ( $\sim 3$  pCi/L) in surface freshwaters from the Savannah River, consistent with strong influence from this localized source.

In localized environmental settings, the most common scenario for finding any detectable  $^{99}\text{Tc}$  is the presence of environmental contamination with “recycled uranium” (RU). RU is uranium that has been formerly used in a plutonium production reactor, where a blanket of uranium captures neutrons with the intention of transmuting  $^{238}\text{U}$  into  $^{239}\text{Pu}$ . During the early part of the Cold War era, the US Government was concerned with a critical shortage of uranium; hence following use in Pu production reactors, as much uranium as possible was recovered and separated from the Pu product. The uranium was then re-used for other nuclear applications,

hence the name “recycled”. RU is characterized by the presence of much higher levels of  $^{99}\text{Tc}$ ,  $^{236}\text{U}$ , and transuranic elements than are found in U of natural origin. The fission product  $^{99}\text{Tc}$  is, to some extent, present with RU as an unintended impurity resulting from the solvent extraction processes intended to separate uranium and plutonium from the fission products, while  $^{236}\text{U}$  and transuranic elements are produced *via* neutron capture processes. The presence of  $^{99}\text{Tc}$  in association with RU at many former military and defense-related activities is well established. One site of this nature is located at Weldon Spring, Missouri (refer to [www.wssrap.com](http://www.wssrap.com)).

One data point generated by USEPA indicates the positive presence of  $^{99}\text{Tc}$ ; a water sample from monitoring well MW-14s, collected by the IEL PRPs in May 2001, revealed the presence of 16.49 pCi/L (0.61 Bq/L)  $^{99}\text{Tc}$ .

The MDA for this particular sample was 15.50. Therefore this result is actually above the MDA. Yet, this fact apparently is currently in contention, with NAREL citing the USEPA IG report to have claimed that, on the contrary, “all the  $^{99}\text{Tc}$  results were at or below the MDA.” Because this finding represents evidence of anthropogenic material buried at the IEL site that has long been denied, it is not a small matter that NAREL is denying this reading as being an actual result and evidence of man-made radiation presence at IEL. Moreover, we are troubled by what appears to be a disingenuous argument, because, while on one hand, the NAREL cites the IG report on this, it seems to ignore completely the IG Table, B-1, appendix B of this report, where the IG pointed out that, in 9 out of 9 samples tested for the  $^{99}\text{Tc}$  at IEL, “the detection limit set in the regulations for that analysis” was exceeded. *The stated preferred detection limit is 5 pCi/L.* Yet, all 9 MDAs were well above that level, with a range of 15.58 to 23.39 pCi/L. These inadequate MDA’s produce the following question: if one well tested at IEL reported 16.49 pCi/L, how many additional wells would have exhibited  $^{99}\text{Tc}$  had analyses with an MDA of 5 pCi/L been used? Instead, USEPA appears to contend that the presence of  $^{99}\text{Tc}$  is immaterial and tolerable as long as an MCL is not exceeded. Since the well in question, MW-14s was sealed, there is currently no way to go back at that specific location and depth in order to re-open the

question of the apparent presence of  $^{99}\text{Tc}$  \*.

It seems likely that EPA has not considered the true significance of the presence of *any level* of  $^{99}\text{Tc}$  in IEL vicinity waters, since their apparent concern is only comparison to an MCL of 900 pCi/L. Nevertheless, a  $^{99}\text{Tc}$  activity of 16.49 pCi/L is *orders of magnitude higher* than could be reasonably expected from thermonuclear testing alone, and therefore implicates the probable presence of a localized, IEL-related source of  $^{99}\text{Tc}$ . The most likely source of  $^{99}\text{Tc}$  is, as discussed above, recycled uranium. In addition to this data point, a number of other data point indicate the presence of relatively high levels of total beta emitters (“gross beta”) that are not accounted for by specific radionuclides. *It is the opinion of the authors of this report that the non-thermonuclear level of  $^{99}\text{Tc}$  in sample MW-14s indicates the probable presence of a “recycled uranium” waste material within the IEL, and that this situation warrants additional prompt and proper investigation by the appropriate US government agencies.*

**Plutonium (Pu,  $^{238}\text{Pu}$ ,  $^{239}\text{Pu}$ , and  $^{240}\text{Pu}$ ).** The isotope  $^{239}\text{Pu}$  is found in minute quantities in Nature, as a result of neutron capture on  $^{238}\text{U}$ ; the source of the neutrons is the spontaneous fission of  $^{235}\text{U}$ .  $^{239}\text{Pu}/^{238}\text{U}$  atom ratios of  $\sim 10^{-12}$  are present in U ores, and Taylor (2001) has estimated the natural concentration of  $^{239}\text{Pu}$  to be about  $10^{-17}$  g  $^{239}\text{Pu}$  /gram Earth’s crust. No evidence to date has indicated the presence of naturally occurring  $^{238}\text{Pu}$  or  $^{240}\text{Pu}$ . Plutonium has been widely distributed, however, on the Earth’s surface from above-ground weapons tests (Beck and Bennett, 2002). Surface concentrations of the most abundant isotopes  $^{239}\text{Pu}$  and  $^{240}\text{Pu}$ , expressed as the sum ( $^{239+240}\text{Pu}$ ), are on the order of  $\sim 1$  Bq/kg or  $\sim 5 \times 10^{-13}$  g Pu / gram Earth’s crust (Ketterer *et al.*, 2004). The environmental behavior of Pu is such that it is mainly associated with particle surfaces; Pu has been thought to exhibit limited mobility in the environment. Levels of plutonium in surface waters, affected by weapons test fallout, are generally very low. Several independent studies (*vide infra*) indicate  $^{239+240}\text{Pu}$  activities in the range of 0.0001 to 0.001 pCi/L. Nevertheless, some recent

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\* In July and October 2005, 13 samples were collected at sites adjoining IEL and detectable  $^{99}\text{Tc}$  was found in every one of the sample (Baskaran and Ketterer, 2006).

evidence indicates that Pu in subsurface environments can be transported in association with colloidal material (Kersting *et al.*, 1999).

Over the history of environmental monitoring at IEL, several sampling episodes have investigated the possible presence of one or more plutonium isotopes in water and borehole soils. The decision tree for the IEL samples was that only samples exceeding screening would move into specific analysis for man-made isotopes such as Pu and  $^{99}\text{Tc}$ . As a result, the total number of samples that were actually subjected to Pu analysis was severely curtailed, mainly due to the questionable screen process. There are some major concerns on the collection, preservation and analysis of samples. For example, total Pu measurements (without any filtration) on acidified samples (acidification immediately after sample collection) would have addressed many of the concerns, the question whether the sample collection and preservation practice followed at the IEL site raises concern on the possible underestimation of the Pu concentration, if any. The EPA's Finished Drinking Water 900 protocol followed in the collection and preservation of water samples were designed for the drinking water where synthetic radionuclides are not expected to be present, and the collection and preservation methodology is not suitable for sites such as IEL, where there is concern about the dumping of synthetic radionuclides. While it is possible to contend that the samples collected in 2000 and 2001 were later preserved in the lab for individual radionuclide analysis, the loss may have well already occurred *before* the samples reached the laboratory. In some cases, the counting time in the alpha spectrometer was too short (179 minutes) and the volume of water samples used for analysis was too small (1 liter). Some of the "positive" test results are shown in Table 2. These results indicate the probable presence of plutonium (of non-weapons testing fallout origin) in the groundwater and solid materials within the IEL. The results indicate several specific water samples containing Pu ( $^{238}\text{Pu}$  and/or  $^{239+240}\text{Pu}$ ) at levels exceeding expected "fallout origin" concentrations in surface waters by at least three orders of magnitude. Moreover, the results apparently indicate  $^{238}\text{Pu}/^{239+240}\text{Pu}$  activity ratios disparate from the  $^{238}\text{Pu}/^{239+240}\text{Pu}$  activity ratio of  $\sim 0.04$  expected from weapons testing fallout in the northern Hemisphere (Mietelski and Was, 1995; Baskaran *et al.*, 1996, 2000). This suggests not only the presence of Pu-

containing waste materials buried at the IEL, but also that this waste material contains an unusual  $^{238}\text{Pu}/^{239+240}\text{Pu}$  activity signature (from a  $^{238}\text{Pu}$ -containing source).  $^{238}\text{Pu}$  is used in thermoelectric generators (El-Genk and Tournier, 2001).

One sample of soil, IE-1-B8-21-92, was collected from a depth of 92 feet during the installation of an exploratory borehole drilled down to the interface of the bedrock on 1/20/1992, at the request of the Ohio EPA. This sample result was provided by USEPA's NAREL (ID IE-1-BS-21-92), and the data deemed to be valid. Results for this sample indicated  $^{239}\text{Pu}$  at  $0.081 \pm 0.027$  pCi/g (wet basis) and  $0.096 \pm 0.027$  pCi/g (dry basis). In a 1992 EPA Fact Sheet, EPA acknowledged that this same borehole sample collected at 92 feet was "retested and showed slight level of plutonium". Subsequent to this, EPA repeated the Pu analysis three times, but those re-analyses showed no measurable Pu activity. If reanalysis involved only the sample that was already processed, then, the reproducibility test is meaningless, as Pu is anthropogenic in origin and will remain sorbed on to particulate surface. Any leaching of solid material with acid will remove quantitatively *all* the *sorbed* Pu and hence re-testing should result in *no Pu* activity. Every time another new aliquot of the sample must be taken for measuring Pu, not re-leaching of the same sample. Thus, the claim by EPA and the data remain highly questionable. Results for this sample indicate  $0.104 \pm 0.029$  pCi/g  $^{239}\text{Pu}$  (note that  $^{239}\text{Pu}$  measured by alpha spectrometry is actually the summed activity of  $^{239+240}\text{Pu}$ ). Also lacking reasonable explanation is why any detectable  $^{239+240}\text{Pu}$  would be found in borings collected from a depth of 92 feet; it is generally recognized that the entire inventory of weapons testing fallout Pu is contained within the top 30 cm of surface soil (Kelley *et al.*, 1999). The activity of  $0.104 \pm 0.029$  pCi/g  $^{239+240}\text{Pu}$  ( $2.81 \pm 0.78$  Bq/kg) would not be unusual in a surface soil horizon, but is inexplicable at a depth of 92 feet. Given the propagated standard deviation is significantly less than the value itself, the data from this site very valid. Given that it is the authors' understanding that this borehole and those drilled earlier and sampled in the fall of 1991 was field filtered, there is serious concern that these reported findings of Pu were underestimated results. In any event, there is no reasonable explanation as to why any detectable  $^{239,240}\text{Pu}$  would be found in borings collected at 92 feet.

There are two additional items that the authors find inexplicable with respect to the apparent presence of non-weapons testing plutonium at IEL. The first is as follows:  $^{239+240}\text{Pu}$  activities shown in Table 2 are lower than the MCL (15 pCi/L based upon the gross alpha MCL) for  $^{239+240}\text{Pu}$  that EPA Region V is apparently considering for the IEL data. Nevertheless, this overlooks the situation that the  $^{239+240}\text{Pu}$  activities in several water samples exceed the water MCL for  $^{239+240}\text{Pu}$  of 0.15 pCi/L that has been applied in the vicinity of the Rocky Flats Site in Colorado (see <http://www.epa.gov/region8/superfund/sites/co/rocky.html>). That USEPA enforces the 0.15 pCi/L MCL for  $^{239+240}\text{Pu}$  at Rocky Flats is evident; USEPA brought an administrative complaint for exceedances of the 0.15 pCi/L MCL for  $^{239+240}\text{Pu}$  at Rocky Flats (see <http://www.epa.gov/oalj/orders/energy.htm>, Docket No. CERCLA-VIII-98-11). *The authors find it completely inexplicable as to why an explicit MCL for  $^{239+240}\text{Pu}$  in water of 0.15 pCi/L can be utilized (and enforced with civil penalties) in one EPA Region, yet in another EPA Region, no standard exists and a gross alpha threshold of 15 pCi/L for  $^{239+240}\text{Pu}$  in water is considered adequate, and protective of human health and the environment.*

The second item relates to the importance of Pu level to the gross alpha activity. It appears that a major factor in the US EPA's assessment regarding the IEL Pu data is based on the questionable contention that unless the Pu values at IEL are higher than the Gross Alpha Finished Drinking Water Limit of 15 pCi/L, the presence of Pu in IEL groundwater is of no concern to EPA. The usage of Gross Alpha and Beta was derived several decades ago as a crude screening measurement for naturally-occurring U-Th series radionuclides, such as radium-226. It was never intended to be used to measure man-made nuclear material, and yet, this appears to be exactly what is meant by this final conclusion. Results from Nevada Test Site (NTS) indicate that only <1% of the Pu is migrating as dissolved Pu and >99% of the total Pu is migrating as colloidal Pu (both intrinsic colloid, Pu oxide, and extrinsic colloids composed of clays (Kersting *et al.*, 1999). The Pu (colloidal + dissolved), however, is very small and represents only a small fraction ( $>10^8$  Ci of radioactive material at NTS and the measured activity of Pu was ~0.3-0.5 pCi/L) of the total Pu associated with the particular test (Benham nuclear test) in the NTS region (Kersting *et al.*, 1999). *If the*

*Pu analytical data reported at the IEL site are “real”, it follows that it is likely that a large amount of Pu is buried at a subsurface environment in the vicinity where the sample was collected. While Pu activity level at the IEL groundwater is low compared to the DWL for gross alpha, this level is about 100 to 10,000 times higher than the value reported for other natural water systems such as ocean water, lake water, river water, etc.*

*It is the opinion of the authors of this report that the elevated levels of Pu in IEL-vicinity waters and the detectable Pu in a borehole at a depth of 92 feet are impossible to reconcile with Pu expected from weapons testing. Therefore, it is probable that site-specific source(s) of Pu exist within the IEL.*

**Uranium:** Uranium in nature consists primarily of three isotopes:  $^{238}\text{U}$  (primordial, half-life of 4.5 billion years, 99.27% abundance),  $^{235}\text{U}$  (primordial, half life of 713 million years, 0.72% abundance), and  $^{234}\text{U}$  (from the  $^{238}\text{U}$  series, half-life of 250,000 years, 0.0055% abundance). An additional isotope,  $^{236}\text{U}$  (half-life of 23 million years) occurs in nature at ultra-trace abundances (Ketterer *et al.*, 2003). Uranium is a naturally occurring constituent of soils and waters; hence its presence alone (even at relatively high concentrations or activities) does not infer an anthropogenic source. Non-naturally occurring U is most efficiently detected by examining the U isotopic composition using modern mass spectrometric techniques (Becker, 2003). Unequivocal indication of non-naturally occurring U can be inferred by the presence of  $^{236}\text{U}$  (Ketterer *et al.*, 2003) and/or deviations from the naturally occurring  $^{238}\text{U}/^{235}\text{U}$  atom ratio (Ketterer *et al.*, 2000).

After review of the IEL-related documentation shown in Table 1, the authors find the evidence inconclusive for the possible existence of non-naturally occurring U at IEL. The evidence neither proves nor disproves non-naturally occurring U, and it is recognized that the activities of  $^{234}\text{U}$  and  $^{238}\text{U}$  in IEL-vicinity waters are relatively low. The activities themselves do not suggest any specific IEL source, but may simply be relatively low on account of local geochemical conditions. Nevertheless, it is apparent that USEPA has failed to adequately conduct appropriate analysis (namely, mass spectrometry) that could readily and definitively examine the U isotope compositions. This is discussed further in the second major section of the

report, and this same point was also brought up in Dr. Gascoyne's previous critique of USEPA's work. USEPA's inadequate U analytical work is perplexing, in light of the widespread existence of capabilities for mass spectrometric U isotope studies in many commercial, academic, and US Government laboratories, including USEPA's own facilities.

**Gross Alpha and Gross Beta Results.** The parameters "gross alpha" (GA) and "gross beta" (GB) are typically used in screening-level radiochemical work in order to test for the possible presence of high levels of radionuclides. The results can be used as indicators to determine if measurements of specific alpha- or beta-emitting isotopes are warranted, and can also be used as control values so that it can be judged that all alpha- or beta-emitting isotopes have likely been accounted for.

These types of test results are reflected in many places in the documents listed in Table 1. A few specific examples indicate GROSS ALPHA and/or GROSS BETA readings that exceed EPA's established MCL's (GROSS ALPHA MCL = 15 pCi/L; GROSS BETA MCL = 50 pCi/L). Several GROSS ALPHA measurements for 1992 samples (analyzed by Betz Laboratories) exhibit GROSS ALPHA levels between 33 and 419 pCi/L. One of EPA's results from the May 2001 IEL sampling indicates GROSS BETA = 62.78 (dissolved) and 64.57 (total) pCi/L in monitoring well MW-17s. One of the high sets of GROSS ALPHA and GROSS BETA results (GROSS BETA = 440 pCi/L) came from the same borehole where  $^{239+240}\text{Pu}$  was found at 92 feet. The presence of these levels of GROSS ALPHA and GROSS BETA would seem to suggest improper accounting for all alpha- or beta-emitting isotopes and/or generally unreliable radiochemical data. Nevertheless, they leave the question open as to the possible presence of non-naturally occurring alpha- and beta-emitting isotopes. High GROSS BETA levels, incongruent with naturally occurring beta emitters such as  $^{40}\text{K}$  or U – Th series radionuclides should be interpreted as indication of synthetic isotopes such as  $^{99}\text{Tc}$ .

**Question 2.** Have the previous environmental sampling and analysis efforts, conducted by USEPA and Ohio EPA, been appropriately designed and conducted so that best available scientific methodology, maximally protective of the environment and human health, has been used to detect any potential anthropogenic radioactivity?

Question 2 has previously been considered elsewhere as part of EPA's own self-scrutiny (Gascoyne, 2004). Gascoyne's report stated the following:

- The radiochemical methods employed in analyzing the groundwater were modified versions of the EPA (900 series) methods for tritium, radium isotopes, potassium, gross alpha and gross beta, Pu, U, Th, Sr and Tc isotopes are not necessarily suitable for determining whether the groundwater is contaminated by small amounts of radioactive waste.
- A significant deficiency in the IEL study was the short periods for which radioactive isotopes were counted. Count times ranged from 30 minutes to about 5 hours but the more important radionuclides (isotopes of Pu) were only counted for 170 minutes each.
- The tests were sufficient to declare that site groundwaters in 2000 and 2001 met the requirements of the drinking water standards with respect to radioactive elements and isotopes. It is not possible, however, to state no radioactive waste is present at the site because, in many cases, the analytical procedures used to detect specific types of radioactivity were insufficiently sensitive to differentiate measured concentrations from background (natural) levels.
- The possible presence of radioactive waste at this site remain unresolved following the 2000 and 2001 sampling, because the analytical methods used were only adequate to show that the groundwaters met drinking water standards.

In all of EPA's previous analytical work, the focus has been to compare activities to drinking water standards (i.e., MCL's). These standards are as follows:

- 5 pico curies per liter (pCi/L) for combined  $^{226}\text{Ra} + ^{228}\text{Ra}$

- 15 pCi/L for gross alpha
- 50 pCi/L for gross beta
- 20,000 pCi/L for  $^3\text{H}$
- 30 microgram per liter ( $\mu\text{g/L}$ ) for total uranium

Only the soluble radionuclides are expected to contribute to the total alpha and beta activities. A majority of the radionuclides that are present in groundwater are soluble and occur naturally (mainly  $^{238}\text{U}$ ,  $^{234}\text{U}$ ,  $^{235}\text{U}$ ,  $^{226}\text{Ra}$  and  $^{224}\text{Ra}$ ). Most of the particle-reactive radionuclides (all radioisotopes of Th, Pa, Pb, Pu) are removed from the solution in relatively short-time scales and hence their activities in the solution phase are extremely low and thus, do not contribute to the gross alpha activity. *The design of EPA's GROSS ALPHA and GROSS BETA methodology, and the rationale for the MCL's, is that these limits were not intended for isotopes such as Pu.* Plutonium is not expected to be present in groundwater systems, as most of the Pu is derived from fallout from nuclear bomb testing and they are retained quantitatively by the soil cover, penetrating up to ~ 30 cm from the surface. Thus, presence of any Pu in groundwater can be attributed to the presence of large amounts of Pu in the subsurface environment. The Pu buried subsurface is relatively immobile owing to its low solubility in groundwater and strong sorption onto aquifer matrix. Nonetheless, colloid-facilitated transport of particle-reactive radionuclides, including Pu has been reported (Buddemeier and Hunt, 1988; Penrose *et al.*, 1990; Kersting *et al.*, 1999). Results from the Nevada Test Site (NTS) indicate that only <1% of the Pu is migrating as dissolved Pu and >99% of the total Pu is migrating as colloidal Pu (both intrinsic colloid, Pu oxide, and extrinsic colloids composed of clays (illite and smectite), zeolites and cristobalite (Kersting *et al.*, 1999). The total Pu activities in water (colloidal + dissolved), however, are very small and represent only a small fraction ( $>10^8$  Ci of radioactive material at NTS and the measured activity of Pu was  $\sim 0.3\text{-}0.5 \times 10^{-12}$  Ci/L) of the total Pu associated with the particular test (Benham nuclear test) in the NTS region (Kersting *et al.*, 1999). The presence of colloidal Pu was found at a distance of ~1.3 km from where the nuclear event took place and the identification of the particular test was done based on the  $^{240}\text{Pu}/^{239}\text{Pu}$  atom ratio. At

IEL, during the 2001 sampling, a total  $^{239+240}\text{Pu}$  level of 0.22 pCi/L was reported in Well MW-15s. Other sites on which Pu was found are as follows: MW-01D – Total Pu was reported to be 0.28 pCi/L; MW-011 – 0.22 pCi/L. While this activity level is low compared to the MCL for GROSS ALPHA, this level is about 100 to 10,000 times higher than the value reported for other natural water systems such as ocean water, lake water, and river water. These natural systems were exposed to the atmosphere and hence received global atmospheric fallout. More recently some of the Pu found in rivers is primarily derived from erosion and subsequent dissolution of previously deposited  $^{239+240}\text{Pu}$  present in surface soils in the watershed.

For comparison, we provide  $^{239+240}\text{Pu}$  activities in several natural water samples and the pertinent reference:

Lake Ontario: ~0.0003 pCi/L – Farmer *et al.* (1976)

Great Lakes (1973-1976): ~0.0005 pCi/L – Alberts and Wahlgren (1981)

Hudson River: ~0.0004 pCi/L – Simpson *et al.* (1980)

Narragansett Bay, RI: ~0.0007 pCi/L – Santschi *et al.*, 1980

Savannah River: ~0.0001 pCi/L – Olsen *et al.*, 1989

Northwest Atlantic Ocean (380 m): ~0.0006 pCi/L – Cochran *et al.*, 1987

Greenland and Barents Sea surface water: ~0.00035 pCi/L – Holm *et al.*, 1986

As can be seen easily, the  $^{239+240}\text{Pu}$  activities in groundwater samples at the IEL CERCLA site are 2-3 orders of magnitude higher than other natural systems. Furthermore, if  $^{239+240}\text{Pu}$  is detected at depths of 92 to 190 feet down from the earth's surface in groundwater system, most likely the Pu (if real) must have been derived from a specific underground source, rather than leaching from soils (fallout Pu) in the surface. **If this value is real, it is likely that the presence of extremely low levels of Pu in the groundwater could imply that a large amount of Pu is buried at a subsurface environment in the vicinity where the sample was collected.**

In addition to this Pu issue, there are some key issues that need to be addressed. They are:

- 1) **Issue#1:** Time elapsed between collection and filtering: While considerable amount of time elapsing between the collection and acidification may not be issue for water-soluble radionuclides (such as radium, uranium isotopes), for particle-reactive radionuclides (such as Pu), this is a serious issue. **While EPA Standard Method (Method-900) was not developed** for cases when there is anthropogenic radiation such as Pu is involved, cases dealing with Pu and other particle-reactive radionuclides require immediate acidification of the sample after collection, which is routinely conducted by the academic scientific community.
  
- 2) **Issue#2:** Issue of the volume of water samples: For the radiological analytical work, a total volume of 7-8 liters of water samples were collected without filtering or preserving and were submitted to ThermoRetec in Oak Ridge, TN. While this volume is adequate for gross alpha and beta measurements, the sample size is >100 times smaller than what is usually used to measure Pu in water samples. For example, Kersting *et al.* (1999) collected large volume of water samples (~200 L). *No meaningful results for particle-reactive radionuclides can be obtained from such small volume of water samples.*
  
- 3) **Issue#3:** The chemical methodology used – The key question that we have undertaken to answer is *not* if radioactive waste was disposed of at the site and poses public health hazard. It is the authors' opinion that the issue is not whether the radiation tests and results conducted so far meets the requirements of the drinking water standards or not, with respect to radioisotopes. For the analysis of Pu,  $^{242}\text{Pu}$  was used as a yield monitor. Although the authors were not given access to USEPA's original raw data on how much  $^{242}\text{Pu}$  spike was added, it was possible to estimate the spike quantity from the alpha spectrum included in Dr. Gascoyne's report. Routinely, in the assay of any nuclide by alpha spectrometry, the internal spike (which implies another alpha emitting isotopes that do not occur in nature and that has different alpha energy than the isotopes of interest) added is comparable to the activity present in the sample. When the spike added is about an order higher than the natural levels, then, generally, the counts in the isotope of interest will be about 10% of the spike counts. When the

sample is counted for less than 180 minutes, then, it will be extremely hard to find any counts in the regions of interest for that particular isotope.

We have analyzed Pu alpha spectra given in Dr. Gascoyne's report. For example, the  $^{242}\text{Pu}$  count from 170 minutes counting for MW-01i is 59 counts. Assuming a chemical efficiency of 50% and alpha detector efficiency of 25%, the calculated activity of the  $^{242}\text{Pu}$  spike is 2.3 pCi. If a one liter water volume was used for the analysis, and if the spike activity is about 10 times higher than the  $^{239+240}\text{Pu}$  value found previously, it is impossible, based upon counting statistics of  $^{239+240}\text{Pu}$ , to produce any meaningful results for any Pu isotopes present in the sample. As discussed, this will result in only ~10% of the counts as that of  $^{242}\text{Pu}$ . It remains enigmatic as to why a very large activity of  $^{242}\text{Pu}$  was added in concert with a short counting time that could not realistically be expected to detect low levels of  $^{238}\text{Pu}$  or  $^{239+240}\text{Pu}$ .

#### 4) **Issue#4:** Alpha Spectrometric Tailing Effects in U and Pu Spectra

In some of the samples, the  $^{238}\text{U}/^{235}\text{U}$  activity ratios, as determined by the alpha spectrometric results, are very different from the activity ratios of natural uranium. For example, in the data reported by S. Cohen and Associates (dated June 23, 1992), one can infer the following: The  $^{238}\text{U}/^{235}\text{U}$  activity ratio is  $10.78 \pm 1.33$ , which is significantly different than the natural U value of 21.76. This discrepancy either speaks to the unreliability of the results, or alternatively, it indicates there is some serious contamination of non-natural uranium at IEL. Having examined the alpha spectrum of the U source, there is considerable tailing between various isotopes of U that have adjacent alpha energies (such as  $^{234}\text{U}$  tailing into  $^{235}\text{U}$  and  $^{235}\text{U}$  tailing into  $^{238}\text{U}$ ). These effects usually originate from poor radiochemical work and improperly prepared alpha sources with deposits of excessive thickness. Thus, the authors contend that all the U data are unreliable, and only high precision isotopic measurements using either thermal ionization mass spectrometry (TIMS) or inductively coupled plasma mass spectrometry (ICPMS) can provide reliable atomic ratios of U. In the case of Pu, we do not have complete information on the blank levels of Pu and hence we are unable to judge the quality of the data.

### **Conclusions and Recommendations:**

Based on our evaluations of the existing data on the radiation levels in the soil and groundwater samples at and around the Industrial Excess Landfill site located in Uniontown, Ohio, we draw the following conclusions:

- 1) The presence of elevated level of tritium in the groundwater sample appears to suggest a local source tritium at the IEL. It appears that the tritium is of non-thermonuclear/cosmogenic in origin and appears to indicate the probable presence of a tritium-containing industrial, governmental source or medical waste within the IEL. This situation warrants additional prompt and proper investigation by EPA.
- 2) Although the  $^{99}\text{Tc}$  found at the IEL site is below the MCL level of 900 pCi/L,  $^{99}\text{Tc}$  seems to be derived from the presence of 'recycled uranium' waste material within the IEL, a finding warranting further investigation<sup>†</sup>. The presence of elevated levels of Pu in IEL-vicinity waters and detectable level of Pu in a borehole at a depth of 92 feet seem to suggest that there is a site-specific source(s) of Pu within the IEL. **If the Pu value is real, it is likely that the presence of extremely low levels of Pu in the groundwater could imply that a large amount of Pu is buried at a subsurface environment in the vicinity where the sample was collected.**
- 3) The methodology adopted (volume of water sample used for the Pu measurements, the amount of  $^{242}\text{Pu}$  spike added, the counting time, problems with the methodology used for the determination of  $^{238}\text{U}/^{235}\text{U}$  activity ratios) for the determination of Pu and U clearly indicate major flaws in the sampling and analytical work.
- 4) USEPA should abandon the finished drinking water method and instead use an approved DOE-method for man-made radionuclides, with good sensitivity, low detection limits, and high precision methods. Analysis must be done on unfiltered

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<sup>†</sup> In every one of the 12 groundwater samples collected adjoining IEL site during July and October 2005,  $^{99}\text{Tc}$  was detected.

samples and the samples must be acidified immediately after collection in the field.

- 5) We strongly recommend that additional core samples be collected in the northeast corner of IEL near the former Borehole 6 is located.

Based on our analysis of all the existing radiation data from IEL, we recommend the following:

- 1) Reopening the sealed test wells (MW 1 I and D, MW 4s, MW 7s, MW 12 d, MW 14s, MW 17s, MW 16 and MW 26 I,) are critical to monitor the anthropogenic radiation levels. The same conclusion is reached by fellow experts (Dr. Julie Rice and Ms. Linda Aller).
- 2) Collect core samples in the Northeast quadrant of the dump (surrounding the ‘eggs’) for anthropogenic radiation testing.
- 3) Conduct proper scientific field sampling, storage and testing for anthropogenic radionuclides following methodology with good sensitivity of those locations where anthropogenic radionuclides have been reported.
- 4) Conduct IR mapping of the surface and subsurface areas in the IEL site to assess how the subsurface water content varies. The heat generated by the anthropogenic radionuclides could dry up the soil in select areas.
- 5) Conduct a scientific investigation on the contents of the eggs using state-of-the-technology.
- 6) Provide CCLT with additional funding to collect split water samples for independent review.

**Table 1. Documents Provided by CCLT and Considered in the Development of this Report.**

1. Ombudsman Report 2004-P-00031, “Review of Actions at Industrial Excess Landfill Superfund Site, Uniontown, Ohio. September 29, 2004.
2. Memo of 4/14/97 to Interested Parties from CCLT RE: Data from the lab “CEP” (Tritium, Pu, Uranium).
3. Memo of July 8, 1993 to Linda Kern, USEPA Region 5 Remedial Project Manager, from Sam T. Windham, Director, National Air and Radiation Environmental Laboratory.
4. Letter of January 30, 2001 to Chris Borello, CCLT, from Ross del Rosario, USEPA Region 5 Remedial Project Manager, RE: Evaluation of August 2000 Radiation Data, Industrial Excess Landfill Site, Uniontown, Ohio.
5. USEPA Method 900.0, Gross Alpha and Gross Beta Radioactivity in Drinking Water.
6. Letter of April 23, 2004 to Chris Borello, CCLT, from Eric Denison, Health Physicist, Ohio Bureau of Radiation Protection.
7. Letter of October 2, 1997 to Norman Martin, Board of Lake Township Trustees, from Timothy Fields, Jr., Acting Assistant Administrator, USEPA, Office of Solid Waste and Emergency Response.
8. Letter of February 26, 1993 to Mary Margaret Rowlands, McNamara and Freeman, from Dr. Elaine B. Panitz, RE: Cause of Death of Mr. Blanton Beltz.
9. Letter of 27 September 2001 to the CCLT from Mark Baskaran.
10. An SAB Report: Review of EPA’s Approach to screening for radioactive waste materials at a superfund site in Uniontown, Ohio. Prepared by the ad hoc Industrial Excess Landfill Panel of the Science Advisory Board, US.EPA, EPA-SAB.EC-94-010, September 1994.
11. A packet of numerous documents containing analytical data and other information as follows: a) May 2001 Sampling Event, Eberline Data – May 2001.xls – 7/19/2001; b) November 2000 Sampling Event; c) Page 3, R. Flaak data of November 1, 1993; “Attachment 5 IEL Recount of Original Samples”, Fax of Feb-09-97; d) Item 8 of

Declaration of John G. Griggs, Chief, Monitoring and Analytical Services Branch, USEPA, NAREL; e) TMA Eberline Report, 1/10/92 for Betz Laboratories; f) S. Cohen and Associates, Radioanalytical Results, dated November 23, 1992; g) Obituary of Klerston Rose Mansfield; h) IEL Radiochemical Results, source unknown, handwritten notes indicating that a borehole soil sample was collected at a depth of 92 feet and contains detectable  $^{238}\text{Pu}$  and  $^{239+240}\text{Pu}$ .

12. Report from the Agency for Toxic Substances and Disease Registry (ATSDR), Department of Health and Human Services, April 5, 1994.
13. Additional data tables from analytical laboratories.
14. Additional data tables from analytical laboratories.
15. Additional data tables from analytical laboratories.
16. Letter dated 3/25/01 from Chris Borello, CCLT, to Ross del Rosario, Uniontown IEL RPM; reply of April 5, 2001 from Ross del Rosario, USEPA Region 5, SR-6J, to Chris Borello, RE: 3/25/01 Fascimile – 3<sup>rd</sup> round of PRP Radiation Testing.
17. Obituary of Klerston Rose Mansfield; discussion of Kittinger deposition (source/author unknown); additional data tables from analytical laboratories.
18. Excerpt of an unknown textbook on occurrence of radon and related radionuclides in water of various US states.
19. Letter of October 6, 2003 to Mike Cook and Christine Baughman, USEPA Inspector General, from Chris Borello, CCLT, RE: Uniontown IEL Superfund Site – Abandonment of 33 Wells.
20. Source unknown, “Table 8. Inventory of IEL Monitoring Wells and Recommendations for their Disposition”, includes tabulated analytical data and lab reports.

**Table 2. Environmental Results Indicating the Presence of Plutonium at the IEL**

Sample Name/Date Reference	Lab	Results	Table	1
Soil 5914E-66 IE-2-SD-90	CEP “invalidated”	$2.08 \pm 1.66$ pCi/g $^{239}\text{Pu}^a$	3	
Soil IE-1-B8-21-92	CEP “invalidated”	$0.007 \pm 0.013$ pCi/g $^{238}\text{Pu}$ $0.104 \pm 0.029$ pCi/g $^{239}\text{Pu}$	9	
Soil 5914E-51 IE-2-SD-11	CEP “invalidated”	$1.04 \pm 0.83$ pCi/g $^{241}\text{Pu}$ $1.97 \pm 1.29$ pCi/g $^{236}\text{Pu}$	12	
MW-01d 11/2000	EPA-NAREL	$< 0.32$ pCi/L $^{238}\text{Pu}$ $0.18$ pCi/L $^{239+240}\text{Pu}$ (MDA = 0.16)	9	
MW-011 11/2000	EPA-NAREL	$< 0.26$ pCi/L $^{238}\text{Pu}$ $0.23$ pCi/L $^{239+240}\text{Pu}$ (MDA = 0.22)	9	
MW-011 05/2001	EPA-NAREL	$0.45$ pCi/L $^{238}\text{Pu}$ (MDA=0.40) $< 0.34$ pCi/L $^{239+240}\text{Pu}$	9	
MW-23s 05/2001	EPA-NAREL	$0.31$ pCi/L $^{238}\text{Pu}$ (MDA=0.30) $0.17$ pCi/L $^{239+240}\text{Pu}$ (MDA=0.12)	9	
MW-26i 05/2001	EPA-NAREL	$0.36$ pCi/L $^{238}\text{Pu}$ (MDA=0.36) $< 0.22$ pCi/L $^{239+240}\text{Pu}$	9	
“Soil”	Unknown <sup>b</sup>	$0.016 \pm 0.013$ pCi/g $^{238}\text{Pu}$ $0.096 \pm 0.027$ pCi/g $^{239}\text{Pu}$	9	

<sup>a</sup>When analyzed by alpha spectrometry, the  $^{239}\text{Pu}$  and  $^{240}\text{Pu}$  peaks are unresolved; hence activity reported as  $^{239}\text{Pu}$  is actually the sum  $^{239+240}\text{Pu}$ . <sup>b</sup>The origin of these data are not known, but they are given in a

computer-generated table entitled "INDUSTRIAL EXCESS LANDFILL RADIOCHEMICAL RESULTS" along with values for two U and four Th isotopes.

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