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TO: Linda C. Modica
FROM: Michael E. Ketterer, Ph.D., Professor Emeritus, Chemistry and Biochemistry
SUBJECT: Report of anthropogenic Pu contamination in Erwin, Tennessee proximity samples collected in 2010

Michael.Ketterer@nau.edu

SUBMITTED BY:



Summary of experimental work. A series of soil and sediment samples were collected from Erwin and vicinity in order to examine whether the Erwin Nuclear Fuel Services facility has contributed to inventories of Pu in the ambient environment.

In August 2010, I worked with NAU student Ms. Kara Saaty and local community members to obtain samples from the following areas:

- * Pond sediment from the Erwin Linear Trail, near footbridge (ER-14)
- * Sediments from North Indian Creek, downstream of the NFS facility (ER-29)
- * Sediments from Davy Crockett Reservoir (DC-1, DC-2, DC-4, DC-5, DC-6)
- * Soil cores from a residential property in the vicinity of Davy Crockett Reservoir (DC-16)
- * Soil core samples from Martins Creek Cemetery in Erwin (ER-24, ER-25) and a former residential lot adjacent to the cemetery (ER-22)

These samples were dried, ground, and dry-ashed prior to obtaining sub-samples for lab analysis of Pu isotopes using inductively coupled plasma mass spectrometry (ICPMS). Samples were leached with nitric acid in the presence of a Pu-242 yield tracer and filtered; after adjustment to Pu(IV) oxidation state, Pu was isolated using TEVA resin. Fractions were then analyzed to determine $^{239+240}\text{Pu}$ activities and the atom ratios $^{240}\text{Pu}/^{239}\text{Pu}$ and $^{241}\text{Pu}/^{239}\text{Pu}$. Additional information about the Pu measurements is given in Ketterer and Szechenyi (2008).

Findings. Results for the $^{239+240}\text{Pu}$ activities and the atom ratios $^{240}\text{Pu}/^{239}\text{Pu}$ and $^{241}\text{Pu}/^{239}\text{Pu}$ are given in Table 1.

Two main observations are evident. First, it is clear that NFS has historically released non-fallout plutonium into the Nolichucky River watershed. Plutonium of identifiably non-fallout origin is detected in sediments of North Indian Creek, of the Erwin Linear Trail pond, and downstream as far as the Davy Crockett Dam. Sediments at North Indian Creek and the Erwin Linear Trail pond exhibit $^{239+240}\text{Pu}$ activities higher than can be reasonably anticipated from background (fallout) sources. Second, it is also evident that NFS has released airborne Pu, that can be detected in soil cores collected from a cemetery adjacent to the facility.

The plutonium from NFS cannot be explained by North American fallout sources, either from the Nevada Test Site or “stratospheric fallout” from Soviet/US nuclear tests in the 1950’s and 1960’s. The measured $^{240}\text{Pu}/^{239}\text{Pu}$ of NFS-affected samples is quite different from that of either Nevada or stratospheric fallout, although the observed $^{240}\text{Pu}/^{239}\text{Pu}$ ratios could conceivably stem from two-component mixing of these two sources (refer to Kelley *et al.*, 1999). However, this cannot be the case when additional information is included *via* the $^{241}\text{Pu}/^{239}\text{Pu}$ atom ratio. Since ^{241}Pu decays with a half-life of 14 years to ^{241}Am , the $^{241}\text{Pu}/^{239}\text{Pu}$ ratio represents a chronometer that reveals the younger (i.e., post-stratospheric testing era) origin of this Pu in NFS-affected samples. Note the manner in which the Erwin-vicinity samples fall significantly off the Nevada-stratospheric fallout mixing line (Kelley *et al.*, 1999), as shown in Figure 1. In this work, the Nevada-stratospheric fallout mixing line ratios were measured for the author’s own authentic samples under the same mass spectrometric conditions as the Erwin samples, and this mixing line measured on the NAU instrument agrees with Kelley’s (1999) published results.

These findings demonstrate, to a reasonable degree of scientific certainty, the presence of an environmental plume of plutonium in Erwin and in the Nolichucky watershed that originates from previous operations at the Erwin, TN Nuclear Fuel Services facility.

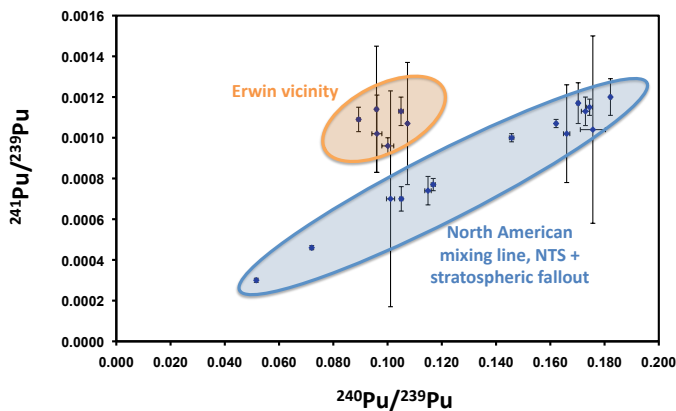


Figure 1. Pu mixing line of $^{241}\text{Pu}/^{239}\text{Pu}$ vs. $^{240}\text{Pu}/^{239}\text{Pu}$ reveals non-fallout origin of environmental Pu detected near Erwin. The blue mixing domain represents samples from US locations spanning mixtures of NTS and stratospheric fallout components. The highest activity Erwin Linear Trail pond sediment provides the best estimate of the isotope composition of NFS-originating Pu.

Sample ID	Description	²³⁹⁺²⁴⁰ Pu Bq/kg	²⁴⁰ Pu/ ²³⁹ Pu	sd 240239	²⁴¹ Pu/ ²³⁹ Pu	sd 241239
ER-14	Erwin Linear Trail pond sediment	28.7	0.089	0.001	0.00109	0.00006
ER-29	North Indian Creek sediment	1.91	0.096	0.002	0.00102	0.00019
ER-22	Residence, soil, near cemetery	0.864	0.147	0.006	Not measured	
ER-24	Soil core, Martins Creek Cemetery	0.375	0.145	0.005	Not measured	
ER-25	Soil core, Martins Creek Cemetery	0.677	0.148	0.007	Not measured	
DC-1	Davy Crockett Lake sediment	0.14	0.095	0.001	0.00079	0.00054
DC-2	Davy Crockett Lake sediment	0.133	0.1	0.002	0.00096	0.00004
DC-4	Davy Crockett Lake sediment	0.149	0.101	0.002	0.0007	0.00053
DC-5	Davy Crockett Lake sediment	0.141	0.105	0.001	0.00113	0.00007
DC-6	Davy Crockett Lake sediment	0.121	0.096	0.001	0.00114	0.00031
DC-16	Davy Crockett lake proximity soil core	0.228	0.107	0.001	0.00107	0.0003

Table 1. ²³⁹⁺²⁴⁰Pu activity (Bq/kg) and atom ratios in Erwin-vicinity soil and sediment samples analyzed at Northern Arizona University.

Literature Cited

Kelley, J.M.; Bond L.A.; Beasley, T.M. "Global distribution of Pu isotopes and ²³⁷Np", *The Science of the Total Environment* **1999**, 237/238:483-500.

Ketterer, M.E.; Szechenyi, S.C., "Review: Determination of plutonium and other transuranic elements by inductively coupled plasma mass spectrometry: A historical perspective and new frontiers in the environmental sciences", *Spectrochimica Acta B* **2008**, 63, 719-737.