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SUBJECT: Source of Neptunium-237 in Air Monitor A41A (Zahn's Corner)

The subject of this memorandum is to share some new insights with the Pike County General Health District, with respect to the origin of the Neptunium-237 (^{237}Np) that was recently reported by the US Department of Energy, at the Zahn's Corner Middle School monitoring location. The conclusions on Air Monitor A41A are augmented by a similar analysis of other data taken from recent DOE ASER documents. This analysis solely uses DOE data, and known reliable references (Moody, 1996; Kelley *et al.*, 1999) that describe the anticipated $^{237}Np/^{239}Pu$ and $^{240}Pu/^{239}Pu$ signatures from PORTS and global fallout.

To a reasonable degree of scientific certainty, the implied source of the ^{237}Np at Zahn's Corner is fugitive dust emissions from the PORTS facility, and not nuclear weapons testing fallout.

As the Pike County General Health District and the local community are aware, the explanation has been offered, to the effect that, the ^{237}Np most likely originates *via* 1950's-1960's "stratospheric fallout" from atmospheric nuclear tests. ***This statement is a hypothesis that is readily tested.*** The air filter sample results in question, from Monitor A41A, is shown on Page 2-24 of the DOE 2017 Annual Site Environmental Report for the Portsmouth Gaseous Diffusion Facility.

The results on Page 2-24 report that ^{237}Np was not detected in three out of four samples from Location A41A, being present in the fourth (*via* process of elimination) at 1.5E-4 pCi/m^3 (i.e., 0.00015 pCi/m^3). The same Table also reports that $^{239+240}Pu$ was not detected in four of four samples at Location A41A, with a maximum detection limit of 5.0E-6 pCi/m^3 (0.0000050 pCi/m^3) being cited. As is stated in the footnotes to the Table, in situations where a substance is not detected, the detection limit itself represents a reasonable estimate of the activity actually present.

The respective airborne activities, in pCi/m^3, of ^{237}Np and $^{239+240}$Pu generate an activity ratio, ^{237}Np/$^{239+240}$Pu of approximately 30 for the sample from A41A where ^{237}Np was detected.

If one assumes a "null hypothesis" (H_0) that the ^{237}Np originates from weapons testing fallout, the ^{237}Np/$^{239+240}$Pu activity ratio should reasonably agree with the ratio observed for this imputed fallout source. A reliable ^{237}Np/$^{239+240}$Pu activity ratio for Northern Hemisphere fallout can be calculated from the data of Kelley *et al.* (1999), a DOE-funded study that measured ^{237}Np/^{239}Pu and ^{240}Pu/^{239}Pu atom ratios in a suite of fallout-containing soils from worldwide locations.

Kelley *et al.* (1999) found consistent ^{237}Np/^{239}Pu and ^{240}Pu/^{239}Pu atom ratios of 0.48 and 0.180, respectively. Using the atom ratios and the known half-lives of these isotopes, the Kelley *et al.* (1999) results generate a fallout ^{237}Np/$^{239+240}$Pu activity ratio of 0.0032.

Notwithstanding the lack of a statistically robust dataset of DOE measurements at A41A, it is immediately apparent that the $(^{237}Np/^{239+240}Pu)_{A41A}$ of 30 greatly exceeds the $(^{237}Np/^{239+240}Pu)_{fallout}$ by ~ four orders of magnitude. It is, therefore, apparent that H_0 must be rejected and therefore, ***a different source of elevated ^{237}Np/$^{239+240}$Pu, besides fallout, must be present.***

The Moody (1995) DOE-funded study of authenticated materials collected within the Portsmouth Gaseous Diffusion Facility reveals a $(^{237}Np/^{239+240}Pu)_{PORTS}$ >> $(^{237}Np/^{239+240}Pu)_{fallout}$.

Given the geographic proximity of the A41A location to PORTS, the obvious disagreement of the A41 sample with $(^{237}Np/^{239+240}Pu)_{fallout}$, the implied $(^{237}Np/^{239+240}Pu)_{PORTS}$, and the lack of any other plausible sources of ^{237}Np in the vicinity, one concludes to a reasonable degree of scientific certainty, that ***PORTS is the source that accounts for the ^{237}Np measured in the 2017 A41A air sample.***

Another way of viewing the rejection of H_0 is as follows: if the A41A ^{237}Np detection of 0.00015 pCi/m^3 was accounted for by fallout, ***the known $(^{237}Np/^{239+240}Pu)_{fallout}$ would imply that the A41 airborne activity of $^{239+240}$Pu would have to be 0.047 pCi/m^3, if fallout were the accountable source of the ^{237}Np.*** This is clearly not the case.

This conclusion is also supported by additional evidence recently reported by the DOE. One can consider that sediment in the creeks draining the facility also exhibit non-fallout, elevated (^{237}Np/$^{239+240}$Pu). The 2017 ASER report does not contain sufficient information to rigorously evaluate $(^{237}Np/^{239+240}Pu)_{sediment}$. Nevertheless, as seen on Page 4-21, some insights can be garnered.

Page 4-21 reports a ^{237}Np activity of 0.00975 pCi/g in a sediment sample from Big Beaver Creek, and reports that the only detection of $^{239+240}$Pu was found in a different location (RM-10S, 0.00961 pCi/g). If one assumes, as is reasonable, that the Big Beaver Creek sediment contains $^{239+240}$Pu ≤ 0.00961 pCi/g, and accordingly, the $(^{237}Np/^{239+240}Pu)_{sediment}$ is ~ unity or greater, which confirms that the PORTS facility has been discharging non-fallout ^{237}Np into the sediments.

An additional data point illustrating, in this case, an airborne pathway of ^{237}Np release from PORTS can be found in the 2016 ASER report. On Page 4-15, results are reported for a soil sample collected adjacent to air monitoring location A12 (east of PORTS on McCorkle Road). This soil had ^{237}Np and $^{239+240}Pu$ activities, respectively, of 0.0197 and 0.0116 pCi/g, respectively. Once again, it is not plausible for the ^{237}Np in the A12 soil to originate exclusively from fallout, as the measured ^{237}Np activity (if from fallout) would require a corresponding $^{239+240}Pu$ activity of 6.2 pCi/g. ***Instead, there is no plausible explanation, other than airborne releases from PORTS, as the source of the vast majority of the ^{237}Np detected in the McCorkle Road soil.***

Literature Cited

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

Moody, K.J., "Forensic Radiochemistry of PUBLIC Site Inspection Samples", Lawrence Livermore National Laboratory **1995**, UCRL-ID-119658.

US Department of Energy, "Portsmouth Gaseous Diffusion Plant: Annual Site Environmental Report - 2016", **2018**, PPPO-03-0813&D1.

US Department of Energy, "Portsmouth Gaseous Diffusion Plant: Annual Site Environmental Report - 2017", **2019**, PPPO-03-5263593-19.

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