Impact of natural organic matter on plutonium vadose zone migration from an NH$_4$PuO$_2$CO$_3$(s) source

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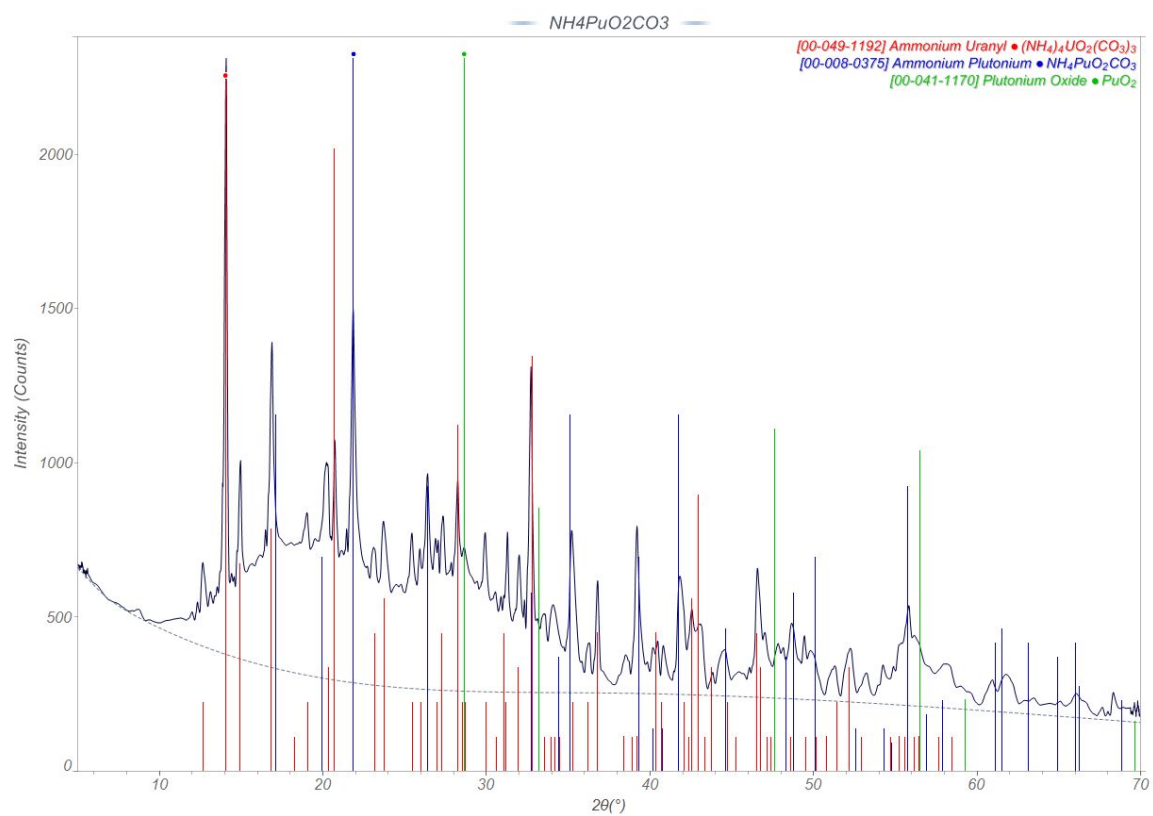
[c] Glenn T. Seaborg Institute, Physical & Life Sciences, Lawrence Livermore National Laboratory, Livermore, CA 94550, USA

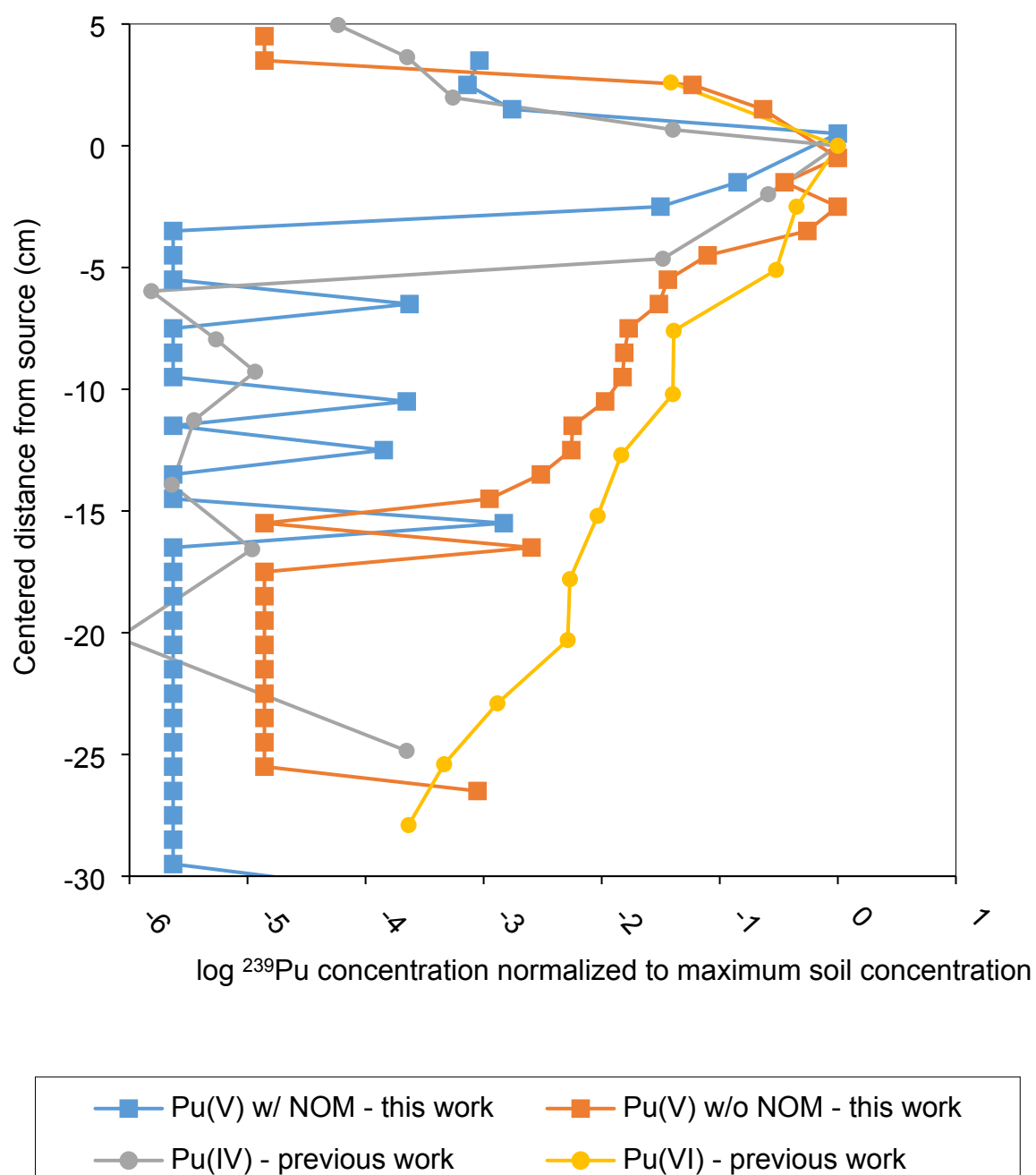[d] Savannah River National Laboratory, Aiken, SC, USA

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ONLINE SUPPORTING MATERIAL

Content: 9 pages including cover sheet

Figure S1: XRD patterns of the initial $NH_4PuO_2CO_3(s)$ realized after synthesis.

Figure S2: Normalized soil plutonium concentrations in the lysimeters containing $NH_4PuO_2CO_3(s)$ sources with and without organic matter as a function of distance from source. Data are compared with previous lysimeters studies deploying Pu(IV) and Pu(VI) sources reported by Kaplan et al.[1]. Detection limits for the OM amended lysimeter and OM free lysimeters are 2.3×10^{-6} and 1.4×10^{-5}, respectively.
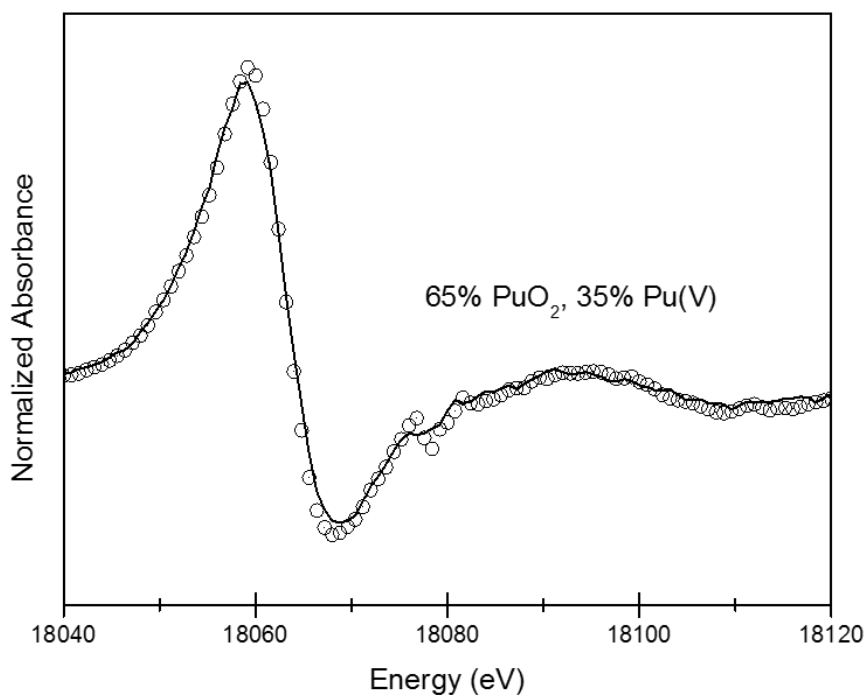
Figure S3: Linear combination fit of the normalized XANES spectrum (first derivative) of the archived NH$_4$PuO$_2$CO$_3$ source after 3 years using the solvent extraction results and Pu(IV) and Pu(V) as references. Rfactor = 0.008.
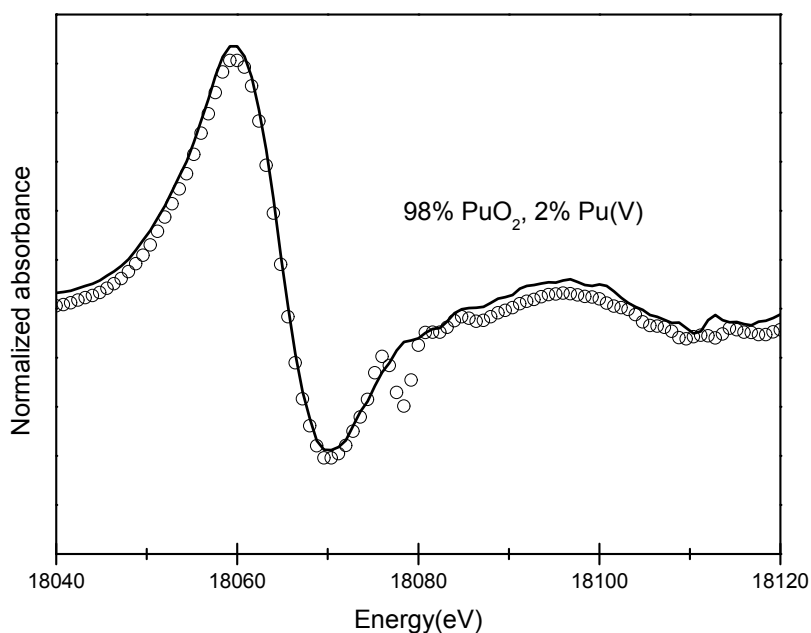

Figure S4: Linear combination fit of the normalized XANES spectrum (first derivative) of the NH$_4$PuO$_2$CO$_3$ source exposed to NOM amended lysimeter for 3 years using the solvent extraction results and Pu(IV) and Pu(V) as references. Rfactor = 0.008.
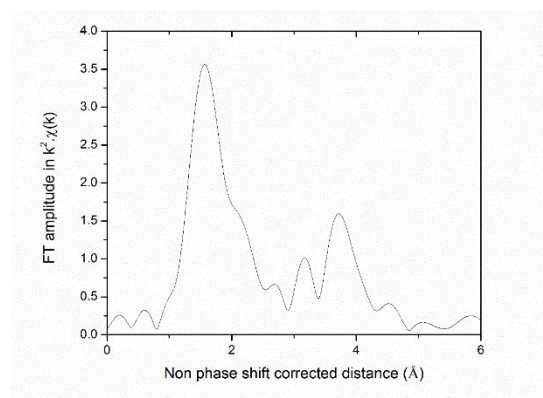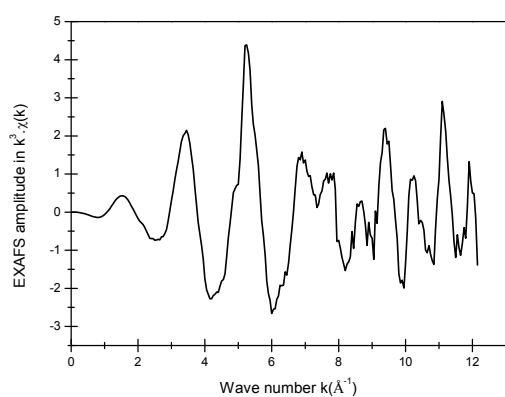
**Figure S5**: EXAFS spectra and its corresponding Fourier transform of the archived $NH_4PuO_2CO_3$ source after 3 years of storage in an inert atmosphere.
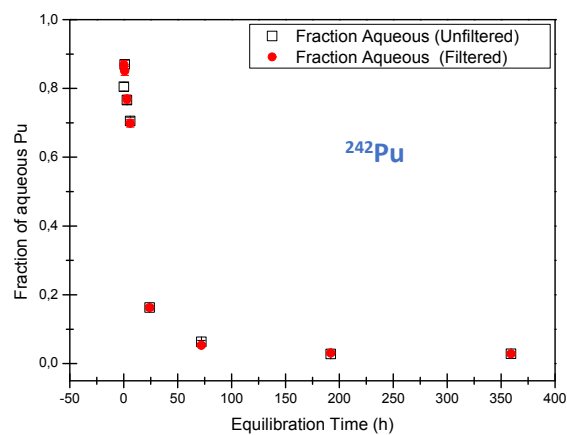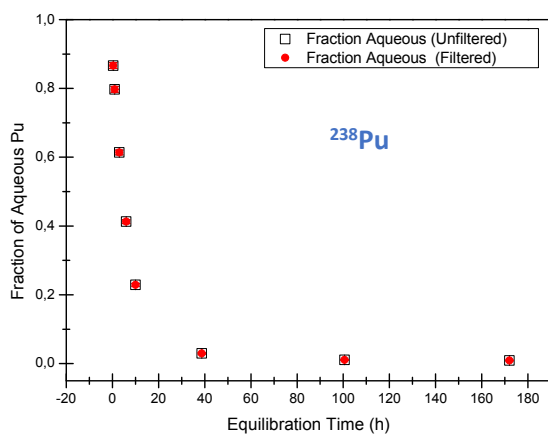

Figure S6: Percentage of Pu remaining in the aqueous phase of the unamended soil suspension, during the sorption step, before and after filtration (3K, MWCO) as a function of time. The sorption was performed at $[^{238}Pu] = 10^{-10}$ M (left) and at $[^{242}Pu] = 3 \times 10^{-7}$ M (right). Suspended solids concentration $= 25g.L^{-1}$.
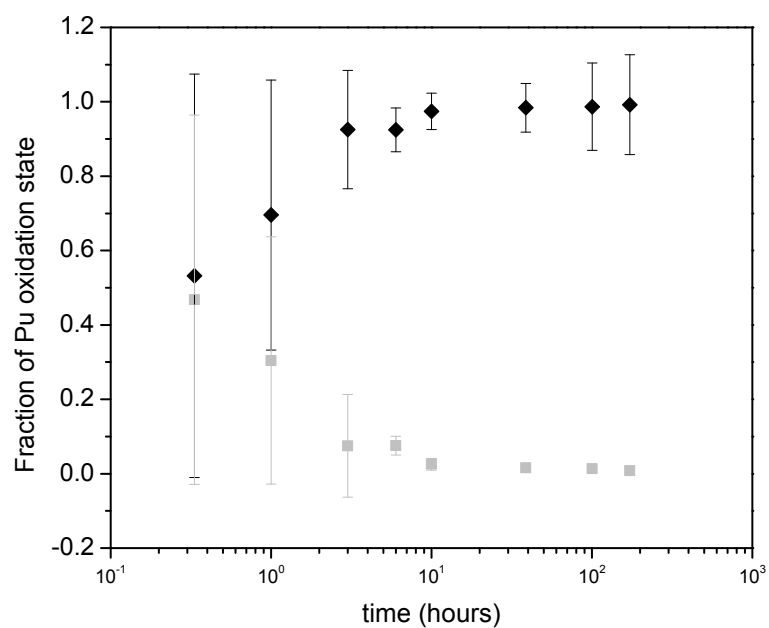
Figure S7: Oxidation state distribution of plutonium (^{238}Pu) leached from unamended soil (suspended solid = 25 g.L^{-1}) as a function of sorption time. Pu(V) fraction is represented by the gray squares and Pu(IV) fraction by the black diamonds.
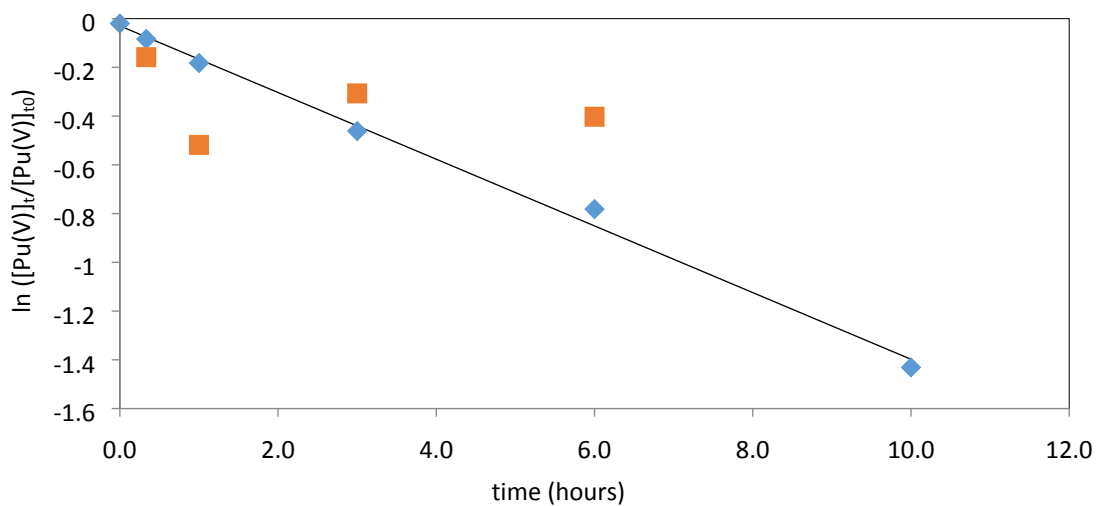
Figure S8: Pseudo-first-order reduction calculation at [^{238}Pu] = 10^{-10}M (blue) and [^{242}Pu] =3 x 10^{-7} M (orange).
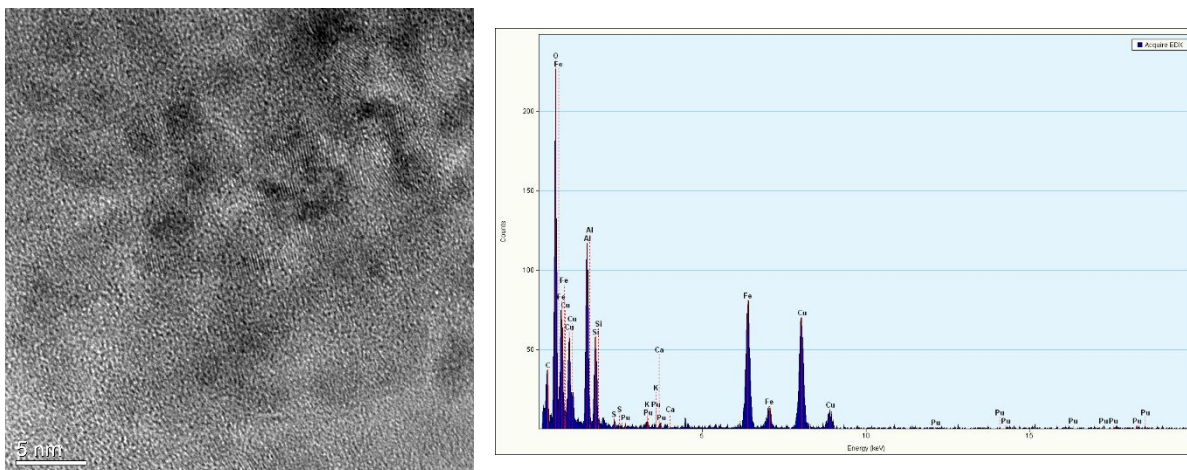
Figure S9: TEM image and associated EDX spectrum of a soil suspension with presence of iron in the phyllosilicates.
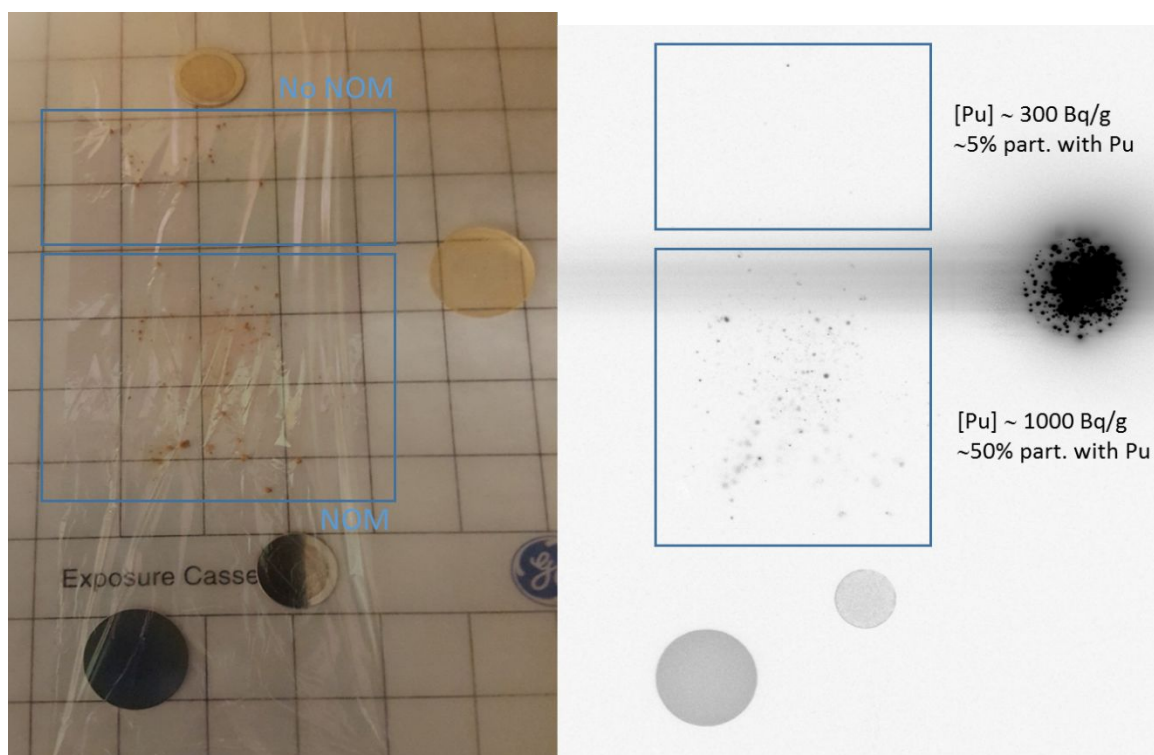
Figure S10: Autoradiogaphy image of soil from lysimeters

REFERENCES

1.   Kaplan, D. I.; Powell, B.; Demirkanli, D. I.; Fjeld, R. A.; Molz, F. J.; Serkiz, S. M.; Coates, J. T., Influence of Oxidation States on Plutonium Mobility during Long-Term Transport through an Unsaturated Subsurface Environment. *Environmental Science and Technology* **2004,** *38*, 5053-5058.