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Natural Organic Matter and Microbial Controls on Mobilization/Immobilization of I and Pu in Soils and Waters Affected by Radionuclide Releases in USA and Japan.

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Examining the short-term deposition and forecasting the long-term fate of plutonium (Pu) is becoming increasingly important as more worldwide military and nuclear-power waste is being generated. Soils from the Fukushima Prefecture provided the opportunity to compare Pu from reactor fallout in the litter layer to that derived from 1960's bomb fallout in the in the underlying soil. Additionally, we used this unique opportunity to explore the long-term behavior of Pu in Nagasaki sediments, where bomb-derived Pu was deposited in 1945. A combination of selective extractions and molecular characterization via electrospray ionization Fourier-transform ion cyclotron resonance mass spectrometry (ESI-FTICRMS) were used in an attempt to resolve what regulated the long-term stabilization of ^{239,240}Pu in Nagasaki bomb residue-containing sediments (>400 cm) and the short-term deposition of nuclear plant accident-derived ^{239,240}Pu in Fukushima Prefecture (<12 cm). In deep Nagasaki sediments, our results demonstrated that 55±3% of the ^{239,240}Pu was preferentially associated with more persistent organic matter compounds, particularly those natural organic matter (NOM) stabilized by Fe oxides ($NOM_{Fe\text{-}oxide}$). Other organic matter compounds served as the secondary sink of these bomb-derived ^{239,240}Pu (31±2% on average), and less than 20% of the bomb-derived ^{239,240}Pu was immobilized by the inorganic mineral particles. In a 9-cm thick, ^{239,240}Pu-enriched layer at ~400cm depth, N-containing carboxyl aliphatic and/or alicyclic molecules (CCAM) in $NOM_{Fe\text{-}oxide}$ and other NOM fractions immobilized the majority of ^{239,240}Pu. Among the cluster of N-containing CCAM moieties, hydroxamate siderophores, the strongest known Pu chelators in nature, were further detected in these "aged" Nagasaki bomb residue-containing sediments. In Fukushima Prefecture and Nagasaki sediments, the NOM still served as the predominant sink for these nuclear plant accident-dervied ^{239,240}Pu (sum of $NOM_{Fe\text{-}}$ oxide and other organic matter). While present long-term disposal and environmental remediation modeling assume that solubility limits and sorption to mineral surfaces control Pu subsurface mobility, our observations suggest that NOM, which is present in essentially all environmental systems, undoubtedly plays an important role in sequestrering Pu. Ignoring the role of NOM in controlling Pu fate and transport is not justified in most environmental systems*.

References:

* Lin, P., Xu, C., Kaplan, D.I., Chen, H.M., Sun, L., Yeager, C.M., Xing, W., Sun, L., Schwehr, K.A., Yamazaki, H., Saito-Kokubu, Y., Hatcher, P.G., Santschi, P.H. 2018. Nagasaki sediments reveal that long-term fate of plutonium is controlled by select organic matter moieties. STOTEN, submitted.