

Molecular Characterization of Organic Compounds Binding $^{239,240}\text{Pu}$ in Nagasaki Soils

Peng Lin¹, Chen Xu¹, Daniel I. Kaplan², Chris M. Yeager³, Kathleen A. Schwehr¹, Peter H. Santschi¹

¹ Department of Marine Science, Texas A & M University at Galveston, Galveston, Texas 77553, United States

² Savannah River National Laboratory, Aiken, South Carolina 29808, United States

³ Los Alamos National Laboratory, Los Alamos, New Mexico 87545, United States

To investigate the organic compounds responsible for the immobilization and binding of bomb-derived plutonium (Pu)-239,240 in Nagasaki soil environments, soil samples containing the highest abundance of $^{239,240}\text{Pu}$ (230-266 Bq/kg) and relatively higher organic matter concentration (1.97%-2.30% for TOC, 0.31%-0.35% for TN) in one Nagasaki soil core were sequentially extracted into two organic fractions, including NaOH-extractable and Fe,Mn-oxide-bound (sodium dithionite, pH=8/NaOH extract) organic matter (> 3 kDa), followed by the quantification of $^{239,240}\text{Pu}$ activity concentration in each fraction. We found that $^{239,240}\text{Pu}$ was predominantly enriched in Fe,Mn-oxide-occluded organic matter fraction, accounting for 55±3% on average of the total soil $^{239,240}\text{Pu}$. In comparison, NaOH-extractable organic matter bound 31±1% of total soil $^{239,240}\text{Pu}$. Purified NaOH-extractable and Fe,Mn-oxide occluded organic matter were both further characterized by ultrahigh resolution ElectroSpray Ionization Fourier-transform Ion Cyclotron Resonance Mass Spectrometry (ESI-FTICR-MS) to examine organic moieties responsible for binding $^{239,240}\text{Pu}$ in Nagasaki soils. Additionally, macromolecules which bind the majority of $^{239,240}\text{Pu}$ in each organic fraction were further separated through an isoelectric focusing (IEF), followed by characterization of ESI-FTICR-MS and comparison with bulk organic matter fractions. The present study provides molecular-level evidence for the important role of selected organic compounds in $^{239,240}\text{Pu}$ immobilization/remobilization in Nagasaki soil environments.