

**Molecular Characterization of
Natural Organic Matter Binding
^{239,240}Pu in the northwestern Fukushima
Prefecture, Japan**

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Plutonium (Pu)-239,240 derived from Fukushima Dai-ichi Nuclear Power Plant (FDNPP) accident and global fallout has been found to be significantly associated with soil organic matter (SOM) in litter and surface layers of northwestern Fukushima Prefecture, even three years after the FDNPP accident-derived emissions. To investigate the potential mobility of ^{239,240}Pu and the organic compounds binding the ^{239,240}Pu in Fukushima Prefecture environments, litter and surface samples in two Fukushima soil cores were re-suspended in artificial groundwater, followed by sequential extraction into two immobile fractions, including Fe,Mn-oxide and its associated organic matter and other organic matter fractions. Our results indicate that the ^{239,240}Pu in Fukushima Prefecture has low mobility, with <1% of ^{239,240}Pu being released into artificial groundwater. Most of immobile ^{239,240}Pu was predominantly enriched in Fe,Mn-oxide and its associated organic matter, accounting for 48±8% on average of the total soil ^{239,240}Pu. In comparison, other organic matter-bound ^{239,240}Pu accounted for 13±7% of total soil ^{239,240}Pu, leaving 39±14% of ^{239,240}Pu not being extracted and bound to residual inorganic mineral particles in Fukushima Prefecture. Purified NaOH-extractable and Fe,Mn-oxide associated organic matter were both further characterized by ElectroSpray Ionization Fourier-transform Ion Cyclotron Resonance Mass Spectrometry (ESI-FTICR-MS) to examine organic moieties immobilizing ^{239,240}Pu in Fukushima Prefecture. The present study not only establishes that FDNPP accident-derived ^{239,240}Pu has low mobility, but also explains the reasons why the SOM can strongly immobilize the ^{239,240}Pu in Fukushima Prefecture.