## **Role of natural organic matter** on iodine and Pu distribution and mobility in environmental samples from the northwestern Fukushima Prefecture

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To assess how environmental factors affect the distribution and transportation of radioiodine and plutonium that were emitted from the Fukushima Dai-ichi Nuclear Power Plant (FDNPP) accident, we quantified iodine and <sup>239,240</sup>Pu concentration changes in soil samples with different land use types (paddy, urban, deciduous forest and coniferous forest), as well as iodine speciation in river water and rainwater, at locations 53 to 63 km northwest of the FDNPP. A ranking of the land uses by their surface soil stable <sup>127</sup>I concentrations was coniferous forest> deciduous forest> urban > paddy, and <sup>239,240</sup>Pu concentrations ranked as deciduous forest> coniferous forest> paddy ≥ urban. Both were quite distinct from that of <sup>134,137</sup>Cs: urban > coniferous forest > deciduous forest> paddy, indicating differences in the sources, deposition phases, and biogeochemical behaviour in these soil systems. Surficial soil <sup>127</sup>I content was well correlated to soil organic matter (SOM) content, regardless of land use type, suggesting that SOM might be an important factor affecting iodine biogeochemistry. Organic carbon (OC) concentrations and Eh were positively, and pH was negatively correlated to <sup>127</sup>I concentrations in surface water and rain samples. It is also noticeable that both forest throughfall and stemflow water consisted exclusively of organo-iodine, suggesting all inorganic iodine in the original bulk deposition (~28.6 % of total iodine) have been completely converted to organo-iodine.

Fukushima-derived Pu is detectable at a distance ~61 km northwest of FDNPP, yet still confined to the litter layer in the forest soil. Pu concentrations were significantly correlated with soil OC and nitrogen contents, indicating Pu may be associated with nitrogen-containing SOM. Together, these finding suggest that natural organic matter (NOM) plays a key role in affecting the fate and transport of I and Pu and may warrant greater consideration for predicting long-term stewardship of contaminated areas and evaluating various remediation options in Japan.