## Analytical methodology and potential applications of a new geochemical tracer of Cs-135/Cs-137 isotope ratio

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The Fukushima Daiichi Nuclear Power Plant (FDNPP) accident released large amounts of radionuclides into the environment. In addition to the atmospheric releases, the release of highly contaminated water from the damaged reactor buildings resulted in the direct discharges of radionuclides into the sea. It has been estimated that the total amount of <sup>137</sup>Cs released from the FDNPP accident to the atmosphere ranged from 15-35 PBq. The released radioactive materials posed radiation threat to human society. Thus, environmental nuclear forensics of radioactive contamination and long-term environmental behavior of released radioactive materials have been important issues of study after the FDNPP accident.

Since the FDNPP accident, intensive studies on the distribution of released fission products, in particular  $^{134}\mathrm{Cs}$  and  $^{137}\mathrm{Cs}$ , in the environment have been conducted, and the activity ratio of <sup>134</sup>Cs/<sup>137</sup>Cs has been widely used as a tracer for contamination source identification. However, due to the short halflife of  ${}^{134}$ Cs (2.06 y), this tracer will become unavailable in the near future. Thus, for studies on the long-term environmental behavior, such as the fate and transport of the released radionuclides, new tracers which can be used in a time scale of several tens- or hundred years are highly required. In this work, we discuss new geochemical tracer of radioactive Cs isotope ratios (135Cs/137Cs) for contamination source identification and long-term environmental behavior studies of Pu and Cs released from the FDNPP accident. To establish the feasibility of the proposed tracer, we identified the isotopic compositions of radioactive Cs and Pu isotopes released from the FDNPP accident, and estimated the released amounts of <sup>135</sup>Cs and Pu isotopes. New development of mass spectrometric methodology for <sup>135</sup>Cs/<sup>137</sup>Cs ratio analysis and the potential applications of the obtained FDNPP-sourced Cs fingerprint in the study of long-term environmental behavior of the released radioactive materials will be discussed.