

Using radiosilver and plutonium isotopes to trace the dispersion of contaminated sediment in Fukushima coastal catchments

Olivier Evrard<sup>a</sup>, Fabien Pointurier<sup>b</sup>, Yuichi Onda<sup>c</sup>, J. Patrick Lacey<sup>a</sup>, Hugo Lepage<sup>a</sup>, Caroline Chartin<sup>d</sup>,  
Maeva Cirella<sup>b</sup>, Anne-Claire Pottin<sup>b</sup>, Amélie Hubert<sup>b</sup>, Irène Lefèvre<sup>a</sup>, Sophie Ayrault<sup>a</sup>

<sup>a</sup> *Laboratoire des Sciences du Climat et de l'Environnement (LSCE/IPSL) – Unité Mixte de Recherche 8212 (CEA-CNRS-UVSQ) – 91 198 Gif-sur-Yvette Cedex (France)*

<sup>b</sup> *CEA, DAM, DIF, 91297 Arpajon (France)*

<sup>c</sup> *Center for Research in Isotopes and Environmental Dynamics (CRIED), Tsukuba University, Tsukuba (Japan)*

<sup>d</sup> *TECLIM, Université catholique de Louvain (Belgium)*

The Fukushima Dai-ichi nuclear power plant (FDNPP) accident in March 2011 resulted in a 3000-km<sup>2</sup> radioactive pollution plume consisting predominantly of radiocesium (<sup>137</sup>Cs and <sup>134</sup>Cs). This plume is drained by several rivers to the Pacific Ocean after flowing through less contaminated, but densely inhabited coastal plains. As the redistribution of radionuclide contaminated sediment could expose the local population to higher radiation rates, novel fingerprinting methods were developed to trace the downstream dispersion of contaminated sediment.

First, the heterogeneous deposition of metastable silver-110 (<sup>110m</sup>Ag) across these coastal catchments was used to investigate sediment migration. In particular, the <sup>110m</sup>Ag/<sup>137</sup>Cs activity ratio was measured in soils and river sediment demonstrating the occurrence of a seasonal cycle of soil erosion during typhoons and spring snowmelt in 2011 and 2012. However, due to the rapid decay of <sup>110m</sup>Ag (half-life of 250 days), alternative methods were required to continue tracking sediment from 2013 onwards. One promising method includes the analyses of plutonium isotopes to further understand sediment migration in the Fukushima region. For example, <sup>241</sup>Pu/<sup>239</sup>Pu atom ratios measured in sediment collected in Fukushima coastal rivers shortly after the accident were shown to be significantly higher (0.0017 – 0.0884) than corresponding values attributed to the global fallout (0.00113 ± 0.00008). Additional analyses were conducted on sediment sampled in 2013 and 2014 after the start of decontamination works. These analyses show that the <sup>241</sup>Pu/<sup>239</sup>Pu atom ratios decreased towards the global fallout values in rivers draining decontaminated paddy fields, demonstrating the effectiveness of remediation works.