

Speciation analysis of the Fukushima accident derived I-129 in the soil using sequential extraction method

HONDA, Maki^{1*} ; MATSUZAKI, Hiroyuki² ; SAITOU, Takumi³ ; NAGAI, Hisao⁴

¹Graduate School of Integrated Basic Sciences, Nihon University, ²Department of Nuclear Engineering and Management, School of Engineering, The University of Tokyo, ³Nuclear Professional School, School of Engineering, The University of Tokyo, ⁴College of Humanities and Sciences, Nihon University

In previous study, we investigated the depth profile of the accident derived ¹²⁹I ($T_{1/2} = 1.57 \times 10^7$ y) and downward migration speed in soils of near field of Fukushima Dai-ichi Nuclear Power Plant (FDNPP), including crop fields and man-made fields. ¹²⁹I in soil was measured by AMS and stable iodine (¹²⁷I) was measured by ICP-MS at MALT (Micro Analysis Laboratory, Tandem accelerator), The University of Tokyo. It was found that ¹²⁹I was concentrated near surface but distributed deeper compared with ¹³⁷Cs ($T_{1/2} = 30$ y). From the estimation of relaxation length using depth profiles, the FDNPP derived ¹²⁹I move 0.6 cm/y downward and ¹³⁷Cs 0.3 cm/y for it. It was also found that ¹²⁹I seems to move downward more quickly than ¹³⁷Cs.

To investigate the adsorption mechanism and the elemental process of migration of the accident derived ¹²⁹I in soil, it is important to know what kind of component the ¹²⁹I combines with.

Recent studies on the X-ray absorption fine structure (XAFS), especially near edge structure (XANES), reported that the stable iodine (¹²⁷I) in soil existed as an organic component^[1]. However, it had not yet been proved that it was also the case with the accident derived ¹²⁹I because it had been incorporated in the soil system only recently and the abundance of ¹²⁹I in soil was more than 8 orders of magnitude smaller than sub-ppm level stable iodine (¹²⁷I).

In this study a progressive sequential extraction method including the dialysis was newly developed to obtain only the iodine sticking to the soil organic component. The advantage of sequential extraction over other method is that stable iodine can be quantified by direct analysis of the fraction and ¹²⁹I can be quantified by AMS method of the fraction added with carrier. The fraction of the organic component for ¹²⁷I and ¹²⁹I can be evaluated respectively by comparing with the other fraction and/or with the total concentration obtained by the bulk analysis (e.g. by the pyrohydrolysis).

Repeatability is 20% for the water soluble, oxides and organic fraction, 10% for Exchangeable fraction and 50% Residue (mainly minerals).

The results show that 60% of the total ¹²⁹I are associated with oxides and 30% associated with organic matter in crop field soil. The former, the oxides bond iodine, it takes a form of iodate (IO_3^-) absorbed in amorphous oxides, especially goethite or delta- MnO_2 . They are formation of monodentate mononuclear outer-sphere species and bidentate, binuclear inner-sphere species^[2]. The latter iodine are linked to organic carbon directly by a covalent bond.

[1] Y. S. Shimamoto et al., 2011, *Environ. Sci. Technol.*, **45**, pp2086-2092

[2] T. Nagata et al., 2010, *Geochim. Cosmochim. Acta.*, **74**, pp6000-6013