Retrospective reconstruction of Iodine-131 distribution through the analysis of Iodine-129

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Abstract. Iodine-131 distribution released from the Fukushima Dai-ichi Nuclear Power Plant accident was reconstructed through the iodine-129 measurements. From nearly 1,000 surface soil samples iodine was extracted by the pyro hydrolysis method. Extracted iodine was then mixed with carrier, purified and finally collected as silver iodide. Silver iodide sample was pressed into the cathode holder and set at the ion source of the MALT facility, The University of Tokyo. The isotopic ratio $^{129}I/^{127}I$ was measured by means of Accelerator Mass Spectrometry. From ^{129}I data obtained, ^{131}I deposition map was constructed. There observed various fine structures in the map which could not estimated neither by the simulation nor ^{137}Cs distribution.

1 Introduction

Among various radioactive nuclides released from the Fukushima Daiichi Nuclear Power Plant (FDNPP) accident, Iodine-131 showed high radioactivity just after the accident. If taken into human body, Iodine-131 concentrates in the thyroid and may cause the thyroid cancer. The recognition about the risk of Iodine-131 dose originated from the experience of the Chernobyl accident based on the epidemiological study [1]. It is thus important to investigate the detailed deposition distribution of I-131 to evaluate the radiation dose due to I-131 and watch the influence on the human health. However I-131 decays so rapidly (half life = 8.02 d) that it cannot be detected several months after the accident. Long lived radioactive isotope I-129 (half life = 1.57E+7 yr,), which is also a fission product as well as I-131, is ideal proxy for I-131 because they are chemically identical. Near 1,000 soil samples collected around FDNPP site have been analyzed and I-131 map was constructed.

2 Experimental procedure

Soil samples were selected from the collection made by Ministry of Education, Culture, Sports, Science & Technology (MEXT), Japan. Experimental procedure is summarized in Figure 1. After homogenization, iodine was extracted by pyro hydrolysis method and trapped into an alkaline solution. Adding iodine carrier, purified by solvent extraction, and finally silver iodide (AgI) precipitation was made. Isotopic ratio of $^{129}/^{127}I$ was measured by Accelerator Mass Spectrometry at MALT, The University of Tokyo. The iodine concentration was determined by ICP-MS performed at Gakushuin University and MALT.

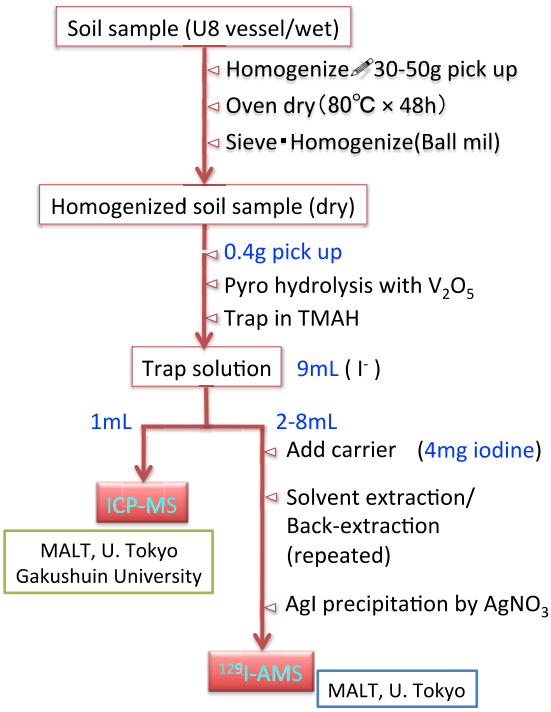

Figure 1. Experimental procedure.

The ^{129}I concentration in soil was calculated according to Equation (1):

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$$n_{129} = \frac{1}{w} R \, w n_{127} \frac{v_1 \; v_q}{v_1} \; b \, R_C b \; \frac{v_1}{v_1 \; v_q}, \qquad (1)$$

here, the each parameter denotes:

w: Dry weight of soil treated.
n_{127}: Stable iodine (^{127}I) concentration in soil.
n_{129}: ^{129}I concentration in soil.
v_1: Weight of trap solution (TMAH-base).
v_q: Aliquot weight for trap solution for ICP-MS.
b: Carrier amount added (as Iodine weight).
R: AMS result (^{129}I/^{127}I ratio = 10^{-12} - 10^{-10} range).
R_C: ^{129}I/^{127}I of the carrier (=1.7×10^{-13}).

The overall analytical error was determined by the AMS measurement which showed typically 1-3%.

3 Results and discussion

3.1 Reconstructed ^{131}I distribution map

Near 1,000 samples were analysed and made distribution map for iodine-131. For the conversion from ^{131}I to ^{129}I the estimated isotopic ration of ^{129}I/^{131}I in the FDNPP reactor was used [2, 3]. The reconstructed map is shown in Figure 2, 3.

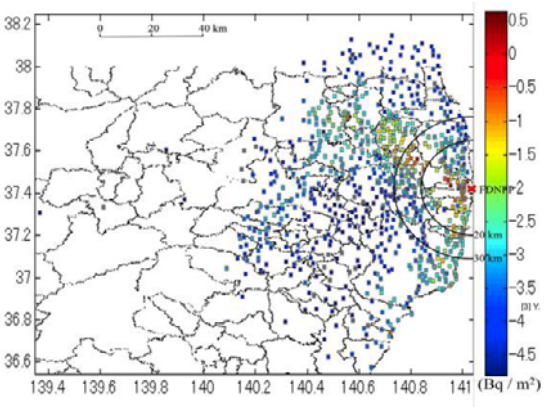


Figure 2. Reconstructed ^{131}I map.

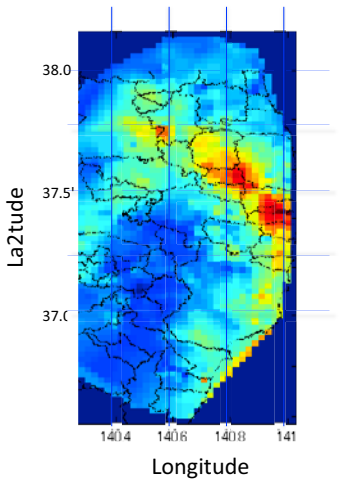
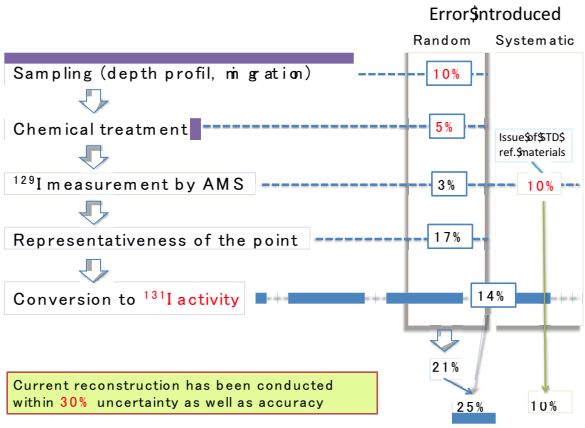


Figure 3. Reconstructed ^{131}I map converted to the mesh distribution for the convenience for the comparison with the simulation. The concentration scale is common with Figure 2.

3.2 Estimation of overall uncertainty

There are several factors that introduce uncertainty to the reconstructed value other than analytical error. Figure 4 shows the factors and introduced uncertainty. It was found that overall uncertainty should be less than 30%.



This is not bad compared to the ^{137}Cs map constructed by the direct measurements.

Figure 4. Estimation of overall uncertainty for the reconstruction of ^{131}I.

In Figure 4, the evaluation of the uncertainty accompanied with "sampling" referred to Honda et al. (2015) [4]. Schnetger and Muramats (1996) was referred for the chemical treatment especially for the extraction yield of iodine at pyro hydrolysis [5]. Issue of the standard reference material for ^{129}I-AMS is described in Matsuzaki et al. (2015) [6]. The "representativeness" is related to the local inhomogeneity of deposition density. In actual measurement, there is always occurred case that even two points close to each other show quite different activity. This causes difficult to evaluate the radiation dose of the residents.

References

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