

## 水-土壌系でのヨウ素の化学形態変化

### Transformation of iodine chemical forms in soil-water systems

嶋本 洋子<sup>1\*</sup>, 高橋 嘉夫<sup>1</sup>

Yoko Shimamoto<sup>1\*</sup>, Yoshio Takahashi<sup>1</sup>

<sup>1</sup>広島大学

<sup>1</sup>Hiroshima University

#### Introduction

Radioactive iodine is released from processing of nuclear fuel, nuclear accident, etc. To predict the fate of radioactive iodine, we have to consider two transport pathways: 1) via atmosphere caused by nuclear accident; 2) from deep underground emitted from underground nuclear-waste repositories. In the case 1, initial iodine chemical forms are  $\text{IO}_3^-$  and  $\text{I}^-$ , while  $\text{I}^-$  is the main initial form in the case 2. In order to compare the difference of initial inorganic iodine forms, column experiments and field observation were performed. Iodine chemical forms in soil and solution were determined by K-edge X-ray adsorption near edge structure (XANES) and high performance liquid chromatography connected to ICP-MS (HPLC-ICP-MS), respectively.

#### Experimental and sampling

Column experiment: The 150 g of dried paddy field soil were packed into the acrylic column. Fifty milliliters of 0.5%  $\text{IO}_3^-$  or  $\text{I}^-$  solution were added to the column. After all the solution flowed out, soil and pore water were taken at 1 cm intervals. The vertical profiles of concentration of iodine in soil and solution were obtained, and simulated using Visual MODFLOW. Adsorption isotherms were also obtained by batch sorption experiments.

Natural soil and water samples: Natural soil and pore water samples were collected at 0 to 12 cm depth in Yoro area, Chiba, Japan. The soil profile was flooded with brine water from a tube well containing iodine (Iodine: 5.8 mg/L). Iodine chemical forms of soil and pore water were determined. Anion exchange column and size exclusion column were equipped to HPLC system to separate  $\text{I}^-$ ,  $\text{IO}_3^-$ , and high molecular organic I. Elemental mapping of iodine in soil grain was obtained at SPring-8 BL37XU.

#### Result and discussion

Column experiment: Upon  $\text{I}^-$  infiltration through the column, that a small amount of  $\text{I}^-$  absorbed on soil. The profile of iodine concentration in pore water was well simulated by assuming equilibrium-controlled Langmuir type adsorption without considering any chemical transformations. For the  $\text{IO}_3^-$  addition system into the column,  $\text{IO}_3^-$  adsorbed to soil to a larger degree, which causes a much larger retardation effect than  $\text{I}^-$ . In addition, reduction of  $\text{IO}_3^-$  to  $\text{I}^-$  was also confirmed in both soil and pore water. The fraction of  $\text{I}^-$  increased toward the deeper end in both phases because of its lower affinity for soil than  $\text{IO}_3^-$ . The mobility of  $\text{I}^-$  is a dominant factor that controls the fate of iodine in the surface environment.

Natural soil and water samples: The depth profile of iodine concentration in soil correlates quite

well with that of organic carbon content. XANES analysis has revealed that iodine in soil exists as organic iodine at all depths. Iodine mapping of soil grain was obtained by micro-XRF analysis, indicates that iodine tends to be bound to organic matter more than clay or Fe rich mineral. Pore water at 0 - 6 cm depth contain 40 - 50% of organic iodine bound to dissolved humic substances and the rest of iodine as  $I^-$ , where as 98% of iodine was  $I^-$  at 9 - 12 cm depth. In the previous paper,  $I^-$  is oxidized to  $I_2$  by a microbial enzyme, and subsequently incorporated into organic compounds (Amachi, Microbes Environ., 2008). We determined the vertical profile of activity of laccase, which is known as iodide oxidase. Laccase activity is higher at the surface rather than that of subsurface. Microbial activity plays an important role for transformation of iodide to organic iodine. Iodine organification is effective for iodine immobilization, especially at the surface environment. The mechanism of dissociation of inorganic iodine from organic matter at subsurface environment should be investigated in the future.

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