Iodine sorption and its chemical form in the soil–soil solution system in Japanese agricultural fields

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Abstract

Iodide (I^-) or iodate (IO_3^-) sorption in two types of Japanese agricultural soil samples was investigated in consideration of microbial effects. A batch sorption test was carried out to observe sorption kinetics under 4 different experimental conditions characterized by temperature or microbial activity. The sorption kinetics results indicated that microbial activity promoted an increase of the soil-soil solution distribution coefficient. In addition, it was observed that the sorption kinetics of IO_3^- was similar to that of I^- for one sample. The results could be attributed to the fact that IO_3^- was transformed to I^- in the soil solution. Therefore, in order to discuss the difference in sorption kinetics between I^- and IO_3^-, it is important to check chemical forms of I in soil solution when IO_3^- is added to the sample.

Key Words

Iodide, iodate, sorption, microbial activity, agricultural soil

Introduction

Long-lived iodine-129 (^{129}I; half-life, 1.57×10^7 y) has been released into the environment during operation of nuclear facilities such as spent fuel reprocessing plants (Buraglio *et al.* 2001). ^{129}I is also one of the dominant radionuclides in transuranium waste repository assessment (JAEA and FEPC 2007). Therefore, investigating its behaviour in the environment is important. Researchers have reported on I behaviour in the soil environment, including the volatilization of I from soil to the air (Whitehead 1981; Whitehead 1984; Bostock *et al.* 2003), the effects of bacteria on I in soil (Amachi *et al.* 2003), and I sorption in soils (Yoshida *et al.* 1992). However, there have been only a few reports on the mechanistic and quantitative effects of microbial activity on I behaviour in soil (e.g., Bostock *et al.* 2003). The major chemical forms of I in the soil environment are iodide (I^-) and iodate (IO_3^-) (Gu and Schulz 1991), and I behaviour in agricultural fields such as I sorption in soil (Yoshida *et al.* 1992) and plant uptake of I (Muramatsu *et al.* 1983) have been reported to depend on the chemical forms. However, knowledge about the effect of chemical form of I on I sorption in soil is still limited. In this study, we observed sorption kinetics of I^- and IO_3^- under 4 types of experimental conditions characterized by temperature or microbial activity. Then, we observed chemical forms of I in the soil solution to check the change of chemical forms of I before and after addition of I^- or IO_3^- to soil samples.

Methods

A batch sorption test was carried out to observe sorption kinetics under 4 different experimental conditions designated ST, CO, AN, and GL (Table 1). Two soil samples were used (Table 2). Methods to determine these properties were based on our previous papers (Tagami *et al.* 2006; Ishikawa *et al.* 2008). Each soil sample and deionized water (solid/liquid ratio: 1g/10 mL) were mixed in a plastic bottle and initially shaken for 24 h. The standard condition ST, in which temperature was 23 ºC, was the basic condition for determination of soil-soil solution distribution coefficient (K_d) (JAEA 2002). The condition CO, in which temperature was 4 ºC, was used to determine the effect of microbial activity at low temperature on I sorption. The other 2 conditions, AN and GL, were used to observe the effects of microbial activity at normal temperature. For AN, 3 types of antibiotics were added to each sample suspension prior to shaking for 24 h to inhibit microbial activity. The initial concentration of each antibiotic was adjusted to 500 mg/L. The condition GL, in which the concentration of glucose was adjusted to 50 g/L after shaking for 24 h, was done to stimulate microbial activity. After all of the sample suspensions were shaken for 24 h, about 10 kBq of ^{125}I ($T_{1/2} = 59.4$ d) as iodide (I^-) or iodate (IO_3^-) were added to each sample bottle. IO_3^- stock solution was prepared using bromine (Muramatsu and Ohmomo 1988).

In order to obtain sorption kinetics under each experimental condition, suspension samples were destructively taken at 3 h, 1, 2, 5, 7, 9, 14, and 21 days after the addition of ^{125}I to the sample suspension. Each sample suspension was centrifuged at 3000 rpm for 10 min (Hitachi, HIMAC CT5L) and then the supernatant was filtered through a 0.45-μm membrane filter. The radioactivities of ^{125}I in the filtrate and the soil sample were measured with a NaI scintillation counter (Aloka, ARC-380). K_d is generally determined by radionuclide concentration in liquid phase (JAEA 2002) because the added radionuclide should exist in the solid phase or liquid phase. However, it was reported that I could be released from soil to the air (Whitehead, 1981).Therefore, in this study K_d (L/kg) was calculated by the following equation:

$$K_d = (C_S/C_L) \tag{1}$$

where C_L (Bq/L) and C_S (Bq/kg-dry) are the radionuclide concentrations in the liquid phase and solid phase, respectively. Measurements by the isotope exchange method (Muramatsu and Ohmomo 1988) showed that the ^{125}I in the filtrate samples at day 7 of contact time for CO and ST conditions was distributed as 3 chemical forms: elemental iodine and organic iodide; I^-; and IO_3^-.

Table 1. Experimental conditions.

Experimental condition	Temperature (°C)	Solution
ST	23	Deionized water
CO	4	Deionized water
AN	23	500 mg/L of streptomycin, tetracycline, and cycloheximide
GL	23	50 g/L of glucose

Table 2. Properties of soil samples.

Properties		Sample No.	
		EP-8	EF-38
Clay	(%)	22	22
EC (H_2O)	(μS/cm)	72	117
pH	(H_2O)	6.0	7.2
CEC	(meq/100g)	11	12
C in soil	(g/kg)	21.2	24.1
I in soil	(mg/kg)	0.6	5.1

Results and discussion

Chemical forms of I in soil solution

Figure 1 shows the percentage of each chemical form of I in the soil solution to the initially added I in each sample at day 7 after contact with I under conditions CO and ST. I in soil solution was mainly present as I^- when I^- was added to the sample bottle, while some IO_3^- became I^- in soil solution when IO_3^- was added to the samples. The chemical form of IO_3^- did not change in the control sample which contained deionized water and IO_3^-, therefore, the soil may affect the chemical form change from IO_3^- to I^- in the soil solution.

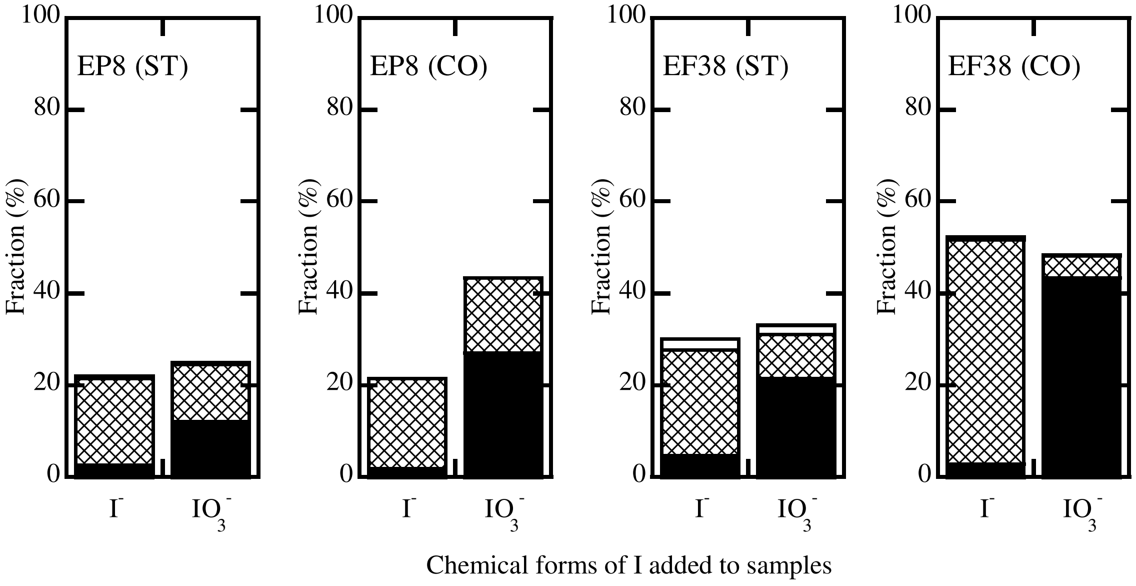

Figure 1. Percentage of elemental iodine and organic iodide (white), I^- (mesh), and IO_3^- (black) in the soil solution to the initially added I at day 7 after contact with I.

Sorption kinetics

The time variation of K_d for each chemical form of I under 4 experimental conditions is shown in Figure 2. For I^- and IO_3^-, the K_d values increased in the order: GL> ST> AN ≈ CO, which is the same order as expected for their microbial activity. This result indicated that microbial activity promoted I uptake by the solid phase.

Comparing the sorption kinetics for I^- and IO_3^-, almost the same results were observed for EP8. For EF38, however, the sorption kinetics for I^- had higher values than for IO_3^-. Indeed, K_d under the ST condition for I^- (24 L/kg) was 3 times higher than that for IO_3^- (8 L/kg). The different results between EP8 and EF38 could be attributed to the chemical forms of I in the soil solution. As shown in Figure 1, for EP8 the amount of I^- in the soil solution when I^- was added to the sample was about the same as the amount of I^- in the soil solution when IO_3^- was added to the sample, which could confirm the similar result of I^- and IO_3^- additions. On the other hand, for EF38, which had different results for I^- and IO_3^-, the amount of I^- in the soil solution when IO_3^- was added to the sample was much lower than the amount of I^- in the soil solution when I^- was added to the sample under the ST condition. Previous studies reported K_d for IO_3^- was lower than that for I^- (IAEA 1994; Yoshida *et al.* 1992). Therefore, it appeared that the difference of sorption kinetics between I^- and IO_3^- was possibly dependent on whether IO_3^- forms I^- in the soil solution; it is recognized that IO_3^- could change its chemical form to I^- in the soil solution. The present results could not identify the major factor contributing to the change of chemical form from IO_3^- to I^- in soil solution; therefore, further studies are needed to identify the mechanisms of change of I chemical forms in soil solution.

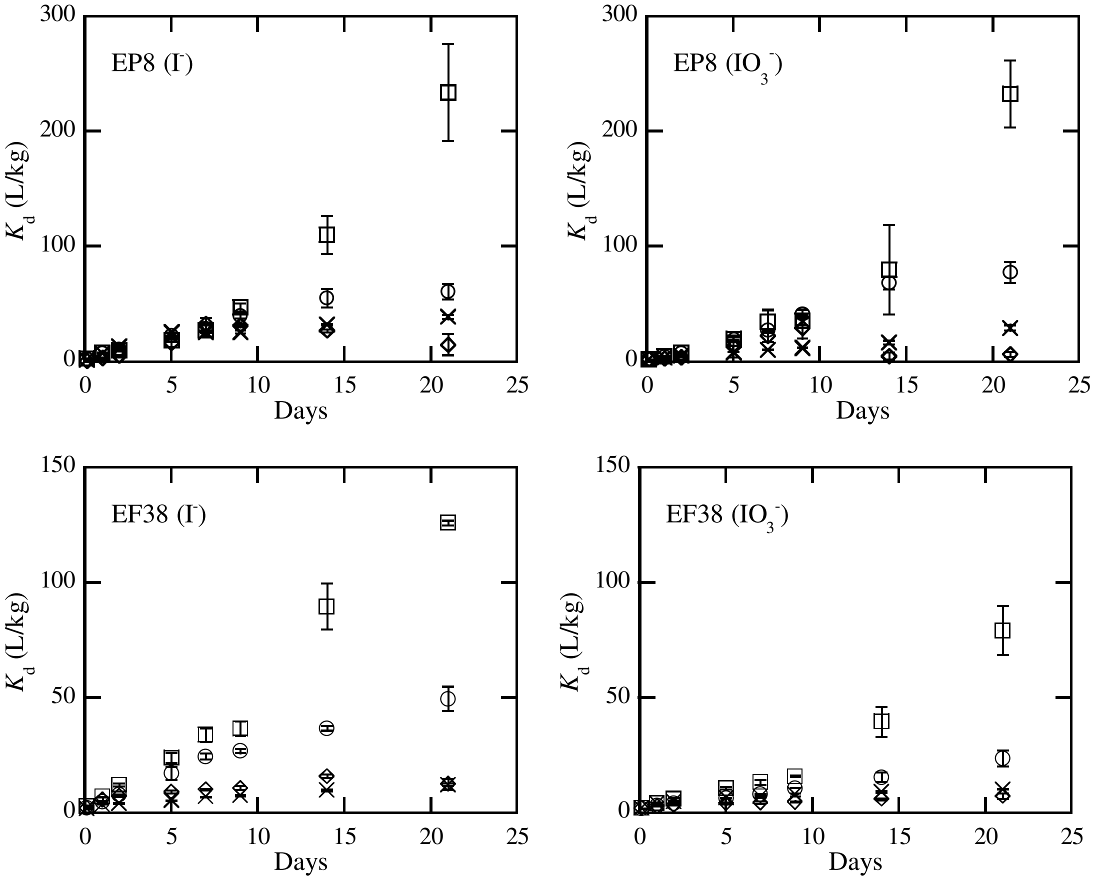

Figure 2. Time variation of the K_d values at 4 different experimental conditions: ST (○),CO (×), AN(◊),and GL (□). Error bars show standard deviation (1σ) of 3 replicates.

Conclusion

We observed the sorption kinetics of I in soil and the chemical forms of I in soil solution when I^- or IO_3^- were added to the soil–soil solution system. For I^- addition, chemical forms of I in soil solution was unchanged, while some IO_3^- in the soil solution became I^-. Our results suggested that the difference in sorption kinetics of IO_3^-possibly depended on whether IO_3^- forms I^- in the soil solution.

Acknowledgements
This work has been partially supported by the Agency for Natural Resources and Energy, the Ministry of Economy, Trade and Industry (METI), Japan. We thank Dr. N. Ishii of National Institute of Radiological Sciences for his advice about this work. In addition, Mr. T. Takamure (Tokyo Nuclear Services Co., Ltd.) is gratefully acknowledged for his technical support.

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