

## Redox reaction of iodine in paddy soil investigated by field observation and the I K-Edge XANES fingerprinting method

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### Abstract

In order to elucidate the cause for the leaching of iodine in a flooded paddy field, we investigated the transformation of an iodine species affected by the water management of the paddy field. The increased concentration of iodide ( $I^-$ ) in soil solution of a flooded paddy field suggested that  $I^-$  was leached from the soil under anaerobic conditions. The post-edge feature of X-ray absorption near-edge structure (XANES) for iodate ( $IO_3^-$ ) spiked to soil totally disappeared after anaerobic incubation of the soils, and  $I^-$  was dissolved in the solution. On the other hand,  $I^-$  in contact with the soil was not likely to be oxidized to  $IO_3^-$  under aerobic incubation. Iodine was leached out in soil solution as  $I^-$  under anaerobic conditions, whereas part of the iodine species was retained by soil as  $I_2$  or organoiodine both under anaerobic and aerobic conditions.

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## 1. Introduction

Reprocessing of nuclear fuel can cause gaseous leaks of trace amounts of radionuclide, including Kr-85, Xe-133, I-131, and I-129. Elevated levels of Kr-85, which is monitored, leaked out from a reprocessing plant and were observed 60 km from the nuclear fuel-reprocessing plant in Tokai-mura, Ibaraki, Japan (Igarashi et al., 2001). The agricultural fields in the vicinity of the reprocessing plants are exposed to radionuclide. Iodine-129 is one of the most hazardous gaseous radionuclides leaked from reprocessing plants due to its very long period of half-decay, 15.9 million years. The background isotope ratio of I-129:I-127 in soil was between  $10^{-9}$  and  $10^{-8}$ , whereas an elevated isotope ratio up to  $10^{-6}$  was found in surface soil taken from the vicinity of the Chernobyl nuclear facility (Handl, 1996). Schmitz and Aumann (1995) reported that I-129 that had recently been added to soil was more mobile than the natural I-127 and, thereby, the transfer factor of I-129 from soil to pasture was higher than that of I-127 (Schmitz and Aumann, 1994). However, the environmental behavior of radioactive I-129 should be basically similar to that of natural I-127. For the sake of food safety and security, it is critical that the behavior of I-127 and I-129 in the agricultural environment be investigated.

Major iodine species in soil include iodide ( $I^-$ ), iodate ( $IO_3^-$ ), and organically bound iodine (Yuita, 1992; Yamada et al., 1999, 2002). The mobility of iodine in soil is strongly controlled by the speciation of iodine. The reduced form of inorganic iodine,  $I^-$ , is the most mobile iodine species in soil. On the other hand,  $IO_3^-$  is relatively immobile in soil since  $IO_3^-$  can be sorbed on the soil components. Furthermore, organically bound iodine is considered as a sink of iodine in soil (Tikhomirov et al., 1980; Yuita, 1992; Yamada et al., 1999, 2002; Whitehead, 1973). The iodine concentration in paddy field soil was substantially lower than that in upland fields and forest soils (Yuita et al., 1982a,b; Muramatsu and Yoshida, 1993). The drop in the redox potential during the flooded period of paddy soil may cause the reduction of  $IO_3^-$  to  $I^-$ , and, as a result, the increased solubility of iodine (Yuita, 1992; Muramatsu et al., 1996). Though the increased concentration of  $I^-$  in a soil solution after incubation of added  $IO_3^-$  under flooded conditions was demonstrated by Yuita (1992), simultaneous evaluation of iodine species in both the solution and solid phases of soils is still required.

Yamada et al. (1999, 2002) proposed a speciation method of iodine in soil. When iodine species are already stabilized in the soil environment, those speciation procedures are quite useful to acquire information of the iodine speciation in soil. However, the time-consuming extraction process of iodine from soil of reduced conditions may result in operational changes in iodine species. Therefore, the direct analysis of the iodine redox state without disturbing the soil conditions is required. X-ray absorption near-edge structure (XANES) has been widely applied to the nondestructive determination of the local structure of elements in environmental samples. When X-ray absorption occurs at a defined energy corresponding to the binding energy for the core electron of the atom of interest, the ejected electron interacts with the surrounding atoms to produce a spectrum. The XANES spectral region is from the X-ray absorption edge up to few tens of electron volts above the X-ray absorption edge. Because the XANES spectrum is unique to the oxidation state of the element of interest, it provides information on the oxidation state of materials. Recent studies successfully determined the oxidation states of chromium (Shaffer et al., 2001) and arsenic (Takahashi et al., 2004) in soil by XANES. Based on the comparison of the post-edge feature of XANES with reference materials, Reed et al. (2002) suggested that the iodine species in a solvent sample from a nuclear fuel-reprocessing plant was possibly an organoiodine species. In addition, the sensitive determination of dissolved  $I^-$  and  $IO_3^-$  has been accomplished by the use of element-specific detection with inductively coupled plasma

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