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Behavior of iodine in sedimentary rocks based on iodine speciation and ^{129}I /^{127}I ratios

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[Introduction]

Japan is the second largest country for iodine production in the world, and iodine is mostly produced from brine. Iodine ages, determined by ^{129}I /^{127}I ratio, in brine are often older than host rocks (Muramatsu et al., 2001). However, formation process of iodine-rich brine is still unclear. On the other hand, radioactive iodine (^{129}I) is one of the most problematic radionuclides contained in nuclear waste, because of its long half-life (15.7 million years) and high mobility. To investigate behavior of iodine in sedimentary rocks is important to evaluate the effectiveness of the natural barrier for nuclear waste repositories. In order to predict behavior of iodine in the environment, speciation of iodine is essential because of different mobility among their species (ex. IO_3^-, I^-, I_2, and organic iodine). In this study, we determined iodine distribution, speciation, and isotope ratios (^{129}I /^{127}I), to investigate long-term migration of iodine in Horonobe area.

[Experimental]

All rock and groundwater samples were collected at JAEA Horonobe underground research center. The region is underlain mainly by Neogene to Quaternary marine sedimentary rocks, the Wakkanai Formation (Wk Fm), and the overlying Koetoi Formation (Kt Fm): siliceous and diatomaceous mudstones. Iodine species in rock samples were determined by iodine K-edge XANES (SPring-8 BL01B1). Thin sections of rock samples were prepared, and iodine mapping were obtained by micro-XRF analysis (SPring-8 BL37XU). Iodine species (IO_3^-, I^-, and organic iodine) in groundwater were separately detected by high performance liquid chromatography (HPLC)-ICP-MS. The ^{129}I /^{127}I ratios in groundwater and rock samples were measured by accelerator mass spectrometry (MALT, Univ. of Tokyo). Iodine in rock samples were separated by pyrohydrolysis and water extraction.

[Results and discussion]

Concentration of iodine in groundwater varied widely and was much higher than that of seawater showing a high correlation with that of chlorine (R^2 = 0.90). Species of iodine in groundwater was mainly I^-. Iodine in rock samples decreased near the boundary between Wk and Kt Fm. Based on iodine K-edge XANES, iodine in rock was a mixture of organic and inorganic I. Iodine mapping showed that iodine accumulated to micro region. Carbon content was also high in iodine rich region, suggesting that iodine existed as organic I. Iodine isotope ratios (^{129}I /^{127}I) were higher in rocks compared with those in groundwater. According to these results, migation of iodine in this area can be expected as follows. During sedimentation of Wk and Kt Fm, iodine accumulated as organic iodine in siliceous sediment. Iodine was released as I^- from the layers deeper than Wk Fm during diagenetic processes. Subsequently, iodine-rich groundwater was distributed to Wk and Kt Fm due to the compaction of the layers. During uplift and denudation processes, both iodine and chlorine were diluted by meteoric water from the surface.

It was suggested that I^- is released to the ground water during the maturation of organic matter. However, the mechanism of dissociation of iodine from organic matter is still unclear. Speciation of carbon in rock at various depths should be investigated in future study.

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