Origin of halogen in hot-spring water in Joban and Hamadori area, northeast Japan, inferred from ^{129}I/I and ^{36}Cl/Cl ratios

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Japan is the second largest iodine producing country in the world, and it is produced from iodine-rich brine. The iodine-rich brine is often generated from fore-arc and back-arc basins. The half-life of long-lived radioisotope ^{129}I is 15.7 Ma so that the ^{129}I/I ratio could be used for the age determination of source formations for iodine. In the previous studies, iodine isotopic ratios of brine frequently showed older ages than those of host formations, however details of migration pathway of iodine had not been fully understood. Joban sedimentary basin is located at offshore of Fukushima Pref., where natural gas used to be produced from the Miocene sandstone layer. In order to understand the origin of groundwater around Joban and Hamadori area, we collected hot-spring water in Fukushima and north part of Ibaraki Pref., and determined ^{129}I/I, ^{36}Cl/Cl, and ^{3}H concentrations.

Iodine and chlorine concentrations in hot-spring waters were 0.4-8.7 mg/L and 722-15400 mg/L, respectively. Iodine concentration was higher than that of seawater, while chlorine concentration was lower than that of seawater. ^{129}I/I and ^{36}Cl/Cl ratios were determined using accelerator mass spectrometry. ^{129}I/I ratios of hot-spring waters were almost constant at around 0.27 x 10^{-12} irrespective of iodine concentration and I/Cl ratio, except for two samples collected at Iwaki city showing 5.8 x 10^{-12} and 1.8 x 10^{-12}. These two samples were considered to be affected by anthropogenic ^{129}I due to the mixing of recent shallow groundwater as indicated by measurable ^{3}H, then these two were omitted from further discussion. Iodine ages were determined to be about 40 Ma, using 1.5 x 10^{-12} as the preanthropogenic initial ^{129}I/I ratio. The homogeneity of iodine ages gives the limit for the source formation of iodine. On the other hand, age determined by ^{36}Cl/Cl ratio varied widely, from about 5 x 10^{4} years to over 1.5 Ma (at secular equilibrium). Chlorine ages are considerably younger than those of iodine. In addition, iodine-rich water was obtained not only from sedimentary rock but also from the Early Cretaceous granite. Because of the large discrepancy between iodine and chlorine ages, migration of iodine is not accompanied with chlorine. There are two possible migration pathways of iodine: 1) iodine originates from the Paleogene layer of Joban basin, because in this case the iodine age is consistent with the sedimentation age; 2) iodine was derived from subducting marine sediments and had migrated with long time to the present location, which is supported by the fact that iodine is also found in granite area. Further investigation is needed to understand the origin of iodine.