

Radium isotopes in Na-Cl type groundwater from the Japan Sea side of Japan, Central Japan

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Radium isotopes, mainly ²²⁶Ra, in groundwater have been investigated worldwide from various viewpoints such as radiation exposure and behavior of Ra isotopes at oil- and gas-fields, search of uranium deposit at underground, Ra isotope contamination of drinking water (Vengosh et al., 2009), and natural analogue study related to radioactive waste disposal. Anomalously high ²²⁶Ra concentrations over several tens of Bq kg⁻¹ have been often found in brine and saline waters from oil-and gas-fields worldwide.

In Japan, ²²⁶Ra contents in groundwater have been measured from 1930's, and many studies have focused on the survey of radioactive springs or analytical methods. However, detailed and systematic studies on Ra isotope transport into groundwater were very scarce. Recently, with the development of more sophisticated drilling techniques, various types of deep groundwaters have been obtained from deep wells over 1000 m in depth drilled in sedimentary basin and coastal areas. These groundwaters are commonly characterized by Na-Cl type groundwaters with intermediate salinity (ca. 1-36‰) and are considered to be fossil seawater in origin from the stable isotope signature, which makes a better understanding of the processes controlling ²²⁶Ra concentrations and ²²⁸Ra/²²⁶Ra activity ratios in these groundwaters scientifically very interesting.

In this paper, Ra isotopes (²²⁶Ra and ²²⁸Ra) in Na-Cl type groundwaters from Ishikawa, Niigata and Toyama (Himi) prefectures, where are located in the central Japan and face the Sea of Japan, were measured together with ²³⁸U and ²³²Th contents of rock samples (vertical borehole rock, rock from outcrop) collected from each area. Transport behavior of Ra isotope into groundwater was mainly investigated, based on ²²⁶Ra concentrations and ²²⁸Ra/²²⁶Ra activity ratios in groundwaters, and ²³⁸U and ²³²Th contents in rocks measured.

Groundwater samples were approximately neutral and their concentrations of total dissolved solids (TDS), which were defined here as the sum of major dissolved ions, ranged from about 1-36 ‰. Relationship between ²²⁶Ra and ²²⁸Ra concentrations in Na-Cl type groundwater samples is shown in Fig. 1. The concentrations of ²²⁶Ra in the groundwater samples ranged widely from 1.8-2143 mBq kg⁻¹, and higher ²²⁶Ra concentrations over 1000 mBq kg⁻¹ were found in some groundwater samples. Activity ratios of ²²⁸Ra/²²⁶Ra

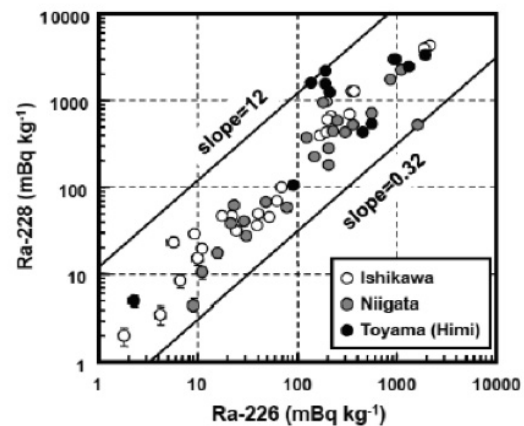


Fig.1 Relationship between ²²⁶Ra and ²²⁸Ra contents in Na-Cl type groundwaters

in water samples varied from 0.32-12, and were similar to or higher than $^{232}\text{Th}/^{238}\text{U}$ activity ratio (0.12-2.3) of rock samples. It is worth noting that groundwater samples from Himi City, Toyama prefecture, showed high $^{228}\text{Ra}/^{226}\text{Ra}$ activity ratio as high as around 10, most of the groundwater being from 1-3. Such high $^{228}\text{Ra}/^{226}\text{Ra}$ activity ratios in the groundwaters are rarely seen.

Radium isotope are supplied into groundwater by multiple processes; (1) decay of dissolved parent nuclides (Th isotope is considered as insoluble element), (2) weathering and/or dissolution of aquifer rock, (3) α -recoil at water-rock interface and (4) desorption reaction at water-rock interface.

For old groundwater, weathering and/or dissolution of Ra-bearing aquifer rock would result in the lower $^{228}\text{Ra}/^{226}\text{Ra}$ ($^{232}\text{Th}/^{238}\text{U}$) activity ratio than those of host aquifer rock due to the difference of their half-lives. On the other hand, α -recoil process would produce the same $^{228}\text{Ra}/^{226}\text{Ra}$ activity ratio in groundwater as the $^{232}\text{Th}/^{230}\text{Th}$ ($^{232}\text{Th}/^{238}\text{U}$) activity ratio of host rock. Desorption reaction at water-rock interface would reflect the same $^{228}\text{Ra}/^{226}\text{Ra}$ ($^{232}\text{Th}/^{230}\text{Th}$ and $^{232}\text{Th}/^{238}\text{U}$) activity ratios in groundwater as those in rock surface. Thus, α -recoil and desorption reaction are considered to be dominant processes to explain the measured $^{228}\text{Ra}/^{226}\text{Ra}$ activity ratio in groundwater by assuming no retardation of ^{228}Ra relative to ^{226}Ra .

Recently, some researchers have pointed out the existence of surface coating enriched in Th isotope at water-rock interface. It is interpreted as given below. Thorium is extremely insoluble element. Thorium isotope ejected into groundwater by weathering of minerals might be rapidly precipitated onto the surface of aquifer grain due to their solubility limit. Radium isotope produced by the decay of such Th isotopes can be ejected easily into water phase by α -recoil and desorption reaction. Thorium-232

accumulated in the water-rock interface, therefore, might be acceptable and possible source leading $^{228}\text{Ra}/^{226}\text{Ra}$ activity ratio in groundwater towards higher level.

As shown in Fig. 2, ^{226}Ra concentrations in groundwater samples show a increasing tendency with increasing TDS contents, indicating strongly that ^{226}Ra concentrations in groundwater samples are much more controlled by adsorption/desorption reaction depending on salinity (Kraemer and Reid, 1984; Sturchio et al., 2001). If this is a main process leading ^{226}Ra concentration in groundwater towards higher level, higher production rate of Ra isotope by α -recoil would be required. According to the equation by Kraemer (1981), such condition can be readily attained if the layer of higher specific activity of Th isotope is present at grain surface. Since half-life of ^{230}Th (7.54×10^4 yr) is short compared with geological scale, the activity of ^{230}Th of surface coating might be supported by U isotope (^{238}U and ^{234}U) in old aquifer. Therefore, both salinity in groundwater and distribution of ^{232}Th and ^{230}Th (^{238}U) at water-rock interface are considered to be very important for the transport of Ra isotope into groundwater.

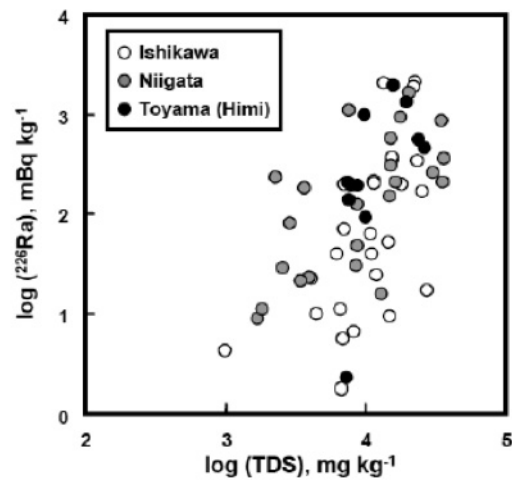


Fig.2 Relationship between ^{226}Ra and TDS contents in Na-Cl type groundwaters