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Longtime behavior of cesium (Cs) in natural spring drinking water

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ABSTRACT

Large amounts of radioactive cesium (Cs^A) have been introduced into the underground environments, through a natural process, as a result of nuclear power plant accidents. It is known that the active Cs sorption onto colloidal-size clay minerals in groundwater is observed and the active Cs can be transported with the colloidal fraction of groundwater by water flows. However, the longtime behavior of radioactive Cs, contained in the flowing groundwater in the aquifers of groundwater source areas, is unknown in terms of the natural water cycle. Herein, we investigate the Cs concentration in natural spring drinking water with the residence time in a groundwater source area of a mountainside composed of volcanic rock, compared with those of other trace elements. This investigation demonstrates that the observed Cs concentration in natural spring drinking water exponentially decreases slowly with the groundwater residence time (∼45 yr), while several trace elements, namely, P, V, Ga, and Ge, increase in concentration with the groundwater residence time through chemical weathering. The findings suggest that Cs^A, contained in flowing groundwater in mountain water source areas, may decrease exponentially at the rate of one-tenth in twenty-two years, by sorption onto the aquifer through rock–water interaction excluding radioactive decay. For the sustainable management of water sources and ecosystems, the long-term (∼50 yr) monitoring of the active Cs in groundwater is needed in mountain water source areas where radioactive cesium has been dispersed at times of nuclear power plant accidents.

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1. Introduction

The longtime (∼100 yr) effect of radioactive cesium (Cs^A), dispersed at the time of nuclear power plant accidents (e.g., von Gunten et al., 1988; Mizuno and Kubo, 2013; Tsuji et al., 2014), is a major environmental issue. As Cs^A is sorptive onto colloidal-size clay in groundwater (Francis and Brinkley, 1976; Comans et al., 1991; Kersting et al., 1999) and the distribution coefficient to the subsurface soil of Cs^A decreases with depth in the soil (Comans et al., 1989), the active Cs can be transported by water flows with the colloidal fraction, with suspended particles of sub micrometer size (von Gunten et al., 1988; Buddemeier and Hunt, 1988; Kersting et al., 1999; Tsuji et al., 2014), and can reach the groundwater surface (Kersting et al., 1999). Although Cs^A sorption onto soil has been reported (Francis and Brinkley, 1976; Comans et al., 1991; Buddemeier and Hunt, 1988), the temporal variation in field groundwater Cs^A by sorption onto rock aquifers is still unknown

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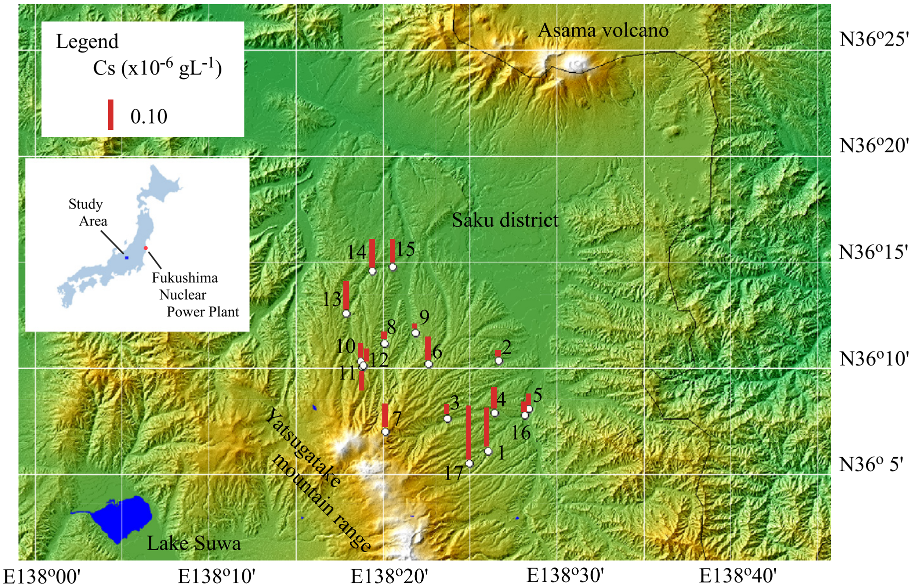

Fig. 1. Study area. Circles indicate sample locations of spring waters. Bars denote cesium concentration of the spring water samples.

in terms of the natural water cycle, and the longtime behavior (10–50 yr) of the Cs^A concentration in groundwater has rarely been observed.

In order to evaluate the longtime behavior of Cs^A in the flowing groundwater of an important source area, this study focuses on the Cs in the spring water of the Yatsugatake Mountains, which form an important source area for drinking water in Central Japan. In the Saku district, a high land area, 650–2500 m in altitude, in the northeastern section of the Yatsugatake Mountains in Nagano, Japan, 280 km southwest of the site of the Fukushima Daiichi Nuclear Power Plant accident (Fig. 1), the groundwater, spring water, and well water in the volcanic rock aquifer have been used for 99%, 47%, and 52%, respectively, of the drinking water and tap water in that district for a long time. According to all the local Government HPs in the Saku district, no Cs^A was detected (detection limit <0.1–10 Bq/kg) from the groundwater for the water works, but Cs^A was detected from the drainage sludge in certain areas. Spring water is still the major source of drinking and tap waters in high land areas of Japan. As the sorption and desorption behavior of the Cs in groundwater should be equal to that of Cs^A (e.g., Cornell, 1993; Yoshida et al., 2004), the longtime behavior (10–50 yr) of the Cs^A concentration in the groundwater flowing through aquifers is expected to be obtainable by field monitoring the relationship between the Cs concentration in groundwater and the residence time of the groundwater in a rock aquifer that includes Cs-bearing minerals. In this study, the Cs concentration in natural spring drinking water was measured with the groundwater residence time, in order to evaluate the longtime behavior of Cs^A in the flowing groundwater of an important source area for drinking water in terms of the natural water cycle.

2. Material and methods

2.1. Sampling locations and geological setting of the study area

Sampling points are located in the northeastern part of the Yatsugatake volcanic area, which is part of the Yatsugatake-Chūshin Kōgen Quasi-National Park. The Yatsugatake volcanic body, formed by Quaternary volcanic activity from 0.8 to 1.2 Ma, is comprised mainly of several thin flows of pyroclastics, including volcanoclastic, basaltic lava, and basaltic andesitic lava (Nishiki and Takahashi, 2012). The slopes are about 0.13 for higher than 1600 m a.s.l., and about 0.08 for lower than 1600 m a.s.l. and for higher than 1000 m a.s.l. Most parts of the study area belong to the Yachiho Group of early Pleistocene, consisting of five formations. These formations are comprised mostly of volcanic sand and gravel with scoria and tuff breccia; namely, pyroclastics, including the volcanoclastic of Nishiki and Takahashi (2012), compose the aquifers of 30–50 m in thickness (Kumai, 1982). After the Yachiho Group was uncomfortably covered and leveled by a gravel formation, the mountain ridges separated from one another due to the erosion of valleys in the geologic body (Kumai, 1982). Kumai (1982) also reported that each hydraulic parameter of the aquifers in the Yachiho Group ranged from 10^{-6} to 10^{-4} m/s in hydraulic conductivity and from 0.06 to 0.4 in effective porosity, respectively.

2.2. Sampling and analysis of spring water

We collected groundwater samples from July to August 2011 from the 17 (cold) springs, 780 m to 1940 m a.s.l. in altitude (Table 1), that are being used for drinking and tap waters as supplied from the local government in the Saku district (Fig. 1).

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