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## Effects of terrigenous He components on tritium–helium dating: A case study of shallow groundwater in the Saijo Basin

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### ABSTRACT

Dating using a combination of  $^3\text{H}$  and  $^3\text{He}$  is believed to be the most practical method for estimating the short residence time of shallow groundwater. However, this method must estimate tritiogenic  $^3\text{He}$  alone and tends to overestimate the residence time of groundwater, if terrigenous  $^3\text{He}$  from the mantle cannot be excluded from the total dissolved  $^3\text{He}$ . We demonstrate the exclusion of terrigenous  $^3\text{He}$  in the Saijo Basin, where mantle He is easily released along the major active fault, Median Tectonic Line. The  $^3\text{He}/^4\text{He}$  ratios suggest that the west bank of the Kamo River, which lies within the basin, has experienced greater emanations of mantle He than the east bank. We estimate the residence times to be 1.1–96 years by the proposed exclusion method.

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### 1. Introduction

Groundwater is one of most indispensable water resources for human activities. As the overuse of groundwater leads to exhaustion, adequate management based on the residence time of groundwater is required to regulate its use. Dating using a combination of  $^3\text{H}$  and  $^3\text{He}$  is believed to be the most practical method for estimating the short residence time of shallow groundwater (Solomon et al., 1993; Aeschbach-Herting et al., 1998, 1999; Solomon and Cook, 2000; Kipfer et al., 2002). The residence time ( $T$ ) of groundwater is estimated by the following equation:

$$T = 17.69 \ln \left[ 4.01 \times (^3\text{He}_{\text{tri}}) / (\text{HTO}) \times 10^{14} + 1 \right] \quad (1)$$

where  $T$  is the residence time of groundwater (years),  $^3\text{He}_{\text{tri}}$  is the accumulated tritiogenic  $^3\text{He}$  produced through  $\beta$  decay of  $^3\text{H}$  in groundwater (ccSTP/g<sub>water</sub>), and  $\text{HTO}$  is the tritium concentration in groundwater (TU).

However, as this method is based on the estimation of only tritiogenic  $^3\text{He}$  produced through  $\beta$  decay of  $^3\text{H}$  (Mahara and Ohta,

2009), the residence time of groundwater tends to be overestimated if the addition of terrigenous  $^3\text{He}$  (i.e. mantle  $^3\text{He}$  supplied from depth and radiogenic  $^3\text{He}$  from the crust) is not appropriately considered. If mantle He is released into shallow groundwater, the contribution of  $^3\text{He}$  that originated in mantle He is greater than that from radiogenic He. Mantle He exhibits  $^3\text{He}/^4\text{He}$  ratios of  $1.1 \times 10^{-5}$  in subduction areas, including Japan (Sano and Wakita, 1985), whereas radiogenic  $^3\text{He}/^4\text{He}$  ratios are typically around  $1 \times 10^{-8}$  (Porcelli et al., 2002). Furthermore, mantle  $^3\text{He}$  released along the volcanic fronts and major active faults of Japan can be easily monitored (Fig. 1), and most Japanese groundwater has been affected by mantle  $^3\text{He}$  supplied from the depths.

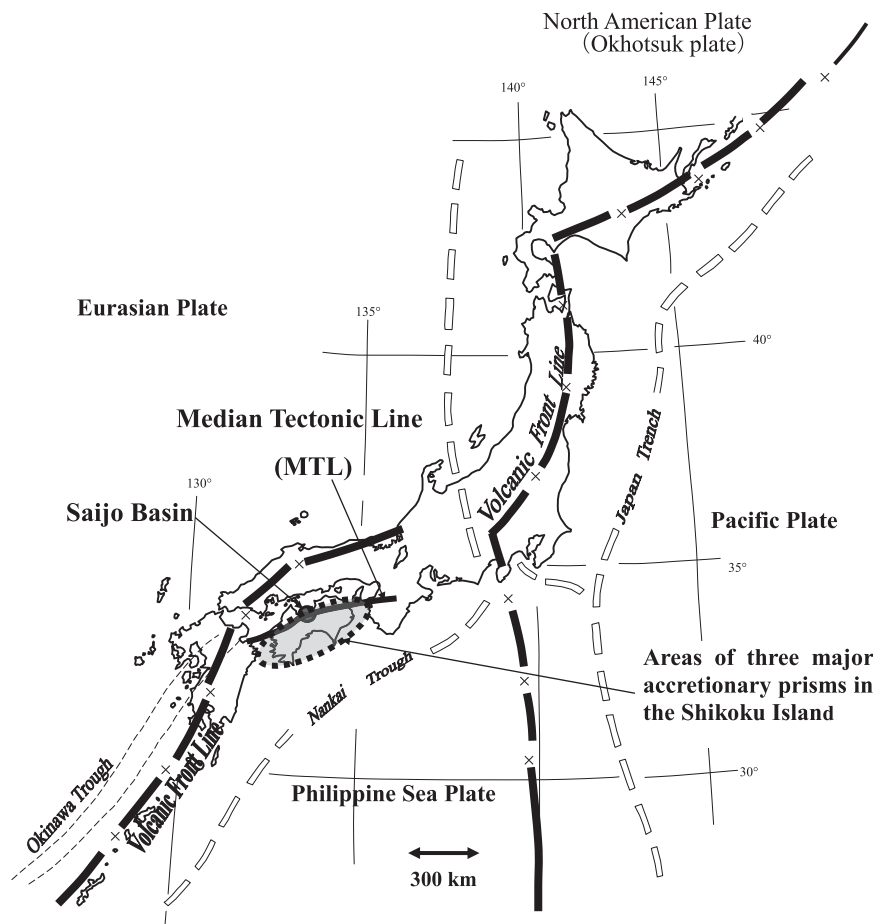
This study proposes a method that incorporates the rest of the  $^3\text{H}$ , which has not yet decayed, and the net tritiogenic  $^3\text{He}$  produced by the  $\beta$  decay of  $^3\text{H}$  in groundwater after subtracting the accumulation of terrigenous  $^3\text{He}$  supplied from other sources (e.g., the mantle He) from the total  $^3\text{He}$  dissolved in groundwater. Then, this study presents an estimation of groundwater residence time in the Saijo Basin, Japan, by the proposed method.

### 2. Exclusion of terrigenous $^3\text{He}$

The total dissolved  $^3\text{He}$  concentration  $^3\text{He}_{(\text{Tot})}$  in groundwater can be expressed as follows:

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**Fig. 1.** Plate tectonic setting of Japan and surrounding areas. Also shown are the locations of the Japan Trench, the Nankai Trough, the Okinawa Trough, and the Median Tectonic Line (MTL). The dark grey solid circle indicates the study area (Saijo Basin). The area enclosed by a dot line indicates three major accretionary prisms (i.e. the Sambagawa metamorphic rock, the Chichibu belt and the Shimanto belt are lined up from north to south).

$${}^3\text{He}_{(\text{Tot})} = {}^3\text{He}_{(\text{At})} + {}^3\text{He}_{(\text{Ex.air})} + {}^3\text{He}_{(\text{Trit})} + {}^3\text{He}_{(\text{Rad})} + {}^3\text{He}_{(\text{Mnt})}, \quad (2)$$

where  ${}^3\text{He}_{(\text{At})}$  represents equilibrated atmospheric  ${}^3\text{He}$  at the recharge temperature, salinity, and pressure,  ${}^3\text{He}_{(\text{Ex.air})}$  represents  ${}^3\text{He}$  from excess air (i.e., air entrapped in groundwater during infiltration in the unsaturated zone),  ${}^3\text{He}_{(\text{Trit})}$  is derived from  $\beta$  decay of  ${}^3\text{H}$  (including natural and man made components),  ${}^3\text{He}_{(\text{Rad})}$  is derived from  $\beta$  decay of  ${}^3\text{H}$  produced through the nuclear reaction of  ${}^6\text{Li}(n, \alpha){}^3\text{H}$  in crustal rocks, and  ${}^3\text{He}_{(\text{Mnt})}$  is derived from the release of mantle  ${}^3\text{He}$ . Then, the net tritogenic  ${}^3\text{He}$  can be calculated from Eq. (2) as follows:

$${}^3\text{He}_{(\text{Trit})} = {}^3\text{He}_{(\text{Tot})} - {}^3\text{He}_{(\text{At})} - {}^3\text{He}_{(\text{Ex.air})} - {}^3\text{He}_{(\text{Rad})} - {}^3\text{He}_{(\text{Mnt})} \quad (3)$$

We estimate  ${}^3\text{He}_{(\text{At})}$  and  ${}^3\text{He}_{(\text{Ex.air})}$  from the recharge temperature of groundwater and excess air effects, which are optimally deduced from the measured correlation between heavy noble gases (Ne, Ar, Kr and Xe) concentrations using the CE-model proposed by Aeschbach-Herting et al. (2000) and iterations. The fourth term  ${}^3\text{He}_{(\text{Rad})}$  is generally negligible in groundwater with a short residence time because the accumulation rates of  ${}^3\text{He}_{(\text{Rad})}$  are typically less than  $10^{-16}$  ccSTP/g<sub>w</sub> y<sup>-1</sup> (i.e., we assumed that the  ${}^3\text{He}/{}^4\text{He}$  ratio for the radiogenic He component is  $10^{-8}$ , and total  ${}^4\text{He}$  accumulation rates range from  $10^{-8}$  ccSTP/g<sub>w</sub> y<sup>-1</sup> to  $10^{-12}$  ccSTP/g<sub>w</sub> y<sup>-1</sup> for igneous and sedimentary rocks, respectively) and can easily be masked by the fifth term  ${}^3\text{He}_{(\text{Mnt})}$  if considerable mantle He release is confirmed in the basin. Here we define terrigenous  ${}^3\text{He}$  ( ${}^3\text{He}_{(\text{Terr})}$ ) which is the sum of mantle  ${}^3\text{He}$  and radiogenic  ${}^3\text{He}$ . Therefore, we can rewrite Eq. (3) as follows:

$${}^3\text{He}_{(\text{Trit})} = {}^3\text{He}_{(\text{Tot})} - {}^3\text{He}_{(\text{At})} - {}^3\text{He}_{(\text{Ex.air})} - {}^3\text{He}_{(\text{Terr})} \quad (4)$$

After subtracting from Eq. (4) the effects of both the saturated atmospheric  ${}^3\text{He}_{(\text{At})}$  at the estimated recharge temperature and the re-equilibrated  ${}^3\text{He}_{(\text{Ex.air})}$  of excess air in groundwater, we can reduce Eq. (4) into Eq. (5) using the new term  ${}^3\text{He}_{(\text{Tot})}^*$  ( ${}^3\text{He}_{(\text{Tot})} - {}^3\text{He}_{(\text{At})} - {}^3\text{He}_{(\text{Ex.air})}$ ) as follows:

$${}^3\text{He}_{(\text{Trit})} = {}^3\text{He}_{(\text{Tot})}^* - {}^3\text{He}_{(\text{Terr})} \quad (5)$$

Additionally, we can graphically estimate the magnitude of  ${}^3\text{He}_{(\text{Terr})}$ , as shown in Fig. 2.

In the graph, the total dissolved  ${}^4\text{He}$  concentration  ${}^4\text{He}_{(\text{Sam})}$  and the  ${}^3\text{He}/{}^4\text{He}$  ratio  $R_{(\text{Sam})}$ , corrected for both the concentration of  ${}^4\text{He}$  and  ${}^3\text{He}$  in atmospheric saturation and the excess effects from the air trapped in the sample water, are expressed as a new parameter  $x = {}^4\text{He}_{(\text{S})}/{}^4\text{He}_{(\text{Sam})}$  ( $0 \leq x \leq 1$ ). This parameter is transformed using the  ${}^4\text{He}_{(\text{S})}$  concentration saturated with the atmospheric  ${}^4\text{He}$  at the estimated recharge temperature under conditions of 1.0 atm and 0‰ salinity, and the corrected  $R_{(\text{Sam})} = {}^3\text{He}_{(\text{Tot})}^*/{}^4\text{He}_{(\text{Sam})}$ .

In the next step, we have to estimate the marginal terrigenous  ${}^3\text{He}/{}^4\text{He}$  ratio accumulated in the groundwater basin. We can estimate the marginal  ${}^3\text{He}/{}^4\text{He}$  ratio  $R_{(\text{Terr})x=0}$  (i.e., it was in secular equilibrium at the end point  $x = 0$  on the terrigenous  ${}^3\text{He}$  accumulation line) for a certain groundwater flow region. We can draw the terrigenous  ${}^3\text{He}$  accumulation line from the starting point ( $x = 1.0$ ,  $R_{(\text{Terr})x=1.0} = 0$ ) to  $R_{(\text{Terr})x=0} = R_{(\text{Terr})0}$  at the end point ( $0$ ,  $R_{(\text{Terr})0}$ ). Using real data, we can estimate the marginal  ${}^3\text{He}/{}^4\text{He}$  ratio  $R_{(\text{Terr})0}$  for  $x = 0$  by extrapolating the terrigenous  ${}^3\text{He}$  accumulation line plotting

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