

1 Estimation of groundwater residence time using the ^{36}Cl bomb pulse

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14
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17 Abstract

18 We propose a methodology for estimating the residence time of groundwater based on
19 bomb-produced ^{36}Cl . Water samples were collected from 28 springs and 2 flowing wells
20 located around Mt. Fuji, Central Japan. $^{36}\text{Cl}/\text{Cl}$ ratios in the water samples, determined
21 by accelerator mass spectrometry (AMS), were between 43×10^{-15} and 412×10^{-15} . A
22 reference time series of the above-background (i.e., bomb-derived) ^{36}Cl concentration
23 was constructed by linearly scaling the background-corrected Dye-3 data according to
24 the estimated total bomb-produced ^{36}Cl fallout in the Mt. Fuji area. Assuming piston
25 flow transport, estimates of residence time were obtained by comparing the measured
26 bomb-derived ^{36}Cl concentrations in spring water with the reference curve. The
27 distribution of ^{36}Cl -based residence times is basically consistent with that of
28 tritium-based estimates calculated from data presented in previous studies, although the
29 estimated residence times differ between the two tracers. This discrepancy may reflect
30 chlorine recycling via vegetation or the relatively small change in fallout rate since
31 ~1975, which would give rise to large uncertainties in ^{36}Cl -based estimates of recharge
32 for the period since ~1975. Given the estimated ages for groundwater from flowing

wells, dating based on a ^{36}Cl bomb pulse may be more reliable and sensitive for groundwater recharged before 1975, back as far as the mid-1950s.

Introduction

Groundwater is generally highly vulnerable to excessive use and contamination, because of its typically long mean residence time. In the case of groundwater depletion or contamination, aquifers require a long time for recovery and purification. Consequently, knowledge of the residence time and flow system of groundwater is crucial for the development and sustainable utilization of groundwater resources.

One of the most promising approaches to estimating the residence time of groundwater is the use of environmental tracers, such as chlorofluorocarbons (CFCs), tritiogenic ^3He , SF_6 , and ^{85}Kr for young groundwater (<60 yr). Bomb-produced ^{36}Cl is a dating tool covering the last ~60 yr (Bentley et al. 1982). In addition to its hydrophilic nature, ^{36}Cl is advantageous for young groundwater because its long half-life (3.01×10^5 yr) means that decay attenuation is negligible on the time scale of decades to centuries.

A ^{36}Cl bomb pulse can be observed in the unsaturated zone of arid and semi-arid regions, and has been used as a tracer to estimate infiltration/recharge rates (e.g. Phillips et al. 1988). In contrast, few studies have applied ^{36}Cl bombs in tracing groundwater, except for detecting the presence of young water (e.g. Andrews et al. 1994). ^{36}Cl has been used in combination with ^3H to estimate the groundwater recharge rate in a fractured rock aquifer (Cook and Robinson 2002) and to deduce the flow velocity and dispersivity in a sandy aquifer (Balderer et al. 2004).

Few studies have investigated the application of ^{36}Cl as a dating tool (Corcho Alvarado et al. 2005; Milton et al. 2003; Tosaki et al. 2007, 2010). One approach to quantitatively investigating the potential use of ^{36}Cl is to reconstruct its fallout rates from measured $^{36}\text{Cl}/\text{Cl}$ ratios in groundwater. The ^{36}Cl fallout rates in Denmark, as reconstructed by Corcho Alvarado et al. (2005), exceeded the estimates based on data from the Dye-3 ice core, Greenland (Synal et al. 1990). The authors attributed this discrepancy to the storage and recycling of chlorine in the biosphere (e.g. Scheffé et al. 1999). The nature of such storage and recycling was thoroughly evaluated at a wet, forested site in Canada by Milton et al. (2003) based on measurements of ^{36}Cl in

groundwater, vegetation, and soil. Their results suggested chlorine retention and subsequent release by vegetation (including bomb-produced ^{36}Cl), which can lead to errors in the application of ^{36}Cl -based methods. In contrast, the ^{36}Cl fallout rates in Germany, as reconstructed by Tosaki et al. (2007, 2010), were consistent with Dye-3 fallout data, suggesting that the successful application of ^{36}Cl is dependent on the conditions of the study site.

No previous study has used ^{36}Cl to quantitatively estimate the residence time of young groundwater. Consequently, the aim of the present study is to develop a methodology for “ ^{36}Cl bomb-pulse dating”. This paper presents the initial results of attempts to estimate the residence time of volcanic spring waters originating from basaltic lava aquifers. The results provide insight into the potential of the proposed method and its limitations.

Study area

The study area lies at the foot of the western, southern, and eastern sections of Mt. Fuji in Central Japan. Mt. Fuji is one of the largest Quaternary stratovolcanoes in Japan (elevation, 3776 m) and consists mainly of alternating basaltic lava flows and coarse-grained pyroclastic rocks (e.g. Tsuya et al. 1988). Mt. Fuji can be structurally divided into three parts: the Komitake, Older Fuji, and Younger Fuji volcanoes, in ascending stratigraphic order. The underlying basement rocks are mainly Neogene marine sediments (e.g. Miyaji et al. 1992).

The large amount of precipitation in the area means that Mt. Fuji contains large reservoirs of groundwater in its main body. Annual precipitation over the Mt. Fuji area ranges from 1500–2000 mm on the northern slope of the mountain to 2750–3000 mm on the eastern slope (Kizawa et al. 1969), with the overall area receiving an annual volume of $\sim 2 \times 10^9 \text{ m}^3$ of rainwater (Yamamoto 1970). Springs located around the foot of the mountain originate mainly from confined groundwater in permeable parts of early Holocene lava flows (e.g. Tsuchi 2007).

Previous studies have estimated the residence times of these spring waters based on ^3H or $^3\text{H}/^3\text{He}$ dating methods. In pioneering work conducted during the 1960s, groundwater in shallow aquifers (i.e. older lava flows of the Younger Fuji volcano) was found to possess high concentrations of ^3H derived from atmospheric nuclear tests, and

residence times were estimated to be on the order of several years (Ochiai and Kawasaki 1970). In contrast, groundwater in deep aquifers (i.e. mudflow deposits of the Older Fuji volcano) possessed low concentrations of ^3H , with estimated residence times of 20–30 years.

Yoshioka et al. (1993) reported relatively high ^3H concentrations (~38 TU) for springs emanating from the Younger Fuji lava at the eastern foot of Mt. Fuji, higher than those measured in the southeastern part of the mountain, and estimated residence times of ~30 yr using an exponential-type model. Using the $^3\text{H}/^3\text{He}$ dating method for samples collected in 1988, Mahara et al. (1993) obtained groundwater ages of ~10 yr for the southeastern foot of Mt. Fuji. Finally, Tsuchi (2007) reported ^3H concentrations of ~5 TU at the foot of the south and southern sections of Mt. Fuji (these sections are defined below) for water samples collected in the mid-1990s, indicating residence times of ~15 yr.

Sampling and analyses

During two sampling campaigns, carried out in August 2006 and March 2007, 30 samples were collected from springs and flowing wells around Mt. Fuji (Figure 1). The first sampling campaign collected 20 samples of spring water and 2 samples of groundwater from flowing wells at the southern foot of Mt. Ashitaka. During the second sampling campaign, samples were collected from eight springs.

Electrical conductivity (EC), pH, and water temperature were measured in the field at the time of sampling. In the laboratory, samples were analyzed for major ions, silica (SiO_2), stable isotopes (D and ^{18}O), and ^{36}Cl . Bicarbonate (HCO_3^-) concentrations were determined by titration with dilute H_2SO_4 solution. Other major dissolved ions (Na^+ , K^+ , Mg^{2+} , Ca^{2+} , Cl^- , SO_4^{2-} , and NO_3^-) were measured by ion chromatography analysis (Ion Analyzer IA-100; Dkk-Toa, Tokyo, Japan) at the Tandem Accelerator Complex, Research Facility Center for Science and Technology, University of Tsukuba, Japan. Dissolved SiO_2 concentrations were determined with an inductively coupled plasma–atomic emission spectroscope (ICP–AES) system (ICAP-757; Nippon Jarrell-Ash, Kyoto, Japan) at the Chemical Analysis Division, Research Facility Center for Science and Technology, University of Tsukuba, Japan. The overall ionic charge balance was better than $\pm 3\%$.

1 Stable isotope ratios of oxygen and hydrogen ($\delta^{18}\text{O}$ and δD) were measured with a
2 stable isotope mass spectrometer (MAT252; Thermo Finnigan, Bremen, Germany) at
3 the Hydrology Lab, Graduate School of Life and Environmental Sciences, University of
4 Tsukuba. Prior to analyses of stable isotopes, samples were equilibrated with CO_2 gas
5 for ^{18}O and H_2 gas with platinum catalysts for D. The analytical errors for $\delta^{18}\text{O}$ and δD
6 were 0.1‰ and 1‰, respectively.

7 For ^{36}Cl analysis, water samples were prepared as AgCl (see Tosaki et al. 2007). The
8 $^{36}\text{Cl}/\text{Cl}$ ratios of the samples were measured with the AMS system at the Tandem
9 Accelerator Complex, Research Facility Center for Science and Technology, University
10 of Tsukuba (Sasa et al. 2010). Also analyzed was the diluted NIST ^{36}Cl standard
11 ($^{36}\text{Cl}/\text{Cl} = 1.000 \times 10^{-11}$; Sharma et al. 1990). The overall precision of the system was
12 better than 2%, and the background level of $^{36}\text{Cl}/\text{Cl}$ measurement was $\sim 1 \times 10^{-15}$ (Sasa
13 et al. 2010). The $^{36}\text{Cl}/\text{Cl}$ ratios reported in this study include the statistical error derived
14 from uncertainties (1σ) in measurements of the sample, the standard, and the blank.

16 Results and discussion

17 *Stable isotopic composition and water chemistry*

18 Tables 1 and 2 summarize chemical and isotopic data, respectively, for the samples
19 collected during the two sampling campaigns. The measured δD and $\delta^{18}\text{O}$ values vary
20 from -66‰ to -46‰ and from -9.9‰ to -7.4‰ , respectively (Table 1). A δD – $\delta^{18}\text{O}$
21 diagram (Figure 2) shows that the δD and $\delta^{18}\text{O}$ values lie slightly below the local
22 meteoric water line ($\delta\text{D} = 8\delta^{18}\text{O} + 15.1$; Yasuhara et al. 2007), but lie directly on a
23 regression line for recharging groundwater, as obtained by Yasuhara et al. (2007) ($\delta\text{D} =$
24 $8\delta^{18}\text{O} + 13.5$). The identical slopes and similar d -values of the samples indicate a
25 meteoric origin and that evaporation during recharge made a negligible contribution to
26 stable isotopic fractionation (Yasuhara et al. 2007).

27 Figure 3 shows the spatial distribution of $\delta^{18}\text{O}$ values in spring water and
28 groundwater. As shown in the figure, the study area is divided into five regions (the
29 eastern, southeastern, southern, and western flanks of Mt. Fuji, and the foot of Mt.
30 Ashitaka) based on the altitude effect on $\delta^{18}\text{O}$ values. The southeastern foot of Mt. Fuji
31 is located around the distal part of the Mishima lava flow, surrounded by the eastern
32 foot of Mt. Ashitaka (1458 m) and the western outer rim of the crater at Mt. Hakone

(~1100 m). The $\delta^{18}\text{O}$ values of the springs at the southeastern foot of Mt. Fuji (-8.5‰ to -8.1‰ ; samples 8–11) are lower than the values obtained around Mt. Ashitaka (-7.4‰ to -7.8‰ ; samples 7 and 12–14). Similar findings were reported by Yoshioka et al. (1993), indicating that the springs at the southeastern foot of Mt. Fuji originate mainly from elevations above those of Mts. Ashitaka and Hakone; i.e., from the foot of Mt. Fuji, passing through the Mishima lava flow (Younger Fuji lava). At the eastern foot of the mountain, samples 1 and 2 have higher $\delta^{18}\text{O}$ values than those obtained at other springs. Considering the locations of these springs, the water probably originated from the foot of Mt. Hakone or from lower mountains. Consequently, these samples were not considered when estimating the residence times.

The data in Table 1 indicate that the majority of the samples are Ca-HCO_3 type, which is indicative of shallow groundwater, as reported previously in the present study area (e.g. Yoshioka et al. 1993). In contrast, several samples from the southern foot of Mt. Fuji have relatively high concentrations of NO_3^- . Some of the spring waters are more enriched in Na^+ than the general composition (e.g. sample 26). High Na^+ contents relative to K^+ are characteristic of groundwater in mudflow deposits of the Older Fuji volcano (Ikeda 1989), and may reflect cation exchange reactions between weathering-induced Ca^{2+} in groundwater and Na^+ in the aquifer matrix (e.g. Chapelle and Knobel 1983), suggesting a relatively long residence time.

Most of the spring waters with high NO_3^- concentrations have higher $\delta^{18}\text{O}$ values than those of nearby springs (Tables 1 and 2; Figure 3), indicating a relatively low recharge elevation and an origin from shallow aquifers. These waters would have been affected by NO_3^- from agricultural sources, and possibly also anthropogenic Cl. Accordingly, samples with high NO_3^- concentrations (>10 mg/L) were excluded from further analyses (cf. Davis et al. 2003).

Estimates of residence time based on bomb-produced ^{36}Cl

Figure 4 shows the $^{36}\text{Cl}/\text{Cl}$ ratios measured in spring waters and groundwater plotted against Cl^- concentrations. The $^{36}\text{Cl}/\text{Cl}$ ratios range from 43×10^{-15} to 412×10^{-15} (excluding the samples with high NO_3^- concentrations). The samples with high NO_3^- concentrations have relatively low $^{36}\text{Cl}/\text{Cl}$ ratios (39×10^{-15} to 62×10^{-15} ; Tables 1 and

2), suggesting the influence of anthropogenic Cl. The $^{36}\text{Cl}/\text{Cl}$ ratios are relatively high in the eastern part of Mt. Fuji. With the exception of the NO_3^- -contaminated samples, the observed variation in ^{36}Cl content can be ascribed to differences in the residence times of the waters. Because we cannot be sure that the other samples did not receive additional Cl after recharge, the obtained ^{36}Cl concentrations are likely to be more reliable than the $^{36}\text{Cl}/\text{Cl}$ values. As discussed below, the mean recharge elevation of the springs is well constrained in the range of 1100–2250 m (Yasuhara et al. 1995), which probably indicates no significant difference among the springs in terms of the influence of evapotranspiration on the observed ^{36}Cl concentrations.

To estimate the groundwater residence time based on bomb-produced ^{36}Cl , we developed a suitable methodology and applied it to the collected samples. The measured $^{36}\text{Cl}/\text{Cl}$ ratios were converted to above-background (bomb-derived) ^{36}Cl concentrations (e.g. Cook and Robinson 2002), by (1) subtracting the background $^{36}\text{Cl}/\text{Cl}$ ratio from measured ratios, and (2) multiplying by the Cl^- concentrations to derive the above-background ^{36}Cl concentrations (atoms/L).

Reference ^{36}Cl data are also required to estimate the residence time. These data were obtained as follows: (1) reduction of Dye-3 fallout data (Synal et al. 1990) using the background ^{36}Cl flux at the Dye-3 site (Synal et al. 1994) to derive a time series of the above-background ^{36}Cl flux; (2) linear scaling of the above-background ^{36}Cl flux using the estimated total bomb-produced ^{36}Cl fallout in Tsukuba (Tosaki et al. 2008) after correcting for the difference in precipitation amount between the two sites (i.e. between the Tsukuba and Mt. Fuji areas); and (3) conversion of the scaled above-background ^{36}Cl flux to the above-background ^{36}Cl concentration by using a mass balance equation for the recharge area (Andrews et al. 1994). To estimate the residence times, the obtained reference curve was then compared with the above-background ^{36}Cl concentrations obtained for the samples.

The background $^{36}\text{Cl}/\text{Cl}$ ratio was assumed to be $(60 \pm 10) \times 10^{-15}$, based on the lower limit of the measured $^{36}\text{Cl}/\text{Cl}$ range (see Figure 4). After subtracting this value from the $^{36}\text{Cl}/\text{Cl}$ ratios measured for the samples, these values were multiplied by each Cl^- concentration to obtain the above-background ^{36}Cl concentration (Figure 5c). In contrast, yearly values of bomb-produced ^{36}Cl fallout (values above the background level) at the Dye-3 site were deduced by subtracting the natural background ^{36}Cl flux

(20 ± 6 atoms $\text{m}^{-2} \text{s}^{-1}$; Synal et al. 1994) from the original Dye-3 fallout data (Synal et al. 1990). These values were then scaled with a scaling factor obtained in the manner described below.

In our previous study (Tosaki et al. 2008), the total bomb-produced ^{36}Cl fallout in Tsukuba, Central Japan ($36^{\circ}04'\text{N}$, $140^{\circ}08'\text{E}$), was estimated to be 2.3×10^{12} atoms m^{-2} , based on depth-profiling of groundwater in an upland area. A revision of the assumed values of parameters used in the calculation (i.e. assumed layer porosities with 10% uncertainties (cf. Yasuhara et al. 1991) and a surface runoff rate of $90 \pm 10\%$ in upland areas) yields an estimated fallout of $(2.4 \pm 0.3) \times 10^{12}$ atoms m^{-2} .

The average annual precipitation in Tsukuba is 1246 ± 178 mm (for 1951–2000; AMeDAS data from the Japan Meteorological Agency), which is approximately half that in the Mt. Fuji area (2500 ± 250 mm, as discussed below; Kizawa et al. 1969). Because the fallout rates of bomb-derived nuclides are expected to depend on the amount of precipitation (Phillips 2000), the difference in precipitation amount needs to be accounted for in our calculations. Here, we employed the following correction scheme proposed by Phillips (2000) for natural ^{36}Cl flux:

$$D_{\text{local}} = \overline{D}(\lambda) + S_{\text{D}}(\lambda)(P_{\text{local}} - \overline{P}(\lambda)) \quad (1)$$

where D_{local} is the measured local ^{36}Cl deposition flux (atoms $\text{m}^{-2} \text{s}^{-1}$), $\overline{D}(\lambda)$ is the mean ^{36}Cl deposition flux for a given latitude band (atoms $\text{m}^{-2} \text{s}^{-1}$), P_{local} is the local mean annual precipitation (mm), $\overline{P}(\lambda)$ is the mean annual precipitation for a given latitude band (mm), and $S_{\text{D}}(\lambda)$ is a parameter that describes the dependence of ^{36}Cl deposition flux on precipitation amount within a given latitude band. Phillips (2000) obtained an $S_{\text{D}}(\lambda)$ value of 0.047 ± 0.006 atoms $\text{m}^{-2} \text{s}^{-1} (\text{mm yr}^{-1})^{-1}$ for the latitude 40°N .

In the present study, Equation (1) was revised for bomb-produced ^{36}Cl fallout, as follows:

$$F_{\text{local}} = \overline{F}(\lambda) + fS_{\text{D}}(\lambda)(P_{\text{local}} - \overline{P}(\lambda)) \quad (2)$$

where F_{local} is the measured ^{36}Cl fallout (atoms m^{-2}), $\overline{F}(\lambda)$ is the mean ^{36}Cl fallout for a given latitude band (atoms m^{-2}), and f is a conversion factor that represents the

ratio of bomb-produced ^{36}Cl fallout (atoms m^{-2}) to natural background ^{36}Cl flux (atoms $\text{m}^{-2} \text{s}^{-1}$) at a given site within the selected latitude band. $\bar{P}(\lambda)$ was assumed to be 818 ± 77 mm, based on the estimated average precipitation amount at $30^\circ\text{--}50^\circ\text{N}$ (Baumgartner and Reichel 1975). As mentioned above, P_{local} was taken as 1246 ± 178 mm for the Tsukuba Upland area and 2500 ± 250 mm for the Mt. Fuji area. The conversion factor f was obtained by dividing the total bomb-produced ^{36}Cl fallout in Tsukuba ($(2.4 \pm 0.3) \times 10^{12}$ atoms m^{-2}) by the natural background ^{36}Cl flux in Tsukuba (32 ± 2 atoms $\text{m}^{-2} \text{s}^{-1}$; unpublished data for bulk precipitation during 2004–2007), yielding a value of $(7.5 \pm 1.0) \times 10^{10}$ atoms m^{-2} (atoms $\text{m}^{-2} \text{s}^{-1}$) $^{-1}$. Because the study area occurs at a latitude of $\sim 35^\circ\text{N}$, it would be reasonable to employ an $S_{\text{D}}(\lambda)$ value of 0.047 ± 0.006 atoms $\text{m}^{-2} \text{s}^{-1}$ (mm yr^{-1}) $^{-1}$, as obtained previously for the latitude 40°N (Phillips 2000). Application of the above correction scheme yielded an estimated fallout of $(6.8 \pm 1.7) \times 10^{12}$ atoms m^{-2} for the Mt. Fuji area.

The time series of bomb-produced ^{36}Cl fallout at the Dye-3 site was scaled according to the ratio of the estimated fallout at the study area ($(6.8 \pm 1.7) \times 10^{12}$ atoms m^{-2}) to the value at the Dye-3 site ($(2.43 \pm 0.03) \times 10^{12}$ atoms m^{-2} , as calculated by integrating the above-background ^{36}Cl flux from 1945 to 1985; Synal et al. 1990, 1994). The scaled fallout values obtained using a scaling factor of 2.8 ± 0.7 were then converted to ^{36}Cl concentrations using the following mass balance equation (Andrews et al. 1994):

$$A = \frac{F \times 3.156 \times 10^7}{P} \left(\frac{100}{100 - E} \right) \quad (3)$$

where A is the ^{36}Cl concentration, F is the ^{36}Cl fallout (atoms $\text{m}^{-2} \text{s}^{-1}$), P is the mean annual precipitation (mm), and E is the mean annual evapotranspiration rate (%).

Mean recharge elevations, estimated from the altitude effect on the stable isotopic composition for springs located on the south, east, and west slopes of Mt. Fuji, are 1100–2000 m, 1250–2200 m and 1600–2250 m, respectively (Yasuhara et al. 1995). P was assumed to be 2500 ± 250 mm, because the precipitation amount at elevations of 1100–2250 m is generally 2250–2750 mm, based on the precipitation map reported by Kizawa et al. (1969).

According to estimates of evaporation based on the fractionation of stable isotopes (Yasuhara et al. 1997), evaporation rates for the investigated slopes are 16% at 2000 m

1 and 21% at 1000 m. These values does not account for transpiration, which would cause
2 little or no isotopic fractionation. The potential evapotranspiration rate was calculated to
3 be $46 \pm 3\%$ at Mishima (elevation, 20.5 m) in the southeastern part of Mt. Fuji, using
4 the Penman equation (e.g. Brutsaert 2005) with an assumed albedo of 0.20 ± 0.05 for
5 vegetated surfaces. Given the estimated recharge elevation of the springs (1100–2250
6 m), the actual evapotranspiration rate in this area is probably between ~20% and ~46%.
7 In this regard, an evapotranspiration rate of 35%, as calculated by Tsuchi (2007) based
8 on the water balance within the Mt. Fuji area, seems to be reasonable; accordingly, E
9 was assumed to be $35 \pm 5\%$. Figure 5a and 5b shows the resulting time series of
10 above-background ^{36}Cl concentrations estimated for the Mt. Fuji area.

11 Most spring waters around Mt. Fuji originate from confined aquifers within the New
12 Fuji lava layers or from an aquifer that overlies the low-permeability Older Fuji mud
13 flow layer (e.g. Tsuchi 2007). Consequently, piston flow would be a reasonable
14 approximation of groundwater flow in this area for an initial interpretation of the data.
15 The residence times of spring waters can be roughly estimated by comparing Figure 5b
16 and 5c. Considering the elevated concentrations of tritium in spring waters during the
17 late 1960s (Ochiai and Kawasaki 1970), most of the spring waters would have been
18 recharged during the post-bomb period. Hence, the estimated residence times were
19 obtained using the post-peak portion of the reference curve shown in Figure 5b. One
20 exception is sample 26, which showed a low $\delta^{18}\text{O}$ value relative to the values for nearby
21 springs, indicating recharge from a higher part of the slope. This sample also has the
22 lowest K/Na ratio among the samples (see Table 1), which is characteristic of the deep
23 Older Fuji aquifer (Ikeda 1989). In addition, its low $^{36}\text{Cl}/\text{Cl}$ ratio, $(53 \pm 7) \times 10^{-15}$,
24 suggests that it was recharged during the pre-bomb period (residence time > 55 yr).

25 Figure 6 shows the distribution of estimated residence times for spring water and
26 groundwater (see also Table 2). Although the distribution is somewhat complex, the
27 residence times estimated for spring water at the eastern foot of Mt. Fuji (22–36 yr) are
28 generally older than those estimated for other areas. Two groundwater samples from
29 flowing wells near Mt. Ashitaka also yielded relatively old ages (23–37 yr). In contrast,
30 spring waters at the southeastern foot of Mt. Fuji have younger residence times (0–31
31 yr), while springs at the southern foot yield variable residence times.

4. Discussion

For the southeastern foot of Mt. Fuji, the residence times estimated in the present study (0–31 yr) overlap with previous estimates obtained using $^3\text{H}/^3\text{He}$ dating (~10 yr; Mahara et al. 1993). Figure 7 shows the available data on ^3H concentrations for selected springs, along with the range of ^3H concentrations in precipitation over Central Japan (see Table 2 for tritium-based ages interpreted from previous tritium data). ^3H concentrations at the foot of the eastern and western sections of Mt. Fuji are higher than those at the southeastern and southern sections. Specifically, samples 4 and 29 from the former sections yield slightly older tritium-based ages (12–17 and 3–16 yr, respectively; Table 2). This difference between the eastern–western and south–southeastern sections is basically consistent with the trend observed for ^{36}Cl .

The estimated residence time for sample 26 (>55 yr) reflects the fact that the spring is located along the Agoyama Fault. The old age indicates that this spring water originates from groundwater in deeper aquifers, including mudflow deposits of the Older Fuji volcano. This interpretation is consistent with the field observation that this spring discharges in a direction away from Mt. Fuji. Other springs at the southern foot of the mountain yield variable residence times, possibly reflecting the specific geological setting of each site.

Figure 8 compares ^{36}Cl -based ages and tritium-based ages. Although the two distributions of residence time are basically in agreement, the ^{36}Cl -based ages are generally much older than the tritium-based ages (Table 2; Figure 8). This discrepancy may reflect (1) dispersion during groundwater flow, (2) the mixing of groundwaters of different ages (e.g. exponential mixing), or (3) chlorine recycling via vegetation. Generally, dispersion acts to broaden the bomb pulse, resulting in an overestimate of residence time for the post-peak portion of the reference data; however, this would have a similar affect on both the ^{36}Cl and tritium pulses, as would exponential mixing. Therefore, these two processes cannot explain the difference in residence time estimated using the two tracers. Because the study area is located in a humid temperate climate, the recycling of chlorine by vegetation may have affected the ^{36}Cl contents of the springs. The estimates are also complicated by the relatively small change in the fallout rate of ^{36}Cl after 1975.

In addition to the above effects, several factors may produce errors in estimates of

1 residence time. Although we assumed a uniform background level for the entire study
2 area, the $^{36}\text{Cl}/\text{Cl}$ ratio is clearly dependent on stable Cl transported from the sea.
3 According to Hiyama et al. (1995), the concentration of Cl^- in precipitation for the
4 southwestern foot of Mt. Fuji varies from 0.59 mg/L at an elevation of 1000 m to 0.21
5 mg/L at 2390 m, indicating reduced Cl transport at higher elevation. In the case of a
6 large difference in recharge elevation among the studied springs, the assumed
7 background of 60×10^{-15} may result in an underestimate of the actual background at
8 higher elevations.

9 Four samples from the western foot of Mt. Fuji yield $^{36}\text{Cl}/\text{Cl}$ ratios of $\sim 100 \times 10^{-15}$
10 (Table 2), which is almost twice the assumed natural background level (60×10^{-15}).
11 However, sample 30 shows especially low concentrations of dissolved ions (Table 1)
12 and a relatively high $\delta^{18}\text{O}$ value (Table 2), which may suggest a short residence time.
13 One reasonable explanation for the discrepancy in residence times is that the natural
14 background level of $^{36}\text{Cl}/\text{Cl}$ at the western foot of Mt. Fuji is higher than that for the
15 other areas considered in the present study. This may reflect differences in Cl^-
16 concentrations within precipitation, as a reduction in the supply of stable chloride
17 results in an increase in the $^{36}\text{Cl}/\text{Cl}$ ratio. In addition, the springs located at the western
18 foot of the mountain are recharged at a higher elevation (1600–2250 m) than the springs
19 at other parts of the mountain (Yasuhara et al. 1995).

20 Given the possibility of chlorine recycling and the minor change in the reference data
21 after 1975, estimates of the residence time of young water would contain relatively large
22 uncertainties. However, it is reasonable that artesian groundwater (from depths greater
23 than 100 m) near Mt. Ashitaka would yield relatively old ages. Therefore, dating based
24 on a ^{36}Cl bomb-pulse may be more reliable and sensitive for groundwater recharged
25 before 1975, back as far as the bomb-peak period.

27 **Conclusions and outlook**

28 The residence times of spring water around Mt. Fuji were estimated based on
29 bomb-produced ^{36}Cl . Dye-3 fallout data were used to construct a reference time series
30 of $^{36}\text{Cl}/\text{Cl}$ values in the Mt. Fuji area. To focus solely on the bomb-derived ^{36}Cl
31 components, all the measured and reference ^{36}Cl data were presented as
32 above-background ^{36}Cl concentrations after subtracting the background levels. The total

1 bomb-produced ^{36}Cl fallout in the Mt. Fuji area was estimated to be $(6.8 \pm 1.7) \times 10^{12}$
2 atoms m^{-2} . The time series of above-background ^{36}Cl concentration at the Dye-3 site
3 was then scaled according to the ratio of the estimated total bomb-produced ^{36}Cl fallout
4 in the study area to that at the Dye-3 site $((2.43 \pm 0.03) \times 10^{12} \text{ atoms m}^{-2})$.

5 Measured $^{36}\text{Cl}/\text{Cl}$ ratios in spring water were also converted to above-background
6 ^{36}Cl concentrations and then compared with the estimated time series to yield estimates
7 of residence time, assuming piston flow. The distribution of ^{36}Cl -based residence times
8 is reasonably consistent with that of tritium-based residence times, although the two
9 tracers yield contrasting residence times, possibly due to chlorine recycling via
10 vegetation and the relatively small change in the reference data after ~1975. This result
11 suggests that ^{36}Cl -based estimates for recharge years after ~1975 would involve large
12 uncertainties. Given the ages estimated for artesian groundwater, ^{36}Cl bomb pulse dating
13 may be more reliable and sensitive for groundwater recharged before 1975, back as far
14 as the bomb-peak period.

15 The natural background $^{36}\text{Cl}/\text{Cl}$ ratio may vary for different slopes upon Mt Fuji,
16 reflecting variations in the Cl^- concentration within precipitation. Therefore, the
17 condition of the recharge area (i.e. the natural background level of $^{36}\text{Cl}/\text{Cl}$, mean annual
18 precipitation, and evapotranspiration rate) must be known as accurately as possible to
19 minimize errors in estimated residence times.

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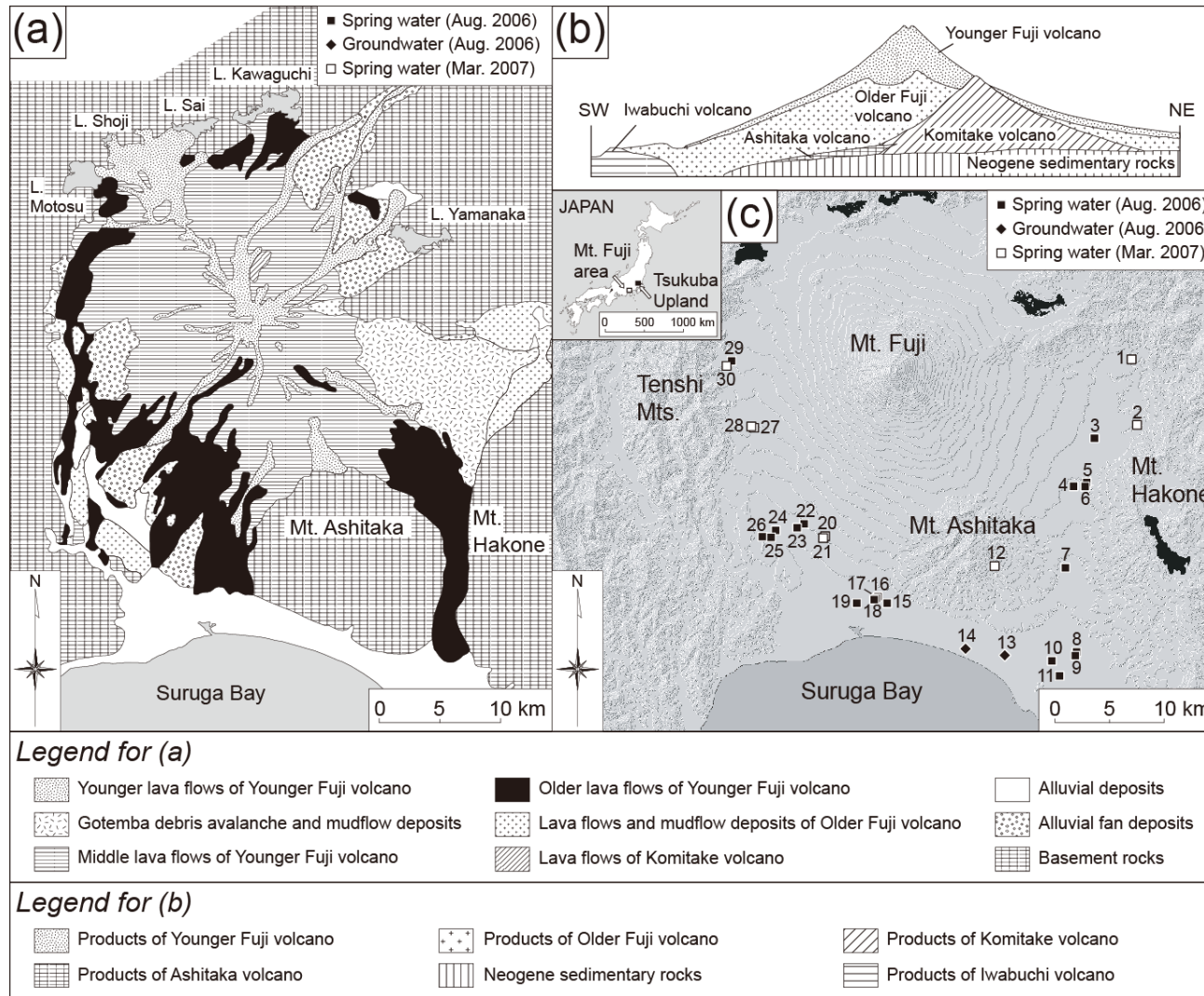


Figure 1. (a) Geological map of the Mt. Fuji area (after Tsuya 1968; Machida 1977). (b) Schematic cross-section through Mt. Fuji (after Tsuya 1940). (c) Topographic map of the Mt. Fuji area. Also shown on (a) and (c) are the sampling locations and sampling dates.

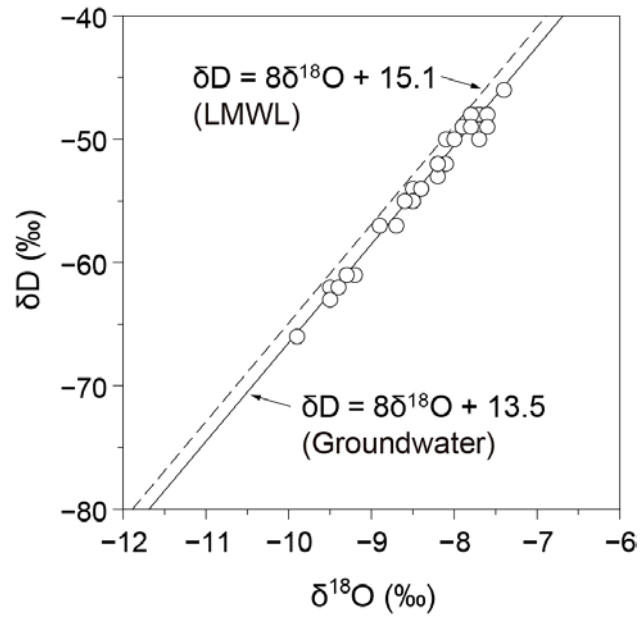


Figure 2. Stable isotopic compositions of samples of spring water and groundwater. Open circles indicate $\delta^{18}\text{O}$ and δD values of the samples. Solid and dashed lines represent regression lines for precipitation (local meteoric water line: LMWL) and groundwater obtained by Yasuhara et al. (2007), respectively.

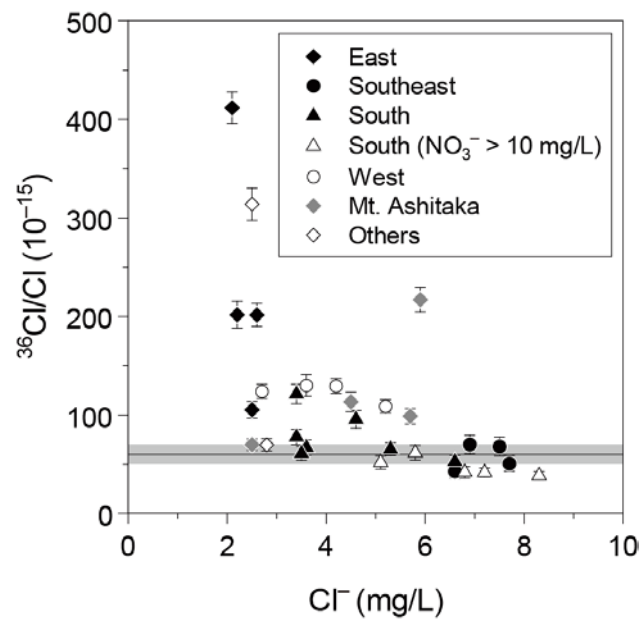
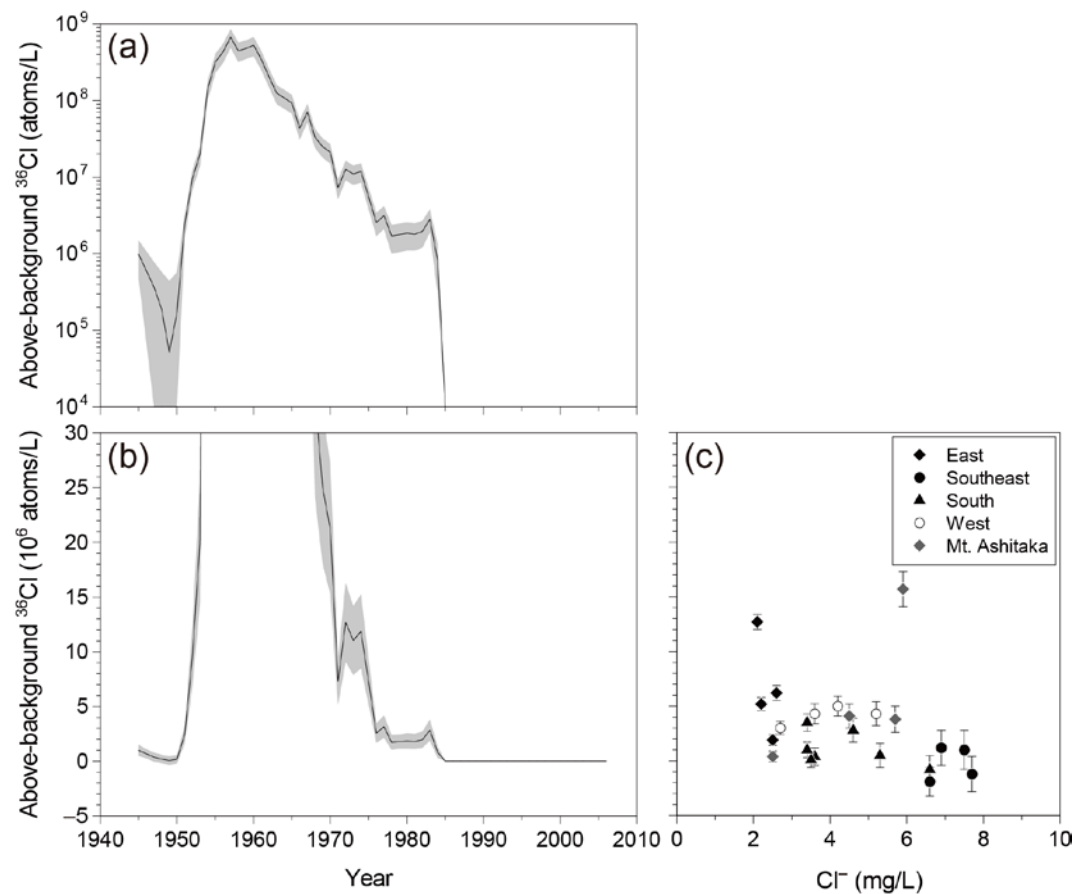


Figure 4. $^{36}\text{Cl}/\text{Cl}$ ratios plotted against chloride concentrations for samples of spring water and groundwater. Solid line and gray band indicate the assumed range of background $^{36}\text{Cl}/\text{Cl}$ ratios in the study area. For an explanation of the geographic regions (e.g., East and Southeast), see Fig. 3.

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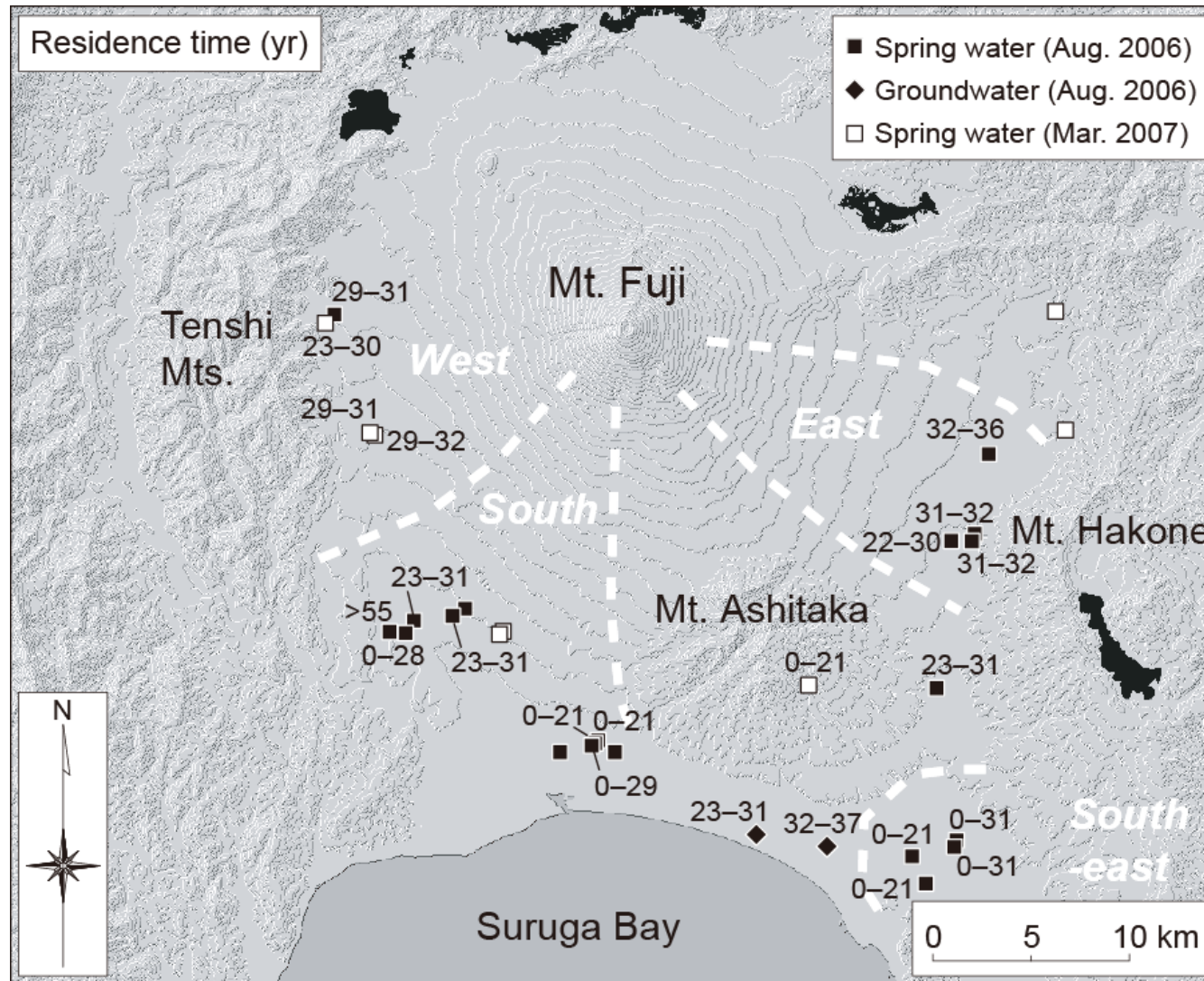


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4 Figure 5. (a) Predicted time series of above-background ^{36}Cl concentration (see the text for details of how these values were calculated) in the Mt.
 5 Fuji area (logarithmic scale). (b) Predicted time series of above-background ^{36}Cl concentration (linear scale). (c) Above-background ^{36}Cl
 6 concentrations of spring water and groundwater. Gray areas shown in (a) and (b) indicate the error estimated from the propagation of uncertainties
 7 arising in the assumed values of parameters. For an explanation of the geographic regions (e.g., East and Southeast), see Fig. 3.

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3 Figure 6. Spatial distribution of the residence time of spring water and groundwater in the Mt. Fuji area, as estimated from ^{36}Cl . The symbol shape
4 indicates the date of sample collection.

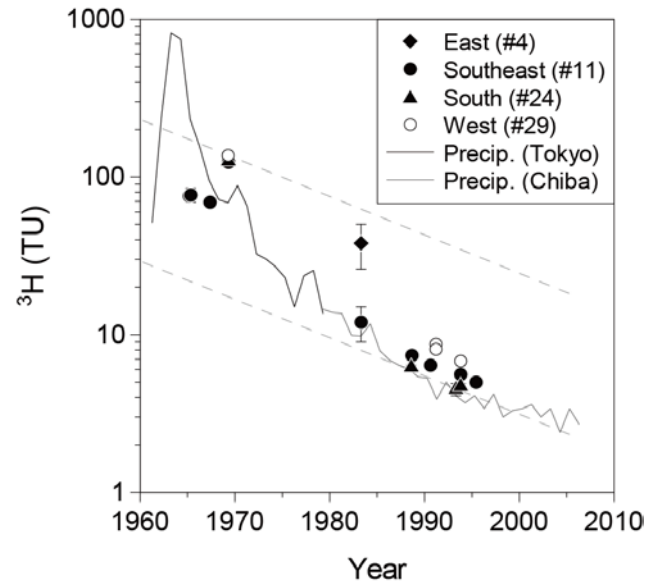


Figure 7. Tritium concentrations in selected springs within the Mt. Fuji area. Data are from Takahashi et al. (1969), Ochiai and Kawasaki (1970), Tsuchi (1992, 1996, 2001), Yoshioka et al. (1993), Mahara et al. (1993), Masuda et al. (1994), Ochiai (1995), Kakiuchi (1995), and Nakai (1996). Each value is expressed as the concentration at the time of sampling. Tritium concentrations in precipitation at Tokyo (1961–1979; IAEA/WMO 2006) and Chiba (1979–2006; NIRS 2006), Central Japan, are shown for comparison. Dotted lines indicate the decay curves for the maximum and minimum tritium concentrations of the samples. The recharge year for each sample can be obtained from the intersection between a straight line that passes through the tritium concentration data and the precipitation data.

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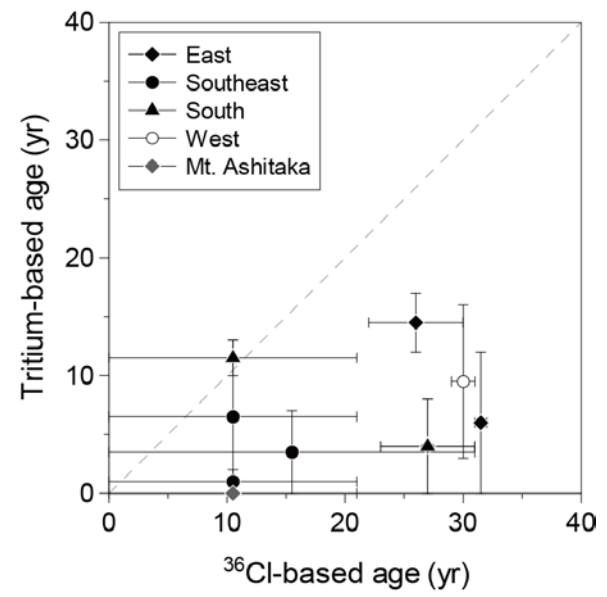


Figure 8. Comparison of ^{36}Cl -based and tritium-based ages.

1 Table 1. Chemical compositions of spring water and groundwater sampled from around Mt. Fuji

Sam ple no.	Name	Type	Elev ation (m)	Sampling date	Na ⁺ (mg/L)	K ⁺ (mg/L)	Mg ²⁺ (mg/L)	Ca ²⁺ (mg/L)	Cl ⁻ (mg/L)	SO ₄ ²⁻ (mg/L)	NO ₃ ⁻ (mg/L)	HCO ₃ ⁻ (mg/L)	SiO ₂ (mg/L)
1	Yubune	Spring	344	5 Mar. 2007	4.7	1.1	5.3	10.2	2.8	3.4	2.2	58.6	52.7
2	Kuwaki	Spring	400	6 Mar. 2007	4.5	1.0	3.7	13.8	2.5	4.6	3.0	58.6	40.3
3	Nimaibashi	Spring	458	24 Aug. 2006	11.3	1.8	4.8	14.5	2.1	18.2	0.6	72.6	25.2
4	Nakashimizu	Spring	385	24 Aug. 2006	6.8	1.6	4.7	12.8	2.5	7.0	3.7	63.4	36.6
5	Numata	Spring	389	23 Aug. 2006	8.4	1.5	5.6	15.0	2.6	14.3	1.9	71.4	32.7
6	Futagosuijin	Spring	380	24 Aug. 2006	7.8	1.5	5.1	13.4	2.2	12.7	2.1	65.9	32.8
7	Hirayama	Spring	185	23 Aug. 2006	7.5	1.5	4.0	12.4	5.7	9.0	5.7	52.5	39.6
8	Komoike	Spring	32	21 Aug. 2006	11.2	2.1	5.9	16.8	7.5	10.9	3.4	82.4	44.9
9	Suisen-en	Spring	32	21 Aug. 2006	9.7	1.8	5.0	14.9	6.9	15.4	6.2	60.4	41.4
10	Fujiyusuichi	Spring	24	21 Aug. 2006	8.2	1.9	4.3	12.4	7.7	9.3	6.3	51.2	42.8
11	Kakitagawa	Spring	20	21 Aug. 2006	9.6	1.9	4.8	14.5	6.6	17.0	4.7	58.6	41.1
12	Mizujinja	Spring	570	8 Mar. 2007	3.4	1.1	0.9	3.6	2.5	1.4	2.3	16.5	30.6
13	Hiyoshijinja	Groundwater	20	21 Aug. 2006	7.3	1.2	4.9	11.2	5.9	1.1	2.6	63.4	46.3
14	Hara	Groundwater	7	21 Aug. 2006	7.2	1.0	2.1	7.0	4.5	2.6	0.5	39.7	39.3
15	Iouji	Spring	23	23 Aug. 2006	7.1	2.7	5.0	12.5	6.8	13.6	16.7	40.3	45.6
16	Kansekien	Spring	33	21 Aug. 2006	6.3	1.5	2.7	7.8	3.6	6.3	5.0	36.0	38.9
17	Takifudou	Spring	24	21 Aug. 2006	6.3	1.6	2.7	7.8	3.5	6.4	5.2	36.0	38.2
18	Youmeiji	Spring	21	21 Aug. 2006	6.7	1.4	2.7	7.7	3.4	6.5	5.1	36.0	37.6

19	Hounji	Spring	9	23 Aug. 2006	8.0	2.3	5.3	14.9	5.1	16.4	19.0	47.6	40.5
20	Sugita (1)	Spring	204	8 Mar. 2007	6.4	5.5	8.2	28.0	8.3	39.7	64.1	22.6	38.1
21	Sugita (2)	Spring	201	8 Mar. 2007	6.6	3.8	6.2	21.2	7.2	31.1	40.6	25.6	38.7
22	Izurimizu	Spring	208	22 Aug. 2006	7.0	2.5	4.9	19.1	5.8	18.9	24.0	47.6	43.7
23	Kamikoizumi	Spring	165	22 Aug. 2006	6.8	1.6	5.1	12.9	3.4	14.2	9.4	50.6	43.6
24	Wakutamaike	Spring	126	22 Aug. 2006	7.5	2.0	4.0	11.7	4.6	9.0	9.4	48.8	43.1
25	Hagoromo	Spring	121	23 Aug. 2006	7.7	2.1	3.5	11.4	5.3	7.4	8.2	48.2	44.3
26	Yoshimaike	Spring	127	23 Aug. 2006	14.2	1.7	4.4	12.6	6.6	18.4	7.6	58.0	44.5
27	Shiraitonotaki	Spring	480	7 Mar. 2007	8.6	1.6	3.5	9.7	4.2	8.9	7.1	44.5	37.0
28	Shiraitoyusui	Spring	485	7 Mar. 2007	9.2	1.9	3.7	10.5	5.2	10.9	8.3	45.8	35.0
29	Inokashira	Spring	730	22 Aug. 2006	5.9	1.0	2.4	8.2	3.6	7.7	3.2	32.9	24.6
30	Jinbanotaki	Spring	700	7 Mar. 2007	4.0	0.9	1.7	7.8	2.7	5.9	3.0	27.5	20.0

1 Table 2. Isotopic composition of spring water and groundwater sampled from around Mt. Fuji

Sample no.	Name	$\delta^{18}\text{O}$	δD	$^{36}\text{Cl}/\text{Cl}$	^{36}Cl concentration	Above-background $^{36}\text{Cl}/\text{Cl}$	Above-background ^{36}Cl concentration	^{36}Cl -based age	Tritium-based age*
		(‰)	(‰)	(10^{-15})	(10^6 atoms/L)	(10^{-15})	(10^6 atoms/L)		
1	Yubune	-7.9	-49	70 ± 6	3.3 ± 0.3	10 ± 12	0.5 ± 0.6	—	—
2	Kuwaki	-8.1	-50	314 ± 16	13.5 ± 0.7	254 ± 19	11.0 ± 0.8	—	—
3	Nimaibashi	-9.9	-66	412 ± 16	14.8 ± 0.6	352 ± 19	12.7 ± 0.7	32–36	—
4	Nakashimizu	-8.9	-57	105 ± 9	4.4 ± 0.4	45 ± 13	1.9 ± 0.5	22–30	12–17
5	Numata	-9.5	-62	201 ± 12	8.8 ± 0.5	141 ± 15	6.2 ± 0.7	31–32	0–12
6	Futagosuijin	-9.4	-62	202 ± 14	7.4 ± 0.5	142 ± 17	5.2 ± 0.6	31–32	—
7	Hirayama	-7.7	-48	99 ± 8	9.6 ± 0.8	39 ± 13	3.8 ± 1.2	23–31	0–8
8	Komoike	-8.1	-52	68 ± 9	8.7 ± 1.2	8 ± 14	1.0 ± 1.8	0–31	—
9	Suisen-en	-8.2	-53	70 ± 9	8.2 ± 1.1	10 ± 14	1.2 ± 1.6	0–31	0–7
10	Fujiyusuichi	-8.2	-52	51 ± 8	6.6 ± 1.0	-9 ± 13	-1.2 ± 1.6	0–21	0–2
11	Kakitagawa	-8.5	-55	43 ± 7	4.8 ± 0.7	-17 ± 12	-1.9 ± 1.3	0–21	0–13
12	Mizujinja	-7.8	-48	70 ± 6	2.9 ± 0.2	10 ± 12	0.4 ± 0.5	0–21	0
13	Hiyoshijinja	-7.4	-46	217 ± 12	21.7 ± 1.2	157 ± 16	15.7 ± 1.6	32–37	—
14	Hara	-7.6	-48	113 ± 10	8.7 ± 0.7	53 ± 14	4.1 ± 1.1	23–31	—
15	Iouji	-8.0	-50	42 ± 6	4.8 ± 0.7	-18 ± 12	-2.1 ± 1.3	—	—
16	Kansekien	-8.4	-54	67 ± 8	4.1 ± 0.5	7 ± 13	0.4 ± 0.8	0–21	10–13
17	Takifudou	-8.5	-54	61 ± 7	3.7 ± 0.4	1 ± 12	0.1 ± 0.7	0–21	—

18	Youmeiji	-8.5	-55	78 ± 8	4.5 ± 0.5	18 ± 13	1.0 ± 0.7	0-29	-
19	Hounji	-8.2	-52	52 ± 7	4.4 ± 0.6	-8 ± 12	-0.7 ± 1.0	-	-
20	Sugita (1)	-7.6	-49	39 ± 4	5.5 ± 0.6	-21 ± 11	-3.0 ± 1.5	-	-
21	Sugita (2)	-7.7	-50	42 ± 4	5.1 ± 0.5	-18 ± 11	-2.2 ± 1.3	-	-
22	Izurimizu	-7.8	-49	62 ± 7	6.1 ± 0.7	2 ± 12	0.2 ± 1.2	-	-
23	Kamikoizumi	-8.4	-54	121 ± 10	7.0 ± 0.6	61 ± 14	3.5 ± 0.8	23-31	-
24	Wakutamaike	-8.5	-55	96 ± 9	7.5 ± 0.7	36 ± 14	2.8 ± 1.1	23-31	0-8
25	Hagoromo	-8.6	-55	65 ± 6	5.9 ± 0.6	5 ± 12	0.5 ± 1.1	0-28	-
26	Yoshimaike	-9.3	-61	53 ± 7	5.9 ± 0.7	-7 ± 12	-0.8 ± 1.3	>55 or 0-21	-
27	Shiraitonotaki	-9.5	-63	129 ± 8	9.3 ± 0.5	69 ± 13	5.0 ± 0.9	29-32	-
28	Shiraitoyusui	-9.2	-61	109 ± 7	9.6 ± 0.6	49 ± 12	4.3 ± 1.1	29-31	-
29	Inokashira	-9.3	-61	130 ± 11	8.0 ± 0.7	70 ± 15	4.3 ± 0.9	29-31	3-16
30	Jinbanotaki	-8.7	-57	124 ± 7	5.8 ± 0.3	64 ± 12	3.0 ± 0.6	23-30	-

*Original tritium concentration data are from Takahashi et al. (1969), Ochiai and Kawasaki (1970), Tsuchi (1992, 1996, 2001), Yoshioka et al. (1993), Mahara et al. (1993), Masuda et al. (1994), Ochiai (1995), Kakiuchi (1995), and Nakai (1996).