Estimation of Groundwater Residence Time Using the 36Cl Bomb Pulse

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Estimation of groundwater residence time using the $^{36}\text{Cl}$ bomb pulse

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Abstract

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

4
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 $^{3}$He, 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 ($3.01 \times 10^5$ 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 $^{3}$H 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 $^{36}$Cl/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. Scheffel 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 $\sim 2 \times 10^9$ 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 $^3$H or $^3$H/$^3$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 $^3$H 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 $^3$H, with estimated residence times of 20–30 years.

Yoshioka et al. (1993) reported relatively high $^3$H 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 $^3$H/$^3$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 $^3$H 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$_2$SO$_4$ 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 ±3%.
Stable isotope ratios of oxygen and hydrogen ($\delta^{18}$O and $\delta^D$) were measured with a stable isotope mass spectrometer (MAT252; Thermo Finnigan, Bremen, Germany) at the Hydrology Lab, Graduate School of Life and Environmental Sciences, University of Tsukuba. Prior to analyses of stable isotopes, samples were equilibrated with CO$_2$ gas for $^{18}$O and H$_2$ gas with platinum catalysts for D. The analytical errors for $\delta^{18}$O and $\delta^D$ were 0.1‰ and 1‰, respectively.

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

Results and discussion

Stable isotopic composition and water chemistry

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

Figure 3 shows the spatial distribution of $\delta^{18}$O values in spring water and groundwater. As shown in the figure, the study area is divided into five regions (the eastern, southeastern, southern, and western flanks of Mt. Fuji, and the foot of Mt. Ashitaka) based on the altitude effect on $\delta^{18}$O values. The southeastern foot of Mt. Fuji is located around the distal part of the Mishima lava flow, surrounded by the eastern foot of Mt. Ashitaka (1458 m) and the western outer rim of the crater at Mt. Hakone.
The $\delta^{18}O$ 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 $\delta^{18}O$ 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 $\delta^{18}O$ 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 $^{36}$Cl/Cl ratios measured in spring waters and groundwater plotted against Cl$^-$ concentrations. The $^{36}$Cl/Cl ratios range from $43 \times 10^{-15}$ to $412 \times 10^{-15}$ (excluding the samples with high NO$_3^-$ concentrations). The samples with high NO$_3^-$ concentrations have relatively low $^{36}$Cl/Cl ratios ($39 \times 10^{-15}$ to $62 \times 10^{-15}$; Tables 1 and
2), suggesting the influence of anthropogenic Cl. The $^{36}$Cl/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 $^{36}$Cl/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 $^{36}$Cl/Cl ratios were converted to above-background (bomb-derived) $^{36}$Cl concentrations (e.g. Cook and Robinson 2002), by (1) subtracting the background $^{36}$Cl/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 $^{36}$Cl/Cl ratio was assumed to be $(60 \pm 10) \times 10^{-15}$, based on the lower limit of the measured $^{36}$Cl/Cl range (see Figure 4). After subtracting this value from the $^{36}$Cl/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 m$^{-2}$ 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°04′N, 140°08′E), was estimated to be $2.3 \times 10^{12}$ 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 ± 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:

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

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

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

where $F_{\text{local}}$ 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 m$^{-2}$ 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°–50°N (Baumgartner and Reichel 1975). As mentioned above, $P_{\text{local}}$ 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 $(32 \pm 2$ atoms m$^{-2}$ 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 m$^{-2}$ s$^{-1}$)$^{-1}$. Because the study area occurs at a latitude of ~35°N, it would be reasonable to employ an $S_{\lambda}(\lambda)$ value of 0.047 ± 0.006 atoms m$^{-2}$ 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 $2.8 \pm 0.7$ were then converted to $^{36}$Cl concentrations using the following mass balance equation (Andrews et al. 1994):

$$ A = \frac{F \times 3.156 \times 10^7}{P \left( \frac{100}{100 - E} \right)} $$  \hspace{1cm} (3)

where $A$ is the $^{36}$Cl concentration, $F$ is the $^{36}$Cl fallout (atoms m$^{-2}$ 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
and 21% at 1000 m. These values do not account for transpiration, which would cause little or no isotopic fractionation. The potential evapotranspiration rate was calculated to be $46 \pm 3\%$ at Mishima (elevation, 20.5 m) in the southeastern part of Mt. Fuji, using the Penman equation (e.g. Brutsaert 2005) with an assumed albedo of $0.20 \pm 0.05$ for vegetated surfaces. Given the estimated recharge elevation of the springs (1100–2250 m), the actual evapotranspiration rate in this area is probably between $\sim20\%$ and $\sim46\%$.

In this regard, an evapotranspiration rate of 35%, as calculated by Tsuchi (2007) based on the water balance within the Mt. Fuji area, seems to be reasonable; accordingly, $E$ was assumed to be $35 \pm 5\%$. Figure 5a and 5b shows the resulting time series of above-background $^{36}\text{Cl}$ concentrations estimated for the Mt. Fuji area.

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

Figure 6 shows the distribution of estimated residence times for spring water and groundwater (see also Table 2). Although the distribution is somewhat complex, the residence times estimated for spring water at the eastern foot of Mt. Fuji (22–36 yr) are generally older than those estimated for other areas. Two groundwater samples from flowing wells near Mt. Ashitaka also yielded relatively old ages (23–37 yr). In contrast, spring waters at the southeastern foot of Mt. Fuji have younger residence times (0–31 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 $^3$H/$^3$He dating (~10 yr; Mahara et al. 1993). Figure 7 shows the available data on $^3$H concentrations for selected springs, along with the range of $^3$H concentrations in precipitation over Central Japan (see Table 2 for tritium-based ages interpreted from previous tritium data). $^3$H 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 effect 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
residence time. Although we assumed a uniform background level for the entire study area, the $^{36}$Cl/Cl ratio is clearly dependent on stable Cl transported from the sea. According to Hiyama et al. (1995), the concentration of Cl$^-$ in precipitation for the southwestern foot of Mt. Fuji varies from 0.59 mg/L at an elevation of 1000 m to 0.21 mg/L at 2390 m, indicating reduced Cl transport at higher elevation. In the case of a large difference in recharge elevation among the studied springs, the assumed background of $60 \times 10^{-15}$ may result in an underestimate of the actual background at higher elevations.

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

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

Conclusions and outlook

The residence times of spring water around Mt. Fuji were estimated based on bomb-produced $^{36}$Cl. Dye-3 fallout data were used to construct a reference time series of $^{36}$Cl/Cl values in the Mt. Fuji area. To focus solely on the bomb-derived $^{36}$Cl components, all the measured and reference $^{36}$Cl data were presented as above-background $^{36}$Cl concentrations after subtracting the background levels. The total
bomb-produced $^{36}$Cl fallout in the Mt. Fuji area was estimated to be $(6.8 \pm 1.7) \times 10^{12}$ atoms m$^{-2}$. The time series of above-background $^{36}$Cl concentration at the Dye-3 site was then scaled according to the ratio of the estimated total bomb-produced $^{36}$Cl fallout in the study area to that at the Dye-3 site $(2.43 \pm 0.03) \times 10^{12}$ atoms m$^{-2}$).

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

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

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**References**


IAEA/WMO. 2006. Global Network of Isotopes in Precipitation. The GNIP Database,


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 $\delta^D$ 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 3. Spatial distribution of $\delta^{18}$O values in spring water and groundwater in the Mt. Fuji area. The symbol shape indicates the date of sample collection. The study area was divided into five sub-regions based on the altitude effect on $\delta^{18}$O values (see the text for details).
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.
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. Fuji area (logarithmic scale). (b) Predicted time series of above-background $^{36}$Cl concentration (linear scale). (c) Above-background $^{36}$Cl concentrations of spring water and groundwater. Gray areas shown in (a) and (b) indicate the error estimated from the propagation of uncertainties arising in the assumed values of parameters. For an explanation of the geographic regions (e.g., East and Southeast), see Fig. 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 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.
Figure 8. Comparison of $^{36}$Cl-based and tritium-based ages.
Table 1. Chemical compositions of spring water and groundwater sampled from around Mt. Fuji

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<td>1 ± 12</td>
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