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An Estimate of Local Bomb-Produced ^{36}Cl Fallout Using the Depth Profile of Groundwater in the Tsukuba Upland, Central Japan

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Abstract:

The depth profile of ^{36}Cl/Cl ratio in groundwater was investigated in the Tsukuba Upland of central Japan. The obtained results clearly show the influence of bomb-produced ^{36}Cl; the highest ^{36}Cl/Cl ratio is about one order of magnitude greater than the natural background ratio (1×10^{-13}). The vertical distribution of ^{36}Cl is consistent with previous observations using ^{3}H and Darcy's law. From the profile, the total bomb-produced ^{36}Cl fallout in the upland is 2.3×10^{12} atoms/m^2 after the correction for surface runoff (c.f. 2.4×10^{12} atoms/m^2 at the Dye-3 site, Greenland) and a scaling factor of 0.96 was obtained (c.f. 2.5 based on the simplified latitudinal fallout distribution model). We then reconstructed the local fallout history of ^{36}Cl based on the Dye-3 data (scaled with a factor of 0.96 for the Tsukuba Upland) and the mean ^{36}Cl flux, produced in the atmosphere from cosmic rays and measured 30 atoms m^{-2} s^{-1} in the upland. The ratio of the maximum bomb-peak fallout to the average natural background flux of meteoric ^{36}Cl is consistent with that of measured data in Nepal. The result implies that the simplified latitudinal distribution model for ^{36}Cl deposition is not easily applicable for the prediction of the bomb-produced ^{36}Cl fallout pattern.

KEYWORDS Bomb-produced ^{36}Cl; fallout history; groundwater; residence time; accelerator mass spectrometry (AMS)

INTRODUCTION

Knowledge of groundwater residence time is crucial for the development of groundwater resources and their continuous use. Various environmental tracers in groundwater (i.e. radionuclides, stable isotopes, anthropogenic substances, etc.) are effective tools in the investigation of groundwater flow and residence time. For modern or young groundwaters (a residence time <50–60 years), ^{3}H (tritium) has been extensively applied because of its large pulse induced during the atmospheric nuclear testing period. However, the ^{3}H bomb pulse becomes increasingly difficult to detect due to its short half-life (12.32 yr); therefore, it is being replaced by such tracers as tritiogenic ^{3}He, CFCs, SF_6 and ^{85}Kr (e.g. Phillips and Castro, 2003).

The application of bomb-produced ^{36}Cl as a groundwater tracer was initially proposed by Bentley *et al.* (1982), which showed the ^{36}Cl depth profile in groundwater. Due to its very long half-life (3.01×10^5 yr) and conservative nature, bomb-produced ^{36}Cl acts as a stable environmental tracer in modern groundwater. With the exception of several studies of the unsaturated zone (e.g. Phillips *et al.*, 1988), few studies have focused on the tracer properties of bomb-produced ^{36}Cl (Balderer *et al.*, 2004; Corcho Alvarado *et al.*, 2005; Tosaki *et al.*, 2007). This may be partly attributed to the scarcity of information on the local fallout history of bomb-produced ^{36}Cl.

According to Green *et al.* (2004), the ^{36}Cl concentration profiles for ice cores from the Dye-3 site (Greenland) (Synal *et al.*, 1990), the Inilchek Glacier (Kyrgyzstan) and the Guiliya Ice Cap (China) have almost identical shapes. From this result, one can assume that the fallout pattern of bomb-produced ^{36}Cl is essentially uniform in the northern hemisphere. Accordingly, the local fallout history of bomb-produced ^{36}Cl can be estimated by scaling the Dye-3 fallout values (Synal *et al.*, 1990), which is currently the only available detailed data on the ^{36}Cl bomb pulse.

Therefore, the present study aims to reconstruct the local fallout history of bomb-produced ^{36}Cl. For this purpose, the ^{36}Cl depth profile of groundwater was investigated in an area of the Tsukuba Upland in central Japan. The total bomb-produced ^{36}Cl fallout was calculated and a tentative scaling factor was derived. The result will contribute to the future application of bomb-produced ^{36}Cl to estimate groundwater residence time.

STUDY SITE

Climate, topography and geology

The study site is located in the central part of the Tsukuba Upland (approximately 60 km northeast of Tokyo) in the Kanto region, central Japan (Figure 1). The climate of the area is humid temperate, with an annual precipitation of 1235.6 mm and an annual mean temperature of 13.5°C (average values for 1971–2000; data from the Aerological Observatory at Nagamine near the study site).

The Tsukuba Upland is a Pleistocene upland surrounded by the Sakura River and Lake Kasumigaura on the east, the Tone River on the south, and the Kokai River on the west. At the northeast of the upland, the

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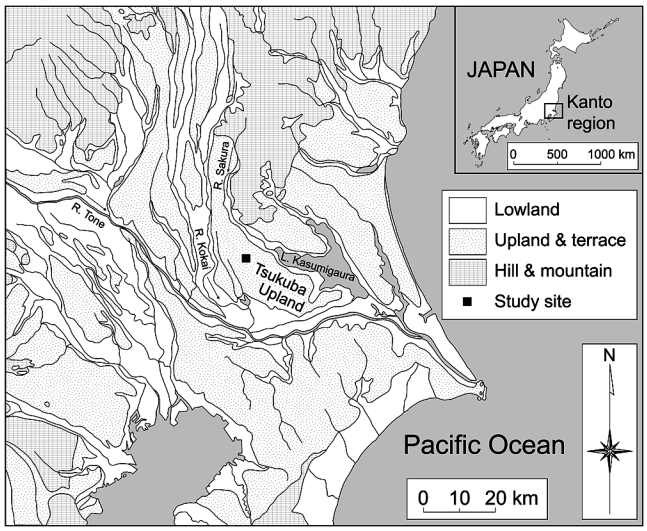

Figure 1. Location of the sampling site in the Tsukuba Upland (after Unozawa *et al.*, 1988).

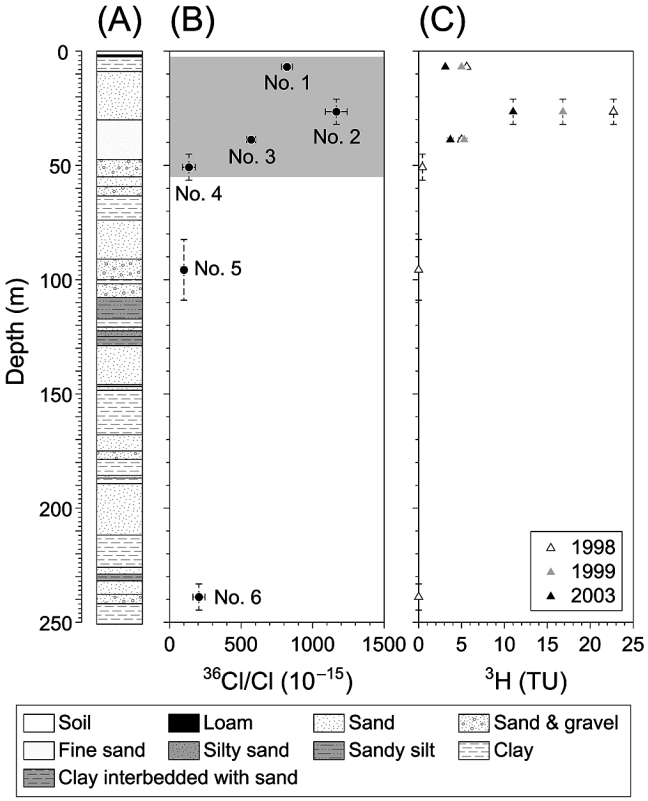

Figure 2. (A) Geologic column for the sampling site in the Tsukuba Upland (after Taguchi, 1981; Unozawa *et al.*, 1988). (B) Depth profile of ^{36}Cl/Cl ratio in the groundwater. Vertical error bars indicate widths of each screen. (C) Depth profiles of tritium concentration.

Tsukuba Mountains (including Mt. Tsukuba, 877 m a.s.l.) extend north and south. Although the upland surface is dissected by small rivers, the elevation typically falls in the range of 20–30 m for the most part (Unozawa *et al.*, 1988).

Geologic information on the upland and its surrounding areas has been described in detail by Unozawa *et al.* (1988). Basement rocks in the area consist of granitic and metamorphic rocks, forming the Tsukuba Mountains. They are overlain by the Kazusa Group sediments of Pliocene to middle Pleistocene age. The overlying Shimosa Group sediments of middle to late Pleistocene age mainly constitute the upland. It consists of six formations (i.e. the Jizodo Formation, the Yabu Formation, the Kamiizumi Formation, the Kamiiwahashi Formation, the Kioroshi Formation and the Joso Formation in ascending stratigraphic order), with the upland surface covered by the Kanto Loam Formation, which is derived from volcanic ash.

The geologic columns for the study site were previously given in Unozawa *et al.* (1988) for 0–62.5 m, and in Taguchi (1981) for 0–300 m (Figure 2A). Underlying the surface soil are a loam layer (1.5–2.5 m; the Kanto loam), a clay layer (2.5–9 m; the Joso clay, which is the upper most part of the Joso Formation), a sand layer (9–30 m), a fine sand layer (30–47.5 m), and a sand & gravel layer (47.5–55 m) in descending order.

Hydrogeology

The water table is generally observed within the Kanto loam or the Joso clay. The lower part of the sand layer (ca. 20–30 m) and the sand & gravel layer (ca. 45–55 m) act as shallow confined aquifers; the latter belongs to dominant aquifers in the area (Unozawa *et al.*, 1988). Several sand or gravel aquifers, respectively confined by clay layers, are additionally observed at greater depth down to ~300 m (ca. 80–110 m, 130–145 m and 230–245 m) (Taguchi, 1981).

According to Yasuhara *et al.* (1991), the Joso clay exhibits remarkably low saturated hydraulic conductivity compared to the overlying Kanto loam (by 2–4 orders of magnitude) and the upper part of the underlying sands (by 3–5 orders of magnitude). The Joso clay, therefore, may have a great influence on the downward flow of groundwater.

Conversely, previous studies have revealed that the hydraulic head of the groundwater decreases with depth (Kayane and Li, 1983; Shimada *et al.*, 1990), to ~55 m (Yasuhara *et al.*, 1990). This observation suggests that the upland surface is essentially acting as a recharge area. Therefore, vertical groundwater movement especially in the central part of the upland can be expected at least for the upper 55 m. Since small rivers dissecting the upland surface are gaining streams for the most part (Unozawa *et al.*, 1988), the shallow aquifers can be assumed to be recharged only from the surface.

METHODS

Groundwater sampling

The observation wells of the Geological Survey of Japan (GSJ), the National Institute of Advanced Industrial Science and Technology (AIST) were used for depth-profile sampling of groundwater. The wells are located on the premises of the GSJ in the central part of the Tsukuba Upland (Figure 1). The six wells used in this study include three deep wells for continuous groundwater level monitoring (Taguchi, 1981).

Groundwater samples for ^{36}Cl analysis were obtained using a bailer sampler in February 2004. Additionally, samples for ^{3}H analysis were collected in 1998, 1999 and December 2003. Each sample mostly corresponds to the upper four layers below the Kanto loam (i.e. the Joso

clay, the sands, the fine sands, and the sands & gravels) and the two deep aquifers (Figure 2). Since the two deeper wells (Nos. 5 and 6) have multiple screens, the corresponding samples were collected around the top screens. Due to the spatial variability of the layer's thickness, well No. 1 is actually screened in the uppermost part of the sands (just beneath the Joso clay). However, when the downward groundwater flow is dominant, it may be reasonable to assume that the sample taken from the well corresponds to the Joso clay.

Analyses of samples

The analyses for chloride (Cl^-) concentration, $^{36}Cl/Cl$ ratio and ^3H concentration were carried out in the laboratory. The samples for Cl^- and ^{36}Cl analyses were filtered through a 0.20 μm membrane filter (DISMIC-25cs, Advantec). The Cl^- concentrations of aliquots were measured by ion chromatography analysis (QIC Analyzer, Dionex). The ^3H concentrations of electrolytically enriched samples were determined by liquid scintillation counting (Tri-Carb 3100TR or 2250CA, Packard).

Water samples containing more than 1 mg of Cl were prepared for ^{36}Cl measurement by AMS (Accelerator Mass Spectrometry). The sample preparation scheme adopted in this study is essentially the same as that described in a previous paper (Tosaki *et al.*, 2007). Each sample was heated on a hot plate for evaporative concentration to obtain 10–20 mL volume. The sample was then acidified with 1 mL of 13 M HNO_3, and AgCl was directly precipitated by adding excess $AgNO_3$. The AgCl precipitate was dissolved once in 3 M NH_4OH, and saturated $Ba(NO_3)_2$ solution was added to the solution. This was allowed to stand overnight and sulfate (SO_4^{2-}) was precipitated as $BaSO_4$. The sample was then filtered through a 0.20 μm membrane filter, and was purified by repeating re-precipitation of AgCl with HNO_3 and re-dissolution in NH_4OH. After being washed with 0.01 M HNO_3 three times and with 99.5% C_2H_5OH twice, the AgCl precipitate was dried in the oven at 130°C for 3 hours.

The $^{36}Cl/Cl$ ratios were measured by the Tsukuba AMS system at the Tandem Accelerator Complex, University of Tsukuba (Nagashima *et al.*, 2000; Sasa *et al.*, 2007). The ^{36}S interference was corrected by using a blank sample prepared from NaCl reagent; typical correction rate was 10% for samples with a $^{36}Cl/Cl$ ratio of ~1×10^{-12}. Relative ratio of $^{36}Cl/^{35}Cl^-$ (counts/μC) for a sample is then normalized to that for a diluted NIST ^{36}Cl standard (with a $^{36}Cl/Cl$ ratio of 4.47×10^{-11}). The obtained $^{36}Cl/Cl$ ratio of the sample includes statistical error derived from uncertainties (1σ) of the sample, the standard and the blank. Details of the measurement and the $^{36}Cl/Cl$ calculations have been given in Nagashima *et al.* (2000, 2004). The overall precision of the system is better than 5%, and the background level of $^{36}Cl/Cl$ measurement is below 2×10^{-14}.

RESULTS AND DISCUSSION

Depth profile of ^{36}Cl in groundwater

Table I summarizes the measured ^{36}Cl and ^3H data. Figure 2B illustrates the depth profile of $^{36}Cl/Cl$ ratio in the groundwater. The obtained results clearly show the influence of bomb-produced ^{36}Cl in the upper ~50 m; the highest $^{36}Cl/Cl$ ratio is about one order of magnitude greater than the natural background level, mainly due to cosmogenic production in the atmosphere. This vertical distribution agrees with the depth profiles of ^3H concentrations (Figure 2C). It is also consistent with the previous observation in the central part of the upland by Kayane and Li (1983), who reported no detectable ^3H in the groundwater at depths greater than 36 m in 1982. In contrast, Yasuhara *et al.* (1990) calculated the downward groundwater flux through the Joso clay as 27 mm/yr based on Darcy's law; it leads to a mean residence time of 30–50 yr in the upper part of the underlying sand layer. The highest $^{36}Cl/Cl$ ratio observed in the sand layer is also consistent with their estimation.

As can be seen from Figure 2C, ^3H concentrations have gradually decreased from 1999 to 2003 due to both radioactive decay and groundwater flow, while $^{36}Cl/Cl$ ratio showed distinct difference from the natural background level (by one order of magnitude) in 2004 (Figure 2B). This suggests the potential usefulness of bomb-produced ^{36}Cl as a tracer in modern groundwater as described in previous studies (Bentley *et al.*, 1982; Balderer *et al.*, 2004; Tosaki *et al.*, 2007).

It can be surmised that the deeper the sample, the greater the uncertainty about the corresponding recharge area. Thus, the deepest sample (No. 6; 233.2–244.6 m) is excluded from further discussion. The natural background $^{36}Cl/Cl$ ratio is then estimated as 1×10^{-13} from the result of sample No. 5 (Figure 2B). This is supported by the $^{36}Cl/Cl$ ratio for a sample from a shallow well screened in the Kanto loam (0.7–2.0 m): $(1.19 \pm 0.48) \times 10^{-13}$ in October 2003; the well is located in the Terrestrial Environment Research Center, University of Tsukuba, which is about 6 km northwest from the sampling site at the GSJ.

Estimation of bomb-produced ^{36}Cl fallout

The total bomb-produced ^{36}Cl fallout at the Tsukuba Upland can be estimated by integrating bomb-produced ^{36}Cl observed in the profile (grayed area in Figure 2B). The following equations were used to calculate the total bomb-produced ^{36}Cl fallout:

Table I. Measured Cl^- concentrations, $^{36}Cl/Cl$ ratios, ^{36}Cl concentrations and ^3H concentrations in the groundwater

Well No.	Screen depth (m)	Cl^- (mg/L)	$^{36}Cl/Cl$ (10^{-15})	^{36}Cl (10^6 atoms/L)	^3H (TU)		
					1998	1999	2003
1	6.75–7.00	6.4	820 ± 38	89.2 ± 4.1	5.6	5.0	3.1
2	21.0–32.0	5.7	1166 ± 76	113.8 ± 7.4	22.7	16.8	11.0
3	38.0–39.5	15.9	570 ± 31	154.2 ± 8.5	5.0	5.3	3.7
4	45.1–56.5	4.0	135 ± 45	9.2 ± 3.1	0.4	–	–
5	82.4–109.0[a]	3.5	100 ± 24	5.9 ± 1.4	< 0.4	–	–
6	233.2–244.6[b]	4.3	205 ± 42	14.9 ± 3.1	< 0.4	–	–

1 TU (tritium unit) means one ^3H atom in 10^{18} ^1H atoms.
[a] Also screened in a deeper aquifer (136.5–140.3 m).
[b] Also screened in deeper layers (255.6–259.4 m and 264.9–272.5 m).

Table II. Ratios of the maximum ^{36}Cl flux to the average natural background flux for the ^{36}Cl bomb pulse at different locations

Location	Method	Max/BG ratio	Reference
Tsukuba, Japan	Estimated	396	This study
Upper Fremont, WY, USA	Measured	21	Green *et al.* (2000)
Nangpai Gosum, Nepal	Measured	416	Green *et al.* (2000)
Dye-3, Greenland	Measured	618	Synal *et al.* (1990, 1994)

$$D = \sum (A_{\mathrm{bomb},\,i} \times h_i \times n_i \times 10^3), \quad (1)$$

$$A_{\mathrm{bomb},\,i} = (R_i - R_{\mathrm{BG}}) \times \frac{[\mathrm{Cl}]_i \times 10^{-3} \times 6.022 \times 10^{23}}{35.45}, \quad (2)$$

where D is the total bomb-produced ^{36}Cl fallout (atoms/m^2), $A_{\mathrm{bomb},\,i}$ is the concentration of bomb-produced ^{36}Cl (atoms/L), h_i is the thickness of the layer (m), n_i is the porosity (%), R_i is the measured ^{36}Cl/Cl ratio, R_{BG} is the natural background ^{36}Cl/Cl ratio, and $[\mathrm{Cl}]_i$ is the Cl^- concentration (mg/L).

In the calculation, we assumed that obtained values are representative for the total thickness of the corresponding four layers below the Kanto loam, and that the porosity is identical to the effective porosity (e.g. Todd and Mays, 2005). The porosity used for the Joso clay was 62%, which is the arithmetic mean of the 19 measured values over the Tsukuba Upland by Yasuhara *et al.* (1991). For other layers, representative values were adopted from Morris and Johnson (1967): 39% for sand (as medium sand), 43% for fine sand, and 34% for sand & gravel (as fine gravel). The natural background ^{36}Cl/Cl ratio was set to 1×10^{-13}, as mentioned in the previous section.

Consequently, Equation (1) gives the total bomb-produced ^{36}Cl fallout of 2.1×10^{12} atoms/m^2. Because the ^{36}Cl deposited onto the ground may be partly lost with surface waters, 2.1×10^{12} atoms/m^2 would be the minimal estimate. General surface runoff rate in Pleistocene upland-alluvial lowland region is ~20% (e.g. Kotoda, 1968; Kondoh, 1985), whereas surface runoff rate measured in an experimental field at the central part of the Tsukuba Upland is ~5% (Itadera and Shimada, 1992). Here we assumed 10% of ^{36}Cl is lost by surface runoff. After incorporation of this loss, we obtained a total bomb-produced ^{36}Cl fallout of 2.3×10^{12} atoms/m^2. Dividing this value by 2.4×10^{12} atoms/m^2 (fallout at the Dye-3 site; Synal *et al.*, 1990), we obtained a scaling factor of 0.96 for the Tsukuba Upland (c.f. 2.5 based on the simplified latitudinal fallout distribution model by Lal and Peters, 1967). It should be noted that the screen length of well No. 2 is wide (11 m) and the thickness of the corresponding sand layer is 21 m (Figure 2A). This indicates that the highest ^{36}Cl/Cl ratio in the profile is fairly representative of the layer. Therefore, the calculation described in this section will not lead to erratic results.

Reconstruction of the local fallout history of ^{36}Cl

The ^{36}Cl flux for 1560–1920 AD at the Dye-3 site is 20 ± 6 atoms m^{-2} s^{-1} (Synal *et al.*, 1994), which is taken to be the natural background flux of meteoric ^{36}Cl. Subtracting this natural background flux from annual values for 1945–1985 (Synal *et al.*, 1990), we obtained yearly values of bomb-produced ^{36}Cl fallout at the Dye-3 site. These values were then scaled using the factor of 0.96 for the Tsukuba Upland.

From the ^{36}Cl measurements of bulk precipitation samples collected monthly at the Natural Sciences building, University of Tsukuba, the average ^{36}Cl flux is ~30 atoms m^{-2} s^{-1} (from April 2004 to March 2006; unpublished data). By adding this natural background flux, we obtained the local fallout history of ^{36}Cl (including both bomb-produced and cosmogenic components) for the Tsukuba Upland.

In order to allow comparison of the reconstructed fallout history with several measured data for ice cores at different locations, the ratios of the bomb-peak flux to the natural background flux of meteoric ^{36}Cl were calculated. It is worth noting that the value determined with the scaling factor of 0.96 reasonably agrees with data from Nepal (Table II). This may imply that the latitudinal fallout distribution (Lal and Peters, 1967) is not easily applicable for the prediction of the bomb-produced ^{36}Cl fallout pattern. The obtained results also suggest that the local fallout history of ^{36}Cl can be reconstructed from groundwater in regions of near vertical recharge. Such studies, in turn, will enable the application of bomb-produced ^{36}Cl to date modern groundwaters.

CONCLUSIONS AND OUTLOOK

The present study investigated the ^{36}Cl depth profile in the groundwater of the Tsukuba Upland, central Japan. The total bomb-produced ^{36}Cl fallout in the upland was derived as 2.3×10^{12} atoms/m^2 using the profile. The local fallout history of ^{36}Cl was estimated using the Dye-3 data with a tentative scaling factor of 0.96 (c.f. 2.5 based on the latitudinal fallout distribution model). The ratio of the maximum bomb-peak fallout to the average natural background flux of meteoric ^{36}Cl is consistent with that of measured data in Nepal. The result implies that the simplified latitudinal distribution model for ^{36}Cl deposition is not easily applicable to predict the fallout pattern of bomb-produced ^{36}Cl.

Prior to the application of bomb-produced ^{36}Cl to estimate groundwater residence time, reconstruction of the local fallout history is necessary. Further study, that is, more data points in the depth profile, is needed to obtain a reliable scaling factor. However, the results of this study will contribute to the future application of bomb-produced ^{36}Cl to date modern groundwaters.

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