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

Tosaki Yuki, Tase Norio, Yasuhara Masaya, Nagashima Yasuo, Sasa Kimikazu, Takahashi Tsutomu


(C) 2008 Japan Society of Hydrology and Water Resources

URL http://hdl.handle.net/2241/00121968
doi: 10.3178/hrl.2.9

| 著者 | 
| --- | --- |
| 西崎義幸 | 
| 正 林 亮 | 
| 長谷原正雅 | 
| 長谷 哲雄 | 
| 水権 | 

| 権利 | 
| --- | --- |
| 前述の著作権に該当する全ての権利を保持します | 

doi: 10.3178/hrl.2.9
An Estimate of Local Bomb-Produced $^{36}$Cl Fallout Using the Depth Profile of Groundwater in the Tsukuba Upland, Central Japan

Yuki Tosaki, Norio Tase, Masaya Yasuhara, Yasuo Nagashima, Kimikazu Sasa and Tsutomu Takahashi

1Geoenvironmental Sciences, Graduate School of Life and Environmental Sciences, University of Tsukuba
2AMS Group, Tandem Accelerator Complex, University of Tsukuba
3Sustainable Environmental Studies, Graduate School of Life and Environmental Sciences, University of Tsukuba
4Research Core for Deep Geological Environments, Geological Survey of Japan, AIST

Abstract:

The depth profile of $^{36}$Cl/$^{36}$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/$^{36}$Cl ratio is about one order of magnitude greater than the natural background (1 x 10$^{-3}$). 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 x 10$^{12}$ atoms/m$^2$ after the correction for surface runoff (c.f. 2.4 x 10$^{10}$ 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 groundwater (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 $^4$He, CFCs, SF$_6$, and $^{41}$Ar (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 x 10$^7$ 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) and the Inlichek Glacier (Kyrgyzstan) and the Guliya 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 Sakurua 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
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 Kamiwahashi 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 uppermost 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 $^3$H 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 concentration of aliquots were measured by ion chromatography analysis (QIC Analyzer, Dionex). The $^3$H concentrations of electrolytically enriched samples were determined by liquid scintillation counting (Tri-Carb 3100TR or 2250CA, Packard). Analyses of samples

The following equations were used to calculate 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 $^3$H concentrations in the groundwater

<table>
<thead>
<tr>
<th>Well No.</th>
<th>Screen depth (m)</th>
<th>Cl$^-$ (mg/L)</th>
<th>$^{36}$Cl/Cl (×10$^{-5}$)</th>
<th>$^{36}$Cl (atoms/L)</th>
<th>$^3$H (TU)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>1998</td>
</tr>
<tr>
<td>1</td>
<td>6.75–7.00</td>
<td>6.4</td>
<td>820 ± 38</td>
<td>89.2 ± 4.1</td>
<td>5.6</td>
</tr>
<tr>
<td>2</td>
<td>21.0–32.0</td>
<td>5.7</td>
<td>1166 ± 76</td>
<td>1318 ± 7.4</td>
<td>22.7</td>
</tr>
<tr>
<td>3</td>
<td>38.0–39.5</td>
<td>15.9</td>
<td>570 ± 31</td>
<td>1542 ± 8.5</td>
<td>5.0</td>
</tr>
<tr>
<td>4</td>
<td>45.1–56.5</td>
<td>4.0</td>
<td>135 ± 45</td>
<td>9.2 ± 3.1</td>
<td>0.4</td>
</tr>
<tr>
<td>5</td>
<td>82.4–109.0$^a$</td>
<td>3.5</td>
<td>100 ± 24</td>
<td>5.9 ± 1.4</td>
<td>&lt;0.4</td>
</tr>
<tr>
<td>6</td>
<td>233.2–244.6$^b$</td>
<td>4.3</td>
<td>205 ± 42</td>
<td>14.9 ± 3.1</td>
<td>&lt;0.4</td>
</tr>
</tbody>
</table>

$^a$ TU (tritium unit) means one $^3$H atom in 10$^{18}$ $^3$H atoms.

$^b$ Also screened in deeper layers (255.6–259.4 m and 264.9–272.5 m).

**RESULTS AND DISCUSSION**

**Depth profile of $^{36}$Cl in groundwater**

Table I summarizes the measured $^{36}$Cl and $^3$H 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 $^3$H 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 $^3$H 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, $^3$H 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$^{-7}$ 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 ± 0.48) × 10$^{-7}$ 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 GSI.

**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>
<thead>
<tr>
<th>Well No.</th>
<th>Screen depth (m)</th>
<th>Cl$^-$ (mg/L)</th>
<th>$^{36}$Cl/Cl (×10$^{-5}$)</th>
<th>$^{36}$Cl (atoms/L)</th>
<th>$^3$H (TU)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>6.75–7.00</td>
<td>6.4</td>
<td>820 ± 38</td>
<td>89.2 ± 4.1</td>
<td>5.6</td>
</tr>
<tr>
<td>2</td>
<td>21.0–32.0</td>
<td>5.7</td>
<td>1166 ± 76</td>
<td>1318 ± 7.4</td>
<td>22.7</td>
</tr>
<tr>
<td>3</td>
<td>38.0–39.5</td>
<td>15.9</td>
<td>570 ± 31</td>
<td>1542 ± 8.5</td>
<td>5.0</td>
</tr>
<tr>
<td>4</td>
<td>45.1–56.5</td>
<td>4.0</td>
<td>135 ± 45</td>
<td>9.2 ± 3.1</td>
<td>0.4</td>
</tr>
<tr>
<td>5</td>
<td>82.4–109.0$^a$</td>
<td>3.5</td>
<td>100 ± 24</td>
<td>5.9 ± 1.4</td>
<td>&lt;0.4</td>
</tr>
<tr>
<td>6</td>
<td>233.2–244.6$^b$</td>
<td>4.3</td>
<td>205 ± 42</td>
<td>14.9 ± 3.1</td>
<td>&lt;0.4</td>
</tr>
</tbody>
</table>

1 TU (tritium unit) means one $^3$H atom in 10$^{18}$ $^3$H 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

<table>
<thead>
<tr>
<th>Location</th>
<th>Method</th>
<th>Max/BG ratio</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>Tsukuba, Japan</td>
<td>Estimated</td>
<td>396</td>
<td>This study</td>
</tr>
<tr>
<td>Upper Fremont, WY, USA</td>
<td>Measured</td>
<td>21</td>
<td>Green et al. (2000)</td>
</tr>
<tr>
<td>Nagpai Gosum, Nepal</td>
<td>Measured</td>
<td>416</td>
<td>Green et al. (2000)</td>
</tr>
<tr>
<td>Dye-3, Greenland</td>
<td>Measured</td>
<td>618</td>
<td>Synal et al. (1990, 1994)</td>
</tr>
</tbody>
</table>

$$D = \sum (A_{\text{bomb},i} \times h \times n \times 10^3),$$  

(1)

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

(2)

where $D$ is the total bomb-produced $^{36}$Cl fallout (atoms/m$^2$), $A_{\text{bomb},i}$ is the concentration of bomb-produced $^{36}$Cl (atoms/L), $h$ is the thickness of the layer (m), $n$ is the porosity (%), $R_i$ is the $^{36}$Cl/$^{36}$Cl ratio in the profile, $R_{\text{BG}}$ is the natural background $^{36}$Cl/$^{36}$Cl ratio, and $[\text{Cl}]$ 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/$^{36}$Cl ratio was set to $1 \times 10^{-3}$, as mentioned in the previous section.

Consequently, Equation (1) gives the total bomb-produced $^{36}$Cl fallout of 2.1 $\times 10^{12}$ atoms/m$^2$. Because the $^{36}$Cl deposited onto the ground may be partly lost with surface waters, 2.1 $\times 10^{12}$ 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 $\times 10^{12}$ atoms/m$^2$. Dividing this value by 2.4 $\times 10^{12}$ 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). 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/$^{36}$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 $20 \pm 6$ 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 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 $2.3 \times 10^{12}$ 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.

ACKNOWLEDGEMENTS

The authors would like to thank the members of the AMS Group, University of Tsukuba for their contributions during $^{36}$Cl measurements. The authors would also like to thank the staff at the Tandem Accelerator Complex, University of Tsukuba for their technical support. Y. T. is grateful to Dr. Makoto Takahashi (GSJ, AIST) and Mr. Seongwon Lee (University of Tsukuba) for their assistance in the groundwater sampling. This work was partly supported by Grants-in-Aid for Scientific Research from the Japan Society for the Promotion of Science.
REFERENCES


