Vertical distribution of $^{10}$Be, $^{26}$Al, and $^{36}$Cl in the surface soil layer of weathered granite at Abukuma, Japan

Yasunori Mahara*, Housho Hohjoa, Takumi Kubota, Tomoko Ohta, Yukihiro Mizuochib, Toshiharu Tashirob, Shun Sekimotoa, Koichi Takamiya, Seiichi Shibata and Kazuhiro Tanakac
(a) Research Reactor Institute, Kyoto University, 2-Chome, Ashashiro-nishi, Kumatori Osaka 590-0494, Japan
(b) Sumiko Consultants Co. Ltd, Ikenohata, Taito Tokyo 110-0008, Japan
(c) Faculty of Science, Yamaguchi University, Yamaguchi City, Yamaguchi 753-8512, Japan
(d) The Japan Atomic Power Company, Kanda, Chiyoda Tokyo, 101-0053, Japan
*Corresponding author, Tel.: +81-72451-2462; Fax: +81-72451-2636, E-mail address: mahara@HL.rrri.kyoto-u.jp

Abstract
The northern Abukuma region of Japan is thickly covered by weathered Mesozoic granite. We investigated the vertical distribution of radionuclides ($^{10}$Be, $^{26}$Al, and $^{36}$Cl) produced by cosmic irradiation of weathered granitic soil collected from this region at a sampling site located 496 m above sea level, at 37°30′52.6″N, 140°40′13.2″E. We mechanically and chemically separated quartz particles from the weathered granitic soil, and purified them. We also analyzed the vertical distribution of $^{26}$Al, $^{10}$Be, and $^{36}$Cl and estimated a long term averaged erosion rates of 0.019 ± 0.001 mm/y from $^{26}$Al, 0.017 ± 0.001 mm/y from $^{10}$Be, and approximately 0.01 ± 0.001 mm/y from $^{36}$Cl. These cosmogenic erosion rates are comparable to the rate estimated from a geomorphological evolution model and with the observed sedimentary deposition rate at Abukuma.

Keywords: AMS measurement, cosmogenic radionuclides, erosion rate, weathered granite

PACS: 92.40.Kf
1. Introduction

Uplift or erosion rates are commonly deduced from the vertical distance between an investigated surface and a dated reference layer such as a coastal terrace, but peneplains are a challenge for this technique if reference layers cannot be found. Therefore, a method independent of geomorphological history must be used to estimate the erosion rate on the peneplain. Such a method should be based on absolute measurements, such as the vertical distribution of cosmogenic radionuclides. Because cosmic irradiation is a phenomenon that affects the entire surface of the earth, these data can be interpreted without the need for comparison with a local standard.

We investigated the vertical distribution of cosmogenic radionuclides ($^{10}$Be, $^{26}$Al, and $^{36}$Cl) produced by cosmic irradiation of weathered granitic soil collected from the Abukuma region, which lies on an inland peneplain typical of those found in Japan. We estimated the erosion rates from cosmogenic radionuclides ($^{10}$Be, $^{26}$Al and $^{36}$Cl) profiles and compared the results with those determined by other methods.

2. Geological Setting

The weathered granitic soil we used in this study was sampled on the northern Abukuma peneplain in Japan at an altitude of 496 m above sea level at 37°30′52.6″N and 140°40′13.2″E (Fig. 1). We did not find any nearby marine sedimentary layers or coastal terrace formations, such as the well-known Shimosueyoshi surface, that could be dated and used as a reference surface for calculation of erosion rates.

The northern Abukuma area is thickly covered by several types of heavily weathered Mesozoic granite. The original granite formed 83–125 million years ago [1]. The area east of the Abukuma River has been uplifted relative to the western area. The Abukuma River and its tributaries, located at the western edge of the uplifting area, have created river terraces by eroding the
uplifting granitic plateaus. The rate of uplift has changed markedly at various times during the formation of these terraces. The flat river terraces are considered to have formed when the rate of uplift was very low [1]. There are five broad, flat, step-like terraces formed along the rivers surrounding the study area. These five terraces have been classified into three groups on the basis of their altitudes. The high surface group (750–1000 m) were formed before the early Miocene, the middle surface group (550–730 m) were formed during the late Miocene, and the low surface group (300–550 m) were formed during the Pliocene and early Pleistocene [2].

3. Materials and Methods

We produced $^{10}$Be and $^{26}$Al targets for acceleration mass spectrometry (AMS) after chemically extracting samples from quartz and purifying them using a conventional technique [3]. For $^{36}$Cl determination, AgCl was isolated from approximately 100 g of quartz separated from the weathered granitic soil, following a recognized procedure [4]. The quartz contained chloride concentrations of $8.16 \pm 0.55$ to $13.46 \pm 10.9 \mu g/g$ (SiO$_2$), as determined by the thermal neutron-activation method after etching three times with the mixture of 300 mL of 1% HF and 1% HNO$_3$ at 95 °C for 9 h. To avoid contamination by chlorine from HCl, we omitted the HCl resolution step in this pretreatment process; therefore, feldspar or other minerals with high chlorine content may have been left in the sample. Measurement of $^{10}$Be and $^{26}$Al by AMS was conducted at the Prime Lab at Purdue University (West Lafayette, IN, USA). $^{36}$Cl was measured at the Swiss Federal Institute of Technology (Zurich, Switzerland).

4. Results and Discussion

We estimated the number of cosmogenic $^{26}$Al, $^{10}$Be, and $^{36}$Cl atoms in SiO$_2$ particles collected from heavily weathered granite from Abukuma in Japan. Table 1 shows the ratios of $^{26}$Al/$^{27}$Al, $^{10}$Be/$^{9}$Be, and $^{36}$Cl/Cl as determined from AMS analysis and the correlation between estimated cosmogenic atoms (n/g (SiO$_2$)) in quartz and sampling depth. The number of $^{26}$Al atoms per gram
of quartz in the surface sample (0 cm depth) was smaller than that in samples collected at greater depths (Table 1). In contrast, the number of cosmogenic $^{36}$Cl atoms in the surface sample was extremely high compared to that estimated from the exponential function. $^{10}$Be exhibited no deviation from the exponential function in the surface sample. The observed deviations of $^{26}$Al and $^{36}$Cl in the surface sample were attributed to the incomplete separation of quartz from the granite parent rock, because the sample was collected from a hard unweathered granite outcrop near the sampling site in order to acquire enough sample for AMS analysis. Unfortunately, trace amounts of residual feldspar could not be completely removed in the separation process for the surface sample, so the measured $^{26}$Al/$^{27}$Al ratio for this layer was very low (2.1 ± 1.0 ×10$^{-15}$), which is close to the AMS detection limit, due to dilution by excess stable $^{27}$Al. On the other hand, the $^{36}$Cl/Cl ratio was increased by both additions of spallogenic $^{36}$Cl from stable $^{39}$K and $^{40}$Ca, and of activated $^{36}$Cl from $^{35}$Cl in the residual feldspar. $^{10}$Be in the surface sample was not affected by contamination from the residual feldspar, because stable beryllium has a very low natural abundance of a few parts per million in quartz and feldspar [5]. For the remaining discussion, the $^{26}$Al and $^{36}$Cl data from the surface sample are disregarded.

We separately estimated three erosion rates based on the three different profiles of $^{10}$Be, $^{26}$Al, and $^{36}$Cl. We analyzed data using a simple erosion model that assumed constant cosmic ray irradiation with a long term averaged erosion rate, because the maximum soil sampling depth was approximately 70 cm (i.e., equivalent to 121 g/cm$^2$ with an average bulk density of 1.68 g/cm$^3$). According to Lal [6] and Nishiizumi et al. [7,8], the model can be expressed as follows:

$$N(x,t) = e^{-\mu \cdot x} \cdot \frac{P(0)}{\lambda + \mu \cdot \epsilon} \quad (1)$$

where $N(x,t)$ is the number of cosmogenic radionuclides in quartz at irradiation time $t$ (y) and soil depth $x$ (cm), $P(0)$ is the production rate (n/g (SiO$_2$)-y$^{-1}$) of cosmogenic radionuclides in quartz at the ground surface per year, $\lambda$ is the radionuclide decay constant (4.33 × 10$^{-7}$ y$^{-1}$ for $^{10}$Be, 9.62 × 10$^{-7}$ y$^{-1}$ for $^{26}$Al, and 2.30 × 10$^{-6}$ y$^{-1}$ for $^{36}$Cl), $\mu$ is the absorption coefficient (cm$^{-1}$) for interacting
nuclear particles (i.e., neutrons and muons) in bulk soil, and $\varepsilon$ is the erosion rate (cm/y). Furthermore, eq. (1) assumes that cosmic rays have adequately irradiated over a much longer time than the effective exposure age $T_{\text{eff}} = 1/(\lambda + \mu \cdot \varepsilon)$, as proposed by Lal [6].

We analyzed profiles of $^{10}$Be, $^{26}$Al, and $^{36}$Cl in quartz at the soil depths shown in Figs. 3 and 4 by using eq. (1). We estimated the apparent absorption coefficients for the interacting nuclear particles at the site by plotting the data on a semi-logarithmic scale. The resulting coefficients were $0.012 \pm 6.4 \times 10^{-5}$ cm$^{-1}$ for $^{10}$Be, $0.011 \pm 3.4 \times 10^{-5}$ cm$^{-1}$ for $^{26}$Al, and $0.007 \pm 5.6 \times 10^{-5}$ cm$^{-1}$ for $^{36}$Cl. Although these values are comparable to or smaller than the value of 0.02 cm$^{-1}$ that is typically used for common rocks [6], the estimated values are acceptable for the heavily weathered granite layer because weathering greatly reduces the bulk density from 2.5 g/cm$^3$ for fresh granite to 1.68 g/cm$^3$ for weathered granite.

We corrected the observed radionuclide production rates for latitude and altitude effects by using Stone’s method [9], which is based on air pressure, spallation, and muon-capture scaling factors. The corrected production rates were $7.1 \pm 0.4$ n/g (SiO$_2$)·y$^{-1}$ for $^{10}$Be and $44 \pm 2.3$ n/g (SiO$_2$)·y$^{-1}$ for $^{26}$Al, using production rates reported by Heisinger and Nolte [10]. In contrast, the corrected production rate for $^{36}$Cl was estimated to be $1.98 \pm 0.18$ n/g (SiO$_2$)·y$^{-1}$ from studies conducted by Stone et al. [11] and Evans et al. [12]. Presumably, the estimated in situ production of $^{36}$Cl may be overestimated, because we assumed that the weathered granite particles contained a maximum of 0.5 wt % residual stable K and Ca on the basis of the highest values (0.3% for Ca and 0.5% for K) obtained from ICP-MS measurements after perfectly resolving sample particles in HF. Furthermore, we estimated that the production rate of $^{36}$Cl by thermal neutron activation was $0.2$–$0.5$ n/g (SiO$_2$)·y$^{-1}$ based on chlorine concentration being approximately $10 \mu$g/g (SiO$_2$). Here, we assumed that the density of thermal neutron on the ground surface was approximately in the range $1$–$2 \times 10^{-3}$ n·cm$^{-2}$ s$^{-1}$, which we deduced from our unpublished data measured in the open air with the He-3 neutron detector at the Research Reactor Institute of Kyoto University ($34^\circ 23'N, 135^\circ 20'E$ and altitude 70 m).
We estimated the erosion rates to be $0.017 \pm 0.001 \text{ mm/y}$ from $^{10}$Be measurements, $0.019 \pm 0.001 \text{ mm/y}$ from $^{26}$Al, and approximately $0.01 \pm 0.001 \text{ mm/y}$ from $^{36}$Cl (Figs. 2 and 3). On the basis of these results, we estimated $0.015 \pm 0.0017 \text{ mm/y}$ as the average cosmogenically derived erosion rate at the site. This erosion rate is comparable to the geomorphologically derived erosion rate of $0.02 – 0.04 \text{ mm/y}$ estimated from the topographic evolution model proposed by Kimura [2] and Koike [1]. In that model, a pyroclastic flow that has partially covered the peneplain was used as the reference surface to estimate the erosion rate. Furthermore, our cosmogenically derived rate is also within the range of the very roughly estimated erosion rate of $0 – 0.3 \text{ mm/y}$ deduced from the rate of sedimentation observed in water reservoirs in the Abukuma area [13].

For our estimation of $T_{\text{eff}}$, we assumed a constant rate of erosion and used $\epsilon = 0.0015 \text{ cm/y}$ and $\mu = 0.012 \text{ cm}^{-1}$ for interacting nuclear particles. The estimated exposure ages ranged from $4.9 \times 10^4$ to $5.4 \times 10^4 \text{ y}$. These estimates suggest that cosmic rays irradiated the site continuously for approximately $5 \times 10^4 \text{ y}$ and that the spallation reaction was overwhelmingly distinguished from other muon effects at depths of up to 70 cm (i.e., $121 \text{ g/cm}^2$) below the surface. However, since near-surface layers can be subjected to high levels of biogenic and hydrological activity, our estimated erosion rate may have been affected by disturbance of the shallow weathered layer by vegetation, worm burrows, rainwater infiltration, and surface runoff.

5. Conclusions

We investigated the vertical distribution of cosmogenic radionuclides $^{10}$Be, $^{26}$Al, and $^{36}$Cl, which are produced by various cosmic ray irradiation processes, at depths of up to 70 cm ($121 \text{ g/cm}^2$) in weathered granite soil from a site on the northern Abukuma peneplain. These radionuclides were distributed unevenly in the shallow weathered layer, with concentrations decreasing exponentially with increasing depth. Using the vertical distribution profiles of the three radionuclides, we estimated three erosion rates in the range of $0.01 – 0.02 \text{ mm/y}$. We concluded that the average erosion rate at the sampling site was $0.015 \pm 0.0017 \text{ mm/y}$. This result
is comparable to rates estimated by different methods. The effective exposure time was roughly estimated to be $5 \times 10^4$ y, assuming constant irradiation by cosmic rays and a constant rate of erosion.

Finally, cosmogenic radionuclide measurement can be useful for estimating rates of erosion in inland areas that lack a dated geological marker surface that can be used as a reference from which to measure uplift or erosion rates over geological time spans.

Acknowledgements

We thank Emeritus Prof. Elmore (Purdue University) for $^{10}$Be and $^{26}$Al analyses and Prof. Suter and Dr. Synal (Swiss Federal Institute of Technology) for $^{36}$Cl analyses. We thank Dr. Cresswell (CSIRO) for $^{36}$Cl sample preparation. We thank all of these individuals for their contribution to this study. We further thank two anonymous reviewers for their constructive comments on the manuscript.

References


**Figure Captions**

Fig. 1: Location of the weathered granitic soil sampling site in the northern Abukuma peneplain in Japan. The sample was collected at 37°30′52.6″N, 140°40′13.2″E, 496 m above sea level.

Fig. 2: Correlations between \(^{10}\)Be and \(^{26}\)Al measured at Purdue University and estimated erosion rates obtained from a simple erosion model based on cosmic irradiation effects.

Fig. 3: Correlation between \(^{26}\)Cl measured at Swiss Federal Institute of Technology and the estimated erosion rates obtained from a simple erosion model based on cosmic ray irradiation.
Sampling Site at Abukuma (37° 30' 52.6"N and 140° 40' 13.2"E)

Fig. 1
Be and Al Atoms/SiO$_2$ (g)

$^{10}$Be = $10^{(5.54 - 0.003 \times x)}$ ($r^2 = 0.91$)

$^{26}$Al = $10^{(6.30 - 0.0028 \times x)}$ ($r^2 = 0.97$)

(Prime Lab.)

Fig. 2
$^{36}\text{Cl} = 10^{(5.32 - 0.0018x)} \ (r^2 = 0.53)$

Fig. 3
Table 1. Ratios of $^{26}$Al/$^{27}$Al, $^{10}$Be/$^{9}$Be and $^{36}$Cl/Cl ($\times 10^{-13}$) and cosmogenic radionuclides atoms (n/g-SiO$_2$)($^{10}$Be, $^{26}$Al and $^{36}$Cl) measured in the quartziferous soil at Abukuma, Japan

<table>
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<tr>
<th>Depth (g/cm$^2$)</th>
<th>$^{26}$Al/27Al ± error</th>
<th>$^{26}$Al ± error</th>
<th>$^{10}$Be/9Be ± error</th>
<th>$^{10}$Be ± error</th>
<th>$^{36}$Cl/Cl ± error</th>
<th>$^{36}$Cl ± error</th>
<th>ETH (Zurich)</th>
</tr>
</thead>
<tbody>
<tr>
<td>0</td>
<td>0.21 ± 0.1</td>
<td>671037 ± 32897.9</td>
<td>2.24 ± 0.28</td>
<td>341917 ± 47317.1</td>
<td>4.82 ± 0.14</td>
<td>872000 ± 34008</td>
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<tr>
<td>14.2</td>
<td>11 ± 1</td>
<td>1790000 ± 163271.2</td>
<td>1.49 ± 0.06</td>
<td>311215 ± 17412.52</td>
<td>2.06 ± 0.21</td>
<td>173000 ± 17819</td>
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<tr>
<td>39.1</td>
<td>10.2 ± 0.9</td>
<td>1560000 ± 139040</td>
<td>2.19 ± 0.12</td>
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<td>2.07 ± 0.17</td>
<td>200000 ± 16400</td>
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<td>78.8</td>
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<td>1330000 ± 139073</td>
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<tr>
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<td>1.42 ± 0.17</td>
<td>113000 ± 13899</td>
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</table>

Prime Lab. (Purdue)

ETH (Zurich)