



# Beryllium isotopic systematics in island arc volcanic rocks from northeast Japan: Implications for the incorporation of oceanic sediments into island arc magmas

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## ABSTRACT

We present beryllium (Be) isotopic ratios ( $^{10}\text{Be}/^{9}\text{Be}$ ) for 55 volcanic rocks from 21 Quaternary volcanoes in the northeast Japan arc, including the Hokkaido and central Japan areas (the NEJH). Although Be isotopic ratios in these rocks are much lower than in other arcs reported in the literature, they are much higher than in control samples (ocean island basalt (OIB) and Tertiary basalts) that have no recent addition of  $^{10}\text{Be}$ . Hence, our results indicate the incorporation of a component derived from subducted sediments in the Japan arc magmas.

Two kinds of Be isotopic variations were observed in the NEJH: 1) variations among lava flows of different ages in the same volcano; and 2) variations among volcanoes in an along-arc or cross-arc direction. In the first case, there are no apparent differences in Be isotopic ratios for lavas of the same age, but relatively large variations among lavas of different ages, and the degree of variation depends on the regional characteristics, indicating that regional differences in near-surface conditions may affect these Be isotopic variations. In the second case, the progressive decrease in Be isotopic ratios in a cross-arc direction suggests the continuous incorporation of subducted sediment, even in the back-arc region. The regional along-arc Be isotopic variations reflect not only the incorporation of sediments into the arc magma, but also regional tectonic and/or geological and geochemical processes.

This is the first systematic study of Be isotopic systems in the NE Japan arc, and we provide direct evidence for the incorporation of sedimentary components from the subducting slab into the arc magma, including in the back-arc region at 300 km from the volcanic front, where the Wadati–Benioff Zone is at a depth of 290 km.

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## 1. Introduction

Although, arc volcanism is closely related to subduction processes, the mechanism and material involved in the formation of arc magmas remain controversial. Both B and  $^{10}\text{Be}$  are useful tracers of the recycling of the sedimentary layer on a subducting slab (e.g., Morris et al., 1990); however, it is difficult to distinguish B of terrestrial origin from that of marine origin because B is concentrated throughout the oceanic sediment column.

Unlike B,  $^{10}\text{Be}$  is a unique tracer in investigating the contribution from the upper part of the sedimentary layer in a subducting slab to an arc magma.  $^{10}\text{Be}$  is a radioactive nuclide with a relatively short half-life (1.36 m.y.: Nishiizumi et al., 2007; 1.39 m.y.: Korschinek et al., 2010), and is produced by cosmic-ray spallation reactions with oxygen and nitrogen in the atmosphere (Arnold, 1956; Lal and Peters, 1967). The  $^{10}\text{Be}$  is adsorbed onto aerosols and quickly removed from the

atmosphere by precipitation (Raisbeck et al., 1981; Monaghan et al., 1986). Because of its strong adsorptive properties, it is concentrated in near-surface pelagic sediments on the ocean floor (average concentration of  $\sim 5 \times 10^9$  atoms/g; Tanaka and Inoue, 1979; Brown, 1984; Bourles et al., 1989; Anderson et al., 1990). Because of its short half-life,  $^{10}\text{Be}$  is only concentrated in the upper part of the sedimentary layer (from about  $4.0 \times 10^9$  atoms/g at the surface to below detection limits at  $>250$  m depth; Dreyer et al., 2010). Therefore, subducting altered oceanic crust (AOC) under a thick sedimentary cover does not contain  $^{10}\text{Be}$ , and concentrations of  $^{10}\text{Be}$  in the mantle are extremely low (Tera et al., 1986; Ryan and Langmuir, 1988). Thus, unlike other isotopes with long half-lives such as Sr and Pb,  $^{10}\text{Be}$  is specifically sensitive to the contribution of recently subducted sediments to arc magmas at convergent margins, because it is not necessary to consider secondary addition from the lower crust or the mantle.

Several studies of  $^{10}\text{Be}$  in island arc systems (Brown et al., 1982; Tera et al., 1986; Monaghan et al., 1988; Morris and Tera, 1989; Dreyer et al., 2010) have demonstrated a component of oceanic sediment from the subducting plate in the source of arc magmas. However, there are only a few preliminary studies of  $^{10}\text{Be}$  in the Japan arc (Imamura et al.,

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1984; Tera et al., 1986; Morris and Tera, 1989), and there are no detailed reports on the relationship between Be isotope ratios and arc volcanism in northeast Japan (including the Hokkaido and central Japan areas, hereafter NEJH).

The Japan arc, which is regarded as a typical island arc, formed where the Pacific Plate and the Philippine Sea Plate are subducting beneath the Eurasian Plate. Most volcanic activity in Japan is strongly related to subduction. An old (~180 Ma), cold plate is continuously subducting beneath the NEJH area (Hasegawa et al., 1994). During the formation of arc magmas, the role of subducting pelagic sediments and AOC with high H<sub>2</sub>O contents is considered essential to reduce the melting point of mantle materials under high-pressure conditions in the upper mantle (e.g., Tatsumi and Eggins, 1995). In investigating the characteristics of and mechanisms for arc magma genesis in the NEJH, it is important to establish whether the oceanic sediments that form the uppermost part of the subducted slab are incorporated into the arc magmas.

Studies of Pb and Sr isotopes in volcanic rocks of the NEJH (Tatsumoto and Knight, 1969; Nohda and Wasserburg, 1981; Notsu, 1983; Notsu et al., 1985) have suggested sediment incorporation into the arc magmas. However, because of the relatively long half-life of their parent radionuclides, Pb and Sr isotopes only indicate sediment incorporation indirectly and they cannot distinguish between recently versus anciently subducted sediments. With its relatively short-half life, <sup>10</sup>Be is much better at revealing a component of subducted sediment in arc magmas.

Furthermore, because the subducted slab in the NEJH is estimated to move at a rate of about 10 cm/yr (Minster and Jordan, 1978; DeMets et al., 1990), <sup>10</sup>Be is a suitable radioactive tracer for investigating variations in the effect of sediment on magmas in a cross-arc direction. The NEJH arc consists of many volcanoes in various geological and tectonic settings; therefore, it is important to investigate how these features affect <sup>10</sup>Be signatures.

Meteoritic <sup>10</sup>Be (~4 × 10<sup>4</sup> atoms/g) can obscure primary magmatic concentrations; therefore, it is important to minimize surface contamination effects as much as possible. In the present study, we report Be isotopic ratios (<sup>10</sup>Be/<sup>9</sup>Be) for the NEJH volcanic rocks determined by the well-established sequential acid leaching method for removing meteoritic <sup>10</sup>Be (Shimaoka, 1999; Shimaoka et al., 2004), and evaluate the contribution of sedimentary components to magma generation in the NEJH.

## 2. Materials and methods

### 2.1. Samples

We analyzed 55 samples from 21 Quaternary volcanoes in the NEJH (Table 1). The localities of the investigated volcanoes are shown in Fig. 1. Considering the effect of in situ produced <sup>10</sup>Be at the land surface, all samples were taken from sites regarded to have been strongly shielded from cosmic rays after the lavas were erupted and exposed.

We analyzed several samples from the same lava flow supplied by different donors to verify the reproducibility of Be ratios (<sup>10</sup>Be/<sup>9</sup>Be); i.e., to assess Be isotopic variations due to sample characteristics such as heterogeneous mineralogy and the location of the sampling site. In some volcanoes, different lava flows were analyzed to compare Be isotopic ratios in lavas erupted at different times.

Because <sup>10</sup>Be is concentrated in the uppermost layer of sediments in the subducting plate, it indicates of which material in the subducting plate is incorporated into arc magma. Therefore, we selected samples from north to south along the arc to compare the effects of subducted materials in different tectonic settings.

Furthermore, to investigate the fate of oceanic sediment in the plate beneath the arc, several samples were taken from back-arc volcanoes. In a study of <sup>10</sup>Be/<sup>9</sup>Be ratios in the Kurile arc, significant <sup>10</sup>Be was identified up to 150 km from the volcanic front (Dreyer et al., 2010). In the NEJH,

the maximum distance from the volcanic front is 120 km (e.g. Ichinomegata) and the depth of the Wadati–Benioff zone is up to 290 km beneath Rishiri.

We also examined a series of samples from Esan and Iwate volcanoes (G and J in Fig. 1, respectively) to study variations in <sup>10</sup>Be/<sup>9</sup>Be ratios in different lavas from the same volcano. Some samples from Esan have extremely high <sup>10</sup>Be/<sup>9</sup>Be ratios (up to 80.8 × 10<sup>-11</sup>; Tera et al., 1986; Morris and Tera, 1989; Morris et al., 1990) compared with other Japanese volcanoes (Imamura et al., 1984). Hence, it is important to establish if such high <sup>10</sup>Be/<sup>9</sup>Be ratios are typical of this volcano. On the other hand, Iwate volcano is situated at the volcanic front and erupted a large volume of basaltic magma during several eruptive periods (Ishikawa et al., 1984; Nakagawa, 1987). They are considered to be minimally affected by near-surface contamination.

As a zero-level control sample, we analyzed an ocean island basalt (OIB) lava from the Loihi Seamount, Hawaii (Table 2), because it is considered to be unaffected by subducted materials. We also analyzed Tertiary basalts from the Nozoki (10–20 Ma) and Aosawa (6.5 Ma) formations (Takaoka, pers. comm.), which are located near the Chokai and Kurikoma volcanoes, respectively (O and P in Fig. 1), to evaluate the effect of contamination by meteoric water on <sup>10</sup>Be and in situ nuclear production by the <sup>7</sup>Li(α, p)<sup>10</sup>Be reaction (Merrill et al., 1960) in U- and Th-containing minerals or through the <sup>16</sup>O(n, 4p3n)<sup>10</sup>Be spallation reaction by cosmic rays (Yokoyama et al., 1977).

### 2.2. Sample preparation

To remove meteoritic <sup>10</sup>Be, all samples were leached using a sequential leaching method (Shimaoka, 1999; Shimaoka et al., 2004). Be isotopic ratios (<sup>10</sup>Be/<sup>9</sup>Be) were determined using a new procedure of Be extraction described below.

Any weathered parts of the rock samples were removed using a saw. Chunks from the interior of each sample were crushed in an iron mortar and pestle, and sieved to a powder size of ≤50 mesh (<300 μm) through a disposable nylon mesh. The powder samples were leached in 1 M HCl for 2–4 h in an ultrasonic cleaner. After leaching, these samples were washed several times with 1 M HCl and finally rinsed with distilled water.

100–150 μg of a Be carrier was added to about 10 g of dried sample. The carrier used in the present study was a Be solution from a commercial product for AA (atomic adsorption) analysis. The sample was then dissolved in a mixture of concentrated HF and HNO<sub>3</sub> in a Teflon beaker by gentle boiling on a hot plate. After complete dissolution, the solution was evaporated. Any trace amount of fluoride was eliminated by fuming with HClO<sub>4</sub> three times. The residue of perchlorate salts was dissolved by adding HCl. Separation of Be from most other elements, such as calcium and aluminium, was achieved by adding NH<sub>4</sub>OH and NaOH to the hydroxide complexes with Fe, such that Be was precipitated with Fe. To remove Fe, isopropyl ether extraction was repeated twice. Acetylacetonate extraction into CCl<sub>4</sub> in the presence of sodium EDTA is effective for eliminating Ti, and many trace elements remained. Subsequently, Be was back-extracted into dilute HNO<sub>3</sub> and the solution was dried in a Teflon beaker on a hot plate. The residue was dissolved in concentrated HCl and passed through a 5 ml Dowex 1-X8, 100–200-mesh anion exchange resin with 11 M HCl. This procedure can be skipped in most cases. Finally, Be was separated from trace amounts of impurities with a 5 ml cation exchange column (using Dowex 50 W-X8, 100–200-mesh cation exchange resin) and recovered in an eluent of 1 M HCl. The Be fraction was checked by AA analysis. In the present study, the chemical yields (80–90%) were determined by AA measurement and comparing with amount of adding Be carrier and Be amount after chemical treatment for Be extraction.

Beryllium hydroxides were precipitated with a few drops of NH<sub>4</sub>OH (aq.) in centrifuge tubes. After washing with distilled water, the precipitates were re-dissolved with 1 M HCl. The Be chloride was ignited to form an oxide (BeO) in a quartz vial at 800 °C for 20 min. Finally, the

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