



Contents lists available at ScienceDirect

Quaternary International

journal homepage: [www.elsevier.com/locate/quaint](http://www.elsevier.com/locate/quaint)

# Re-identification of Shishimuta-Pink tephra samples from the Japanese Islands based on simultaneous major- and trace-element analyses of volcanic glasses

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## ARTICLE INFO

### Article history:

Received 23 August 2016

Received in revised form

31 January 2017

Accepted 21 February 2017

Available online xxx

### Keywords:

Tephra

Volcanic glass

LA–ICP–MS

Multi-element analysis

## ABSTRACT

The Shishimuta-Pink (Ss-Pnk) tephra erupted from the Shishimuta caldera on Kyushu Island, Japan, at 1 Ma, and it is distributed extensively over the Japanese Islands. We quantitatively analyzed 58 elements in volcanic glass shards of this tephra, which sampled at 9 points, using laser ablation–inductively coupled plasma–mass spectrometry (LA–ICP–MS). The nine samples have been correlated as Ss-Pnk tephra in previous studies, based on various geologic and petrographic properties. The element patterns of the tephra samples are similar, except for that of the Kuratama–Rindo O7 sample obtained in the Kazusa Group at the Boso Peninsula, located ~900 km from the source volcano. The results of the Tukey–Kramer multiple comparison test suggest that 62%–71% of the concentrations of the 58 elements in the Kuratama–Rindo O7 sample differ significantly from the element concentrations in the other eight Ss-Pnk samples, whereas only 2%–31% of the element concentrations of the eight Ss-Pnk samples vary significantly. The element pattern of the sample of the Osaka Group Pink (OGPK), which is treated as being representative of the Ss-Pnk tephra, can be clearly distinguished from five other widespread marker tephtras erupted from Kyushu Island. These results strongly suggest that the multi-element analysis of volcanic glass shards from tephtras can be used as a basis for the identification and correlation of widespread Quaternary tephtras.

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## 1. Introduction

Determining the identity of distal tephtras and cryptotephtras dispersed at distances of more than 100 km from the source volcano is important for understanding highly explosive eruptions of the Quaternary (e.g., Lowe, 2011; Ponomareva et al., 2015). Therefore, analytical techniques for tephrochronology are being developed to characterize or ‘fingerprint’ distal tephtras in more detail and with greater degrees of reliability (e.g., Kimura et al., 2015; Maruyama et al., 2016a, b).

On the Japanese Islands, many deposits from highly explosive eruptions have been found in the representative geologic groups of the Plio-Pleistocene, including the Osaka, Kobiwako, Tokai,

Kakegawa, Uonuma, and Kazusa groups (e.g., Satoguchi, 1997; Satoguchi et al., 1999; Machida and Arai, 2003; Satoguchi and Nagahashi, 2012). These geologic groups contain layers of widespread tephtras, which are important stratigraphic markers across the Japanese Islands. In particular, the Pink volcanic ash of the Osaka Group (OGPK) in the Kinki district of Japan (e.g., Yoshikawa, 1976) is one of the most important markers.

The OGPK has been identified as the co-ignimbrite ash of the Yabakei pyroclastic flow deposit (YFPD; Danhara et al., 1992a, 1993). The source of the YFPD is the buried Shishimuta caldera, which was identified on the basis of a negative Bouguer gravity anomaly, drillhole data, and geochronology (Kamata, 1989). Danhara et al. (1997) determined the fission track (FT) ages of the OGPK ( $1.00 \pm 0.04$  Ma) and the YFPD ( $1.04 \pm 0.05$  Ma). Uto and Sudo (1985) estimated the K–Ar age of the YFPD as  $0.99 \pm 0.03$  Ma. The FT and K–Ar ages correspond to the period of the Jaramillo subchron ( $1.07$ – $0.99$  Ma; e.g., Shackleton et al., 1990; Berggren et al., 1995). Machida and Arai (2003) recognized as the widespread tephtra that erupted from the Shishimuta caldera at 1 Ma

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(the late Early Pleistocene) and named the Shishimuta-Pink (Ss-Pnk) tephra. The Shishimuta caldera is also regarded as the source volcano of two widespread tephra that erupted shortly after the ejection of the Ss-Pnk tephra, namely, the Azuki volcanic ash in the Osaka Group distributing around the Osaka City and the Ku6C tephra in the Kazusa Group at the Boso Peninsula of the Kanto district ~900 km from the Shishimuta caldera (Kamata and Danhara, 1994; Kamata et al., 1994, 1997). Machida (1999) named these tephra together as the Shishimuta-Azuki (Ss-Az) tephra, and this tephra, like the Ss-Pnk tephra, is considered to be an important volcanic ash marker of the late Early Pleistocene in Japan.

The Dochu volcanic ash layer found in the northwestern part of Shikoku Island is related to the OGPK (Mizuno, 1987, 1992; Morie et al., 2010). The OGPK is correlated to several volcanic ash layers on Honshu Island. There are the Shiratsuchidani volcanic ash layer of the Kobiwako Group (Yamasaki et al., 1996; Itoh and Danhara, 2008), the Tara volcanic ash layer of the Tokai Group (Yoshikawa et al., 1988, 1991), the Ogi volcanic ash layer of the Uonuma Group of Niigata Prefecture (Tomita and Kurokawa, 1994), and the O7 volcanic ash layer of the Otadai Formation of the Kazusa Group (Kikkawa et al., 1993). In addition, a B943-3 tephra layer at 635.1 m of the 1400-m-long bored under Lake Biwa in central Honshu (Danhara et al., 2010). The distribution of the Ss-Pnk tephra is almost identical to that of the Ss-Az tephra (Machida, 1999).

The FT ages of the Dochu, Shiratsuchidani, and B943-3 tephra have been estimated to be  $1.3 \pm 0.2$  Ma (Akojima and Suyari, 1989),  $1.07 \pm 0.17$  Ma (Yamasaki et al., 1996), and  $1.00 \pm 0.08$  Ma (Danhara et al., 2010), respectively. The Tara volcanic ash layer in the uppermost Tokai Group correlates with the Yoro volcanic ash layer in the Chikarao Formation, whose FT age has been estimated as  $0.98 \pm 0.19$  Ma (Yoshida et al., 1990). Tsuji et al. (2005) estimated the age of the O7 tephra to be 1.056–1.050 Ma based on oxygen isotope stratigraphy using benthic foraminifers. The FT ages and the oxygen isotope stratigraphic age of these tephra are almost identical to those of the YPF and the OGPK.

The paleomagnetic directions of the YPF, OGPK, and O7 tephra were determined by Hayashida et al. (1996). The tephra are characterized by ~30° inclinations of normal polarity with slightly westerly declinations, achieved by normal magnetization during the period of the Jaramillo subchron. The Dochu (Mizuno, 1987), B943-3 (Danhara et al., 2010), Tara (Yoshida et al., 1990), and Ogi (Tomita and Kurokawa, 1994) tephra also have a normal paleomagnetic polarity corresponding to the Jaramillo subchron.

The above-mentioned relationships are based mainly on various petrographic properties, tephrostratigraphy, magnetostratigraphy, FT dating, and biostratigraphy. The petrographic properties include the morphological correlation, the refractive indices, the chemical compositions of volcanic glass shards, and the abundance ratios of all mineral and glass components and those of heavy mineral species such as amphibole and pyroxene. Kikkawa et al. (1993) used inductively coupled plasma–atomic emission spectrometry (ICP–AES) to determine the chemical compositions of volcanic glass shards in the O7 and OGPK tephra. Nagahashi et al. (2015) analyzed the concentrations of eight trace elements of tephra samples obtained from the Osaka Group, including the OGPK and core samples from Lake Biwa, using ICP–AES, and the concentrations of the major oxide components using electron-beam analytical techniques.

The chemical compositions of the major components (e.g., Al<sub>2</sub>O<sub>3</sub> and MgO) of the volcanic glass shards in the tephra obtained from electron-beam analyses have, in particular, been widely used for conducting various tephrochronologic investigations and for identifying tephra (e.g., Kuehn et al., 2011; Lowe, 2011). However, the chemical compositions of the major oxides of the volcanic glass shards of some tephra are quite similar to those of other tephra

and are occasionally insufficient for tephra characterization or ‘fingerprinting’ (e.g., Stokes and Lowe, 1988; Brendryen et al., 2010; Kaufman et al., 2012). Therefore, some researchers have analyzed the concentrations of the trace elements of the volcanic glass shards to obtain stronger geochemical data for identifying tephra (e.g., Eastwood et al., 1999; Pearce et al., 2004a; Lane et al., 2013).

Kimura et al. (2015) reported the compositions of the major elements and various trace elements in volcanic glass shards sampled from 80 widespread tephra from the Japanese Islands. However, those authors analyzed a single representative sample of each tephra, and the fact that the samples correlated as a single tephra occur in various locations was not necessarily considered. This points out another problem of previous tephrochronologic investigations, which is that a reidentification is needed of the individual tephra samples correlated as single tephra by previous studies.

In this study, we used laser ablation–inductively coupled plasma–mass spectrometry (LA–ICP–MS) to determine the element abundances and patterns of 58 major and trace elements in volcanic glass shards sampled from the Ss-Pnk tephra. Recently, many elements and isotopes having wider concentration range covering 9–10 orders of magnitude can be measured by LA–ICP–MS (e.g., Becker, 2007; Dussubieux et al., 2016). Combination of pulse counting and analogue current monitoring protocols used in ICP–MS makes it possible to carry out simultaneous major- and trace-element quantitative analyses of volcanic glass shards. In addition, we compared the element abundances and patterns of the Ss-Pnk samples with those of five samples of widespread tephra from Kyushu Island, which are important marker tephra in Japan. As shown by Maruyama et al. (2016a, b), the element patterns of volcanic glass shards can be useful for identifying and correlating tephra samples and can be used to distinguish between tephra dispersed from the same source volcano.

## 2. Samples and analytical methods

### 2.1. Tephra samples

The abbreviations of the tephra referred to in the present study are based on the work of Machida (1999). Nine tephra samples previously correlated as Ss-Pnk (Table 1) were prepared for major- and trace-element analyses of the volcanic glass shards. The Ss-Pnk tephra sampling locations are shown in Fig. 1. The Yoro River O7 (Yoro) and the Kuratama–Rindo O7 (Kuratama) tephra samples were obtained from locations furthest away from the Shishimuta caldera (sampling points 8 and 9 in Fig. 1). Ss-Pnk tephra has not yet been found in the Tohoku district of northeastern Honshu Island nor on Hokkaido Island (Fig. 1).

The tephrochronology of the Ss-Pnk tephra-bearing geological units examined in this study is shown in Fig. 2. The petrographic properties of the YPF are summarized in Danhara et al. (1992a, 1993). The layer of YPF in the Kaguma region of Yabakei in Oita Prefecture (~120 m thick) is directly covered by the Imaichi pyroclastic flow (~30 m thick). A densely welded and devitrified layer (~100 m thick) is dominant in the YPF found in Kaguma. The uppermost layer (<10 m thick) of the YPF in Kaguma is not welded and devitrified, but it may have been affected by the high-temperature Imaichi pyroclastic flow. The lowermost layer (<15 m thick) is glassy and welded and occurs as a solid obsidian-like layer. This lowermost layer presumably formed as a result of the high temperature shortly after the eruption and the pressure of the thick pyroclastic flow deposits. However, the lowermost layer contains characteristic glass shards of bubble-wall or pumice types. The sample of the YPF analyzed in this study is obsidian-like glass obtained from the lowermost layer of the YPF at Kaguma.

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