

Constraints on the timing of explosive volcanism at Aso and Aira calderas (Japan) between 50 and 30 ka: New insights from the Lake Suigetsu sedimentary record (SG14 core)

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Key Points:

Refined eruption stratigraphy for Japan (between 50 and 30 ka) through the identification of cryptotephra layers in the varved Lake Suigetsu sediment core.

Aso caldera was particularly active between 35 and 30 ka, producing at least 5 tephtras that were dispersed >500 km from source.

A large eruption occurred from Aira 1.3 ka before the cataclysmic VEI 7 caldera-forming AT eruption that blanketed Japan in ash.

Abstract

Volcanoes in the East Asian/Pacific region have been the source of some of the largest magnitude eruptions during the Late Quaternary, and accurately evaluating their eruptive histories is essential for hazard assessments. To overcome difficulties in resolving and precisely dating eruptions in the near-source realm, the high-resolution (varved) sediments of Lake Suigetsu (central Honshu, Japan) were examined for the presence of non-visible (cryptotephra) layers from 50 ka up until the 30 ka Aira-Tanzawa (AT) caldera-forming event of Aira volcano. Cryptotephra layers are four times more frequently preserved than visible markers in the Suigetsu sediments, meaning that this archive provides a unique and unprecedented record of eruptions that were dispersed over the densely populated regions of central Honshu. Major and trace element volcanic glass chemistry is used to fingerprint the ash layers and pinpoint their volcanic origin. Tephra layers are found throughout the investigated sediments, but the highest abundance of ash fall events are recorded between 39 and 30 ka, capturing a period of intense volcanism at calderas on Kyushu Island (Japan). The augmented Suigetsu tephrostratigraphy records at least seven eruptions from Aso caldera (southern Kyushu) that post-date the widespread ACP-4 Plinian eruption (ca. 50 ka), and four explosive events from Aira (central Kyushu) that occurred leading up to the catastrophic caldera-forming AT eruption (ca. 30 ka).

1. Introduction

Volcanoes distributed across the heavily populated East Asian/Pacific region have been the source of some of the largest known caldera-forming events [Simkin and Siebert, 2000], blanketing Japan and the surrounding seas in ash [e.g., Machida and Arai, 2003; Smith *et al.*, 2013; Mahony *et al.*, 2016; Schindlbeck *et al.*, 2018; McLean *et al.*, 2020]. Accurately reconstructing the tempo and magnitude of pre-historic eruptions at these volcanoes is an essential component of risk and hazard assessments [Kiyosugi *et al.*, 2015]. Unfortunately, the burial, destruction and subsequent erosion of eruption deposits in near-source areas contribute to an incomplete geological record, hampering our ability to accurately reconstruct eruption histories. Piecing together fragmentary records close to the source volcano is further complicated by the fact that many Japanese volcanoes erupt successive deposits with geochemically very similar compositions [e.g., Machida and Arai, 2003; Smith *et al.*, 2013; Kimura *et al.*, 2015; McLean *et al.*, 2018]; consequently, deciphering the true number of events is challenging and can result in an under-recording of eruptions [e.g., Albert *et al.*, 2018]. Furthermore, precisely evaluating the tempo and recurrence intervals of past eruptions using near-source volcanic successions is difficult, and for many volcanoes in Japan this remains a significant obstacle, particularly at low-K (tholeiitic) and medium-K (calc-alkaline) centres where mineral phases are largely unsuitable for $^{40}\text{Ar}/^{39}\text{Ar}$ dating. Consequently, dating such eruptions from the last 50 ka is heavily reliant upon the sporadic discovery of charcoal fragments buried within eruption units, that are suitable for ^{14}C dating.

Fortunately, volcanic ash (tephra) deposits are routinely identified in distal sedimentary (palaeoclimate and archaeological) archives in and around Japan [e.g., Machida and Arai, 1983; 2003; Furuta *et al.*, 1986; Moriwaki *et al.*, 2016; McLean *et al.*, 2018; Albert *et al.*, 2019; Chen *et al.*, 2019], which provide critical insights into the tempo, behaviour and ash dispersals of

highly-explosive volcanism in East Asia. The Lake Suigetsu sedimentary archive provides a key tephrostratotype of explosive events for Japan [e.g. *Smith et al.*, 2013], and the unique high-precision chronology [e.g. *Bronk Ramsey et al.*, 2012] ensures that tephra layers identified in the radiocarbon timeframe are robustly dated. The sediments also preserve numerous non-visible (cryptotephra) layers throughout the sequence [e.g. *McLean et al.*, 2018], which are derived from more distal sources and/or lower magnitude events.

This paper explores the cryptotephra record preserved in the ca. 14 m of the varved (annually laminated) Lake Suigetsu sediments (SG14 core) dated between 50 and 30 ka [*Bronk Ramsey et al.*, 2012; *Schlolaut et al.*, 2012]. While much more is known about recent eruptions from the proximal volcanological record, the periodicity and repose intervals during the 50 and 30 ka window is at present poorly understood. During this period, the high-resolution sediments of Lake Suigetsu can provide unprecedented insight into the timing and frequency of large eruptions and their associated ash fall events dispersed over the now heavily populated regions of East Asia. Several large calderas situated downwind of Lake Suigetsu (i.e., those on the Ryukyu-Kyushu arc; Figure 1), and Aira and Aso in particular, are thought to have been very active between 50 and 30 ka, based on the record preserved in the proximal volcanic stratigraphies [e.g., *Nagaoka et al.*, 2001; *Miyabuchi*, 2009; 2011]. Despite this, almost no distal ash equivalents have been identified. Currently, most eruption ages established during this period are poorly constrained and are either broadly extrapolated based on their stratigraphic position relative to the ~30 ka Aira-Tanzawa (AT) ash from Aira, or radiocarbon dated via palaeosols developed during periods of quiescence. A more thorough eruption chronology can aid our understanding of the behaviour of these centres prior-to (e.g., Aira) or following (e.g., Aso) large caldera-forming events.

2. Study Site and Potential Tephra Sources

2.1 Lake Suigetsu, central Japan

Lake Suigetsu is the largest of the ‘Mikata Five Lakes’ (covering an area of ca. 4.2 km²), located in a small tectonic basin on the western side of the Mikata fault, Honshu Island, central Japan (Figure 1; 35°35’0’’N, 135°53’0’’E, 0 m above present sea level; Figure 1a). The main tributary is the River Hasu, which enters on the southeast side of Lake Mikata and flows through a narrow channel into Lake Suigetsu (Figure 1b). This unique configuration of the lakes and hydrological setting means that cryptotephra layers are well preserved within the fine and laminated (varved) sediments. Suigetsu is also adequately situated away from the large calderas of Hokkaido and Kyushu, and so is not inundated with locally sourced volcanic glass, which would also preclude the identification of cryptotephra layers with low concentrations of glass shards.

The ‘SG06’ sedimentary core (Figure 1a) was extracted from Lake Suigetsu in 2006, with the composite sequence spanning ca. 73.2 m. The SG06 core was obtained from four overlapping boreholes A, B, C and D, which were situated ca. 20 m apart; [*Nakagawa et al.*, 2012]. The lake was re-cored in 2014 (‘SG14’), to provide additional material for scientific analysis and to prepare sediments for display at the *Fukui Prefectural Varve Museum*. The sediments were obtained from new boreholes E, F, G and H, which were located ca. 320 m further east of those

from SG06. The SG14 sediments were utilised for the cryptotephra investigations outlined in this study.

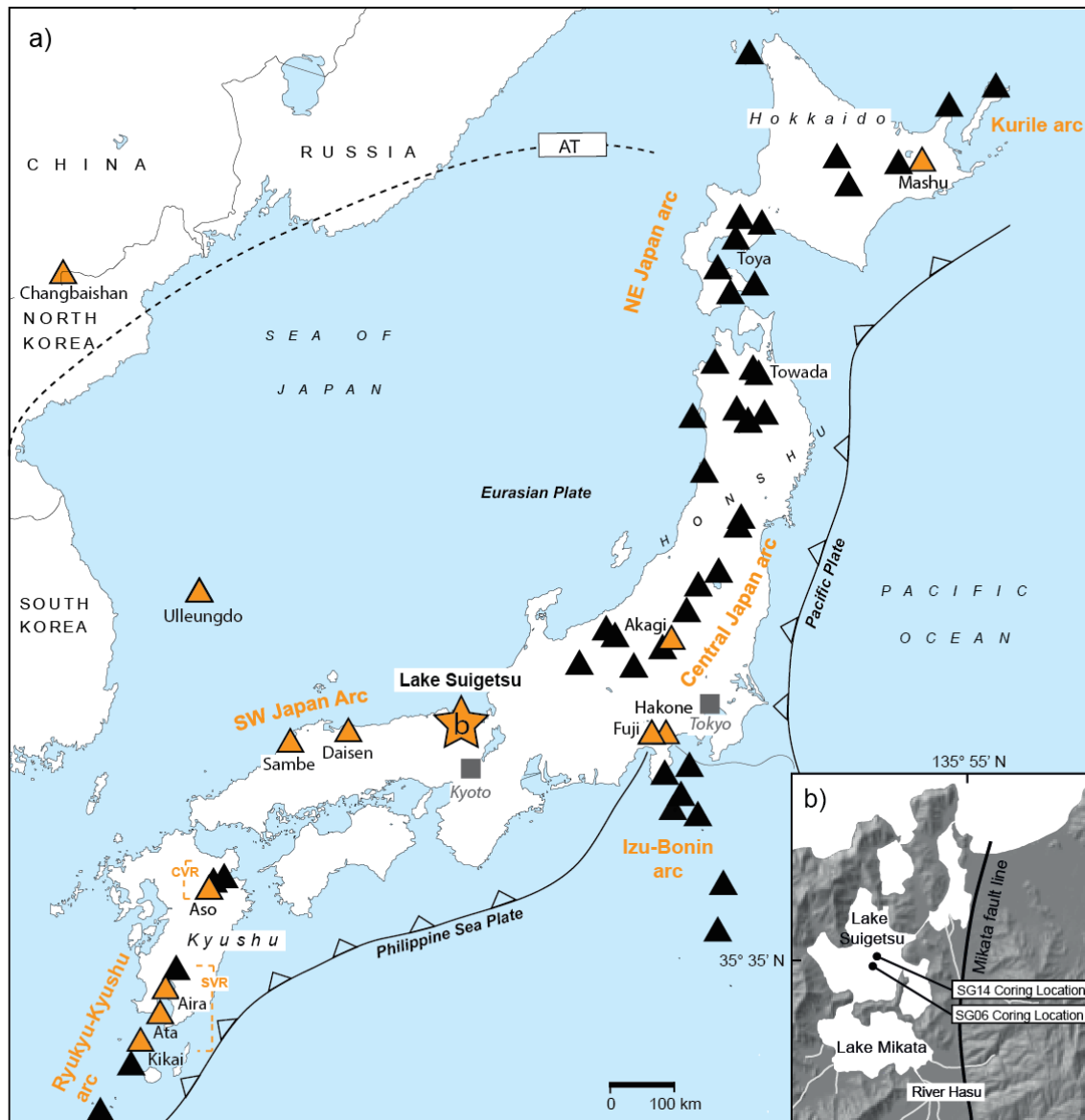


Figure 1. (a) Volcanoes in Japan, North Korea/China (Changbaishan) and South Korea (Ulleungdo) (black and orange triangles) known to have been active during the Late Quaternary (data from *Machida and Arai* [2003]). Volcanoes mentioned within the text are coloured orange, and a star denotes the location of Lake Suigetsu. The visible ash dispersal for the AT (Aira volcano; 30 ka) tephra is marked by a dashed line (as defined by *Machida and Arai* [2003]). (b) Location of the five Mikata lakes, which are situated west of the active Mikata fault, adjacent to Wakasa Bay. The positions of coring campaigns SG06 and SG14 are marked on Lake Suigetsu (modified after *Nakagawa et al.* [2005]).

2.2 Sources of Visible Tephra Dispersed to Lake Suigetsu

Thirty-one visible ash layers (named according to their SG06 Composite Depths (CD)) were originally identified in the SG06 core [*Smith et al.*, 2011, 2013; *McLean et al.*, 2016], most of which originating from arc volcanoes south of Suigetsu, situated in the Kyushu Central and Southern Volcanic region (Kikai, Aira, Aso and Ata volcanoes; Figure 1; see *Albert et al.*, 2019). Four visible tephra layers in the Suigetsu sequence (SG06-0226, SG14-1091, SG06-1288, SG14-3216) originate from intraplate volcanoes Ulleungdo (South Korea) and Changbaishan (North Korea/China) (Figure 1; *Smith et al.*, [2011]; *McLean et al.*, [2016, 2018, 2020]).

The visible tephrostratigraphic sequence investigated here (50 to 30 ka), is composed of five key markers (see Table 1; Figure 2), which include the Central Pumice Cone 4 (ACP4) eruption from Aso caldera (ca. 50 ka; correlated to Suigetsu tephra SG06-3912), the Ikeda (SI) tephra erupted from Sambe (ca. 46 ka; SG06-3668), an eruption from Ko-Fuji (ca. 44 ka; SG06-3485), the Ulleung-Yamato (U-Ym) tephra erupted from Ulleungdo (ca. 40 ka; SG14-3216) and the AT tephra (ca. 30 ka; SG06-2650) erupted from Aira [*Smith et al.*, 2013; *Albert et al.*, 2018; 2019; *McLean et al.*, 2020]. Since cryptotephra analysis is typically able to integrate eruption stratigraphies from more distal volcanic sources [e.g., *Jensen et al.*, 2014; *Bourne et al.*, 2015; *Lane et al.*, 2015; *Mackay et al.*, 2016; *van der Bilt*, 2017; *Cook et al.*, 2018], it is possible that cryptotephra layers identified within these sediments may also originate from other more distal sources (> 1,000 km) across East Asia. For instance, ash erupted from the Izu-Bonin and NE Japan arc are preserved as low-concentration cryptotephra layers in the Holocene sediments of Lake Suigetsu [*McLean et al.* 2018].

3. Materials and Methods

3.1 Cryptotephra Extraction Techniques

The master SG14 core (composite sequence) was contiguously sub-sampled at a ca. 5 cm resolution, from the ACP4 tephra (39.1 m CD) to the base of the AT tephra (26.5 m CD), avoiding the known high-energy event layers (e.g., flood horizons; *Schlolaut et al.*, 2014). If elevated peaks in shard concentrations were observed in the ca. 5 cm scan sample, the sediment was resampled at a 1 cm resolution to more precisely determine the stratigraphic positioning of the peak. All samples were wet sieved through a 25 μm mesh, and processed using the heavy liquid floatation method outlined by *Turney* [1998] and *Blockley et al.* [2005]. The extraction residues were mounted on slides using Canada Balsam, and glass shards were counted via microscopic examination to quantify the number of shards per gram of dried sediment (s/g). Glass shards were extracted from samples chosen for geochemical analysis and hand-picked from a wetted-slide using a micromanipulator [see *Lane et al.*, 2014]. These shards were mounted in Epoxy resin stubs, which were sectioned and polished to expose a flat surface, and carbon coated for electron microprobe analysis.

3.2 Major Element Analysis of the Glass Shards

Major and minor element compositions of individual glass shards from the SG14 tephra layers and near-source reference material were measured using a JEOL-8600 wavelength-dispersive electron microprobe (WDS-EMP) at the Research Laboratory for Archaeology and History of

Art (RLAHA), University of Oxford. Analyses used an accelerating voltage of 15 kV, beam current of 6 nA and 10 μm -diameter beam. Peak counting times were 12 s for Na, 50 s for Cl, 60 s for P, and for 30 s for all other elements. The electron microprobe was calibrated using a suite of mineral standards and the PAP absorption correction method was applied for quantification. The accuracy and precision of these data were assessed using analyses of the MPI-DING reference glasses (ATHO-G-1, StHs6/80-G and GOR132-G) from the Max Plank Institute [Jochum *et al.*, 2006], which were run as secondary standards. Analyses of these secondary standards lie within two standard deviations centered on the preferred values and are presented in the Supporting Information. Data were filtered to remove non-glass analyses, and those with analytical totals < 93%. The raw values (given in the Supplementary Material) were normalised to 100 % to better enable comparison and to account for variable hydration, and are presented as such in all tables and figures in the main text.

3.3 Trace Element Analysis of the Glass Shards

Trace element compositions for the glass shards obtained from the SG14 sediments were measured by laser ablation inductively coupled plasma mass spectrometry (LA-ICP-MS) at the Department of Solid Earth Geochemistry, Japan Agency for Marine-Earth Science and Technology (JAMSTEC). The analytical equipment used include the deep-ultraviolet (200 nm) femtosecond laser ablation system (DUV-FsLA) of OK-Fs2000K (OK Laboratory, Tokyo, Japan) connected to the modified high-sensitivity sector field ICP-MS of Element XR (Thermo Scientific, Bremen, Germany). All analyses used a 25 μm crater diameter and depth, and conditions followed those reported by Kimura and Chang [2012]. Ten major elements including P_2O_5 and 33 trace elements were analysed for each sample, and were also run alongside several MPI-DING reference glasses [Jochum *et al.*, 2006] and the BHVO-2G standard provided by the Geological Survey of Japan. Accuracies of the BHVO-2G glass analyses are typically < 3 % for most elements, < 5 % for Sc, Ga, Sm, Eu, Gd, and U and < 10 % for Ni, Cu, and Lu. Full trace element datasets and secondary standard analyses are provided in the Supporting Information.

Trace element compositions for proximal units Aso-Kpfa, A-Fm and A-Kn were performed using an Agilent 8900 triple quadrupole ICP-MS (ICP-QQQ) coupled to a Resonetics 193nm ArF excimer laser-ablation system in the Department of Earth Sciences, Royal Holloway, University of London, using analytical procedures and data reduction (Microsoft Excel) methods outlined by Tomlinson *et al.* [2010]. MPI-DING glasses (StHs6/80-G and ATHO-G-1; Jochum *et al.*, [2006]) were analysed alongside the tephra deposits to monitor the accuracy. Full operating conditions used to analyze these reference samples are provided in the Supporting Information, along with the analyses of the MPI-DING glasses.

3.4 Chronology of the Lake Suigetsu Sediment Cores

A detailed Bayesian age model was used to determine the deposition age of ash layers identified in the Suigetsu sediments. The composite Suigetsu sedimentary sequence (correlation model ver. 08 May '16) was modelled on to the IntCal13 timescale [Reimer *et al.*, 2013] implementing three successive cross-referenced Poisson-process ('P_Sequence') depositional models using OxCal [ver. 4.3; Bronk Ramsey, 2008, 2017]. These include 775 AMS ^{14}C dates obtained from terrestrial plant macrofossils from the upper 38 m (SG06-CD) of the SG93 and SG06 cores [Kitagawa and Van der Plicht, 1998a; 1998b, 2000; Staff *et al.*, 2011, 2013a, 2013b] and varve counting between 12.88 and 31.67m SG06 CD [Marshall *et al.*, 2012; Schlolaut *et al.*, 2012].

SG06 Event Free Depths (EFDs; model ver. 29th Jan '11) were used within the age model, which excludes instantaneous deposits > 5 mm in thickness, (i.e., high-energy event layers, such as floods) [see *Staff et al.*, 2011; *Scholaut et al.*, 2012].

4. Results

4.1 Tephrostratigraphy

Twenty-five ash layers (five visible and twenty cryptotephra layers) are identified in the SG14 varved sediments spanning 50 to 30 ka (39.1 m CD to 26.5 m CD) (Figure 2; Table 1). These ash layers are herein labelled using their SG14 composite depth in cm (SG14 correlation model 30 May '17) that is rounded to the nearest integer (Table 1). For continuity, SG06 labels are used for the four visible tephra layers that were previously identified and geochemically characterised in the SG06 cores (Table 1; Figure 2).

Cryptotephra layers are clearly identified within the sediments as they are characterised by significant peaks in glass shard concentrations, which range from 1,400 s/g to > 20,000 s/g (Figure 2; Table 1). Glass shard characteristics (e.g., shard size, morphology and colour) vary considerably between isochrons (Table 1), but remain relatively uniform in each layer. This provides a first order discrimination between primary and secondary ash layers, since ash is well-sorted during dispersal, but reworking processes tend to mix glass from several sources (as observed in flood layers; *McLean et al.* [2018]). On average, one ash layer per metre is observed between 39 and 30 m CD (Zones 3 and 2; Figure 2), whereas fifteen ash layers are identified in the 4 m prior to the AT ash (30 – 26 m CD; Zone 1). Several of the cryptotephtras identified in Zone 1 are closely spaced; such as SG14-2841 and SG14-2838, which are only separated by 3 cm of sediment that is equivalent to ca. 50 years.

Despite the relatively high glass concentrations in primary isochrons, very low levels of background glasses (i.e., those identified consistently through the sediments) are observed, which indicates very little reworking of material in the lake catchment through this period. As expected, background glass concentrations are lowest in regions furthest from visible ash horizons, for example throughout Zone 1 they remain below 100 s/g (Figure 2). Following (i.e. stratigraphically above) the 1 mm thick visible layer SG06-3912 (ACP4, Aso), similar shard characteristics and glass compositions are observed for ca. 1 m of sedimentation. Higher background concentrations are observed during the Holocene sediments (typically between 500 and 5,000 s/g), due to the presence of several much thicker visible ash layers at 10 ka and 7.3 ka (e.g., U-Oki and K-Ah tephra), which were more easily reworked and continuously in-washed from the local catchment [*McLean et al.*, 2018].

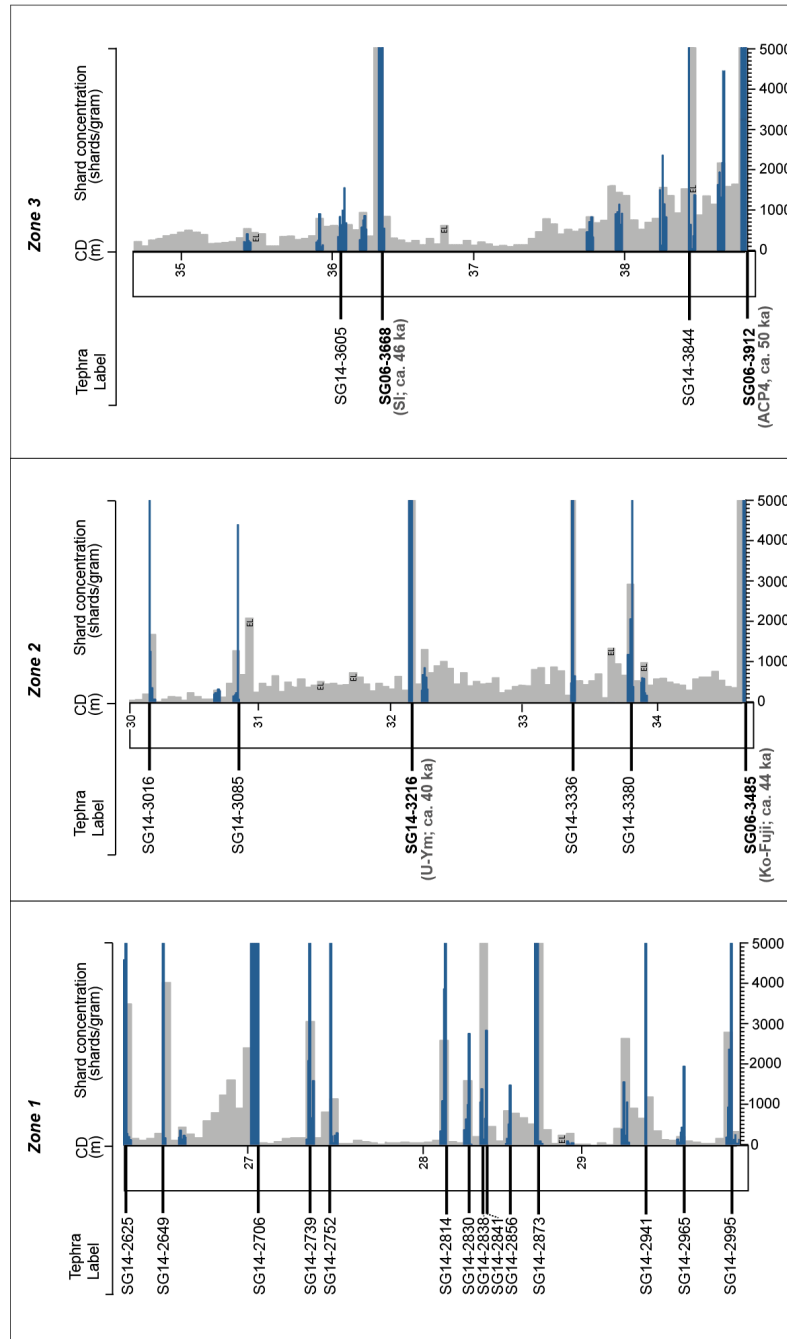


Figure 2. Glass shard concentrations (shards per gram of dry sediment; capped at 5,000) in the annually laminated sediments of the Lake Suigetsu SG14 core (50 to 30 ka). The tephrostratigraphy is divided into three Zones (1 - 3) for illustrative purposes. Concentration of low-resolution (5 cm) samples are shown in grey and high-resolution samples (1 cm) are overlain in blue. Tephra isochrons are labelled using their SG06/SG14 composite depth, with visible layers in bold. Peaks in shard concentrations that coincide with visible, high-energy event layers (EL) are not considered primary. SG06 visible tephra correlations and ages after *Albert et al.* [2018; 2019] and *McLean et al.* [2020].

Table 1. Summary of the tephra layers identified the sediments of the Lake Suigetsu (SG14 core) between 50 and 30 ka.

Suigetsu Tephra Label	V / C	SG14 master core and position	SG14 Composite Sampling Depth (cm)	Shards per gram	Glass shard morphology	Major element glass compositions (wt. %)				Trace element analysis n	Eruption Age (IntCal. yrs BP; 95.4 % Confidence Interval)	Compositional Group	Source
						SiO ₂	K ₂ O	CaO	n				
SG06-2650	V	G-6-base	2614.9	>20,000	PL	77.02 - 78.41	3.24 - 3.65	1.03 - 1.20	35¹	19²	30,174 - 29,982²	3	AT, Aira¹
SG14-2625	C	E-23 (6.0 - 7.0 cm)	2624.2 - 2625.2	9,838	MI, C, V	60.51 - 65.12	2.61 - 3.88	3.57 - 5.48	21	-	30,272 - 30,055	2	Aso
SG14-2649	C	E-23 (30.0 - 31.0 cm)	2648.2 - 2649.2	23,117	MI, PL	74.14 - 77.75	3.69 - 4.63	1.00 - 1.70	15	-	30,602 - 30,395	2	Aso
SG14-2706	C	F-28 (29.8 - 30.8 cm)	2705.4 - 2706.4	>20,000	F, V	78.54 - 79.35	3.54 - 4.20	0.93 - 1.10	49	-	31,631 - 31,391	3	Ryukyu-Kyushu arc
SG14-2739	C	F-28 (62.8 - 63.8 cm)	2738.4 - 2739.4	8,367	PL, MI	63.70 - 65.18	3.58 - 3.92	3.39 - 4.20	16	-	32,380 - 32,111	2	Aso
SG14-2752	C	F-28 (75.8 - 76.8 cm)	2751.4 - 2752.4	10,067	F, C, MV	68.84 - 70.11	3.95 - 4.50	1.66 - 2.02	17	9	32,647 - 32,376	2	Aso
SG14-2814	C	F-29 (35.0 - 36.0 cm)	2813.3 - 2814.3	7,125	F	77.23 - 78.09	3.72 - 4.08	0.79 - 1.01	33	9	33,977 - 33,716	3	Aira
SG14-2830	C	E-30 (14.0 - 15.0 cm)	2828.6 - 2829.6	2,767	PL	73.91 - 75.80	2.70 - 2.94	1.43 - 2.49	16	8	34,368 - 34,111	3	Ryukyu-Kyushu arc
SG14-2838	C	E-30 (22.0 - 23.0 cm)	2836.6 - 2837.6	1,400	PU, MI	-	-	-	5	-	34,562 - 34,301	2	-
SG14-2841	C	E-30 (25.0 - 26.0 cm)	2839.6 - 2840.6	2,880	PL, MI	61.23 - 70.12	2.97 - 5.723	1.74 - 5.13	28	8	34,635 - 34,372	2	Aso
SG14-2856	C	E-30 (40.0 - 41.0 cm)	2854.6 - 2855.6	1,482	C, V	77.56 - 78.86	3.50 - 3.95	0.81 - 1.30	15	-	34,984 - 34,712	3	Aira
SG14-2873	C	E-30 (57.0 - 58.0 cm)	2871.6 - 2872.6	12,575	F, V	75.46 76.76	2.85 - 3.73	0.95 - 1.64	22	9	35,354 - 35,082	3	Aira
SG14-2941	C	E-31 (16.0 - 17.0 cm)	2939.6 - 2940.6	-	MI, P	-	-	-	-	-	36,633 - 36,329	-	-
SG14-2965	C	E-31 (40.0 - 41.0 cm)	2963.6 - 2964.6	1,950	V, MV, MI	74.14 - 74.65	0.74 - 0.81	2.56 - 2.80	4	-	37,085 - 36,755	4	Kurile arc
SG14-2995	C	E-31 (70.0 - 71.0 cm)	2993.6 - 2994.6	>20,000	V	76.55 - 78.42	3.09 - 3.42	1.04 - 1.58	20	-	37,604 - 37,242	3	Ryukyu-Kyushu arc
SG14-3016	C	F-31 (24.0 - 25.0 cm)	3014.6 - 3015.6	6,567	PL, F	75.15 - 76.15	2.76 - 3.68	1.44 - 1.67	10	-	37,944 - 37,567	3	Ryukyu-Kyushu arc
SG14-3085	V	G-08 (52.0 - 53.0 cm)	3083.5 - 3084.5	4,400	PU	57.89 - 68.16	1.88 - 4.55	2.56 - 6.95	28	12	39,010 - 38,634	2	Aso
SG14-3216	V	G-09-06	3216.2	>20,000	MV	59.46 - 62.92	5.79 - 7.23	1.14 - 1.55	37⁴	9⁴	40,332 - 39,816⁴	1	U-Ym, Ulleungdo⁴
SG14-3336	C	F-34 (29.0 - 30.0 cm)	3335.1 - 3336.1	5,750	PU, MI	70.63 - 80.17	1.45 - 2.97	0.90 - 3.16	19	-	42,231 - 41,804	4	-
SG14-3380	V	E-35 (39.5 - 40.5 cm)	3378.9 - 3379.9	18,383	MV, F	65.45 - 67.95	5.31 - 5.67	0.85 - 1.56	16 ⁴	10	42,750 - 42,323 ⁴	1	B-Sg-42; Changbaishan ⁴
SG06-3485	V	E-36-04	3488.6	>20,000	F, V, PL	53.11 - 55.43	0.33 - 0.67	8.50 - 10.58	13¹	13²	44,013 - 43,413²	-	Fuji
SG14-3605	C	E-37 (58.3 - 59.3 cm)	3603.6 - 3604.6	2,200	MI, G	62.27 - 70.28	2.62 - 4.47	0.38 - 5.53	11	10	46,229 - 45,419	2	Aso
SG06-3668	V	E-37-13	3638.6	>20,000	PU, MV, F	70.62 - 78.69	2.22 - 5.19	0.50 - 2.90	36^{1,5}	15³	46,713 - 45,877²	-	Si, Samba²
SG14-3844	C	F-39 (25.0 - 26.0 cm)	3842.9 - 3843.9	16,013	F, V, MI	77.69 - 78.12	3.54 - 4.03	0.84 - 1.06	9	6	49,913 - 48,424	3	Aira
SG06-3912	V	F-39-07	3878.9	>20,000	F, C	69.64 - 73.63	4.46 - 4.90	1.05 - 2.33	20^{1,5}	16²	50,227 - 49,559²	2	ACP4, Aso²

References 1) Smith et al. (2013); 2) Albert et al. (2019); 3) Albert et al. (2018); 4) McLean et al. (2020); 5) This study

V / C = Visible / Cryptotephra layer
 PU = Pumiceous, PL = Platy, C = Cuspate, F = Fluted, V = Vesicular, MV = Microvesicular, MI = Microlite inclusions, G = Green
 n = number of analyses

^a Sampling range for cryptotephra layers and base of visible ash layer.

^b General shard length in the fraction extracted, i.e. in the population > 25 μm.

^c Tephra marked with a dash have not been able to be robustly correlated to a particular eruption or volcano.

^d Tephra SG06-3912 was modelled on the 'SG062012' chronology as reported in Albert et al. (2019).

4.2 Glass Geochemical Compositions

Major and minor element glass compositions of the ash layers identified between 50 and 30 ka are presented in Table 2 and in Figure 3. The major element compositions of SG14-2941 and SG14-2838 could not be analysed due to the high abundance of microlite inclusions within the glass. Trace element compositions were obtained for several cryptotephra layers with suitable shard sizes to further characterise and assist correlations (Table 2; Figure 4). The cryptotephra layers can be categorised using their glass affinities into four compositional groups, described below and outlined in Table 1.

Compositional Group 1 spans a wide SiO₂ (58 – 72 wt. %) and K₂O (2 – 5 wt. %) range, and include seven cryptotephra layers and one visible (SG06-3912) tephra layer (Table 1). Four of these tephra layers have homogeneous compositions (SG14-2625, SG14-2649, SG14-2739 and SG14-2752), and three are heterogeneous (SG14-2841, SG14-3085 and SG14-3605) with both silicic (population a) and less evolved (population b) end-members (Figure 4a – b). When normalised to the primitive mantle, the lower SiO₂ population glasses are less enriched in Large Ion Lithophile Elements (LILE; e.g., Rb, Ba, K) and High Field Strength Elements (HFSE), compared to the silicic population, and possess very similar Rare Earth Element (REE) profiles with light REE enrichment relative to the heavy REEs (La/Yb ratio). Compositional Group 1 shows strong depletions in Nb and Ta (Figure 4) consistent with subduction related genesis.

Compositional Group 2 is characterised by highly silicic glass compositions (> 75 wt. % SiO₂) which all have > 2.7 wt. % K₂O (Figure 3). Four of these tephra layers contain glasses that are very homogenous and compositionally similar (SG14-3814, SG14-2856, SG14-2873 and SG14-3844), which are also distinguished by their low CaO (0.79 – 1.64 wt. %) content. Furthermore, SG14-2814, SG14-2873 and SG14-3844 all have identical mantle normalised profiles, which can be easily distinguished from Compositional Group 1 by more pronounced Eu anomalies and steeper REE profiles (La/Yb; Table 2; Figure 4). This sub-group is also less enriched in HFSE, in particular Nb (Figure 4c – d). In comparison, silicic glasses from SG14-2830 have similar mantle normalised profiles, but lower in light REE, and therefore exhibit a flatter REE profile (La/Yb ratios).

Compositional Group 3 consists of tephra layers SG14-3336 and SG14-2965, which contain medium and low-K glasses, respectively. SG14-3336 glass compositions are particularly heterogeneous, with SiO₂ from 70.5 to 80.0 wt. %, K₂O from 1.45 to 2.97 and CaO from 0.90 to 3.16 wt. % (Figure 3). The four geochemical analyses for SG14-2965 (and several other low-total results; Supporting Information) show that these are very distinctive from the other SG14 glasses presented here, with SiO₂ from 74.14 to 74.65 wt. %, K₂O from 0.74 to 0.81 wt. % and CaO from 2.56 to 2.80 wt. %. Unfortunately, trace element compositions could not be determined for SG14-3336 and SG14-2965 as the shape and size of the shards meant that there was not sufficient area for analysis.

Compositional Group 4 comprises two alkali-rich (Na₂O + K₂O = > 10 wt. %) tephra layers, named SG14-3380 and the visible deposit SG14-3216, which are classified as phonolites and trachytes, respectively (Figure 3c). When normalised to the primitive mantle [Sun and McDonough, 1989], both tephra layers show significant depletions in Ba, Sr and Eu, typical of K-

feldspar fractionation (Figure 4e – f). SG14-3216 has a Y/Th ratio of 1.04 ± 0.70 , whereas for SG14-3380 it is 3.62 ± 0.23 (Table 2). The paucity of depletions in Nb and Ta within these volcanic glasses is inconsistent with subduction related volcanism (e.g., *Kimura et al., 2015*; Figure 4).

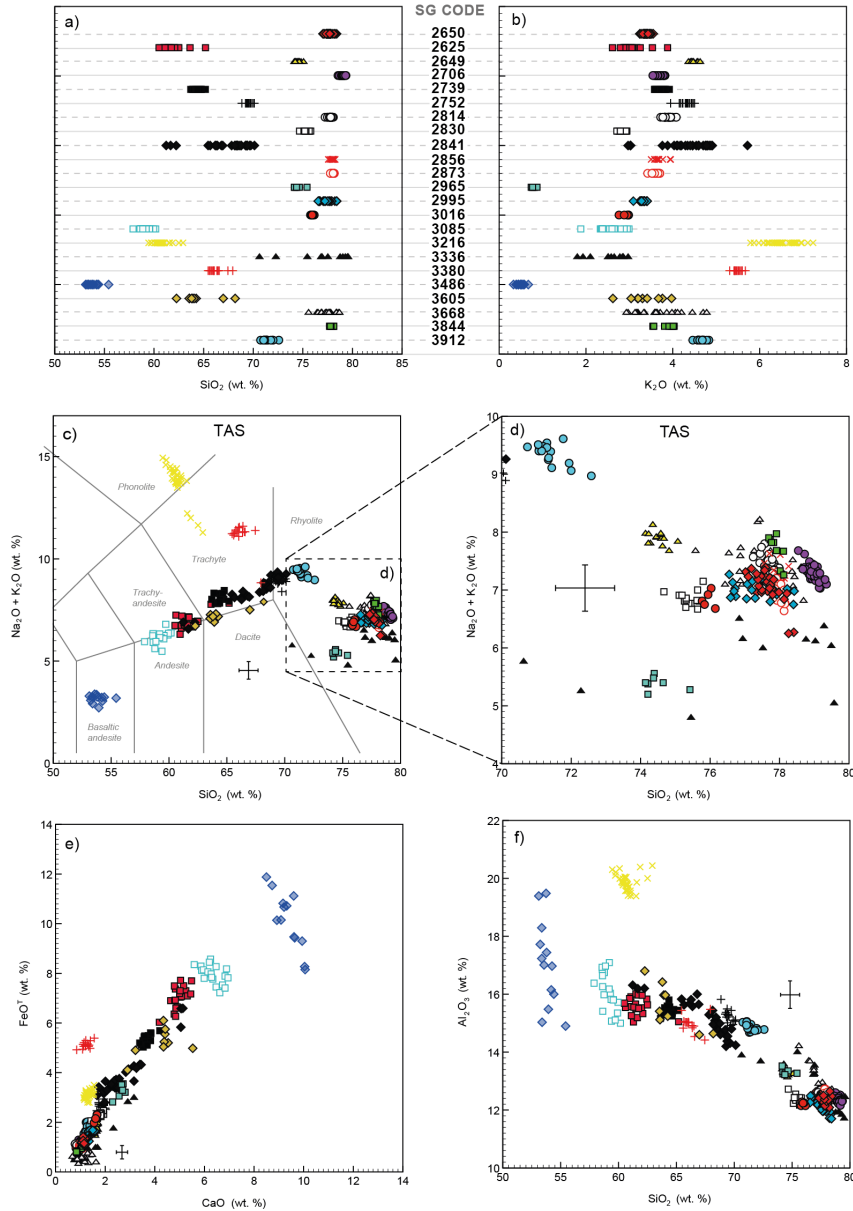


Figure 3. Major element compositions of the tephra layers preserved within the laminated sediments of Lake Suigetsu (SG14 core) between 50 and 30 ka. These are labelled using their SG06/SG14 composite depths. (a, b) SiO₂ and K₂O compositions of tephra layers versus their stratigraphic position. (c, d) Total alkalis versus silica plot (TAS; with whole-rock classification based on *Le Bas et al., [1986]*). (e, f) Bivariate plots for FeO^T, CaO, SiO₂ and Al₂O₃ (error bars represent 2 x standard deviation of repeat analyses of the StHs6/80-G MPI-DING standard glass).

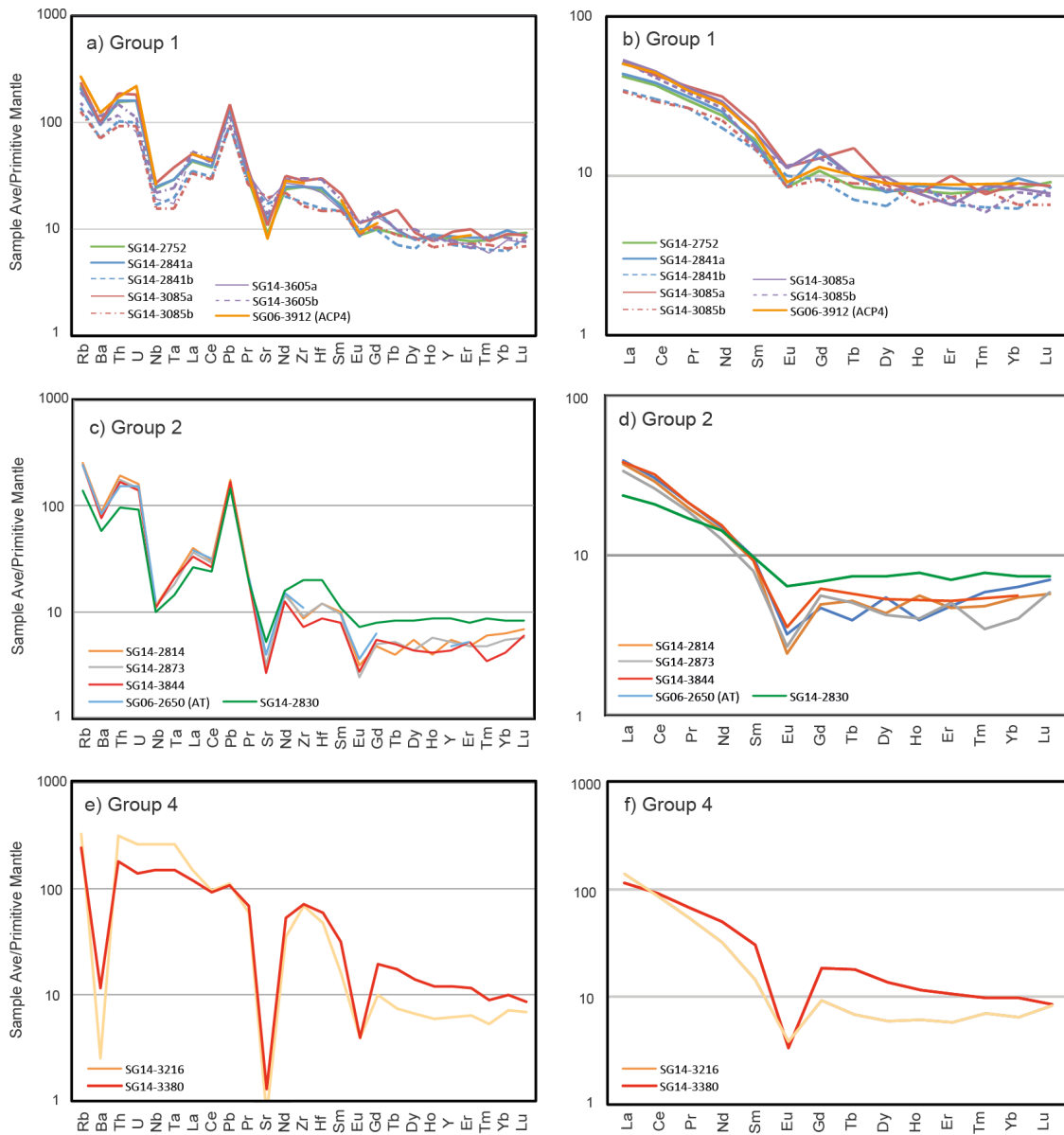


Figure 4. Average Primitive Mantle normalised glass compositions [of *Sun and McDonough* 1989] of the tephra layers (Compositional Groups 1, 2 and 4) preserved within the laminated sediments of Lake Suigetsu (SG14 core) between 50 and 30 ka. Trace element data for SG06-3912 (ACP4) and SG06-2650 (AT) are from *Albert et al.* [2019], and SG14-3216 and SG14-3380 are from *McLean et al.* [2020].

Table 2. Major and trace element glass compositions (normalised) of the newly identified ash layers preserved in the SG14 core between 50 and 30 ka (FeO^T = all Fe reported as FeO). Raw datasets and secondary standards are included in the Supporting Information. Major and trace element data for SG14-3380 and SG14-3216 are from *McLean et al.* [2020].

wt. (%)	SG14-2625		SG14-2649		SG14-2706		SG14-2739		SG14-2752		SG14-2814		SG14-2830		SG14-2841a	
	Avg.	±1σ	Avg.	±1σ	Avg.	±1σ	Avg.	±1σ	Avg.	±1σ	Avg.	±1σ	Avg.	±1σ	Avg.	±1σ
SiO ₂	61.78	1.05	74.51	0.26	78.97	0.16	64.41	0.36	69.54	0.32	77.76	0.19	75.38	0.29	68.14	1.36
TiO ₂	1.21	0.11	0.42	0.07	0.12	0.03	0.97	0.04	0.74	0.05	0.11	0.03	0.60	0.04	0.84	0.06
Al ₂ O ₃	15.56	0.30	13.25	0.30	12.40	0.10	15.45	0.13	15.24	0.22	12.49	0.09	12.40	0.13	15.05	0.54
FeO ^T	6.95	0.57	1.77	0.15	1.14	0.08	5.34	0.21	2.67	0.12	1.04	0.07	2.29	0.10	3.60	0.41
MnO	0.15	0.07	0.04	0.03	0.05	0.04	0.09	0.05	0.12	0.04	0.04	0.02	0.06	0.03	0.12	0.04
MgO	2.11	0.22	0.37	0.04	0.12	0.02	1.56	0.08	0.62	0.06	0.10	0.03	0.45	0.05	0.89	0.23
CaO	4.90	0.42	1.63	0.06	1.03	0.04	3.70	0.19	1.85	0.10	0.90	0.06	1.78	0.13	2.45	0.53
Na ₂ O	3.91	0.16	3.42	0.11	3.62	0.11	4.34	0.13	4.64	0.16	3.54	0.13	4.03	0.11	4.17	0.20
K ₂ O	3.04	0.27	4.48	0.08	3.68	0.07	3.73	0.10	4.29	0.14	3.89	0.08	2.82	0.08	4.51	0.41
P ₂ O ₅	0.41	0.02	0.05	0.01	0.02	0.02	0.41	0.04	0.13	0.02	0.02	0.02	0.09	0.02	0.19	0.06
Cl	-	-	-	-	-	-	-	-	0.14	0.03	0.12	0.01	0.11	0.02	0.11	0.01
total	99.00	0.81	95.74	1.31	95.01	0.62	98.66	0.69	94.68	2.13	95.90	1.03	95.93	1.45	98.11	1.02
n	21		14		48		16		17		33		14		25	
ppm																
Rb	-	-	-	-	-	-	-	-	131.84	6.28	153.31	10.16	82.56	2.97	133.45	28.86
Sr	-	-	-	-	-	-	-	175.86	17.17	54.79	6.53	54.79	101.52	2.55	225.17	106.11
Y	-	-	-	-	-	-	-	34.69	2.40	23.75	2.21	23.75	37.16	1.26	37.04	3.62
Zr	-	-	-	-	-	-	-	260.52	12.63	92.34	7.03	92.34	205.41	7.77	272.92	43.04
Nb	-	-	-	-	-	-	-	15.89	0.87	7.48	0.92	7.48	6.49	0.68	17.01	2.80
Ba	-	-	-	-	-	-	-	623.29	23.55	568.58	56.15	568.58	387.48	14.19	648.38	57.94
La	-	-	-	-	-	-	-	62.48	3.19	51.23	5.00	51.23	39.18	2.18	67.14	8.71
Ce	-	-	-	-	-	-	-	7.36	0.52	5.18	0.64	5.18	4.71	0.37	7.98	1.08
Pr	-	-	-	-	-	-	-	29.94	2.30	18.61	2.58	18.61	19.96	0.69	32.04	5.39
Nd	-	-	-	-	-	-	-	6.85	0.79	3.96	1.56	3.96	4.44	0.45	6.71	2.13
Sm	-	-	-	-	-	-	-	1.30	0.16	0.55	0.29	0.55	1.11	0.29	1.24	0.27
Eu	-	-	-	-	-	-	-	5.60	1.62	2.54	2.75	2.54	4.20	1.42	7.54	2.06
Gd	-	-	-	-	-	-	-	5.51	0.83	3.62	0.73	3.62	5.54	0.87	5.54	1.50
Dy	-	-	-	-	-	-	-	3.29	0.66	2.07	0.63	2.07	3.49	0.48	3.83	0.87
Er	-	-	-	-	-	-	-	3.69	0.41	2.79	0.93	2.79	3.67	0.80	4.33	0.90
Yb	-	-	-	-	-	-	-	6.56	0.68	3.34	0.61	3.34	5.70	0.49	7.25	1.47
Hf	-	-	-	-	-	-	-	1.04	0.21	0.77	0.13	0.77	0.53	0.09	1.17	0.31
Ta	-	-	-	-	-	-	-	12.30	0.68	15.15	1.44	15.15	7.76	0.65	13.47	2.47
Th	-	-	-	-	-	-	-	3.27	0.15	3.26	0.36	3.26	1.83	0.16	3.47	0.82
U	-	-	-	-	-	-	-	2.82	0.20	1.58	0.17	1.58	4.81	0.35	2.79	0.30
Y/Th	-	-	-	-	-	-	-	21.20	0.85	6.12	0.48	6.12	26.57	1.83	20.36	1.24
Zr/Th	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-
n								9		9		9	8		7	

wt. (%)	SG14-2841b		SG14-2856		SG14-2873		SG14-2965		SG14-2995		SG14-3016		SG14-3085a		SG14-3085b	
	Avg.	±1σ	Avg.	±1σ	Avg.	±1σ	Avg.	±1σ	Avg.	±1σ	Avg.	±1σ	Avg.	±1σ	Avg.	±1σ
SiO ₂	61.95	0.40	77.97	0.18	78.00	0.13	74.32	0.20	77.36	0.61	75.95	0.15	67.68	0.68	59.14	0.58
TiO ₂	1.11	0.03	0.11	0.02	0.14	0.03	0.23	0.03	0.35	0.04	0.59	0.02	0.88	0.30	1.05	0.07
Al ₂ O ₃	16.20	0.01	12.33	0.16	12.53	0.12	13.34	0.10	12.14	0.24	12.21	0.06	14.44	0.31	16.06	0.67
FeO ^T	6.20	0.53	1.07	0.11	1.06	0.07	3.31	0.21	1.47	0.16	2.18	0.14	4.39	0.29	7.93	0.38
MnO	0.15	0.01	0.05	0.04	0.03	0.03	0.16	0.06	0.05	0.03	0.10	0.04	0.10	0.04	0.17	0.03
MgO	2.15	0.10	0.12	0.02	0.12	0.02	0.25	0.03	0.27	0.05	0.42	0.03	1.00	0.07	2.78	0.36
CaO	5.06	0.01	1.05	0.09	1.03	0.05	2.69	0.09	1.34	0.15	1.63	0.05	2.89	0.40	6.37	0.40
Na ₂ O	3.70	0.22	3.61	0.10	3.35	0.12	4.61	0.14	3.70	0.15	3.93	0.17	3.88	0.19	3.61	0.29
K ₂ O	3.03	0.01	3.68	0.13	3.60	0.07	0.78	0.03	3.31	0.07	2.91	0.09	4.39	0.12	2.54	0.26
P ₂ O ₅	0.36	0.04	0.02	0.02	0.02	0.02	0.06	0.01	0.02	0.01	0.07	0.02	0.26	0.03	0.29	0.03
Cl	0.08	0.01	-	-	0.12	0.02	0.25	0.04	-	-	-	-	0.09	0.01	0.07	0.02
total	98.18	0.32	95.58	0.75	95.03	1.10	95.18	2.23	95.53	0.89	93.69	1.64	97.89	1.05	97.31	1.55
n	2		14		18		5		20		5		5		20	

ppm	SG14-2841b		SG14-2856		SG14-2873		SG14-2965		SG14-2995		SG14-3016		SG14-3085a		SG14-3085b	
	Avg.	±1σ	Avg.	±1σ	Avg.	±1σ	Avg.	±1σ	Avg.	±1σ	Avg.	±1σ	Avg.	±1σ	Avg.	±1σ
Rb	85.47	-	-	-	144.54	7.06	-	-	-	-	-	-	142.34	4.71	74.48	8.48
Sr	325.70	-	-	-	62.68	4.05	-	-	-	-	-	-	211.75	21.88	409.30	70.34
Y	31.15	-	-	-	21.77	1.56	-	-	-	-	-	-	40.21	3.03	30.52	3.18
Zr	195.62	-	-	-	96.33	3.54	-	-	-	-	-	-	297.08	8.27	167.36	18.16
Nb	11.36	-	-	-	7.48	0.83	-	-	-	-	-	-	17.54	1.58	10.08	0.89
Ba	480.77	-	-	-	523.60	22.10	-	-	-	-	-	-	663.24	35.11	464.95	23.96
La	23.04	-	-	-	24.04	1.45	-	-	-	-	-	-	33.09	2.34	20.80	2.65
Ce	52.20	-	-	-	48.17	2.30	-	-	-	-	-	-	71.68	2.18	48.11	4.20
Pr	6.21	-	-	-	4.81	0.40	-	-	-	-	-	-	8.99	0.22	6.14	0.83
Nd	25.04	-	-	-	17.70	3.68	-	-	-	-	-	-	39.35	2.52	26.13	3.30
Sm	7.29	-	-	-	3.78	1.24	-	-	-	-	-	-	8.55	0.60	6.13	1.12
Eu	1.99	-	-	-	0.37	0.27	-	-	-	-	-	-	1.75	0.20	1.38	0.36
Gd	5.46	-	-	-	2.64	2.01	-	-	-	-	-	-	6.97	0.61	5.31	1.96
Dy	3.97	-	-	-	2.95	0.60	-	-	-	-	-	-	6.21	1.99	5.39	1.18
Er	2.46	-	-	-	2.03	0.45	-	-	-	-	-	-	4.38	0.32	3.07	0.69
Yb	2.94	-	-	-	2.40	0.78	-	-	-	-	-	-	3.96	0.38	2.75	0.77
Hf	4.14	-	-	-	3.40	0.66	-	-	-	-	-	-	8.36	0.74	4.16	0.54
Ta	0.86	-	-	-	0.67	0.17	-	-	-	-	-	-	1.40	0.29	0.59	0.20
Th	8.88	-	-	-	14.28	0.61	-	-	-	-	-	-	15.00	1.78	7.21	0.71
U	2.16	-	-	-	2.94	0.48	-	-	-	-	-	-	3.70	0.12	1.82	0.29
Y/Th	3.51	-	-	-	1.53	0.13	-	-	-	-	-	-	2.69	0.15	4.24	0.31
Zr/Th	22.02	-	-	-	6.76	0.38	-	-	-	-	-	-	19.96	1.94	23.25	1.70
n	1		14		9		5		20		5		3		20	

wt. (%)	SG14-3216		SG14-3336		SG14-3380		SG14-3605a		SG14-3605b		SG14-3844	
	Avg.	$\pm 1\sigma$	Avg.	$\pm 1\sigma$	Avg.	$\pm 1\sigma$	Avg.	$\pm 1\sigma$	Avg.	$\pm 1\sigma$	Avg.	$\pm 1\sigma$
SiO ₂	60.75	0.67	76.89	3.01	66.26	0.70	70.18	0.15	64.51	1.86	77.90	0.16
TiO ₂	0.39	0.07	0.21	0.06	0.60	0.09	0.82	0.03	1.16	0.12	0.10	0.03
Al ₂ O ₃	19.85	0.28	12.77	0.91	14.97	0.28	13.86	0.09	15.58	0.77	12.27	0.11
FeO ^T	3.12	0.16	1.46	0.77	5.12	0.14	3.68	0.17	5.21	0.57	0.85	0.09
MnO	0.18	0.05	0.10	0.04	0.15	0.04	0.10	0.01	0.11	0.04	0.04	0.03
MgO	0.17	0.07	0.85	0.95	0.25	0.06	0.96	0.02	1.54	0.33	0.10	0.02
CaO	1.34	0.12	1.69	0.78	1.24	0.17	2.44	0.08	4.23	0.77	0.91	0.08
Na ₂ O	7.28	0.96	3.34	0.52	5.67	0.66	3.23	0.17	3.88	0.19	3.83	0.09
K ₂ O	6.50	0.33	2.50	0.39	5.49	0.09	4.47	0.01	3.35	0.41	3.87	0.19
P ₂ O ₅	0.05	0.03	0.09	0.03	0.09	0.03	0.31	0.07	0.31	0.07	0.02	0.02
Cl	0.37	0.10	0.11	0.02	0.16	0.02	0.13	0.03	0.13	0.03	0.10	0.01
total	97.03	1.62	95.78	0.88	95.90	1.54	97.02	0.80	96.97	2.72	93.89	0.88
<i>n</i>	37		11		14		2		9		9	

ppm	SG14-3216		SG14-3336		SG14-3380		SG14-3605a		SG14-3605b		SG14-3844	
	Avg.	$\pm 1\sigma$	Avg.	$\pm 1\sigma$	Avg.	$\pm 1\sigma$	Avg.	$\pm 1\sigma$	Avg.	$\pm 1\sigma$	Avg.	$\pm 1\sigma$
Rb	195.42	19.06	-	-	143.55	8.91	123.00	25.59	88.93	3.68	146.33	11.47
Sr	14.31	10.20	-	-	25.01	24.41	273.84	99.60	351.22	31.88	52.25	7.96
Y	26.04	2.94	-	-	51.11	3.94	30.10	5.46	32.64	2.49	18.19	2.52
Zr	709.63	104.31	-	-	741.36	45.81	267.43	78.60	268.35	14.59	74.00	14.87
Nb	175.17	21.16	-	-	96.98	5.52	13.67	1.58	12.65	0.82	7.11	0.54
Ba	16.29	13.34	-	-	77.31	57.26	695.85	75.80	649.66	20.93	508.70	28.90
La	98.17	9.04	-	-	76.42	4.75	34.25	2.72	32.95	0.47	21.73	1.19
Ce	161.66	15.70	-	-	154.34	9.55	71.11	6.93	68.96	5.34	43.75	2.77
Pr	14.46	1.24	-	-	17.05	1.14	8.14	0.94	8.23	0.35	4.56	0.52
Nd	43.92	4.69	-	-	65.57	6.37	33.59	5.09	33.51	3.63	15.62	1.57
Sm	6.41	0.70	-	-	12.91	1.91	5.91	2.74	6.96	0.43	3.22	0.88
Eu	0.59	0.24	-	-	0.61	0.20	1.58	0.36	1.81	0.18	0.41	0.11
Gd	5.45	1.93	-	-	10.69	1.42	7.36	1.12	6.97	2.07	3.01	1.89
Dy	4.44	0.61	-	-	9.26	0.89	5.83	1.28	5.54	1.28	2.88	0.31
Er	2.77	0.62	-	-	5.08	0.73	2.61	0.75	3.41	1.10	2.23	0.25
Yb	3.09	0.70	-	-	4.30	0.59	3.11	0.83	3.71	0.54	1.78	0.23
Hf	13.39	2.09	-	-	16.92	1.24	6.80	1.97	6.40	0.74	2.45	0.58
Ta	9.72	1.09	-	-	5.56	0.46	0.79	0.16	0.64	0.31	0.77	0.22
Th	25.23	3.23	-	-	14.14	1.05	12.17	2.75	9.49	0.88	13.63	1.07
U	5.29	0.86	-	-	2.79	0.34	2.21	0.46	1.64	0.16	2.86	0.21
Y/Th	1.04	0.07	-	-	3.62	0.23	2.59	0.69	3.45	0.26	1.35	0.26
Zr/Th	28.14	2.30	-	-	52.56	3.32	23.48	8.80	28.39	1.67	5.50	1.41
<i>n</i>	8		-	-	10		5		5		6	

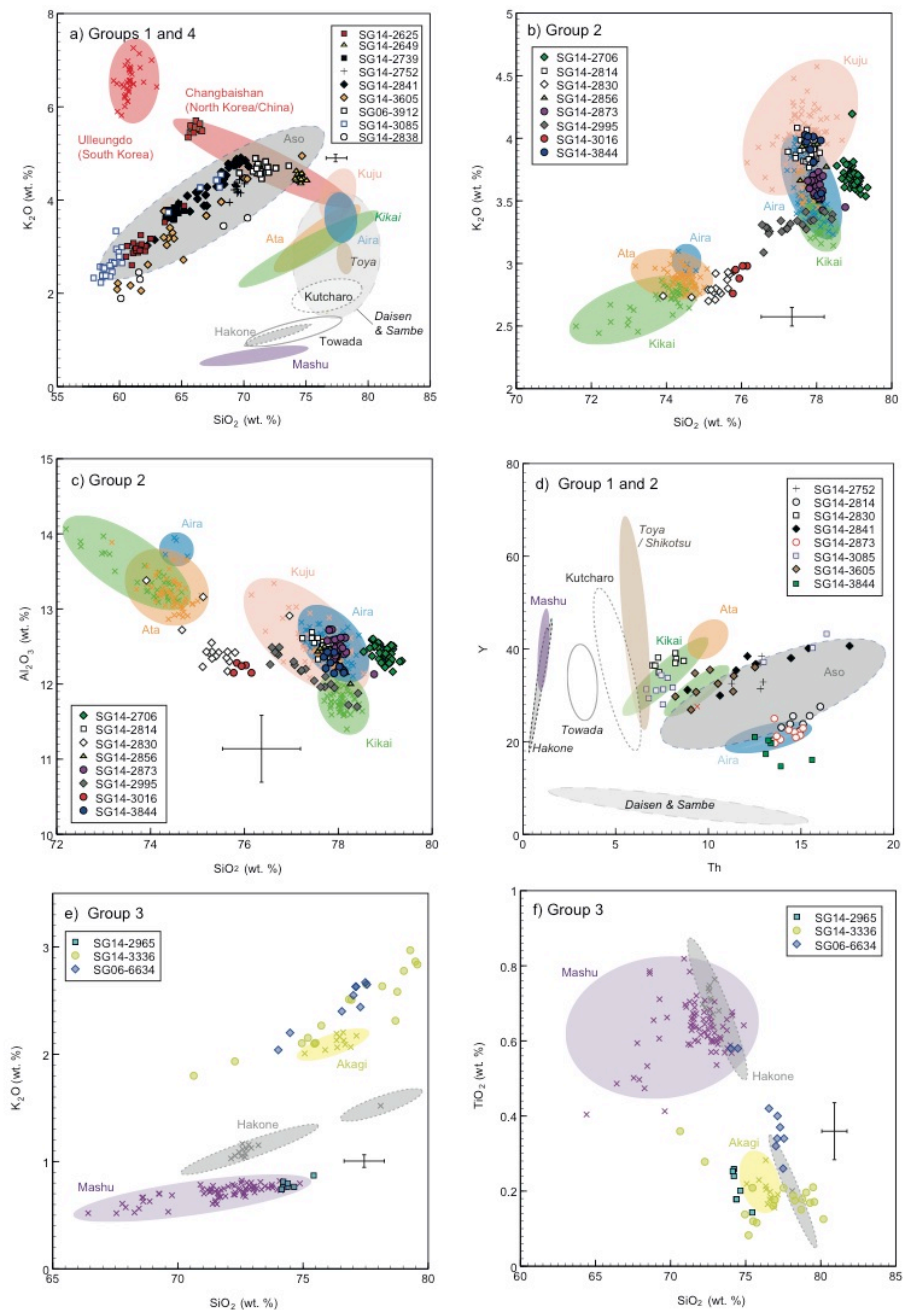


Figure 5. Major and trace element compositions of the SG14 cryptotephra layers (50 to 30 ka) in comparison to those established for major eruptive centres in East Asia. Proximal geochemical fields are shown for Japanese volcanoes [data from *Albert et al.*, 2019], Changbaishan volcano (North Korea/China; *McLean et al.* [2016; 2020]; *Chen et al.* [2016]) and Ulleungdo volcano (South Korea; *Smith et al.* [2011]; *McLean et al.* [2018; 2020]). Error bars represent 2 x standard deviation of repeat analyses of the StHs6/80-G MPI-DING standard glass.

5. Provenance of the Suigetsu cryptotephra identified between 50 and 30 ka

5.1 Geochemically Discriminating Volcanic Sources

Geochemical glass datasets for key widespread ash layers in East Asia are well established [e.g., *Machida and Arai*, 2003; *Okuno et al.*, 2011; *Kimura et al.*, 2015; *Nakamura*, 2016; *McLean et al.*, 2018; *Schindlebeck et al.*, 2018; *Albert et al.*, 2019, and references therein], and their characteristics can be used to help provenance the tephra layers preserved in the Lake Suigetsu sequence.

It is relatively straightforward to distinguish tephra erupted from different arc sources distributed along the length of Japan, which can typically be discriminated by their SiO₂ to K₂O ratios (Figure 5a – b). For instance, Late Quaternary glasses erupted from the Kyushu Central Volcanic Region (CVR; e.g., Aso) and Southern Volcanic Region (SVR; e.g., Aira, Ata, Kikai) typically contain medium to high-K compositions, and are clearly distinguishable at a major element level from tephra deposits erupted at volcanoes along the SW and NE Japan arcs [see *Kimura et al.*, 2015; *Albert et al.*, 2019; Figure 5]. Vitreous tephra deposits erupted from Aso have the highest known K₂O content produced at a Japanese volcano (> 3 wt. % K₂O at 66 wt. % SiO₂), and the most evolved (~70 – 72 wt. % SiO₂) extend to 6 wt. % K₂O, and are therefore some of the most compositionally distinct. However, some centres (e.g., Kikai, Ata and Aira) erupt very compositionally similar tephra over tens to hundreds of thousands of years, with only slight major element variances. Therefore, trace element compositions, which reflect more subtle source characteristics and magmatic processes [e.g., *Allan et al.*, 2008; *Tomlinson et al.*, 2010], can be used to successfully fingerprint these sources. Vitreous tephra derived from all sources in Japan are enriched in LILE (e.g., Rb, Ba, K) relative to REE (e.g., La to Yb) [*Kimura et al.*, 2015; *Albert et al.*, 2019], which is expected given their subduction genesis. Since, LILE concentrations are linked to K₂O, these elements are also very useful discriminators between different arc sources (Figure 5d - f). Furthermore, Y and Th can clearly separate glass erupted from the Kyushu calderas situated in the fore-arc (e.g., Kikai, Ata and Aira) (*Albert et al.* [2019]; Figure 5d). Only intraplate volcanoes Ulleungdo (South Korea) and Changbaishan (North Korea/China) are known to have dispersed alkaline tephtras across Japan during the Quaternary [*Machida and Arai*, 2003; *Lim et al.*, 2013; *Sun et al.*, 2014; *Chen et al.*, 2019; *McLean et al.*, 2020] and are therefore easily discriminated from those originating from Japan and other nearby arc settings.

In summary, the SG14 cryptotephra layers identified are provenanced to the following source regions or volcanoes.

Group 1: Kyushu Central Volcanic Region (CVR), Japan: Aso caldera, (*n* = 8 layers preserved in Lake Suigetsu between 50 and 30 ka)

Group 2: Kyushu Southern Volcanic Region (SVR), Japan: Aira, and other southern Kyushu sources (*n* = 3)

Group 3: Central and northern Honshu/Kurile arc tephtras, (*n* = 2)

Group 4: Ulleungdo (South Korea) and Changbaishan (North Korea/China), (*n* = 2)

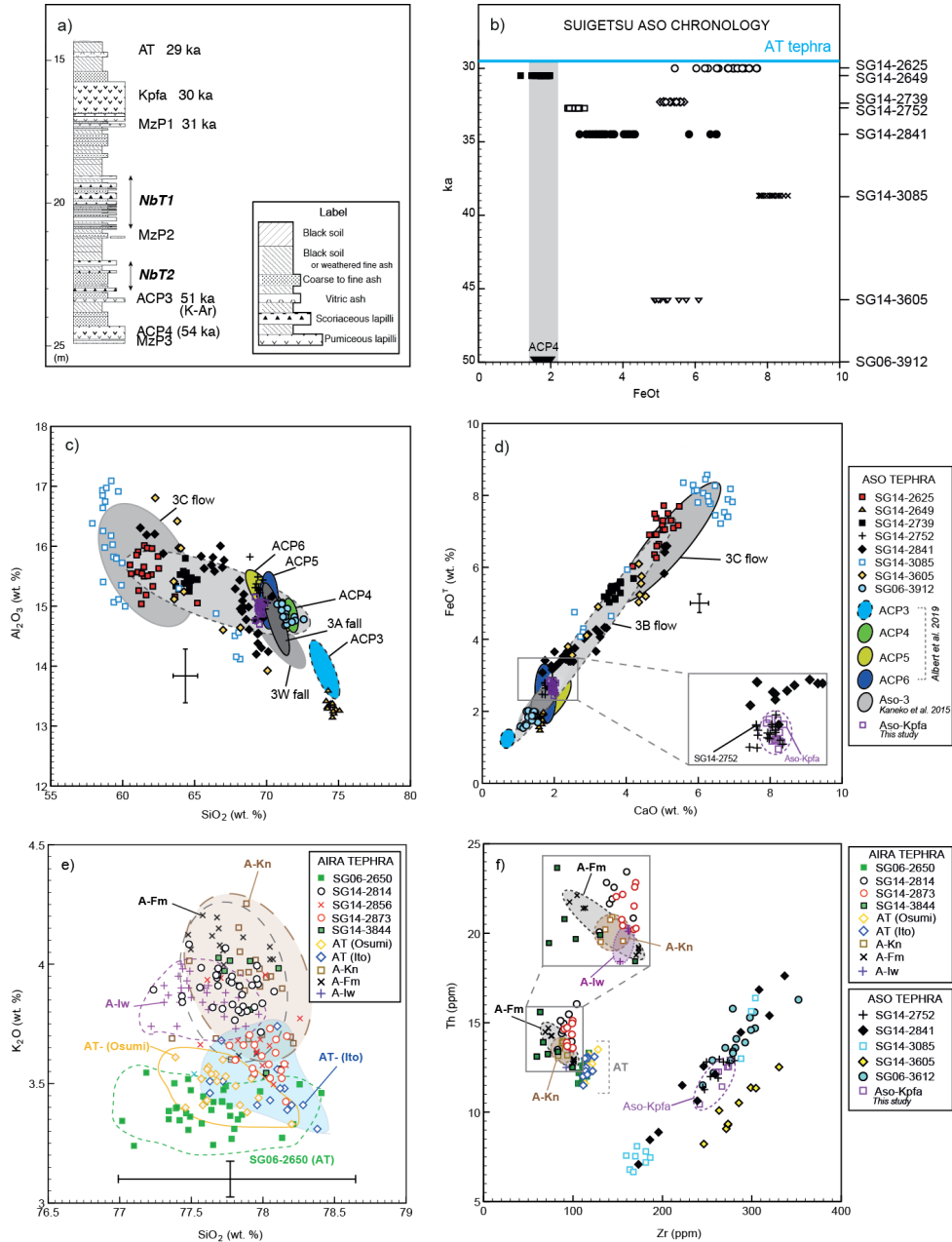


Figure 6. (a) Proximal volcanic stratigraphy compiled at outcrops at Aso caldera [Miyabuchi, 2011], (b) FeO^T content versus Suigetsu age for Aso derived tephra layers, (c – d) Major element glass compositions of compositional Group 2 (Aso derived tephra layers) in comparison to other proximal deposits derived from Aso outcrops [Albert et al., 2019] including fields (in grey) for glass compositions of the Aso-3 (3W, 3C and 3A) event [Kaneko et al., 2015], (e – f) Major and trace element compositions for Aira derived tephra (compositional Group 3) in relation to Aira proximal glass chemistries [Smith et al., 2013; Albert et al., 2019; this study].

These correlations and groups are discussed in detail below, as well as other tephras that have not been correlated to known eruptions or volcanoes. Group 4 tephras that are derived from intraplate volcanic sources west of the Japanese arc (e.g., SG14-3216 and SG14-3380) are comprehensively reviewed by *McLean et al.* [2020], in order to place these eruptions within the known eruptive records of Changbaishan and Ulleungdo volcanoes.

5.2 Group 1: Aso caldera, Kyushu CVR (Japan)

Aso caldera (32°53'04''N, 131°06'14''E) is situated 530 km SW from Lake Suigetsu on the Kyushu CVR arc (Figure 1). Aso is one of the largest active volcanoes on Earth, and the caldera was formed by a series of at least four VEI 6 - 7 eruptions (oldest to youngest): Aso-1 (ca. 270 ka), Aso-2 (ca. 140 ka), Aso-3 (ca. 112 – 134 ka) and Aso-4 (ca. 86 – 87 ka; correlated to SG06-4963 in Lake Suigetsu; *Aoki*, [2008]; *Albert et al.*, [2019]). Post-caldera cones that formed after the Aso-4 eruption cycle, are thought to have produced small, frequent explosive episodes, that have generated deposits over 100 m thick on the eastern side of the caldera [*Miyabuchi*, 2009]. The largest magnitude post-Aso-4 eruptions are associated with the Aso Central Cone [*Miyabuchi*, 2009], the most significant of which producing the Aso Central Cone Pumice 4 (ACP4; VEI 4; *Takada* [1989]) tephra deposit, which is correlated to tephra SG06-3912 (50.3 ka; *Albert et al.* [2019]). This ash layer marks the basal depth of the cryptotephra investigations presented here.

Miyabuchi [2009; 2011] provides details of proximal evidence of other post-caldera forming events that occurred between the deposition of the ACP4 (ca. 50 ka; SG06-3912) and AT ash (ca. 30 ka) (summarised in Figure 6a). Notably, a period of intense silicic activity is estimated to have occurred between 31 and 30 ka, directly following the formation of a thick soil, and prior to the AT eruption [*Miyabuchi* 2009; 2011; Figure 6a]. This activity includes the Aso-Kusasenrigahama pumice (Kpfa; *Watanabe et al.*, [1982]) which is found 1 m below the AT ash in the proximal stratigraphy, and is thought to be the most voluminous of the fall units erupted from the Kusasenrigahama crater (2.21 km³ bulk volume; *Miyabuchi* [2011]). Proximally, the Aso-Kpfa is characterised by six fall units at outcrops to the east of the caldera (E1 – E6), and five units to the west (W1 – W5) [*Miyabuchi et al.*, 2003] implying numerous eruptive phases and a fluctuating dispersal axis [*Miyabuchi*, 2009].

As outlined above, glass compositions from Aso are easily discriminated from other centres across Japan and, importantly, those situated further south in the Kyushu SVR (e.g., Aira, Kikai, Ata) (Figure 5a), due to their clear HKCA affinity. Characteristically, glass erupted from Aso has a wide compositional range, from basalt to rhyolite (Figure 5c - d), which has been sequentially erupted during caldera forming events (e.g., Aso-3 eruption; *Kaneko et al.* [2015]). Whole rock data of the proximal units identified by *Miyabuchi* [2009; 2011] between 67 to 30 ka, suggest that explosive mafic (basaltic to basaltic andesitic; SiO₂ = 48 – 56 wt. %) eruptions were dominant, with infrequent silicic pumice and ash fall events [e.g., Aso-Kpfa; *Ono*, 1989; *Miyabuchi*, 2009]. Detailed proximal volcanic glass datasets derived from the eruptive products of Aso during the 50 to 30 ka timeframe are not yet available, apart from the Aso-Kpfa deposit, which is characterised within this study. We find the Aso-Kpfa glass compositions are exclusively rhyolitic, containing ca. 69.5 wt. % SiO₂ 4.3 wt. % K₂O (Figure 6c – e). Trace element glass compositions are also typical of those erupted from Aso and are relatively

homogeneous (e.g., Th = 11.9 ± 1.6 ppm; Y = 33.8 ± 4.5 ppm) compared to the other characterised Aso units (Figure 6f).

Seven cryptotephra layers identified in the Lake Suigetsu sediments (SG14-2625, SG14-2649, SG14-2739, SG14-2752, SG14-2841, SG14-2997, SG14-3085) have HKCA glass compositions and are characteristic of those derived from Aso (Figure 6b – d). These glasses range from mafic (e.g., SG14-2625), intermediate (e.g., SG14-2841) and silicic (e.g., SG14-2649, SG14-2649) in composition, or in the case of SG14-3085 and SG14-3605, the glasses span the entire compositional range. Trace element signatures of SG14-2752, SG14-2841, SG14-3085 and SG14-3605 are enriched in the LILE (Rb) and HFSE (Th, U, Zr), typical of Aso glasses (Figure 4a). Due to the lack of detailed proximal volcanic glass datasets it is not yet possible to correlate the majority of the identified SG14 cryptotephra to specific units identified in the near source realm. However, here we are able to show that only the glasses of SG14-2752 geochemically overlap those of the newly analysed Aso-Kpfa proximal deposit (Figure 6c, d, f).

The Aso-Kpfa eruption age is constrained at the caldera by its stratigraphic position beneath the AT tephra, as well as several radiocarbon dates obtained from bulk sediment samples of the underlying palaeosols [Miyabuchi, 2009]. Palaeosol ^{14}C ages dated by Miyabuchi, [2009] can be re-calibrated using IntCal13 to 28,720 – 29,615 and 31,110 – 31,940 cal. yrs BP. In contrast, SG14-2752 is dated using the Lake Suigetsu chronology to 32,647 – 32,376 cal. yrs BP (95.4 % confidence interval), which is significantly older than the maximum radiocarbon ages. This means that either; i) SG14-2752 represents a pre-Aso-Kpfa eruption at Kusasenrighama/Aso caldera that has not yet been geochemically characterised at source, or (ii) the bulk soil ^{14}C dates from the palaeosol underlying the Aso-Kpfa deposit are not accurate, and were perhaps contaminated by younger material. One of the calibrated palaeosol ages is younger than the known eruption age of the overlying AT tephra (30,174 – 29,982 cal. yrs BP; Albert *et al.*, [2019]), which provides further support that the radiocarbon measurements of the soil may not be accurate. Using this evidence we tentatively suggest that SG14-2752 represents the distal fallout of at a significant phase of the Aso-Kpfa eruptive sequence that occurred ca. 32.5 ka.

In summary, the Aso-derived tephrostratigraphy preserved at Lake Suigetsu is in agreement with proximal evidence, suggesting that a productive period of activity occurred prior to the AT event. Five separate ash fall events from Aso are preserved in the Lake Suigetsu archive between 35 and 30 ka, indicating that these eruptions dispersed ash over 500 km from source.

5.3 Group 2: Aira and Other Sources South of Suigetsu

5.3.1 Aira, Southern Kyushu (Japan)

Aira caldera is situated at the northern end of Kagoshima Bay in southern Kyushu ($31^{\circ}40'\text{N}$ $130^{\circ}40'\text{E}$; Figure 1). The flooded collapse structure was formed by the enormous VEI 7 eruption that dispersed the AT ash across the majority of Japan [Aramaki, 1984; Machida and Arai, 2003]. The AT eruption ejected approximately 463 km^3 of bulk tephra, and is preserved as a > 30 cm thick sand-sized tephra (SG06-2650) in Lake Suigetsu [Fukushima and Kobayashi, 2000; Machida and Arai, 2003; Smith *et al.*, 2013]. This eruption is most precisely dated using the Lake Suigetsu chronology to 30,174 – 29,982 IntCal13 yrs BP (95.4 % confidence interval);

Albert et al. [2019]) and forms the upper boundary of the cryptotephra record presented here. The caldera-forming event began with the Plinian eruption (Osumi pumice fall), which was followed by the Tsumaya and Ito pyroclastic flows (Aramaki, 1984).

In the context of this timeframe (50 – 30 ka), outcrops from around Aira indicate that the centre produced three pyroclastic successions between the 58 ka A-Iwato formation (A-Iw; VEI 6) and the 30 ka AT (VEI 7). These successions are separated by well-developed palaeosols, and include the ca. 33 ka Otsuka pumice fall deposit (A-Ot; VEI 4), the ca. 31 ka Fukaminato tephra formation (A-Fm; VEI 5) and the ca. 30 ka Kenashino tephra formation (A-Kn) [*Machida and Arai, 2003*]. *Nagakoa et al.* [2001] constrained the age of these eruptions using bulk sediment ¹⁴C ages of palaeosols between the A-Ot, A-Fm and A-Kn units. Here, we incorporate these radiocarbon dates within an OxCal Bayesian phase model [ver. 4.3.2; *Bronk Ramsey, 2017*] to generate calibrated age estimates for these events. This model is further constrained by inserting the Suigetsu-derived AT eruption age, which is known to post-date these deposits. Using this phase model, the A-Ot eruption is dated to 33,216 – 30,730 IntCal13 yrs BP, A-Fm to 30,933 – 29,865 IntCal13 yrs BP, and A-Kn to 30,437 – 29,545 IntCal13 yrs BP (95.4 % confidence interval).

Importantly, volcanic glass from Aira can be distinguished from other sources distributed along the Kyushu SVR arc by their lower Y/HREE and higher Th contents (Figure 5d; *Albert et al.* [2019]). Glasses erupted from Aira are typically homogenous and trace element compositions are required to robustly distinguish tephra from sequential eruptions. Indeed, *Albert et al.* [2019] showed that glass compositions of the A-Iw eruption (ca. 58 ka) are subtly elevated in K₂O (ca. 0.3 wt. %) compared to ash erupted during the 30 ka AT event, but could be more confidently discriminated using concentrations of the HFSE. Specifically, the older A-Iw glasses displayed lower Zr content than the AT glasses, most likely relating to a greater degree of zircon fractionation (Figure 6f). Here, geochemical characterisation of the intervening Aira eruption units (A-Kn and A-Fm) indicate that they also display the same offsets with respect to the AT eruption deposits, again with ca. 0.4 wt. % higher K₂O (Figure 6e), and lower Zr content than the AT tephra. The A-Fm trace element compositions are more heterogeneous in comparison to those of the A-Kn and the older A-Iw unit, with Zr ranging from 70 – 100 ppm (Figure 6f).

Four cryptotephra layers in the Suigetsu sediments, SG14-3844, SG14-2873, SG14-2856 and SG14-2814, have compositions which are distinctive of those erupted from Aira (Table 1; Figure 5). The trace element compositions of glass from these four cryptotephra layers are more subtly depleted in Zr and more enriched in Th, compared to the AT (Oshumi and Ito) tephra, and are generally more consistent with those of A-Kn, A-Fm and A-Iw (Figure 6f). Whilst, subtle offsets exist between the trace element compositions of the proximal Aira deposits (A-Kn and A-Fm; Figure 6f) and the distal SG14 ash layers, this is attributed to slight differences in the analytical setup between the two instruments used. Indeed, by correcting the proximal and distal Aira trace element glass data based on differences between the measured and known contents of the StHs6/80-G reference glass, routinely measured on the respective instruments, the chemical similarity between the proximal and distal glasses is clear.

Since these pre-AT tephra units from Aira are compositionally similar we must use the chronostratigraphy to facilitate correlations. Since SG14-2856 and SG14-2873 are separated by less

than 400 years using the Lake Suigetsu varve chronology, then it is possible that their near-source counterparts would not be separated by a prominent palaeosol. Therefore, it is possible that in the proximal stratigraphy these would be interpreted as separate phases of one of the pre-AT eruptive formations (A-Kn, A-Fm or A-Ot). Based purely on stratigraphic grounds and the eruptive formations recognised at the volcano [*Nagako et al. 2001*], we tentatively suggest that SG14-2814 relates to the A-Kn formation, SG14-2856/SG14-2873 relates to the A-Fm formation, and SG14-3844 is the distal equivalent of the A-Ot formation. Whilst these correlations are not entirely consistent with the existing ^{14}C dates of the palaeosols between the A-Kn, A-Fm and A-Ot formations [*Nagako et al. 2001*], it is possible that the ^{14}C dates are erroneously young. This is supported by the fact that the paleosol age of the A-Kn deposit (30,437 – 29,545 cal. yrs BP), overlaps that of the known age of the AT eruption (30,174 – 29,982 cal. yrs BP; *Albert et al. [2019]*), despite these units being separated by a well-developed palaeosol in the proximal sequence. Such soils are also developed between the underlying eruptive units in a period of just 3 ka.

5.3.2 Uncorrelated Tephra from Kyushu

Tephra layers SG14-2830 (ca. 34.2 ka), SG14-2995 (ca. 37.4 ka) and SG14-3016 (ca. 37.8 ka) are also attributed to explosive volcanism on the Kyushu arc (Figure 5b – c). Considering the compositional overlap, and equivalent shard textures (platy, bubble-walled shards), SG14-2830 and SG14-3016 at least are highly likely to originate from the same source. Trace element compositions obtained for SG14-2830 are less depleted in Nb, Sr and Eu compared to glass erupted from Aira, and therefore more consistent with deposits from Ata and Kikai that are elevated in HFSE (Figure 4c – d; *Albert et al., [2019]*). Ata in particular is known to have been active between 50 and 30 ka [*Machida and Arai, 2003; Nagaoka et al., 2001*] but further proximal samples are required to build geochemical datasets and secure a correlation for these events.

5.4 Group 3: Uncorrelated Tephra from Central and Northern Japan

5.4.1 Uncorrelated Tephra from Central Honshu

SG14-3336 (ca. 42.0 ka) is compositionally similar to the older visible tephra layer SG06-6634 (ca. 130 ka) identified in the Suigetsu core [*Smith et al., 2013; Albert et al., 2019*], although glass compositions are ca. 0.5 wt. % lower in FeO^{T} . Heterogeneous and medium-K series eruptions are known from stratovolcanoes in central Honshu, including Akagi, Haruna and Asama (Figure 5e – f). Chronologically, the ca. 45 ka eruption of Haruna (Haruna-Hassaki ash; Hr-Hs) and the ca. 44 ka eruption of Akagi (Akagi-Kanuma; Ag-KP) are possible correlatives [*Suzuki, 1996; Geshu and Oishi, 2011*]. However, proximal samples obtained from these units do not span the entire geochemical range of SG14-3336, precluding a robust correlation at this time (Figure 5e – f).

5.4.2 Uncorrelated Tephra from Hokkaido and the Kurile arc

SG14-2965 (ca. 36.9 ka) glass shards are very distinctive and contain < 1 wt. % K_2O , suggesting that this deposit was erupted from a source from NE Hokkaido or Kurile arc. The only known low-K source to have dispersed ash to central Honshu is Mashu (Ma-b tephra), situated in eastern Hokkaido [*McLean et al., 2018*]. However, SG14-2965 glass compositions can be distinguished

from Mashu tephra (e.g., Ma-b and older deposits), by CaO (ca. 0.5 wt. % lower) and other minor elements including TiO₂ (Figure 5e – f).

6. Discussion

6.1. Tempo, Origin and Dispersal of Ash Fall Events Reaching Central Honshu

The augmented Lake Suigetsu tephrostratigraphic record (50 to 30 ka) indicates that, on average, an ash fall event reached central Honshu at least once every 1,300 years (Figure 7). However, between 39 and 30 ka the average return interval was significantly higher, with at least one eruption recorded every ca. 500 years, which is in close agreement with that observed during the Holocene [McLean *et al.*, 2018]. The record is dominated by ash fall events erupted from sources south of Suigetsu on the SVR and CVR ($n = 18$; Figure 7), and most of which ($n = 8$) are derived from explosive activity from Aso (CVR).

Four eruption events are identified from different arc regions in Japan (e.g., central Honshu) and other nearby volcanic settings (Changbaishan and Ulleungdo), which highlights that other sources also have the potential to disperse ash more widely than previously anticipated. Interestingly, these eruption events are clustered between 45 and 39 ka, when no other ash layers are recorded from productive centres southwest of Suigetsu. However, it is not possible to attribute the cause of this change, due to the number of controlling parameters and preservation biases that affect the presence of cryptotephra layers. For example, this may represent a change in eruption styles and mechanisms (e.g., explosivity; efficient fragmentation of magma; Cassidy *et al.*, [2018]) or external differences during eruptions (e.g., wind direction and strength; Carey and Sparks [1986]).

Several volcanoes in northern Honshu and Hokkaido are known to have been very active between 50 and 30 ka (e.g., Towada, Shikotsu), yet distal ash deposits from these large magnitude events (VEI 5 – 7) have not been identified in this study of the Suigetsu sediments (Figure 7). It is possible that they are preserved as lower concentrated peaks of glass shards in the sediments, or are hidden in the reworked ‘tails’ that follow visible tephra layers. Identifying these markers would be of significant value for tephrochronology, and help to further constrain their eruption histories. The identification of other primary cryptotephra peaks in the sediments may be achieved through detailed studies of shard morphologies, supplemented by further geochemical analysis. This was shown to be possible in the Holocene sedimentation, where the distinctive vesicular To-Cu (Towada) shards were clear within the platy K-Ah remobilised glass [McLean *et al.*, 2018].

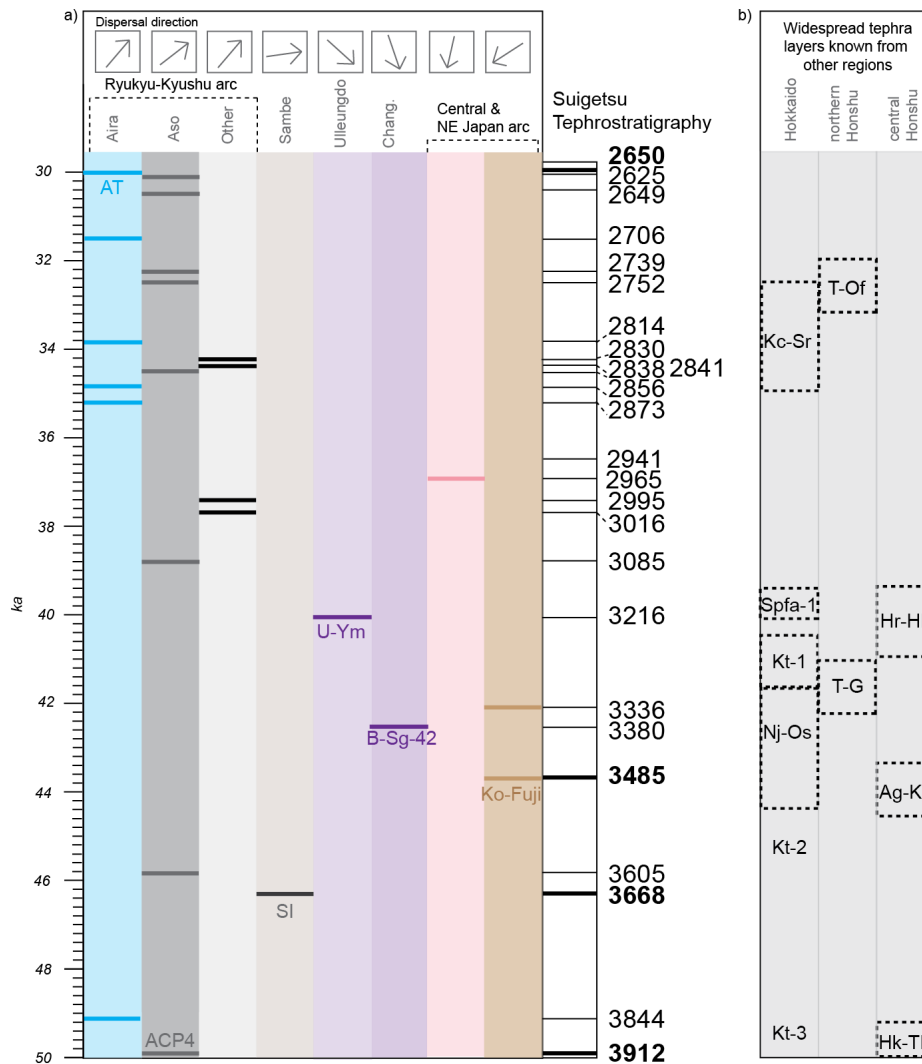


Figure 7. (a) Summary of the augmented Lake Suigetsu (SG14) tephrostratigraphy (visible and cryptotephra layers) spanning 50 to 30 ka. The distal tephra layers are correlated to their volcanic sources and, where possible, specific eruptions, using the compositions of the volcanic glass. The Suigetsu sediments preserve evidence of a significant period of activity from centres on the Ryukyu-Kyushu arc (e.g., Aira and Aso) prior to the AT caldera-forming eruption dated to ca. 30 ka. (b) The timing of other key widespread and large magnitude (VEI 7 – 5) eruptions known from other regions north of Lake Suigetsu, which were not identified in the sediments (data from Machida and Arai, 2003). Approximate eruption ages are shown by dashed boxes, as reported by Suzuki [1996]; Machida and Arai [2003]; Yamamoto *et al.* [2010]; Geshu and Oishi [2011]; Uesawa *et al.* [2016]; Matsu'ura *et al.* [2017].

The Japanese cryptotephra layers identified within the sediments of Lake Suigetsu are the first known distal occurrences of these eruption events. Therefore further ash deposits between Suigetsu and their sources (e.g., in other high-resolution lake and marine cores) are required to better constrain the magnitude of these events, and integrate other eruption stratigraphies. Nevertheless, it is clear that these must have had a VEI of at least 4, in order to have been dispersed as far as Lake Suigetsu (located 300 km from the nearest potential source) and have been preserved as a cryptotephra layer. Cryptotephra ash fall events are likely to be thin and perhaps of limited consequence so far from source, but such fine ash (typically < 80 μm) may have implications for air traffic, air and water quality and critical infrastructure [e.g., *Horwell et al.*, 2006].

6.2. Constrained eruption chronology for Aso and Aira

The Suigetsu tephrostratigraphy provides detailed insight into the behaviour of two of the most active calderas in Japan, Aso and Aira, which are central to hazard assessments for Japan and even possible candidates for the next VEI 7 eruption [*Newhall et al.*, 2018]. Here, we confirm that both centres were very active between 50 and 30 ka (as informed by their proximal stratigraphies), producing moderate to large explosive events within short temporal scales (ca. 300 years for both). Almost all of these eruptions have yet to be the subject of detailed petrological investigations, so the glass compositions, and constrained eruption ages allow a new insight and comparisons into the magma generation and storage pre- (Aira) and post- (Aso) caldera-forming events.

Prior-to the AT eruption (VEI 7; *Machida and Arai*, [2003]) of Aira, the Suigetsu record indicates that at least four large eruption events occurred and are dated to ca. 33.8 ka, 34.8 ka, 35.2 ka and 49.2 ka (Table 1; Figure 7). The Suigetsu chronology also indicates that these eruptions occurred as frequently as 3,600 years before the enormous AT event. These erupted characteristically high silicic glass compositions (75.5 – 78.9 wt. % SiO_2) that are compositionally similar on major elements [*Machida and Arai*, 2003; *Albert et al.*, 2019], but we show that they can be geochemically distinguished from the AT ash using their trace element compositions (e.g., Zr, Y, Th).

The Suigetsu sediments indicate that Aso erupted compositionally different magmas through time, ranging from rhyolitic to andesitic in composition, and producing at least eight widespread tephra units over 20 ka (Table 1; Figure 7). Furthermore, Aso was also very active prior to the AT eruption at ca. 30 ka, with at least one eruption every 600 years between ca. 32.6 and 30.3 ka. The bulk volumes estimated for the Aso eruption events [*Miyabuchi*, 2009; 2011] in some cases are small, yet they are preserved over 600 km from the vent. This could imply that perhaps the eruption volumes are underestimated, the transport mechanism is highly efficient, and/or the eruption generated very fine ash suitable for widespread ash dispersal.

7. Conclusions

New insights into the frequency and timing of East Asian volcanism between 50 and 30 ka are elucidated through the identification of cryptotephra layers preserved in the laminated (varved) sediments of Lake Suigetsu (Japan). Four times more microscopic ash (cryptotephra) layers are

preserved relative to visible layers, meaning that this tephrostratigraphy provides an unprecedented record of ash that has been dispersed over central Honshu. Major and trace element compositions of the glass shards allow most of these distal tephras to be correlated to their volcanic source, however additional datasets from proximal outcrops are pivotal to integrate these records and learn more about these individual eruption events.

Of the twenty-five ash layers presented in the augmented tephrostratigraphy, nineteen of these were dispersed from eruptive centres southwest of Suigetsu, which is unsurprising considering the prevailing westerly winds. However, the record indicates that other sources have the potential to reach central Honshu, including Ulleungdo (500 km NW of Suigetsu) and Changbaishan (1000 km NW). Explosive activity from Aso and Aira are most frequently recorded in the record, and indicate that these calderas were very active between 35 and 30 ka. Aso erupted compositionally different magmas through time, producing eight widespread events over 20 ka. Here, we also show that Aira erupted explosively 1.3 ka before the catastrophic VEI 7 caldera-forming eruption that blanketed Japan in the AT ash.

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