

REFINING THE ERUPTIVE HISTORY OF ULLEUNGDO AND CHANGBAISHAN VOLCANOES (EAST ASIA) OVER THE LAST 86 KYRS USING DISTAL SEDIMENTARY RECORDS

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Highlights:

- Distal records show eruptions are more frequent and widespread
- At least 8 Changbaishan eruptions produced widespread ash over the last 86 kyrs
- Explosive eruption of Changbaishan at ca. 42.5 ka dispersed ash >1000 km

- 4 Ulleungdo eruptions are now precisely dated using the Lake Suigetsu chronology
- U-Ym tephra is identified in Suigetsu and dated to 40,332 – 39,816 IntCal13 yrs BP

Abstract

The eruptive histories of Ulleungdo (South Korea) and Changbaishan (North Korea/China border) volcanoes are not well constrained since their proximal stratigraphies are poorly exposed or largely inaccessible. However, determining the past behaviour of these volcanoes is critical since future eruptions are likely to disperse ash over some of the world's largest metropolitan regions. Alkaline tephra deposits erupted from both centres are routinely identified in marine cores extracted from the Sea of Japan, as well as high-resolution lacustrine records east of the volcanoes. Here, we review the distal ash occurrences derived from Ulleungdo and Changbaishan and provide new data from the Lake Suigetsu (central Honshu, Japan) sediment core, in order to provide a more complete and constrained eruption framework. The intensely-dated Lake Suigetsu archive provides one of the most comprehensive distal eruption records for both centres, despite being located ca. 500 km E of Ulleungdo and ca. 1000 km SSE of Changbaishan. The Suigetsu record is utilised to precisely date and geochemically fingerprint (using major, minor and trace element glass compositions) ash fall events that reached central Honshu. Here, we identify a new non-visible (cryptotephra) layer in the Suigetsu sediments, which reveals a previously unreported explosive event from Changbaishan at 42,750 – 42,323 IntCal13 yrs BP (95.4 % confidence interval). This event is chronologically and

geochemically distinct from the B-J (Baegdusan-Japan Basin) tephra reported in the Sea of Japan (ca. 50 ka). Furthermore, we also confirm that the widespread U-Ym tephra erupted from Ulleungdo reached central Japan, and is herein dated to 40,332 – 39,816 IntCal13 yrs BP (95.4 % confidence interval). This terrestrial ¹⁴C-derived age of the U-Ym can be used to constrain the chronology of marine records containing the same marker layer. This reviewed and integrated tephrostratigraphic framework highlights the pivotal role that distal sedimentary records can play in evaluating the eruptive histories and hazard potential of Ulleungdo and Changbaishan.

Keywords: Ulleungdo, Changbaishan; Glass geochemistry; Eruption history; Sedimentary archives; Lake Suigetsu

1. Introduction

Intraplate volcanoes Ulleungdo (South Korea) and Changbaishan (North Korea/China border) are responsible for two of the largest Holocene eruptions (\geq Volcanic Explosivity Index (VEI) 6; Newhall and Self, 1982) in East Asia, blanketing large parts of Japan and the surrounding seas in ash (Figure 1; Machida and Arai, 2003). Fine ash from the AD 946 'Millennium Eruption' (ME) (Hakozaki et al., 2017; Oppenheimer et al., 2017) of Changbaishan has also been identified ca. 9000 km from its source in northern Greenland (Sun et al., 2014a), which demonstrates the enormous potential of the volcano to cause major disruption to airspace across the East Asian and Pacific region. Yet, the complete eruptive histories of Ulleungdo and Changbaishan are not well

constrained since proximal eruption deposits are poorly exposed and are largely inaccessible.

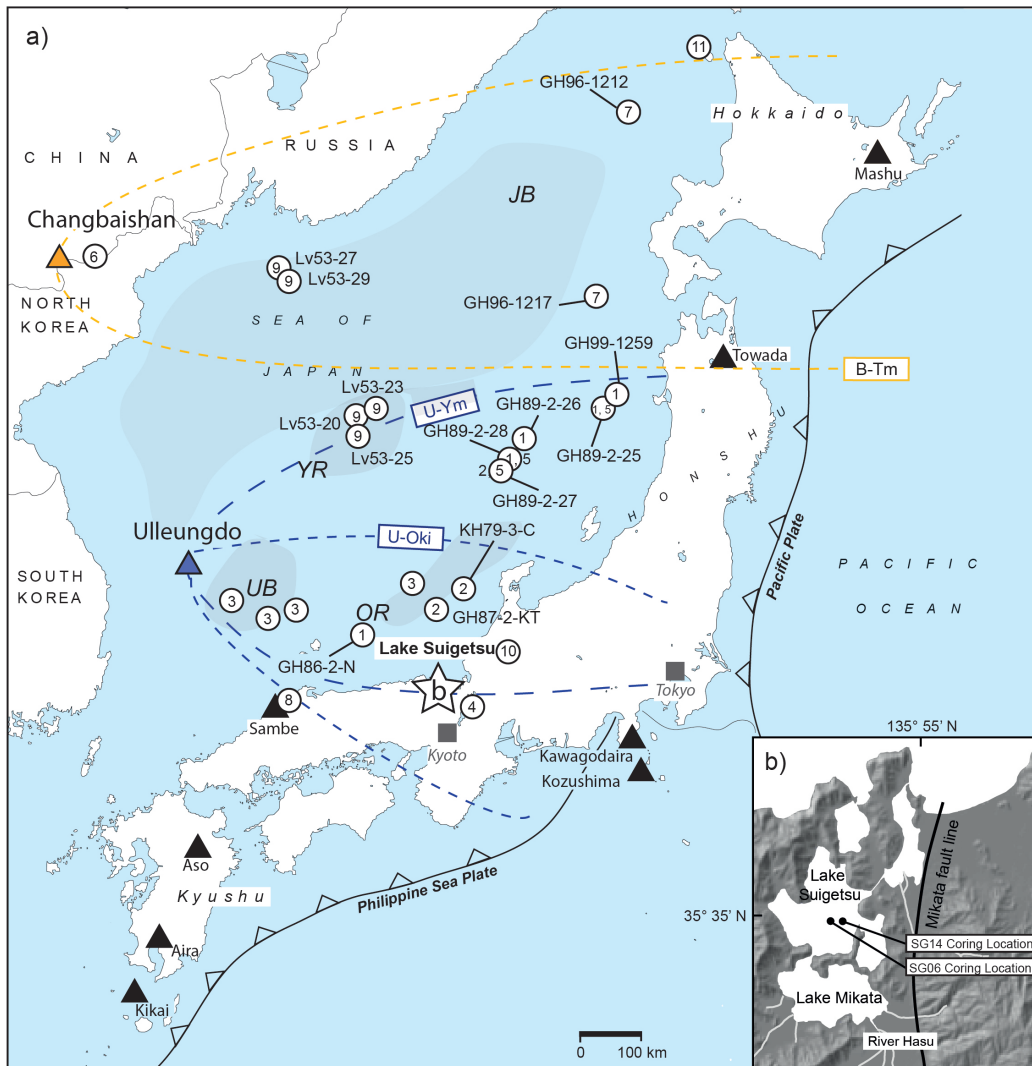


Figure 1. (a) Location of Ulleungdo (South Korea; blue triangle) and Changbaishan (North Korea/China; orange triangle) and other sources of Japanese tephras outlined in the text (black triangles). Distal sites mentioned in the text are marked by white circles; 1 = Marine cores; Lim et al. (2013); 2 = Marine cores; Arai et al. (1981); Chun et al. (2007); 3 = Marine cores; Chun et al. (2007); 4 = Lake Biwa; Nagahashi et al. (2004); 5 = Marine cores; Ikehera et al. (2004); 6 = Yuanchi Lake; Sun et al. (2018); 7 = Marine cores; Ikehara (2003); 8 = Lake Hane; Sawada et al. (1997); 9 = Marine cores; Derkachev et al. (in press); 10 = Hakusan volcano; Higashino et al. (2005); 11 = Lake Kushu; Chen et al. (2016, 2019). A white star notates the location of Lake Suigetsu, and ocean basins are marked in grey (JB= Japan Basin; YR= Yamato Rise; OR= Oki Ridge; UB = Ulleungdo Basin). Dispersal boundaries of the B-Tm (AD 946; Oppenheimer et al., 2017), U-Oki (ca. 10 ka) and U-Ym (ca. 40 ka) are marked by dashed lines (B-Tm and U-Oki as defined by Machida and Arai, 2003). (b) Location of Lake Suigetsu, which is the largest of the five Mikata lakes, adjacent to Wakasa Bay. The positions of coring campaigns SG06 and SG14 are marked on Lake Suigetsu (modified after Nakagawa et al., 2005).

It is likely that proximal evidence of older eruptions, especially those of low- to mid-intensity, have been destroyed, or are now completely buried, following more recent large magnitude Holocene events. At Changbaishan clear depositional breaks and soil horizons are not well documented within proximal eruption successions (e.g., Chen et al., 2016; Sun et al., 2017), making it unclear how many eruption deposits are preserved.

Distal sedimentary records (e.g., marine and lacustrine sequences) have proved very important archives of past explosive eruptions, and can be used to help constrain the frequency and dispersal of tephra-forming events (e.g., Wulf et al., 2004; Albert et al., 2013; 2018; Smith et al., 2013; Tomlinson et al., 2014; Ponomareva et al., 2018). Ulleungdo and Changbaishan are the only sources known to have dispersed alkaline tephra across Japan (Machida and Arai, 2003; Kimura et al., 2015; Albert et al., 2019), and their distal deposits can be easily discriminated from other intraplate sources in the back-arc (e.g., Doki and Jeju volcanoes; Brenna et al., 2014). Tephra layers preserved in marine cores extracted from the Sea of Japan (Oki ridge, Yamato and Japan basins; Figure 1), indicate that both Ulleungdo and Changbaishan have been very active during the Late Quaternary, however the number and precise timing of these events remains uncertain. This is partly since successive eruption deposits are difficult to geochemically distinguish, and because marine cores in some localities are susceptible to reworking processes (e.g., turbidites; Albert et al., 2012; Cassidy et al., 2014), and often cannot be precisely dated (i.e., due to variations in the marine radiocarbon reservoir; Ikehara et al., 2013).

In order to provide new insight into the eruptive histories of Ulleungdo and Changbaishan, this study provides a detailed review of the distal occurrences of alkaline ash deposited in sedimentary records (marine and lacustrine cores) spanning the last 86 kyrs (i.e., post-dating the widespread Aso-4 tephra that is dated to 86.4 ± 1.1 ka using the $^{40}\text{Ar}/^{39}\text{Ar}$ method; Albert et al., 2019). We also provide new tephra data from the intensely-dated Lake Suigetsu archive (central Honshu, Japan), a record that has significant potential to develop a comprehensive eruption history for both centres (despite being located 500 km E of Ulleungdo and 1000 km SSE of Changbaishan). Using the lake sediments, we identify and geochemically characterise two new ash layers erupted from Ulleungdo and Changbaishan, allowing these eruptive events to be precisely dated for the first time. New trace element data are also generated for the previously identified marker layers preserved as cryptotephra in the Holocene sediments (McLean et al., 2018), offering new possibilities to discriminate between successive eruption deposits. This reviewed and integrated distal eruption framework for Ulleungdo and Changbaishan permits critical new insight into the hazard potential of these active centres.

2. Regional setting and proximal volcanic deposits

2.1. Ulleungdo Island, South Korea

Ulleungdo Island (12 km x 10 km) is the sub-aerial portion of a Quaternary stratovolcano located in the mid-western part of the Sea of Japan ($37^{\circ}30'\text{N}$, $130^{\circ}52'\text{E}$), 130 km east of the Korean Peninsula (Figure 1; Kim, 1985).

Ulleungdo is the youngest volcano in the back-arc basin, and is known to have erupted intermittently from the Pliocene until the mid-Holocene (Kim et al., 1999; Okuno et al., 2011; Im et al., 2012). Nari caldera is located at the centre of the island (2.8 km in diameter) and is the source of the most recent phase of activity (< 19 ka; Kim et al., 2014), erupting rocks that range from alkali basalt to trachyandesite in composition (Kim, 1985; Brenna et al., 2014; Chen et al., 2018).

The most recent activity of Ulleungdo is exposed at several outcrops near or within Nari caldera. Machida et al. (1984) defined seven pyroclastic units at extra-caldera outcrops in the north (named in ascending order: U-7 to U-1), which are comprised of trachytic or phonolitic ash and pumice that were emplaced as fall deposits and/or by pyroclastic flows (Figure 2). The Holocene stratigraphy (U-4 to U-2) was further subdivided by Okuno et al. (2011) and Shiihara et al. (2011) at exposures in the southeast, where the units have also been geochemically analysed and radiocarbon dated (Figure 2). Two widespread Japanese tephra marker layers erupted from volcanoes of southern Kyushu Island are found within the soils that formed between these pumice falls, and are named the Aira-Tanzawa (AT; ca. 30.0 ka) and Kikai-Akahoya (K-Ah; ca. 7.3 ka) ash. The U-7 to U-5 eruption units are stratigraphically identified below the AT tephra, and the K-Ah ash is positioned between the U-3 and U-2 eruptions (Shiihara et al., 2011). Radiocarbon dates obtained from buried soils (ca. 2 cm thick) between units, and charred tree material preserved in the Holocene deposits suggest that the U-3 eruption occurred ca. 8.3 or 9 ka BP, respectively, and the U-2 eruption at ca. 5.6 ka BP.

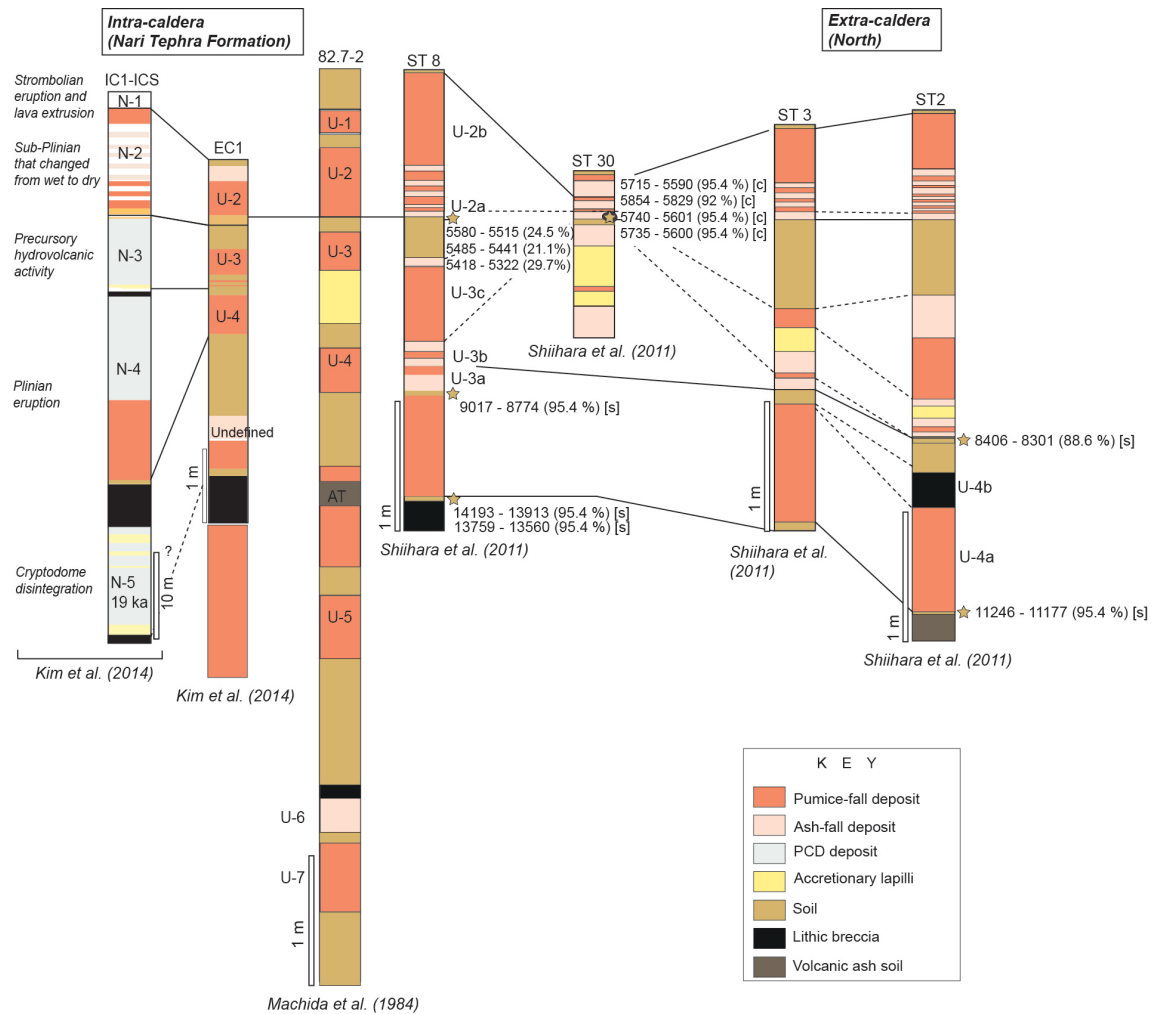


Figure 2. Sedimentological and stratigraphic characteristics of the intra- and extra-caldera outcrops on Ulleungdo Island (Machida et al., 1984; Okuno et al., 2010; Shiihara et al., 2011; Kim et al., 2014). The radiocarbon ages (s = soil; c = charcoal) reported by Okuno et al. (2010) have been recalibrated using IntCal13 (IntCal13 yrs BP). Two Japanese tephra layers erupted from volcanoes in Kyushu are identified within the soils of the extra-caldera sequences, which include the AT (30 ka) and K-Ah (7.3 ka) ash.

Major element glass compositions for Holocene eruptions U-4 to U-2 are known to be geochemically similar (Machida et al., 1984; Martin Jones, 2012; Shiihara et al., 2011). Slight geochemical differences between some subunits are reported by Shiihara et al. (2011), who show that U-4a and U-3c contain glass with lower Al_2O_3 and higher CaO and FeO^T . Furthermore, subunits U-3a and U-

2a are characterised by slightly lower CaO, TiO₂ and FeO^T contents compared to the other units.

Intra-caldera outcrops at Nari are ca. 70 m thick, and are composed of unwelded pyroclastic and epiclastic deposits spanning the last 19 kyrs (Figure 2; Im et al., 2012; Kim et al., 2014). This sequence is named the Nari Tephra Formation, and consists of five key eruptive units (in ascending order, N-5 to N-1), some of which exhibit signs of weathering and soil formation (Figure 2). Several radiocarbon ages have been obtained from this formation by Im et al. (2012), which have been used to correlate the intra- and extra-caldera Holocene deposits (U-4 to U-2, and N-4 to N-2) as shown in Figure 2. Kim et al. (2014) have proposed a detailed succession of eruption styles for the last 19 ka, and suggest that only a few of the events generated sustained eruption columns or pyroclastic density current (PDC) deposits large enough to overtop the caldera wall, and therefore extra-caldera sequences may underestimate the eruption frequency.

2.2. Changbaishan, North Korea/China border

Changbaishan (also referred to as Baitoushan, Paektusan, or Hakutozan) is an intraplate stratovolcano situated on the border of North Korea and China (41°00'N, 128°03'E; Figure 1), located on a Neogene trachybasalt lava shield (the Gaema Plateau). Activity at Changbaishan began in the Middle Pleistocene, and has been divided into three main episodes: early shield building, middle cone construction, and a late explosive stage (Wei et al., 2007; 2013). The most recent of which (< 20 ka) culminated with the caldera-forming

Millennium Eruption (ME; VEI 7) which ejected ca. 100 km³ of tephra (Dense Rock Equivalent ca. 25 km³), blanketing the northernmost regions of Japan in ash (Horn and Schmincke, 2000; Zou et al., 2010; Wei et al., 2013; Sun et al., 2014b; McLean et al., 2016) and injecting 45 Tg of sulphur into the atmosphere (Iacovino et al., 2016). This eruption produced a ca. 4.5 km wide caldera, which today contains Lake Tianchi (meaning “Heavenly Lake”; Machida et al., 1990). The age of the ME has been precisely dated to AD 946, by combining dendrochronology with the presence of a closely related (AD 994) ‘Miyake event’ (pronounced radiocarbon peak) preserved in charred tree deposits (Hakozaki et al., 2017; Oppenheimer et al., 2017). The hazard potential of Changbaishan is considerable and is particularly concerning given that there has been recent seismic unrest at the crater (Stone, 2010; Xu et al., 2012; Wei et al., 2013).

The most comprehensively studied proximal outcrop at Changbaishan is at Twianwenfeng peak, which is on the northern Chinese flank of the summit (e.g., Chen et al., 2016; Pan et al., 2017; Sun et al., 2017; 2018). There are many inconsistent interpretations of these Late Quaternary eruption deposits, even amongst those assigned to the ME (see Pan et al., 2017). Sun et al. (2017) identify and geochemically characterise five sequential deposits (oldest to youngest, named NS-1 to NS-5) at Twianwenfeng peak, and suggest that the three uppermost units (NS-3 to NS-5) are associated with the ME, due to the geochemical (major element glass chemistry) overlap with distal ash deposits, and that no depositional break is evident between units NS-4 and NS-5. This is in contrast to other studies that suggest that the youngest unit (NS-5) may

correlate to post-ME events, which are suggested by historical records in AD 1668 or AD 1702 (Cui et al., 1995; Liu et al., 1998).

The ME had two explosive phases, with the initial main phase (ca. 95% by volume) associated with a ca. 25 km-high Plinian column, producing a widespread rhyolitic pumice fall unit (Machida et al., 1990; Horn and Schmincke, 2000), which equates to NS-3 of Sun et al. (2017). This fall unit is overlain by partially-welded PDC deposits attributed to the partial collapse of the Plinian column. Trachytic magma was erupted in a late phase of the eruption (i.e., NS-4 and NS-5), forming moderately welded PDC units that overlie the rhyolitic fall and PDC deposits (Horn and Schmincke, 2000). Chen et al. (2016) report trace element compositions for the ME at Twianwenfeng peak (therein named units C-3 to C-1), and show that the rhyolitic fall deposits (C-3 to C-2) had higher contents of incompatible trace elements (e.g., Th, Ta, Nd, Y) and lower contents of compatible elements (e.g., Ba, Sr) relative to the upper trachyte unit (C-1).

Sun et al. (2017) identify two pre-ME pyroclastic fall deposits at Twianwenfeng peak, NS-1 (grey fall unit) and NS-2 (yellow fall unit), which are compositionally distinct from the ME deposits. These units are estimated to have been erupted between 4 – 5 ka based on $^{40}\text{Ar}/^{39}\text{Ar}$, uranium series disequilibrium, ^{14}C and optically stimulated luminescence (OSL) methods (Liu et al., 1998; Wan and Zheng, 2000; Wang et al., 2001; Yang et al., 2014).

A large “lava flow” landform, named the Qixiangzhan Comendite that is 5 km long and 400-800 m wide, is observed on the northern summit of Changbaishan (Yang et al., 2014; Sun et al., 2017). This is widely considered as another pre-ME event, although $^{40}\text{Ar}/^{39}\text{Ar}$ ages generated from this deposit span several thousand years (e.g., Singer et al., 2014; Yang et al., 2014) and its stratigraphic relationship to the units preserved at Twianwenfeng peak is unclear (Sun et al., 2017). Major element glass compositions of the Qixiangzhan comendite overlap with those of the rhyolitic phase of the ME (Sun et al., 2018).

2.3. Distal marine and lacustrine tephra records

The Sea of Japan (East Sea) is a semi-enclosed marginal sea located between the Japanese islands and the Asian continent, and is the product of the rear-arc extension (Figure 1). Due to the prevailing westerly winds, tephra erupted from Ulleungdo and Changbaishan is typically dispersed to the east and deposited in the surrounding marine basins (Arai et al., 1981; Chun et al., 1997, 2007; Ikehara, 2003; Machida and Arai, 2003; Ikehara et al., 2004; Lim et al., 2013, 2014; Derkachev et al., in press). Furthermore, several Japanese tephra layers erupted from volcanoes on Kyushu Island have been dispersed to, and deposited in the Sea of Japan, including the K-Ah (Kikai), AT (Aira), SAN1 (Kuju) and Aso-4 eruption deposits (Machida and Arai, 2003; Albert et al., 2019). The marine sediments across the Sea of Japan are characterised by alternations of light and dark coloured sediments, which have been attributed to millennial-scale palaeoenvironmental changes associated with changes in the East Asian summer monsoon (Tada, 1999; Ikehara, 2003). These organic rich

dark-layers are commonly used to date tephra layers that are preserved in the marine sediments (Tada et al., 1999). It has proved very difficult to correlate between the proximal eruption successions exposed at Ulleungdo and Changbaishan with those in distal records (Shiuhara et al., 2011; Kim et al., 2014; Chen et al., 2016; Pan et al., 2017). Typically, only the largest Holocene eruptions that reached the Japanese islands have been correlated to specific eruption units within proximal stratigraphies of these two volcanoes.

The most widespread tephra layer from Changbaishan is associated with the AD 946 ME, and is distally named the Baegdusan-Tomakomai (B-Tm tephra). The B-Tm tephra was named and characterised using glass refractive indices and major element compositions (Machida and Arai, 1983; McLean et al., 2016) at a distal type-locality in Tomakomai Port, Hokkaido (northern Japan), where it was identified above the Tarumai-c (ca. 50 BC) and below the Tarumai-b (AD 1667) tephra layers from the nearby Tarumae volcano (Machida and Arai, 1983). The B-Tm tephra has since been identified in numerous marine, lacustrine and archaeological sequences across northern Japan, northeast China and coastal regions of Russia (see Sun et al., 2014b; McLean et al., 2016) and B-Tm glass shards have been identified in the Greenland ice cores (Sun et al., 2014a).

The most widespread tephra erupted from Ulleungdo is the Ulleung-Oki (U-Oki), which is correlated to the proximal U-4 deposits on the island (Machida et al., 1984; Okuno et al., 2010; Shiuhara et al., 2011; Smith et al., 2011; Kim et al., 2014). The U-Oki tephra has been identified in several marine cores in the Sea

of Japan, and in archives on the islands of Japan, including Lake Biwa, Lake Suigetsu and Lake Hane (Chun et al., 1997; Domitsu et al., 2002; Nagahashi et al., 2004; Smith et al., 2011; Figure 1). As outlined further below, several of these archives contain a younger phonolitic/trachytic ash that post-dates the U-Oki tephra, and are thought to be distal correlatives of the U-3 eruption of Ulleungdo.

One of the most comprehensive records of East Asian volcanism is the Lake Suigetsu sedimentary archive, which is located ca. 500 km E of Ulleungdo and ca. 1000 km SSE of Changbaishan (35°35'0"N, 135°53'0"E, 0 m above present sea level; Figure 1). The sequence spans approximately 150 ka (Nakagawa et al., 2012), and contains a detailed record of visible and non-visible (cryptotephra) layers derived from Ulleungdo and Changbaishan eruptions, as well as over thirty visible tephra layers erupted from sources that span the length of Japan (Smith et al., 2013; McLean et al., 2016, 2018; Albert et al., 2018, 2019). Despite the difficulties of identifying non-visible layers in a productive arc setting, cryptotephra layers are precisely preserved and identified in Lake Suigetsu, partially due to its unique hydrological setting. Suigetsu is a tectonic lake, adequately situated away from the large calderas in Hokkaido and Kyushu, and so is not inundated with locally sourced volcanic glass which would preclude the identification of cryptotephra layers deposited during large distally occurring eruptions. Furthermore, no rivers flow directly into Lake Suigetsu (Figure 1b) with the water level controlled by input into the other connected lakes. The fine-grain sedimentation in the lake is often interrupted by deposits of coarse volcanic ash that fall through the water column.

Since the Lake Suigetsu sediments have been extensively radiocarbon (^{14}C) dated, and seasonal laminae (varves) are preserved between ca. 10 and 50 ka (Staff et al., 2011; Bronk Ramsey et al., 2012; Marshall et al., 2012; Schlolaut et al., 2012), eruptions within the radiocarbon timeframe can be precisely dated if their associated tephra layers are identified. The Lake Suigetsu tephrostratigraphic record is therefore utilised in this study to precisely date ash fall events of Ulleungdo and Changbaishan that reached central Honshu, and integrate their tephrostratigraphies.

3. Tephra identification and analytical methods

3.1. New tephra identification in Lake Suigetsu

The high-resolution and intensely dated sediments of Lake Suigetsu (SG06 and SG14 cores) have been re-investigated for the presence of thin (i.e., sub millimetre in thickness) and cryptotephra layers, in order to supplement the visible tephrostratigraphy as introduced above, and published by Smith et al., (2011, 2013) and Albert et al. (2018, 2019). Cryptotephra extraction procedures (modified from Turney, 1998; Blockley et al., 2005) were undertaken through the 12 m of Holocene sediments (≤ 10 ka; McLean et al., 2016, 2018) and more recently the 14 m of annually laminated (varved) sediments dating to between ca. 50 and 30 ka, These sections were chosen for analysis as they were expected to contain low-background levels of volcanic glass, which would not obscure primary cryptotephra peaks (see McLean et al., 2018). On average,

through these investigated sediments cryptotephra layers are four times more frequently preserved than visible ash layers. Identified tephra layers in the Suigetsu sediments are named, and are referred to using their SG06 (correlation model 06 June '17) or SG14 (correlation model 30 May '16) core composite depth(s) in cm.

The Suigetsu Bayesian age model (Staff et al., 2011; Bronk Ramsey et al., 2012) was used to determine the age for ash layers preserved in the sediments. The composite Suigetsu sedimentary sequence 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 ¹⁴C dates obtained from terrestrial plant macrofossils from the upper 38 m (SG06-Composite Depth (CD) of the SG93 and SG06 cores (Kitagawa and van der Plicht, 1998a; 1998b, 2000; Staff et al., 2011, 2013a, 2013b; Bronk Ramsey et al., 2012) and varve counting between 12.88 and 31.67m SG06 CD (Marshall et al., 2012; Schlolaut et al., 2012). Outside of the varve-counted depth interval, SG06 event-free depth(s) (EFD, ver. 29th Jan '11) were used within the age model, which excludes instantaneous deposits > 5 mm in thickness, (e.g., floods, and tephra deposits; Staff et al., 2011; Schlolaut et al., 2012).

3.2. Major and trace element analysis of the glass shards

Major and minor element compositions of individual glass shards extracted from the Suigetsu visible and cryptotephra layers 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. All glass 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 from the Max Plank Institute (Jochum et al., 2006), which were run as secondary standards. Analyses of these secondary standards lie within the standard deviation of the preferred values and are presented in the Supplementary Material. All these data were filtered to remove non-glass analyses, and those with low analytical totals <93%. The raw values were normalised (to 100 %) for comparative purposes and to account for variable glass hydration, and are presented as such in all tables and figures.

Trace element compositions of the glass shards >25 μm (i.e. large enough for analysis) 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 United States Geological Survey. Accuracies of the BHVO-2G glass analyses are typically $< 3 \%$ for most elements, $< 5 \%$ for Sc, Ga, Sm, Eu, Gd, U and $< 10 \%$ for Ni, Cu, Lu. Full trace element datasets and secondary standard analyses are provided in the Supplementary Material.

4. Results

4.1. *Suigetsu tephrostratigraphy*

To date, thirty-three visible tephra layers (Smith et al., 2011, 2013; Albert et al., 2018; 2019; McLean et al., 2016, 2018) and thirty-four cryptotephra layers (between 50 to 30 ka, and > 10 ka; McLean et al., 2018) have been identified and geochemically fingerprinted in the Lake Suigetsu sediments. The distinctively high alkali content of glass shards ($Na_2O + K_2O = > 9$ wt. %; Figure 3) of eight of these tephra layers indicates that they are not from the Japanese arc volcanoes (Machida and Arai, 2003; Kimura et al., 2015; Albert et al., 2019), and are correlated by McLean et al. (2016, 2018; $n = 6$) and herein ($n = 2$) to eruptions from Ulleungdo and Changbaishan.

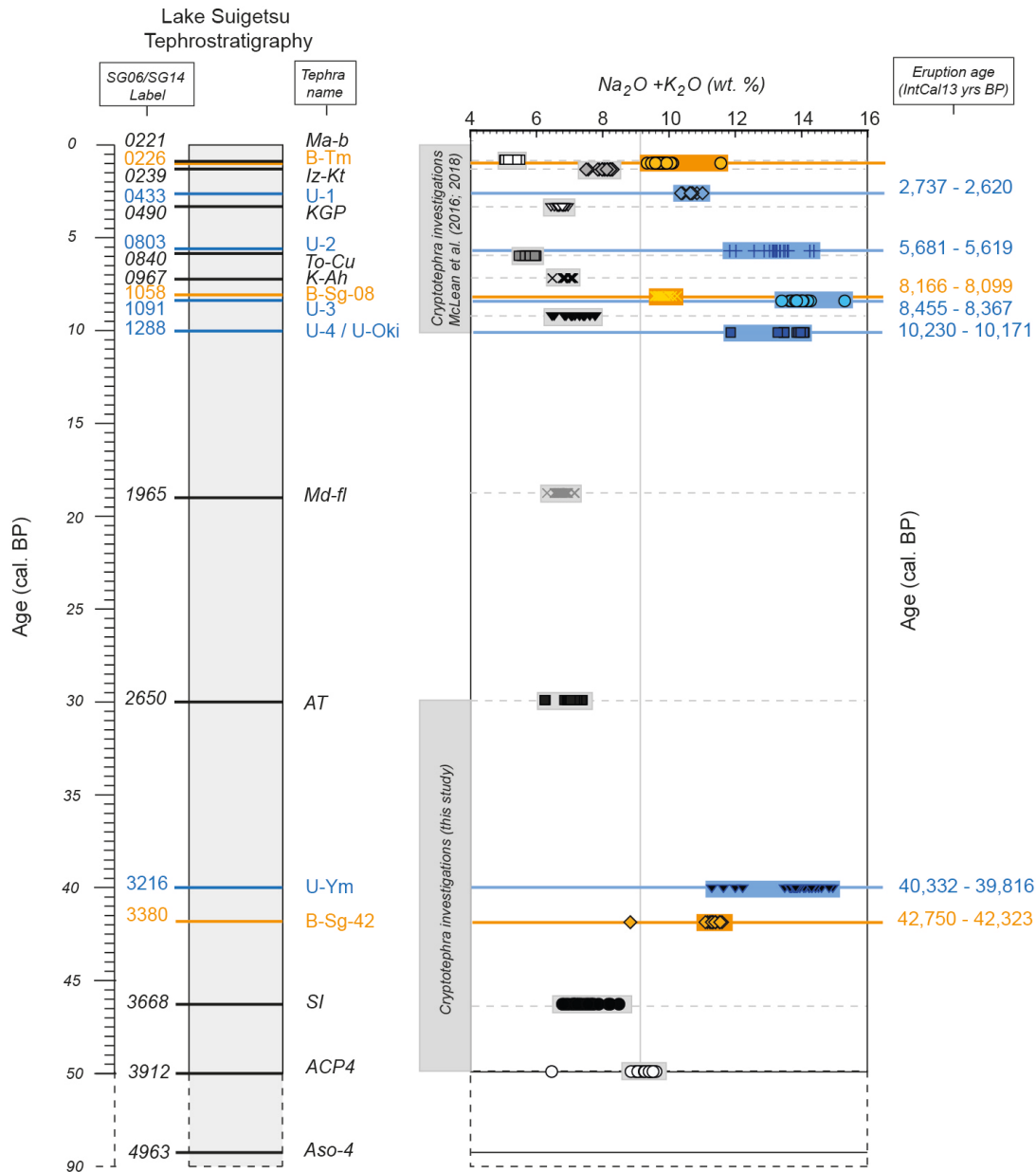


Figure 3. The composite Lake Suigetsu tephrostratigraphy and the positioning of Ulleungdo (blue lines), Changbaishan (orange) and other key Japanese (black/grey) tephra layers preserved through the sequence (Smith et al., 2013; McLean et al., 2016, 2018; Albert et al., 2018; 2019). The glass shard total alkali content ($\text{Na}_2\text{O} + \text{K}_2\text{O}$) of these layers is also plotted against eruption age, with Ulleungdo and Changbaishan tephra containing > 9 wt. %.

4.1.1. New Ulleungdo and Changbaishan deposits

As part of detailed cryptotephra investigations through the Suigetsu sediments dated between ca. 50 to 30 ka, two new alkaline ash layers named SG14-3380 and SG14-3216 have been identified. These are positioned between the Samba-Ikeda (46.4 ka; Albert et al., 2019) and AT (30.0 ka; Smith et al, 2013; Albert et al., 2019) tephras (Figure 3; Table 1). SG14-3380 is a highly concentrated cryptotephra horizon erupted from Changbaishan, and contains over 18,000 shards per gram of dried sediment (Figure 4a.). This eruption is dated to between 42,750 – 42,323 IntCal13 yrs BP (95.4 % confidence interval) using the Suigetsu age model. SG14-3216 is a thin visible (ca. 1 mm) white ash layer (Figure 4b) that is ca. 1.6 m above SG14-3380, and represents an Ulleungdo eruption between 40,332 – 39,816 IntCal13yrs BP (95.4 % confidence interval).

4.1.2. Previous identifications of Ulleungdo and Changbaishan deposits

As previously reported by McLean et al. (2018), the Lake Suigetsu Holocene tephrostratigraphy contains three eruptions from Ulleungdo: SG06-1288, SG14-1091 and SG14-0803 that are dated to 10,230 – 10,171 IntCal13 yrs BP, 8,455 – 8,367 IntCal13 yrs BP and 5,681 – 5,619 IntCal13 yrs BP (95.4 % confidence interval), respectively (Table 1; Figure 3; Smith et al., 2011; McLean et al., 2018). A younger ash layer that also has glass compositions that are similar to eruptions from Ulleungdo (SG14-0433) is dated to 2,737 – 2,620 IntCal13 yrs BP (Table 1; Figure 3).

Table 1. Summary of the Ulleungdo and Changbaishan derived tephra layers identified in the Lake Suigetsu (SG06 & SG14) sequence (in bold and shaded grey), along with their stratigraphic positioning relative to key Japanese marker layers. Tephra correlations for SG06 tephra layers are discussed by Smith et al. (2013), McLean et al. (2016) and Albert et al. (2018, 2019) and SG14 tephra layers are correlated in McLean et al. (2018, this study).

SG Label	Tephra code	Tephra name	Source volcano	Source location	¹⁴ C date (AD / IntCal13 yrs BP)
SG14-0221	Ma-b	Mashu-b	Mashu	Kurile arc, Japan	AD 960 - 992 ¹
SG06-0226	B-Tm	Baegdusan-Tomakomai	Changbaishan	North Korea / China	AD 946²
SG14-0239	Iz-Kt	Izu-Kozushima-Tenjosan	Kozushima	Izu arc, Japan	AD 838 ³
SG14-0433	U-1	Ulleung-1	Ulleungdo	South Korea	2,737 - 2,620¹
SG14-0490	KGP	Kawagodaira Pumice	Kawagodaira	Izu arc, Japan	3,227 - 3,129 ¹
SG14-0803	U-2	Ulleung-2	Ulleungdo	South Korea	5,681 - 5,619¹
SG14-0840	To-Cu	Towada-Chuseri	Towada	Northern Honshu, Japan	5,986 - 5,899 ¹
SG06-0967	K-Ah	Kikai-Akohoya	Kikai	Southern Kyushu, Japan	7,307 - 7,196¹
SG14-1058	B-Sg-08	Baegdusan-Suigetsu-08	Changbaishan	North Korea / China	8,166 - 8,099¹
SG14-1091	U-3	Ulleung-3	Ulleungdo	South Korea	8,455 - 8,367¹
SG14-1185	1185	-	-	-	9,372 - 9,301¹
SG06-1288	U-Oki / U-4	Ulleung-4 / Ulleung-Oki	Ulleungdo	South Korea	10,230 - 10,171¹
SG06-1965	Md-fl	Sambe-Midorigaoka fl	Sambe	SW Japan	19,631 - 19,471 ⁴
SG06-2650	AT	Aira-Tanzawa	Aira	Southern Kyushu, Japan	30,174 - 30,078 ⁴
SG14-3216	U-Ym	Ulleung-Yamato	Ulleungdo	South Korea	40,332 - 39,816⁵
SG14-3380	B-Sg-42	Baegdusan-Suigetsu-42	Changbaishan	North Korea / China	42,750 - 42,323⁵
SG06-3668	SI	Sambe-Ikeda	Sambe	SW Japan	46,566 - 46,162 ⁴
SG06-3912	ACP4	Aso-Central Pumice 4	Aso	Central Kyushu, Japan	50,311 - 49,637 ⁴
SG06-4963	Aso-4	Aso-4	Aso	Central Kyushu, Japan	86.4 ± 1.1 ⁴⁰ Ar/ ³⁹ Ar ⁴

1) McLean et al. (2018); 2) Oppenheimier et al. (2017); Hakozaiki et al., 2017; 3) Tsukui et al. (2006); 4) Smith et al. (2013); Albert et al. (2019); 5) This study

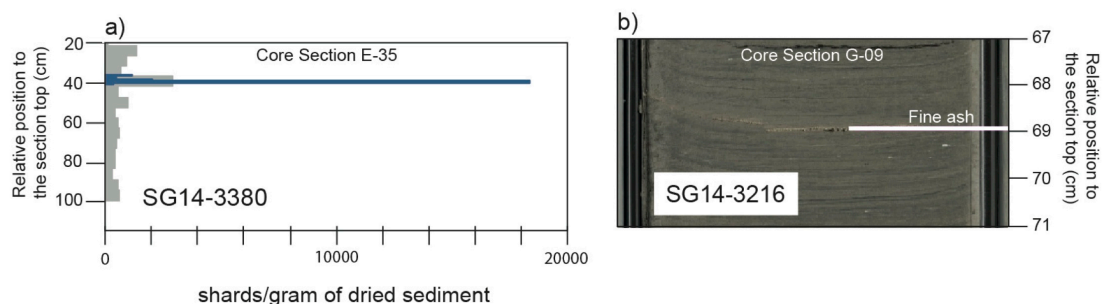


Figure 4. (a) Glass shard concentrations (shards per gram of dry sediment) preserved in SG14 core E-35 and the positioning of cryptotephra SG14-3380. Concentration of low-resolution (5 cm) samples are shown in grey and high-resolution samples (1 cm) are overlain in blue. Shard counts for the other Holocene cryptotephra layers are published by McLean et al. (2018). (b) Photograph of visible tephra layer SG14-3216 in Lake Suigetsu Core G-09.

Two Holocene Changbaishan eruptions are preserved in the sediments: SG14-1058 at 8,166 – 8,099 IntCal13 yrs BP (McLean et al., 2018); and SG06-0226, which has been correlated to the AD 946 B-Tm tephra from the ME (McLean et al., 2016; Hakozaki et al., 2017; Oppenheimer et al., 2017). Several other widespread markers have been identified in the Holocene sediments, which are able to stratigraphically separate eruption events from Ulleungdo and Changbaishan (Figure 3; Table 1). SG14-1185 stratigraphically separates the SG06-1288 and SG14-1091 Ulleungdo layers, and the K-Ah (7.3 ka; Kikai volcano) and To-Cu (5.9 ka; Towada) tephra layers separate SG14-1091 and SG14-0830 (McLean et al., 2018; Table 1; Figure 3).

4.2. Major and trace element volcanic glass geochemistry

4.2.1. Ulleungdo glass geochemistry

The newly analysed glass of SG14-3216 geochemically overlaps with the other previously identified Ulleungdo derived tephra layers preserved in the Suigetsu tephrostratigraphy (e.g., SG06-1288, SG14-1091 and SG14-0803). Collectively they straddle the phonolitic/trachytic boundary on the basis of the Total Alkalis versus Silica (TAS) classification (Le Bas et al., 1986) and contain 60 – 63 wt. % SiO₂, ca. 7 wt. % K₂O and 19 – 20 wt. % Al₂O₃ (Table 2; Figure 5). These glasses are characterised by < 2.5 wt. % CaO and contain between 2.5 and 3.5 wt. % FeO^T.

When normalised to the primitive mantle (Sun and McDonough, 1989), we find the newly obtained trace element compositions of SG14-3216 and SG14-1091 show enrichments in the Light Rare Earth Elements (LREE) relative to the Heavy Rare Earth Elements (HREE) ($\text{La/Yb} = 30 - 35$ ppm) and significant depletions in Ba, Sr and Eu that reflect K-feldspar fractionation (Figure 6). The paucity of a depletion in Nb and Ta content within these volcanic glasses, when normalised to the primitive mantle, is inconsistent with subduction related volcanism (Figure 6).

The four tephra layers with Ulleungdo compositions are difficult to distinguish using their major element glass compositions, but we find that the younger glasses of SG14-0803 are more elevated in CaO (by ca. 0.5 wt. %), compared to the early Holocene and SG14-3216 glass (Figure 5c). In addition, the alkaline glasses of SG14-3216 (59.5 – 62.5 wt. % SiO_2 and total alkalis [$\text{Na}_2\text{O} + \text{K}_2\text{O}$] of 11.6 – 14.9 wt. %) can be discriminated from SG14-1091 by larger feldspar-related depletions in Sr, Ba and Eu, that are normalised to primitive mantle compositions (Figure 6). The alkaline glass shards of SG14-0433 ($\text{Na}_2\text{O} + \text{K}_2\text{O} = 10.4 - 11.0$) are also likely to derive from Ulleungdo, but contain ca. 2.5 wt. % lower K_2O , and ca. 2 wt. % higher CaO compared to the older eruption events outlined here (Figure 5a; 5c).

Table 2. Average major, minor and trace element glass compositions of the Ulleungdo and Changbaishan tephra layers in the Lake Suigetsu sediment core.

wt. (%)	SG14-0433		SG14-0803		SG14-1091		SG06-1288		SG14-3216	
	<i>McLean et al. 2018</i>		<i>McLean et al. 2018</i>		<i>McLean et al. 2018</i>		<i>Smith et al. 2011</i>		<i>this study</i>	
	Mean	± 1 σ	Mean	± 1 σ	Mean	± 1 σ	Mean	± 1 σ	Mean	± 1 σ
SiO ₂	61.76	0.16	60.54	0.63	60.52	0.24	60.85	0.42	60.75	0.67
TiO ₂	0.79	0.05	0.62	0.07	0.51	0.08	0.50	0.07	0.39	0.07
Al ₂ O ₃	16.66	0.05	19.48	0.30	19.87	0.22	19.55	0.17	19.85	0.28
FeO ^T	5.65	0.17	3.16	0.42	2.77	0.24	3.16	0.19	3.12	0.16
MnO	0.23	0.05	0.14	0.10	0.15	0.03	0.14	0.05	0.18	0.05
MgO	1.09	0.06	0.48	0.12	0.23	0.03	0.30	0.06	0.17	0.07
CaO	6.07	0.18	6.60	0.60	6.95	0.40	1.61	0.17	1.34	0.12
Na ₂ O	2.57	0.08	1.99	0.33	1.48	0.14	6.51	0.79	7.28	0.96
K ₂ O	4.59	0.10	6.61	0.21	7.03	0.19	7.07	0.28	6.50	0.33
P ₂ O ₅	0.36	0.04	0.17	0.05	0.05	0.04	0.10	0.03	0.05	0.03
Cl	0.23	0.03	0.21	0.04	0.40	0.08	0.24	0.03	0.37	0.10
Analytical total	96.85		96.84		97.81		99.72		97.03	
<i>n</i>	8		19		24		12		37	

wt. (%)	SG06-0226		SG14-1058		SG14-3380	
	<i>McLean et al. 2016</i>		<i>McLean et al. 2018</i>		<i>this study</i>	
	Mean	± 1 σ	Mean	± 1 σ	Mean	± 1 σ
SiO ₂	74.89	0.21	75.01	0.18	66.26	0.70
TiO ₂	0.22	0.04	0.20	0.03	0.60	0.09
Al ₂ O ₃	10.27	0.10	10.28	0.11	14.97	0.28
FeO ^T	4.05	0.14	3.89	0.10	5.12	0.14
MnO	0.08	0.05	0.07	0.03	0.15	0.04
MgO	0.02	0.03	0.01	0.01	0.25	0.06
CaO	0.22	0.02	5.30	0.16	1.24	0.17
Na ₂ O	5.36	0.15	0.20	0.02	5.67	0.66
K ₂ O	4.38	0.09	4.50	0.06	5.49	0.09
P ₂ O ₅	-	-	0.01	0.01	0.09	0.03
Cl	0.50	0.03	0.52	0.02	0.16	0.02
Analytical total	96.19		95.54		95.90	
<i>n</i>	29		24		14	

ppm	SG06-0226		SG14-1058		SG14-1091		SG14-3216		SG14-3380	
	<i>this study</i>		<i>this study</i>		<i>this study</i>		<i>this study</i>		<i>this study</i>	
	Mean	± 1 σ	Mean	± 1 σ	Mean	± 1 σ	Mean	± 1 σ	Mean	± 1 σ
Rb	396.4	9.8	383.9	23.9	188.7	10.1	195.4	19.1	143.5	8.9
Sr	2.2	0.6	4.4	0.9	59.5	36.2	14.3	10.2	25.0	24.4
Y	143.3	3.9	149.8	11.4	23.5	2.0	26.0	2.9	51.1	3.9
Zr	2415.2	80.0	2302.5	189.8	626.0	50.9	709.6	104.3	741.4	45.8
Nb	286.1	8.9	261.1	21.1	169.8	11.5	175.2	21.2	97.0	5.5
Ba	6.4	0.9	6.6	2.5	99.1	83.5	16.3	13.3	77.3	57.3
La	138.9	4.5	125.2	14.6	86.4	4.3	98.2	9.0	76.4	4.7
Ce	286.9	13.5	243.2	12.3	144.8	6.9	161.7	15.7	154.3	9.6
Pr	31.9	1.2	28.3	1.8	12.8	0.9	14.5	1.2	17.1	1.1
Nd	115.8	4.8	102.3	6.9	39.3	3.5	43.9	4.7	65.6	6.4
Sm	27.6	1.2	26.1	4.5	5.6	1.0	6.4	0.7	12.9	1.9
Eu	0.3	0.1	0.3	0.3	1.5	0.3	0.6	0.2	0.6	0.2
Gd	26.7	1.5	28.0	3.3	5.0	1.4	5.4	1.9	10.7	1.4
Dy	25.3	1.8	25.5	2.2	3.8	0.6	4.4	0.6	9.3	0.9
Er	14.0	0.6	14.0	1.6	2.5	0.5	2.8	0.6	5.1	0.7
Yb	11.4	0.5	11.2	2.0	2.6	0.4	3.1	0.7	4.3	0.6
Hf	55.6	2.3	55.6	5.4	12.0	1.1	13.4	2.1	16.9	1.2
Ta	17.0	1.1	16.4	1.9	9.8	0.7	9.7	1.1	5.6	0.5
Th	49.9	3.1	49.6	5.0	23.5	1.9	25.2	3.2	14.1	1.1
U	10.8	0.7	9.7	0.9	4.8	0.5	5.3	0.9	2.8	0.3
Y/Th	2.9	0.2	3.0	0.1	1.0	1.1	1.0	0.1	3.6	0.2
Zr/Th	48.5	2.5	46.5	2.0	26.6	27.5	28.1	2.3	52.6	3.3
La/Yb	12.1	0.4	11.3	1.4	34.4	6.5	32.7	4.9	18.0	2.4
<i>n</i>	8		5		13		8		10	

(FeO^T = all Fe reported as FeO). Raw dataset and secondary standards are included in the Supplementary Material.

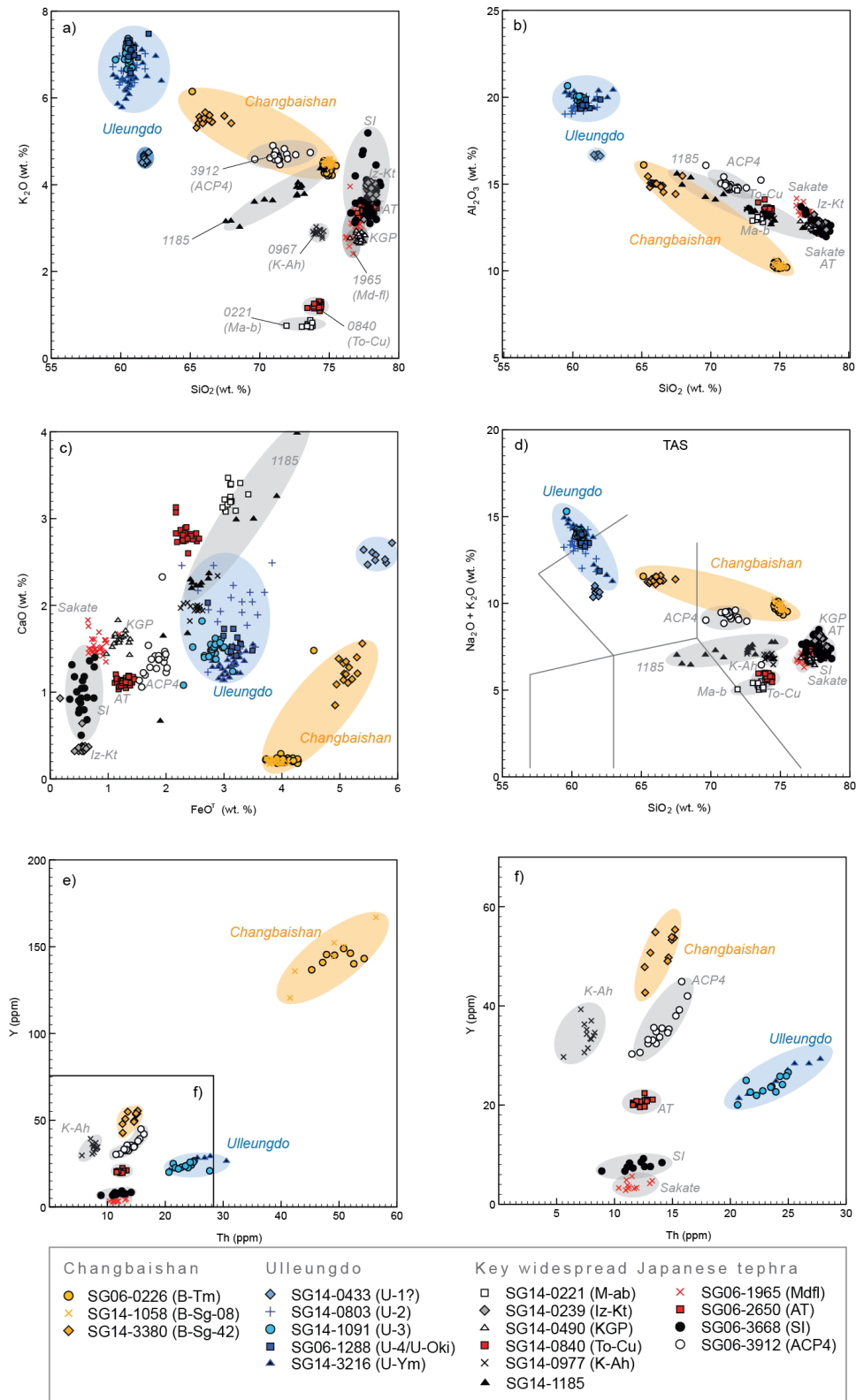


Figure 5. Glass shard major (5a – d) and trace (5e – f) element compositions of Ulleungdo (South Korea; shown in blue), Changbaishan (North Korea/China; shown in orange), and key Japanese tephras that are preserved in the Lake Suigetsu sediments (shown in grey and black) (Smith et al., 2013; McLean et al., 2016, 2018; Albert et al., 2018, 2019). (d) Total alkali versus silica plot (TAS) with whole-rock classification based on Le Bas et al., 1986).

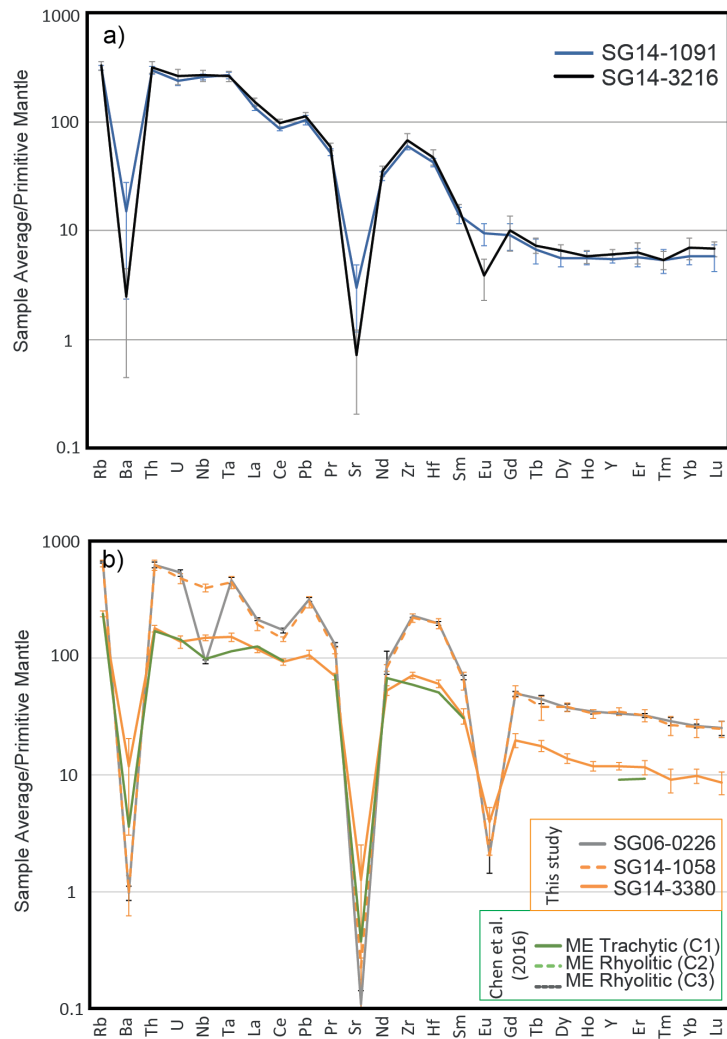


Figure 6. Primitive mantle normalised trace element compositions of glasses of: (a) Ulleungdo tephra layers (SG14-1091 and SG14-3216) in the Lake Suigetsu sequence; (b) Changbaishan tephra layers (SG06-0225, SG14-1058 and SG14-3380) in comparison to proximal ME rhyolitic (C2 – C3) and trachytic (C1) proximal deposits (Chen et al., 2016). Primitive mantle values follow Sun and McDonough (1989).

4.2.2. Changbaishan glass geochemistry

The three Changbaishan derived tephra layers (e.g., SG14-3380, SG14-1058 and SG06-0226) contain both rhyolitic and trachytic glass compositions ($\text{Na}_2\text{O} + \text{K}_2\text{O} = > 9.5 \text{ wt. } \%$; Le Bas et al., 1986), with SiO_2 ranging from 65.5 to 75.5 wt. % (Table 2; Figure 5d). They contain $< 1.6 \text{ wt. } \%$ CaO, and between 3.7 and 5.4 wt. % FeO^{T} (Figure 5c). New trace element analyses indicate that when

normalised to primitive mantle compositions, all glasses are enriched in LREE relative to HREE, with La/Yb ratios higher in those of SG14-3880, relative to SG14-1058 and SG06-0226 (Table 2) and show pronounced negative feldspar-related anomalies in Ba, Sr and Eu (Figure 6).

The newly analysed glasses of SG14-3380 are exclusively trachytic (65.4 – 67.9 wt. % SiO₂, 14.4 – 15.5 wt. % Al₂O₃, and Na₂O + K₂O = 8.7 – 11.7 wt. %) and are compositionally similar to the single trachytic analysis from SG06-0226. Trace element compositions for SG14-3380 are homogenous with 14.1 ± 1.1 ppm Th, 144 ± 9.0 ppm Rb, and 51 ± 4 ppm Y (Table 2). Unfortunately, no trace elements could be obtained for the trachytic end-member of SG06-0226 for further comparison with SG14-3380.

As discussed by McLean et al. (2018), the rhyolitic SG06-0226 glass compositions overlap with SG14-1058 for all major elements (Table 2; Figure 5). Both tephra contain glass compositions that are geochemically homogenous, with ca. 75 wt. % SiO₂, 10.3 wt. % Al₂O₃ and ca. 4.4 wt. % K₂O, and are characterised by very low CaO concentrations (< 0.3 wt. %). The newly generated trace element compositions of SG14-0226 and SG06-1058 show significant overlap, and are more enriched in incompatible elements (e.g., Th, Ta and Y), whilst depleted in compatible elements (e.g., Sr, Ba, and Eu) relative to the trachytic glass of SG14-3380 (Table 2; Figure 6b). SG06-0226 and SG14-1058 have similar mantle normalised profiles and levels of incompatible trace element enrichment (Figure 6). Glasses of both tephra layers have greater feldspar-related depletions in Ba, Sr and Eu relative to SG14-3380. The SG06-0226 glasses show a significant depletion in Nb, which is not observed in

SG14-1058 and SG14-3380, which given the intraplate setting of the volcano may relate to late stage, high-level fractionation processes.

5. Review of Ulleungdo and Changbaishan eruption framework

Here, the distal ash deposits erupted from Ulleungdo and Changbaishan are outlined and reviewed using the relative stratigraphy, geochemical glass compositions and eruption chronology. Published occurrences are centred on the Lake Suigetsu tephrostratigraphy to provide an integrated framework that is constrained by numerous widespread ash layers erupted from Japanese volcanoes (Figure 7). There are no pre-50 ka visible ash layers in Lake Suigetsu with Ulleungdo or Changbaishan compositions, but we should highlight that cryptotephra extraction techniques have not yet been carried out on these older sediments. It is possible that there are other pre-50 ka Ulleungdo or Changbaishan layers preserved cryptically in Lake Suigetsu.

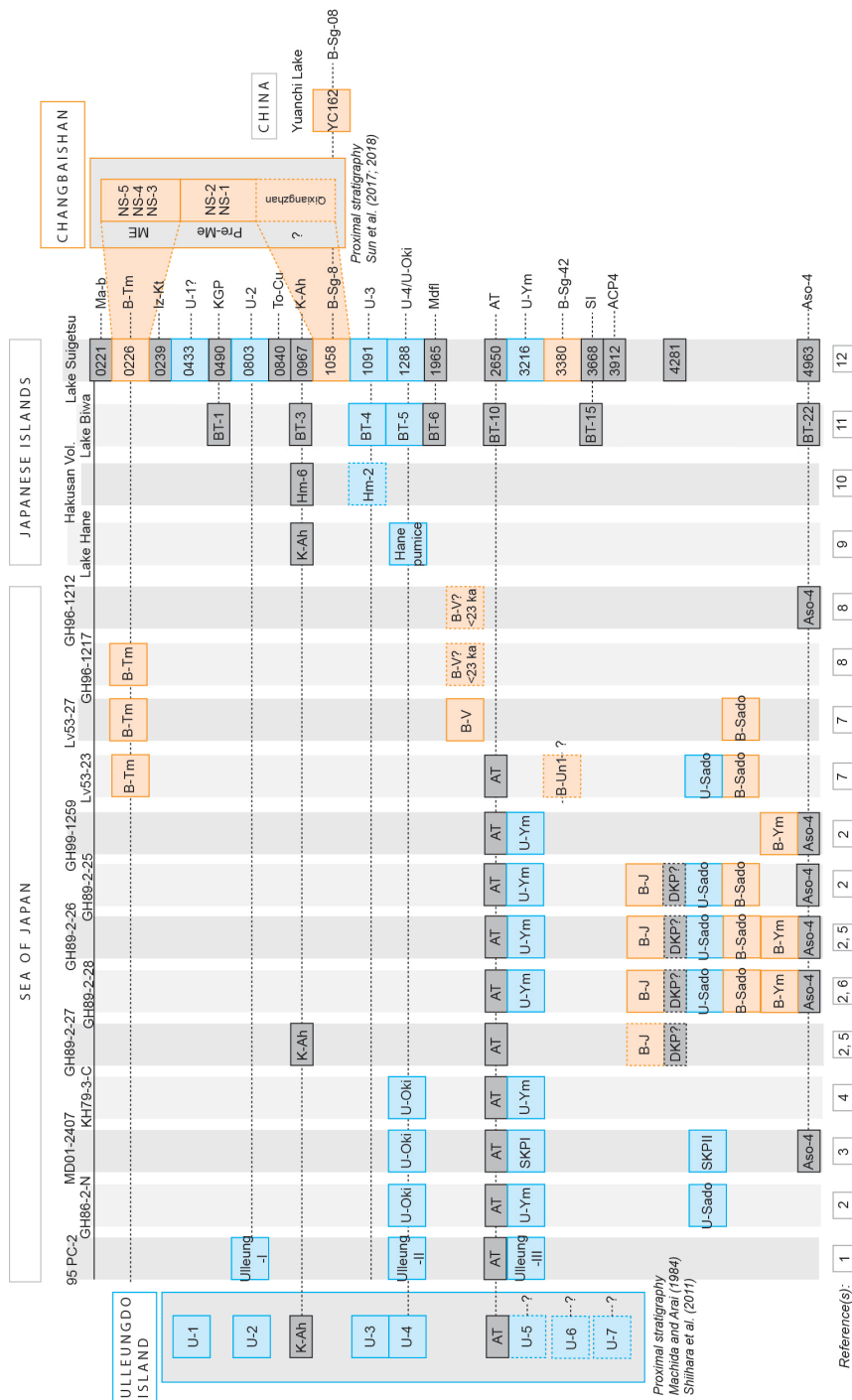


Figure 7. Summary tepхроstratigraphy of proximal and distal archives that contain ash erupted from Ulleungdo (blue boxes) and Changbaishan (orange boxes) (see Figure 1 for archive location). Widespread Japanese tephra layers, which provide valuable chronostratigraphic markers, are also shown in grey boxes. Alkali-rich ashes erupted from Ulleungdo and Changbaishan are routinely identified in marine cores extracted from the Sea of Japan (East Sea) and high resolution lake archives in Japan. 1 = Chun et al. (1997); 2 = Lim et al. (2013); 3 = Chun et al. (2007); 4 = Arai et al. (1981); 5 = Nakajima et al. (1996); 6 = Ikehara et al. (2004); 7 = Derkachev et al. (in press); 8 = Ikehara (2003); 9 = Sawada et al. (1997); 10 = Higashino et al. (2005); 11 = Nagahashi et al. (2004); 12 = Smith et al. (2011; 2013), Albert et al. (2018; 2019), McLean et al. (2016; 2018; *this study*). Due to the paucity of glass data from pre-Holocene eruptions of Ulleungdo and Changbaishan, these units cannot be correlated to distal ash deposits elsewhere.

5.1. Ulleungdo eruption history

Ulleungdo has erupted explosively at least five times over the last 86 kyrs (since the eruption of the Aso-4 tephra) with associated widespread ash fall events recognised by the ca. 60 – 61 ka U-Sado tephra (Lim et al., 2013); the ca. 40.1 ka U-Ym tephra (*this study*); the ca. 10 ka U-Oki/U-4 tephra (Smith et al., 2011; 2013); ca. 8.4 ka U-3 tephra (McLean et al., 2018); and the ca. 5.7 ka U-2 tephra (McLean et al., 2018). The known distal deposits of these events and possible proximal correlations are illustrated in Figure 7 and discussed further below.

5.1.1. Post 86 ka (Aso-4) Ulleungdo eruptions

Distal ash layers erupted from Ulleungdo were identified stratigraphically below the AT tephra (30 ka) in marine sediments obtained from the Oki ridge (Arai et al., 1981) and Yamato Basin (Ikehara et al., 2004). These pre-AT Ulleungdo tephra layers were originally considered to be from a single eruption, but the age was controversial. However, Chun et al. (2007) clarified the issue by identifying two separate alkaline ash deposits in marine core MD01-2407 (Figure 1), which they named SKPI and SKPII, and were dated to 40 – 41 ka and 60 – 61 ka, respectively, based on correlations with the regional-scale thinly-laminated marine stratigraphy (Tada, 1999; Chun et al., 2007).

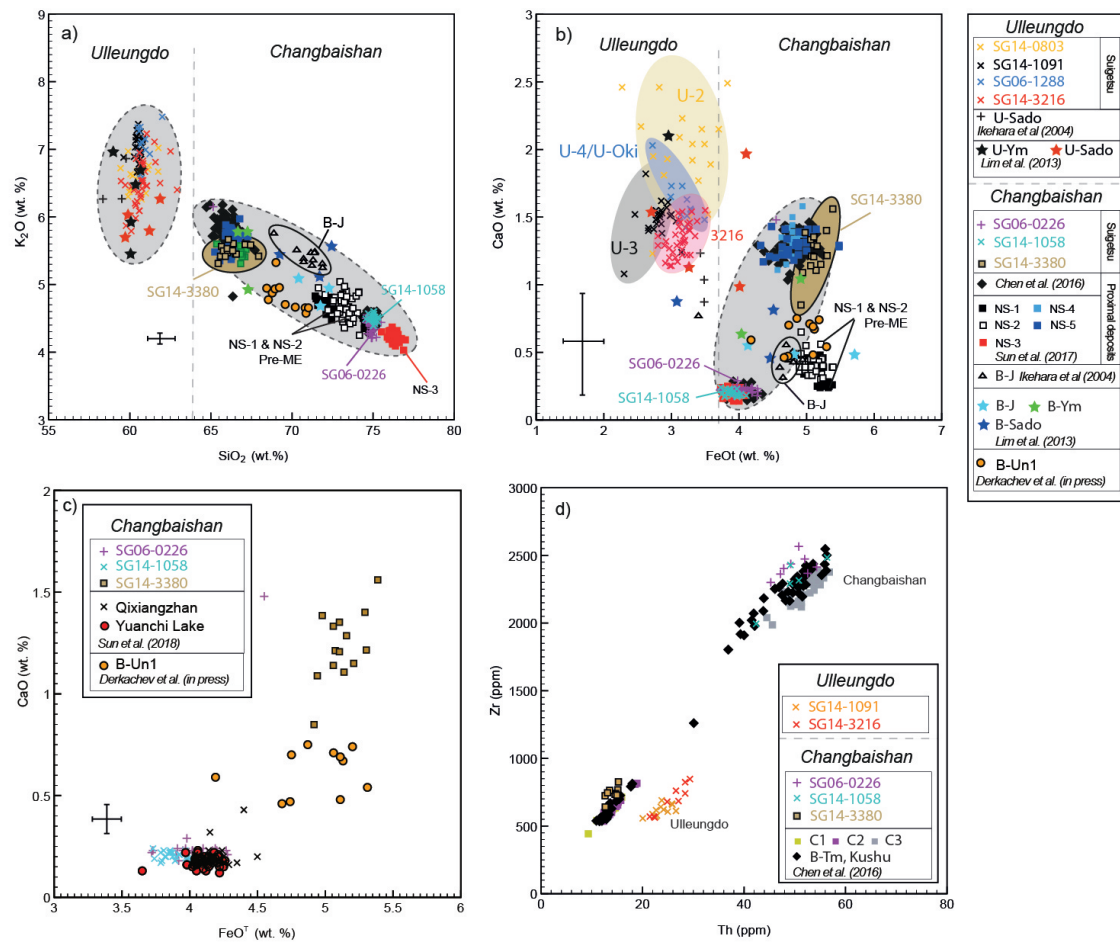


Figure 8. Glass shard major and trace element compositions of Ulleungdo (SG14-0803; SG14-1091; SG06-1288; SG14-3216) and Changbaishan (SG06-0226; SG14-1058; SG14-3380) tephra layers preserved in the Lake Suigetsu archive, compared to other proximal (Chen et al., 2016; Sun et al., 2017, 2018) and distal occurrences (Ikehara et al., 2004; Lim et al., 2013; Derkachev et al. (in press)). Error bars represent 2 x standard deviations of repeat analysis of the StHs6/80-G MPI-DING reference glass analyses, error bars for (d) are smaller than the data symbols.

Lim et al. (2013) also identified two equivalent cryptotephra layers in several other marine cores northeast of Ulleungdo (e.g., GH86-2-N, GH89-2-25, GH89-2-26 and GH89-2-28), which were stratigraphically positioned between the rhyolitic Aso-4 and AT tephra. These distal tephra layers were therein named the Ulleung-Yamato (U-Ym) and Ulleung-Sado-Oki (U-Sado), and are considered to be equivalent to SKP-I and SKP-II, respectively (Figure 7).

The Lake Suigetsu sediments verify that an ash fall event from Ulleungdo occurred at 40,332 – 39,816 IntCal13 yrs BP (95.4 % confidence interval). This 1 mm thick ash layer (SG14-3216; Figure 4) contains volcanic glass that compositionally overlaps the other Ulleungdo-derived tephra deposits preserved in the Suigetsu sequence (e.g. SG06-1288, SG14-1091), and other distal and proximal occurrences of the U-Oki tephra (Figure 5; Figure 8; Furuta et al., 1986; Nagahashi et al., 2004; Chun et al., 2007; Park et al., 2003, 2007).

Although grain-specific glass compositional datasets for the U-Ym tephra preserved in the Sea of Japan have not been published for comparison, the broad geochemical and chronological data and the stratigraphic position is consistent with SG14-3216, meaning this ash must also correlate to the same eruption of Ulleungdo. The Suigetsu-derived deposit age of 40,332 – 39,816 IntCal13 yrs BP (95.4 % confidence interval) provides the most precise eruption age, and this date can now be imported into other site-specific age models that contain this marker.

No other distal ash occurrences have been reported that are chronologically or geochemically consistent with the ca. 19 ka eruption (proximal unit N-5; Kim et al., 2014), suggesting that this eruption of Ulleungdo was probably not widespread.

5.1.2. Holocene Ulleungdo eruptions

As previously outlined, the largest known Plinian eruption from Ulleungdo generated the U-Oki tephra layer that is dated to 10,230 – 10,171 IntCal13 yrs BP (95.4 % confidence interval; Smith et al., 2011, 2013). The U-Oki ash is found in several high-resolution sedimentary records in Japan, including Lake Biwa (BT-4; Nagahashi et al., 2004) and Lake Suigetsu (SG06-1288; Smith et al., 2011) (Figure 7). This U-Oki tephra is the only distal tephra that has been correlated to proximal deposits on Ulleungdo, and equates to the proximal U-4 unit of Shiihara et al. (2011).

Lake Suigetsu tephra layers SG14-1091 (ca. 8.4 ka) and SG14-0803 (ca. 5.7 ka) overlay the U-Oki tephra and are considered distal equivalents of proximal deposits U-3 and U-2, respectively, due to their close agreement to the proximal radiocarbon dates of soils between fall units (Okuno et al., 2010). Furthermore, the K-Ah tephra is stratigraphically positioned between the U-3 (SG14-1091) and U-2 (SG14-0803) deposits (Shiihara et al., 2011). Distal equivalents of the U-3 eruption have been reported in the Sea of Japan (TRG1 sediment core; Domitsu et al., 2002), Lake Biwa (Nagahashi et al., 2004), and close to Hakusan volcano in central Honshu (Higashino et al., 2005). In comparison,

SG14-0803 is the only known distal equivalent of the U-2 tephra, indicating that it was either a lower magnitude event or the eruption plume was dispersed in a different direction.

The proximal deposits of the youngest U-1 eruption of Ulleungdo suggest it was a small strombolian-type eruption with a lava dome extrusion (Kim et al., 2014). Whole rock trace element data reported for this youngest event have a distinct tephriphonolite composition (Brenna et al., 2014). Similarly, distal SG14-0433 glass compositions are distinct from those of other Ulleungdo derived tephras in Suigetsu. Unfortunately, the lack of proximal glass chemistry for the U-1 unit means that this correlation cannot be confirmed but it is likely that the SG14-0433 layer at 2,737 – 2,620 IntCal13 yrs BP (95.4 % confidence interval) correlates to the U-1 eruption.

5.2. Changbaishan eruption history

The newly identified Changbaishan-derived tephra layer outlined here, in addition to the previously recognised layers, indicate that at least eight explosive eruptions have produced widespread ash dispersals over the last 86 kyrs. These include the: ca. 85.8 ka B-Ym tephra (Lim et al., 2013); ca. 67.6 ka B-Sado tephra (Lim et al., 2013); ca. 50.5 ka B-J tephra (Ikehara et al., 2014; Lim et al., 2013); ca. 42.5 ka B-Sg-42 tephra (*this study*), ca. 38 ka B-Un1 tephra (Derkachev et al., in press), ca. 25 ka B-V tephra (Machida and Arai, 2003); ca. 8.1 ka B-Sg-08 tephra (McLean et al., 2018); and AD 946 B-Tm tephra associated with the ME (McLean et al., 2016; Hakoziaki et al., 2017;

Oppenheimer et al., 2017). All the known distal ash deposits associated with eruptions at Changbaishan and possible correlations to proximal units on the volcano are summarised in Figure 7 and are discussed below.

5.2.1. Post 86 ka (Aso-4) Changbaishan eruptions

To date, at least four individual tephra layers originating from Changbaishan have been recognised in marine cores stratigraphically positioned between the Aso-4 and AT tephra layers (Figure 7). The two oldest, Baegdusan-Yamato Basin (B-Ym; ca. 85.8 ka) and Baegdusan-Sado-Oki (B-Sado; ca. 67.6 ka) tephtras, have been identified as cryptotephra horizons in both the GH89-2-26 and GH89-2-28 marine cores (Figure 7; Lim et al., 2013). Lim et al. (2013) report that the B-Ym and B-Sado glass shards are trachytic in composition (Figure 8). More recently, Derkachev et al. (in press) also identify visible deposits in several marine cores across the Yamato and Pervenets Rise (e.g., cores Lv53-25, Lv53-20, Lv53-27, and Lv53-29) that they correlate to the B-Sado tephra. The constructed age models for these marine cores suggest an eruption age ca. 71 ka (Derkachev et al., in press).

The Baegdusan-Japan (B-J) tephra is found between Ulleungdo U-Ym and U-Sado tephra layers (Figure 7), and estimated to have been erupted at ca. 50 ka based on correlations with the regional-scale thinly laminated layer stratigraphy (Ikehara et al., 2004; Lim et al., 2013; Derkachev et al., in press). Ikehara et al. (2004) and Lim et al. (2013) report a homogenous rhyolitic composition for the B-J tephra, with ca. 71.2 wt. % SiO₂, ca. 12.0 wt. % Al₂O₃,

and total alkalis of 11.1 wt. % (Figure 8). It contrasts with the exclusively trachytic glass compositions of the older ca. 67.6 ka B-Sado tephra (Lim et al., 2013).

Derkachev et al. (in press) report a 5 mm thick volcanic ash layer in a sequence (Lv53-23 at 211 cm) from the Yamato Rise, about 270 km SE of Changbaishan, in the Sea of Japan. This deposit is therein named the Baegdusan-Unknown (B-Un1) tephra and represents another explosive event dated to around 38.3 ka. The glass chemistry of this layer is somewhat distinct, containing ca. 73.9 wt. %, ca. 13.5 wt. % Al_2O_3 , and total alkalis of 8.2 wt. %

The Lake Suigetsu sediments also provide evidence of another explosive eruption of Changbaishan chronologically occurring between the B-J and B-Un1 event. A cryptotephra layer (SG14-3380) is found ca. 1.6 m below the U-Ym tephra (SG14-3216) and this depth corresponds to a date of 42,750 – 42,323 IntCal13 yrs BP (95.4 % confidence interval). The glass compositions of SG14-3380 are exclusively trachytic and geochemically overlap with proximal units assigned to the late phase of the ME (e.g., NS-4 and NS-5 proximal deposits; Sun et al., 2017) (Figure 8) and other distal occurrences of the trachytic end member of the B-Tm ash (e.g., Okuno et al., 2011; Hughes et al., 2013; Sun et al., 2015; Chen et al., 2016). SG14-3380 does not geochemically overlap with the reported composition of the ca. 38 ka B-Un1 tephra reported in the Yamato Rise (Derkachev et al., in press), or the ca. 50.5 ka B-J tephra (Ikehara et al., 2004; Lim et al., 2013) clearly indicating that they represent separate eruptions of Changbaishan. Here, we name the Suigetsu distal ash of this Changbaishan

eruption (ca. 42 ka) as the Baegdusan-Suigetsu-42 (B-Sg-42) tephra, following the convention of naming proximally undefined distal deposits using the type-locality.

A post-AT distal tephra named the Baegdusan-Vladivostok (B-V) ash was found in the Primorye regions of Russia, and in the north-eastern part of the Japan Sea (Figure 1; Machida and Arai, 2003; Ikehara, 2003; Derkachev et al., in press). The eruption age is estimated to ca. 29 ka (Derkachev et al., in press).

5.2.2. Holocene Changbaishan eruptions

An eruption from Changbaishan was identified in Lake Suigetsu (SG14-1058) at 8,166 – 8,099 IntCal13 yrs BP (95.4 % confidence interval; McLean et al., 2018) and was the first known discovery of a large early Holocene eruption from this volcano. A visible patchy grey peralkaline tephra has since been identified in Lake Yuanchi, located ca. 30 km east of Changbaishan in China, which is dated to a similar age as the Suigetsu layer, 8,831 – 8,100 IntCal13 yrs BP (95.4 % confidence interval; Sun et al., 2018). The major element glass compositions of this Yuanchi tephra broadly overlap with those of SG14-1058 (Figure 8c), although some offsets, which are close to instrumental/analytical uncertainty, are observed. Sun et al. (2018) also suggest the Suigetsu SG14-1058 and Yuanchi tephra are distal deposits from the eruption that produced the Qixiangzhan comendite lava flow, but it is not known if there was an explosive phase associated with this eruption, and the stratigraphic relationship between the Qixiangzhan comendite and the explosive pre-ME fall deposits is not

known. Furthermore, the chronological uncertainty on $^{40}\text{Ar}/^{39}\text{Ar}$ ages for the Qixiangzhan comendite mean that it could be a separate eruption (e.g., Singer et al., 2014; Yang et al., 2014). When normalised to mantle concentrations, we find that SG14-1058 only shows a minor depletion in Nb, unlike SG06-0226 (B-Tm ash) and C-3 unit (Chen et al., 2016), which could help identify the proximal deposit. We suggest that the distal ash erupted from Changbaishan at ca. 8.1 ka (e.g., SG14-1508) is named Baegdusan-Suigetsu-08 (B-Sg-08).

No distal ash deposits have been identified that geochemically or chronologically overlap with the pre-ME proximal deposits of NS-4 and NS-5 that are dated to ca. 4 – 5 ka (Sun et al., 2017). Similarly, even in the high-resolution archives in northern Japan (e.g., Lake Kushu; Chen et al., 2019) there are no clear isochrons representing post-ME ash eruptions of Changbaishan.

6. Conclusions

Distal records can provide useful information on past eruption activity from volcanoes whose deposits are inaccessible for various reasons, e.g., burial or deposition into dynamic ocean environments. The new occurrences reported here and considered with other known distal alkali-rich ash units found in marine and lacustrine cores (spanning the last 86 kyrs) in the East Asian/Pacific region provide an improved eruption framework for intraplate volcanoes, Ulleungdo and Changbaishan. This framework shows that there are numerous

explosive eruptions responsible for distal ash fall events that are only cryptically recorded in the geological record.

Ulleungdo has erupted explosively at least five times over the last 86 kyrs (since the deposition of the Aso-4 tephra) and these are: the 60 – 61 ka U-Sado tephra (Lim et al., 2013); the ca. 40.1 ka U-Ym tephra (*this study*); the ca. 10 ka U-Oki/U-4 tephra (Smith et al., 2011; 2013); ca. 8.4 ka tephra (U-3; McLean et al., 2018); and the ca. 5.7 ka U-2 tephra (McLean et al., 2018). Furthermore, it is likely that a younger eruption from Ulleungdo occurred ca. 2.7 ka, but chemical analyses of proximal deposits are required to confirm the correlation. This age would be consistent with an eruption repose interval of <3 ka throughout the Holocene.

The new Changbaishan-derived tephra layers identified in the Suigetsu sediments indicate that at least eight explosive eruptions have produced significant ash dispersals over the last 86 kyrs which include the: ca. 85.8 ka B-Ym (Lim et al., 2013); ca. 70 ka B-Sado (Lim et al., 2013; Derkachev et al., in press); ca. 50.5 ka B-J tephra (Ikehara et al., 2014; Lim et al., 2013; Derkachev et al., in press); ca. 42.5 ka B-Sg-42 (*this study*), ca. 38 ka B-Un1 (Derkachev et al., in press), ca. 25 ka B-V (Machida and Arai, 2003); ca. 8.1 ka B-Sg-08 (McLean et al., 2018); and AD 946 B-Tm tephra associated with the ME (Hakozaki et al., 2017; Oppenheimer et al., 2017). It is possible that additional ash fall events will be discovered in other distal records in the future, as there are some proximal units near Changbaishan (e.g., the compositionally distinct

NS-4 and NS-5 layers; Sun et al., 2017) that have not yet been correlated to distal markers.

Even though Lake Suigetsu is located ca. 500 km E of Ulleungdo and ca. 1000 km SSE of Changbaishan (i.e., not downwind of the current prevailing winds), tephra from these volcanoes is clearly preserved in the sediments. The eruptions responsible for the B-Sg-42 and B-Sg-08 distal tephra must have been large eruption events (i.e., greater than VEI 5-6), based on the shard concentrations preserved in Suigetsu (>18,000 shards per gram of dried sediment). Unfortunately, it is not possible to get better constraints on volume and magnitude of these events given that they have not yet been found as visible layers and have not been identified in multiple locations. The precise ages provided in this paper from the Lake Suigetsu chronology may help locate these deposits in other records, which may provide more information about the eruptions and the dispersal of the events. Critically, these tephra occurrences demonstrate that both Ulleungdo and Changbaishan have been more active than previously thought, and the ash plumes from these explosive eruptions were widespread.

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