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24 Abstract

Far-travelled ash layers from explosive volcanic eruptions can provide invaluable marker horizons for dating and correlating regional to global sedimentary archives. Here, we present a new cryptotephra associated with the ~5.9 cal ka BP Towada Chuseri eruption (To-Cu) in a peat sediment record from northeast China. This tephra exhibits a rhyolitic glass composition that can be distinguished from other widespread tephra layers around the region of Japan and northeast China. Our findings extend the known range of this ash significantly, making it now traceable about 1,200 km from its source, Towada volcano, Japan. Notably, this tephra provides an important isochron for synchronizing palaeoenvironmental studies during the mid-Holocene period from the western Pacific, central Japan, Japan Sea, and northeast China.

1. Introduction

Volcanic ash can form regional to global time-equivalent marker horizons, and thus provide a unique dating method for synchronizing studies of regional palaeoclimatic and palaeoenvironmental archives (Lowe 2011). The development of cryptotephra (invisible volcanic ash) analysis has extended the applications of tephrochronology to ultra-distal regions several thousand kilometers away from the volcanic vent, yielding trans-continental marker horizons (Bourne et al., 2016; Cook et al., 2018; Jensen et al., 2014; Lane et al., 2013; Mackay et al., 2016; Pyne-O'Donnell et al., 2016; Sun et al., 2014, 2021; van der Bilt et al., 2017). Japan has one of the largest number of subaerial volcanoes of the Earth, and explosive volcanic eruptions are very frequent here (Machida 2002). Tephras of Japanese origin have been identified in ultra-distal regions; for example, Hokkaido and Tohoku tephras have been reported from eastern North America and Greenland ice cores (Bourne et al., 2016; Mackay et al., 2016). Sun et al. (2021) found tephras from the ~7.3 cal ka BP Kikai and ~12.8 cal ka BP Sakurajima eruptions in Huguangyan Maar Lake in southern China. However, whether Japanese tephras can be traced across northeast China is still unknown, and due to limited applications of tephrochronology in this region.

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In this study, we demonstrate the potential for long-distance sedimentary linkages through the identification of a Japanese tephra in Gushantun peat sediment from northeast China. Its glass shard composition indicates that corresponds to the ~5.9 cal ka BP Towada-Chuseri (To-Cu) eruption. The occurrence of distal To-Cu tephra in continental northeast China, >1,200 km away from the volcanic vent, implies that this tephra can be used to link palaeoclimatic records from central Japan's Lake Suigetsu to northeast China, and it can potentially be used as a mid-Holocene dating horizon within marine sediments from the Japan Sea.

2. Material and methods

2.1 Study site

Our study site, Gushantun bog (42°18′ N, 126°17′ E; Fig.1), is located in the Huinan Country of Jilin Province, western flank of the Changbai Mountains. The bog is about 1 km in diameter, and is surrounded by a basaltic platform. Peat sediments were collected from near the center of the bog using a Russian peat corer to a depth of 6.45 m. The section of sediment used in this study (2.70– 4.20 m) consists of fibrous peat with some plant remains.

73 2.2 Tephra separation

The cryptotephra separation method developed by Turney (1998) and Blockley et al. (2005) was used in this study. First, contiguous 10 cm intervals were burned at $^{\circ}$ C for 4 hours, and then sieved through 80 and 30 μ m meshes, retaining the fraction 30–80 µm. Secondly, 2.15 g/cm³ and 2.5 g/cm³ sodium polytungstate (SPT) was used to float glass shards from the sediments. A polarizing microscope was used to check for the presence of glass shards. Where shards were located, further samples were prepared at 1 cm sampling resolution; these samples were treated using the same approach to determine the peak concentration of glass shards. Further samples from levels corresponding to shard concentration peaks were treated using acid digestion (Dugmore et al., 1992) to remove organic materials in preparation for major element

geochemical analysis. Samples were mounted on glass slides and covered in Buehler
EpoxyCure2 resin, which were then ground and polished to expose fresh glass
surfaces for electron microprobe analysis (EPMA).

2.3 Geochronology

Plant remains were hand-picked and submitted for accelerator mass spectrometry
(AMS) ¹⁴C dating to the Beta Analytic Radiocarbon Laboratory, Florida, USA (Table
1; Fig. 2A, 2B). A Bayesian age-depth model was established using the P-sequence
depositional model in OxCal 4.4 (Bronk Ramsey 2008; 2009) with the IntCal20
calibration dataset (Reimer et al., 2020).

95 2.4 Geochemical analysis

We measured major and minor element oxide concentrations using wavelength dispersive spectrometer electron microprobe analysis (WDS-EPMA) on a JEOL JXA 8100 at the State Key Laboratory of Lithospheric Evolution of the Institute of Geology and Geophysics, Chinese Academy of Sciences (IGGCAS), Beijing. Ten elements (Na, Mg, Al, Si, K, Ca, Fe, Ti, Mn, and P) were analyzed with an accelerating voltage of 15 kV, a beam current of 6 nA, and a beam diameter of 5 µm. The Na content was determined at the start of the analysis, and peak counting times used were 20 s for all elements except for Na (10 s). Secondary standard glass of MPI-DING including ATHO-G and StHs6/80-G were used to check the precision and accuracy of the data (Jochum et al., 2006). Data presented in figures have been normalized to an anhydrous basis.

3. Results and discussion

A cryptotephra layer with 160 shards/0.5g dry sediments was identified at a depth of 314 cm in the Gushantun core, and thus the tephra layer was labelled GST-314. This tephra layer is composed of colourless and vesicular glass shards (Fig. 2C), of which seven were analyzed. All glass shards exhibit a consistent rhyolitic composition, with SiO₂ concentrations ranging from 73.59 to 75.48 wt%, and low K₂O concentration (1.11 to 1.26 wt%) (Table 2, Fig. 3, Fig. 4). The age of GST-314 is
constrained by the age model to between 6030 and 5676 cal a BP (95% probability).

To identify the source of GST-314, we considered the geochemical compositions of tephra from the surrounding volcanic region. Gushantun bog is located in the Longgang volcanic field (LVF), and the composition of the volcanic products from this region is basaltic with no known silicic products (e.g., Fan et al., 2000; Liu et al., 2009; Sun et al., 2016). Therefore, the rhyolitic GST-314 tephra was not from a local eruption of the LVF. Changbaishan volcano on the border between China and North Korea, ~120 km to the west, experienced frequently felsic volcanic eruptions during the Quaternary period (Chen et al., 2016; Liu et al., 2015; Sun et al., 2014, 2017, 2018; Yi et al., 2021; Zhang et al., 2018). However, Changbaishan tephras predominantly with trachytic and rhyolitic glass compositions exhibit higher potassium content than GST-314, which excludes it as the source (Fig. 3). Ulleungdo is a Quaternary volcanic island in the Japan Sea that is known to have produced many explosive eruptions (Kim et al., 2014; McLean et al., 2018, 2020), including the U2 tephra (5681-5619 cal a BP) which has a similar age to GST-314. With an even higher potassium content than the Changbaishan tephras, Ulleungdo can also be ruled out as the source for the GST-314 tephra (Fig. 3).

Considering other coincident large explosive (volcanic explosivity index: VEI >5) eruptions from southeast to east Asia that have widespread tephra dispersals, there are also some potential source candidates for GST-314: the 7307-7196 cal a BP Kikai-Akahoya eruption (K-Ah, Kikai volcano, Japan), the 6079-5004 cal a BP Pinatubo (Philippines) eruption, the 4894-4827 cal a BP SG14-0704 tephra recorded in Lake Suigetsu (unknown eruption), the 7340-7180 cal a BP KS₂ tephra from Ksudach caldera (Russia), the 6375-6220 cal a BP IAv12 (AV₄) tephra from Avachinsky volcano (Russia), the 7920-7620 cal a BP KHG tephra from Khangar volcano (Russia), and the 5986-5899 cal a BP Towada-Chuseri eruption (To-Cu, Towada volcano, Japan) (Ishimura and Hiramine 2020; McLean et al., 2018; Plunkett et al., 2015; Ponomareva et al., 2017; Portnyagin et al., 2020; Smith et al., 2013; Sun et al., 2021). Geochemically, the GST-314 tephra has a diagnostic glass composition

with low potassium content and high silicon content which is consistent only with the
To-Cu tephra and clearly separable from other broadly contemporary tephras (Fig. 3
and Fig. 4). Consequently, on the basis of comparable glass composition and eruptive
age, we attribute GST-314 in Gushantun bog to the To-Cu eruption.

Towada is an active volcano located in northern Tohoku, Honshu Island, northeast Japan (Fig. 1). The Towada-Chuseri (To-Cu, ~5.9 cal ka BP) and Towada-a (To-a, AD 915) tephras were produced by the largest volcanic events of Towada during the Holocene. The To-Cu eruption has been attributed a VEI of 5 based on a total estimated tephra volume of 9.18 km³ (Hayakawa, 1985; Newhall and Self 1982). To-Cu can be divided into three units: Chuseri pumice (Cu), Kanegasawa pumice (Kn), and Utarube ash (Ut), of which the Cu pumice is the lowermost bed and most widely distributed (Hayakawa, 1985; Ishimura and Hiramine 2020). While there is some overlap in their glass composition, Ishimura and Hiramine (2020) have shown that the units are separable on the basis of shard morphology and major element geochemistry, especially Ut, the youngest unit which is the product of phreatic eruptions. On the basis of previous investigations, To-Cu dispersal was mainly towards the south of Towada, and tephra has been identified in various sedimentary archives, the most distal occurrence being in Lake Suigetsu ~700 km southwest of the vent (Hirose et al., 2014; Ishimura et al., 2017; Kariya et al., 2016; McLean et al., 2018). The GST-314 tephra exhibits a homogeneous glass composition which cannot be separated from the Cu and Kn pumices and which is also similar to the records in Lake Suigetsu (Fig. 4), but it does not show the heterogeneous geochemistry of the Ut ash. Therefore, the GST-314 tephra is likely related to the pyroclastic fall deposits (Cu or Kn) of the To-Cu eruption rather than its late stage phreatic eruption (Ut).

¹⁶⁸ ¹⁴C dating has been widely used to constrain the eruptive timing of To-Cu: for ¹⁶⁹ example, charcoal within the tephra deposit yielded an age of 5390 ± 140 BP ¹⁷⁰ (GaK-9761, 6487–5898 cal a BP, 95% probability, calibrated using IntCal20) ¹⁷¹ (Hayakawa 1983). Recently, the identification of the tephra in Lake Suigetsu has ¹⁷² enabled a precise age of 5986–5899 cal a BP (95% probability) to be attributed to this ¹⁷³ eruption based on varve chronology combined with ¹⁴C dating (Bronk Ramsey et al.,

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174 2012; McLean et al., 2018; Staff et al., 2011). The precisely constrained Lake
175 Suigetsu age makes this tephra a valuable age marker in sedimentary sequences.
176 When we apply the Suigetsu age of To-Cu to the Gushantun age-model, age
177 uncertainty is reduced considerably during this period (Fig 2B).

During the Holocene, major tephras reported from Japan Sea marine sediments are mainly visible beds (e.g., Furuta et al., 1986; Machida 2002), amongst which ash from ~7.3 cal ka BP K-Ah and AD 946 Changbaishan Millennium Eruption (B-Tm) are the most commonly reported layers (Furuta et al., 1986; Machida 2002). The presence of To-Cu in northeast China increases the known dispersal of this tephra to more than 1,200 km from the vent, and suggests that it too may have been deposited in the Japan Sea, but perhaps only as cryptotephra. The To-Cu tephra also was found in marine sediments from the Pacific coast of the Tohoku region (Ikehara et al., 2017). Thus, it can provide a high-precision mid-Holocene age marker linking the western Pacific Ocean, central and north Japan, very likely the Japan Sea, and northeast China (Fig. 1).

The isopachs of proximal To-Cu deposits show a mainly eastern distribution (Hayakawa 1985). However, identification of this tephra in distal sedimentary archives from central Honshu confirms a southwestern dispersal (Ikehara et al., 2017; Ishimura et al., 2017; Ishimura and Hiramine 2020; Kariya et al., 2016; McLean et al., 2018). The finding of To-Cu in northeast China now also shows that tephra was transported to the west during this eruption. Most tephras around northeast Asia have eastern dispersals, reflecting the prevailing westerly winds in the region (Lane et al., 2017; Machida 2002). During the summer season, however, the Asian Summer Monsoon brings wind from the Pacific Ocean towards the Asian continent. This weather pattern is consistent the south-westerly dispersal of To-Cu tephra. Consequently, the eruptive season of To-Cu is unlikely to be winter, assuming a similar weather pattern as today.

202 4. Conclusions

Our discovery of the ~5.9 cal ka BP To-Cu tephra in Gushantun bog, northeast

China, extends the known dispersal of this tephra to ~1,200 km west of Towada. Its
presence in China suggests that it can be used as a mid-Holocene age marker horizon
for various sedimentary archives in the wider region that will facilitate the dating and
synchronization of mid-Holocene palaeoenvironmental records around northeast Asia.
The tephra also raises the possibility that other tephra layers of Japanese origin may
be detected here, emphasizing the potential for cryptotephrochronological
applications across northeast Asia.

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Figure 1. Locations of the study site (Gushantun bog), Towada volcano, and major active volcanoes around northeast Asia. The black solid line represents the confirmed dispersal of Towada-Chuseri (To-Cu) tephra. The known distribution of other major Holocene tephras around this area are highlighted by dashed lines (data source: U-Oki and K-Ah is from McLean et al., 2018; QEA/B-Sg-08 is from Sun et al., 2018; B-Tm is generated from Chen et al., 2016, McLean et al., 2016, and Sun et al., 2015). The locations of other key sites mentioned in the text are also shown (SHL: Sihailongwan; YC: Yuanchi, SG: Suigetsu; CBS: Changbaishan).

 Figure 2. Tephrostratigraphy and age-depth model for the section of Gushantun peat discussed in this paper (A). Light grey shading is the number of glass shards per 0.5 g of dried sediment in 10 cm increments, and black bands refer to glass shard concentrations in the 1 cm samples. When the initial age for the GST-314 was found to be consistent with To-Cu, the age-depth model was re-run with the precisely constrained age for the To-Cu from Lake Suigetsu as determined by McLean et al. (2018) (B). Photos of the colourless glass shards from the GST-314 tephra (C).

Figure 3. Bivariable plots that compares the glass shard compositions of GST-314
with the discussed known eruptions. Data sources: K-Ah, SG14-0704, Ulleungdo:
McLean et al., 2018; Pinatubo: Sun et al., 2021; KHG: Cook et al., 2017; IAv12:
Ponomareva et al., 2017; KS₂: Plunkett et al., 2015 and Ponomareva et al., 2017;
Changbaishan: Sun et al., 2017, 2018.

Figure 4. Biplots comparing glass shard composition of GST-314 recorded in
Gushantun peat, with To-Cu recorded in Lake Suigetsu (SG14-0840; McLean et al.,
2018), and three proximal units of To-Cu tephra from Japan (Ishiruma and Hiramine,
2020).

Table 1. Radiocarbon dates from plant remains from Gushantun peat. Radiocarbon
ages were calibrated using the IntCal20 curve in OxCal 4.4 (Bronk Ramsey 2008,
2009; Reimer et al., 2020).

Depth (cm)	Lab code Beta-	δ ¹³ C (‰)	¹⁴ C Age (yr BP)	Calibrated Age (cal a BP, 95% probability)
282	448787	-29.6	4490 ± 30	5297–5040, 5001–4986
311	448788	-27.7	5010 ± 30	5895–5805, 5794–5783, 5769– 5653, 5623–5607
408	448789	-27.3	7580 ± 40	8450-8327, 8234-8225

Table 2. WDS-EPMA analyzed glass shards for the tephra from Gushantun peat (GST-314). All the data have been normalized to an anhydrous basis (original analytical totals shown). Comparative glass data of the To-Cu tephra in Lake Suigetsu is from McLean et al. (2018).

Sample	SiO ₂	TiO ₂	Al_2O_3	FeO	MnO	MgO	CaO	Na ₂ O	K ₂ O	P_2O_5	Analyt tota
GST-314											
	74.67	0.43	13.34	2.47	0.09	0.61	2.68	4.44	1.18	0.08	99.9
	74.20	0.45	13.48	2.51	0.17	0.59	2.78	4.63	1.15	0.03	97.8
	74.73	0.40	13.33	2.46	0.20	0.59	2.80	4.18	1.24	0.08	98.3
	74.63	0.42	13.32	2.46	0.11	0.62	2.65	4.50	1.21	0.09	96.9
	74.18	0.48	13.45	2.42	0.10	0.65	2.73	4.57	1.26	0.17	100.
	73.59	0.49	13.64	2.79	0.09	0.64	2.87	4.72	1.11	0.06	99.0
	75.48	0.44	13.21	1.88	0.07	0.50	2.54	4.59	1.23	0.05	98.′
ATHO-G											
Average (n=3)	75.39	0.24	12.29	3.33	0.10	0.09	1.68	4.23	2.64	0.01	
2σ	0.57	0.04	0.56	0.34	0.19	0.05	0.01	0.17	0.07	0.05	
Preferred value	75.60	0.26	12.20	3.27	0.11	0.10	1.70	3.75	2.64	0.03	
Uncertainty (95%)	0.70	0.02	0.20	0.10	0.01	0.01	0.03	0.31	0.09	0.00	
StHS6/80-G											
Average (n=3)	63.22	0.71	17.92	4.51	0.11	1.94	5.21	4.95	1.28	0.15	
2σ	0.28	0.06	0.40	0.14	0.04	0.05	0.08	0.51	0.04	0.10	
Preferred value	63.70	0.70	17.80	4.37	0.08	1.97	5.28	4.44	1.29	0.16	
Uncertainty (95%)	0.50	0.02	0.20	0.07	0.00	0.04	0.09	0.14	0.02	0.02	
Suigetsu Lake SG14-0840											
Average (n=25)	74.15	0.47	13.54	2.33	0.11	0.61	2.81	4.57	1.22	0.08	
2σ	0.45	0.06	0.36	0.23	0.11	0.12	0.21	0.30	0.10	0.05	



Figure 1

203x122mm (300 x 300 DPI)







Figure 4

194x78mm (300 x 300 DPI)