

United States Department of the Interior

U.S. GEOLOGICAL SURVEY VOLCANO SCIENCE CENTER

4230 University Drive, Suite 100 Anchorage, Alaska 99508

MEMORANDUM

To: Richard Staff

From: Kristi L. Wallace

Subject: REVIEW COMMENTS FOR GEOCHRONOLOGY MANUSCRIPT:

Late Holocene cryptotephra from Cascade Lake, Alaska: supporting data for a 21,000-year multi-chronometer Bayesian age model by Lauren Davies, Britta Jensen, and Darrell Kaufman

July 26, 2021

Richard,

Below, are my review comments on the subject manuscript. All of my specific comments and suggestions for revisions are written directly on the manuscript that were done digitally using the Adobe Acrobat review tool on the PDF file that was made available to me. I have attached the commented version to the referee comment record.

GENERAL COMMENTS: This is a very good paper and makes use of ensemble modelling using multiple chronometers to improve an age model. An impressive amount of work has gone into developing an age model for this Arctic lake sequence and I commend the authors for their diligent work. I am not an expert in age models, so all of my comments are related to the tephra-related aspects of this manuscript. The identification and sampling and geochemical analysis of the tephras is robust and follows best practice guidelines (<u>https://doi.org/10.5281/zenodo.3866266</u>). In theory tephrochronology can be a very powerful tool for precisely and accurately dating events. My most significant concern is the fact that there is not a robust suite of reference materials from Alaska sources from which to make confident correlations. Considering the general lack of glass geochemical data and fully characterized tephra data for Alaska tephra it makes sense to attempt correlations with what is available and to draw geochemical fields for volcano-like geochemistry but I worry that false correlations using limited data may propagate errors into the literature by refining age models using low-confidence correlations.

I agree with the interpretations of the authors regarding pure deposits (representing primary eruptions) and mixed deposits (likely representing multiple eruptions) but there needs to be more discussion about homogenous vs heterogenous glasses so make this point as not many Alaska eruptions are homogenous. There is no discussion about correlation techniques in the methods or elsewhere – are all of the correlations based on glass geochemistry alone? How do the glass shard morphologies compare, glass characteristics (microlitic, clean etc.)? Based on the general lack of glass reference data it is hard to make a confident case for correlation without a multiparameter approach (using multiple sample characteristics) and I worry that a geochemistry alone is not very robust – a good starting point but in some cases not strong enough to confidently link to a known dated event. The manuscript would benefit from more discussion regarding the limited suite of reference materials available and evaluation of the quality of the correlations made. The Aniakchak CFE II and White River Ash (WRA) cases are more compelling based on there being more published data to support this correlation (although it is not clear why distal samples of WRA would have higher wt. % SiO2...) but I am skeptical about the Ruppert correlations as it implies an unknown source that erupted in the late Holocene but that has only been identified as cryptotephra in very distal areas and a very large footprint yet no visible layers or source has been identified. This would make sense where ice has removed older records, but this is a young deposit where preservation should not be a problem and we should see this layer more often in Holocene sediment sequences. If there were more discussion regarding the "robustness" of each correlation it would allow the reader to evaluate if the age model is really improved by the addition of the tephra correlations. I suggest the authors review the community established best practice recommendations for correlation (https://doi.org/10.5281/zenodo.3866266 -Tephra Correlation Best Practice Guidelines v3.xlsx, specifically tab F) to help further discuss the

quality or confidence of their correlations. Other than these over arching comments I believe the authors to be very careful in their evaluation of the available tephra data and think the manuscript would be improved by an evaluation of the quality of correlations that highlights the current realities in available data.

SPECIFIC COMMENTS & TECHNICAL CORRECTIONS: All such comments are documented on the reviewed version of the PDF of the manuscript and not listed here.

If you have any questions about this review please do not hesitate to contact me,

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Late Holocene cryptotephra from Cascade Lake, Alaska: supporting data for a 21,000-year multi-chronometer Bayesian age model

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- 10 Abstract. Multiple chronometers can be employed for dating Holocene palaeoenvironmental records, each with its own inherent strengths and weaknesses. Radiocarbon dating is one of the most widely used techniques for producing chronologies, but its application at high-latitude sites can be problematic. Here, cryptotephra identified in the Late Holocene portion of a core from Cascade Lake, Arctic Alaska, resolve a divergence
- 15 identified between radiocarbon and paleomagnetic secular variation (PSV) data in the top 1.5 m of the sediment sequence. Identifiable geochemical populations of cryptotephra are shown to be present in detectable concentrations in sediment from the north flank of the Brooks Range for the first time. Major element glass geochemical correlations are demonstrated between ultra-distal cryptotephra and reference samples from the Late Holocene caldera
- 20 forming eruption of Opala, Kamchatka, as well as three eruptions in North America: the White River Ash (northern lobe), Ruppert tephra and the Late Holocene caldera forming eruption of Aniakchak. The correlated ages of these cryptotephra support the PSV ages reported in Steen et al. (this volume) and provide evidence for an old-carbon effect in Cascade Lake. Chronological data from the Cascade Lake were then combined using a
- 25 Bayesian approach to generate an age-depth model that extends back to 21,000 cal yr BP.

1 Introduction

The accuracy and precision of ages and chronological models produced from sedimentary records directly impacts the utility and value of the associated proxies used for palaeoenvironmental reconstructions. In Arctic North America, the majority of Holocene to

30 late Pleistocene palaeoenvironmental reconstructions are produced from lake and peat deposits (e.g. Kaufman et al., 2016), and often rely on radiocarbon (¹⁴C) dating to develop age models.

However, there are several issues that can affect the application and interpretation of ¹⁴C ages in Arctic regions. Firstly, there may be a lack of organic material in lake sediment

35 cores or the terrestrial macrofossils that are often preferred for dating (e.g. Oswald et al., 2005; Turney et al., 2000) may be absent. This can be a particular problem for sediments that accumulated during colder periods. Secondly, high-latitude regions often have an abundance





of old carbon due to slow rates of decomposition in cold, typically nutrient poor soils (e.g. Gaglioti et al., 2014; Schuur et al., 2008), erosion from the surrounding sediments or

40 bedrock, and the reworking and redeposition of older, well-preserved macrofossils (e.g. Kennedy et al., 2010).

More broadly, ¹⁴C samples can also be affected by issues relating to sample selection, remobilisation, the hard-water effect and contamination (for a general review of these topics see, e.g. Olsson, 1974; Lowe and Walker, 2000). These factors can contribute to complicated

45 resulting age models for Arctic sediments that require careful independent verification. For example, the use of bulk sediments for dating has been shown to incorporate organic fractions of varying ages (e.g. Brock et al., 2011; Nelson et al., 1988) and hard-water effects have long been known in North American lakes (e.g. Abbott and Stafford, 1996; Karrow and Anderson, 1975; Moore et al., 1998).

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The combination of multiple chronometers has been successfully used to highlight differences between chronological methods and produce more accurate final age models for lacustrine and peat cores (Davies et al., 2018; Tylmann et al., 2016). Two additional techniques that have been applied in Arctic areas are discussed here - palaeomagnetic secular variation (PSV) and tephrochronology.

55 1.1 Palaeomagnetic chronologies

In recent years there have been an increasing number of studies looking to improve chronologies of late Quaternary Arctic sedimentary sequences by using palaeomagnetic data (e.g. Barletta et al., 2008; Deschamps et al., 2018; Lund et al., 2016; Ólafsdóttir et al., 2013). Sediments at high-latitude sites can be sensitive to palaeomagnetic secular variation (PSV) –

- 60 small directional changes in the geomagnetic field (Cox, 1970) that are preserved in sediment through the alignment of magnetic mineral grains with Earth's ambient field around the time of deposition. Tie-points, identified using peaks and troughs, can then be dated and used as correlative chronostratigraphic tools. These ages can be produced from both individual site measurements and geomagnetic model calculations. PSV correlation techniques are useful as
- 65 they can produce more frequent data points and be applied beyond the limits of ¹⁴C dating, or where organic material is not preserved. Their use, however, is limited geographically as high-latitude geomagnetic field dynamics are spatially complex (e.g. Stoner et al., 2013).

Steen et al. (this volume) report PSV-correlated ages for cores from Cascade Lake,

Alaska, that have substantial offsets during the Late Holocene from ¹⁴C ages from the same

70 sediment. Over the top 175 cm of the core, ¹⁴C ages are up to ~2000 years older than





palaeomagnetic correlated ages. When using multiple chronometers from the same sediment there is not always coherence or clear agreement between the results and additional chronological information is required. Here, a third chronostratigraphic technique – tephrochronology – was applied to Cascade Lake sediments to resolve the offset.

75 1.2 Cryptotephra chronologies

Cryptotephra - non-visible horizons of volcanic ash from distal sources – have been studied globally (see, e.g. Davies, 2015; Lowe et al., 2017) and are a useful chronostratigraphic tool (Pilcher et al., 1995; Plunkett, 2006; Swindles et al., 2010). Where correlations can be made with well-dated tephra (e.g. historical eruptions, or tephra preserved

- 80 within annually resolved records), tightly constrained associated ages can be included in agedepth models (e.g. Schoning et al., 2005). They can also be used as an independent test of other chronological methods applied to the same record (e.g. Davies et al., 2018; Oldfield et al., 1997).
- In Alaska and northern Canada the majority of tephra studies have been limited to areas where visible tephra are present and only a few studies have identified cryptotephra (e.g. de Fontaine et al., 2007; Lakeman et al., 2008; Monteath et al., 2017; Payne et al., 2008; Zoltai, 1989). However, there is significant potential for cryptotephra to be found in Alaska as it is downwind of a large number of volcanoes known to have been active over the Holocene. Of Alaska's 130 volcanoes and volcanic fields, 96 have been active either
- historically or within the Holocene (Miller et al., 1998) and historical observations show that more than 50 volcanoes have been active since ~ 1760 AD alone (Alaska Volcano)
 Observatory, 2016). Here, key tephra are from historical eruptions, or eruptions that produced regionally widespread tephra within Alaska and have precise age estimates (Davies et al., 2016).
- 95 While there are currently no published occurrences of Kamchatkan tephra within Alaska, the large number of Kamchatkan-Kurile volcanoes active in the Holocene can also be considered as a potential source of distal cryptotephra, given prevailing wind directions and the large number of recorded major explosive eruptions (e.g. Braitseva et al., 1997; Kyle et al., 2011; Ponomareva et al., 2017). Transcontinental distribution of tephra from non-super
- 100 eruptions has been established (e.g. Cook et al., 2018; Jensen et al., 2014), and Kamchatkansourced tephra have been traced to Greenland, Svalbard and the east coast of North America (van der Bilt et al., 2017; Cook et al., 2018; Mackay et al., 2016, Jensen et al., submitted).





Here, ages from Cascade Lake for three different chronostratigraphic techniques were visually compared and then modelled using Bayesian statistics to produce a composite agedepth model. Bayesian statistical techniques have been utilised in a wide range of fields to produce detailed age-depth models based on a relatively small number of dates (e.g. Christen et al., 1995; Litton and Buck, 1995) and, through their inclusion of additional (prior) information, they provide more precise interpolations than using raw dates alone (e.g. Blaauw

110 2 Materials and Methods

and Christen, 2005; Bronk Ramsey, 2008).

Cascade Lake (68°22'48" N, 154°38'00" W; 990 m asl) lies on the north-central slope of the Brooks Range, the northernmost mountain range in Alaska (Fig. 1). Overall, the Brooks Range is located almost entirely above the Arctic Circle and represents a significant topographic barrier that divides the climatic influences of the Arctic and Pacific. The lake has

115 an area of ~ 1 km² and a maximum depth of ~ 40 m in the main northwestern basin (Fig. 1b) with a total catchment size of ~10 km². It presently has no significant inflow and one small outflow, west to Kurupa Lake (~ 920 m asl).



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Figure 1: Location map showing Cascade Lake, coring sites, and other relevant locations and volcanoes mentioned in the text. Grey circles = active Holocene volcanoes (Global Volcanism Program, 2013); black triangles = volcanic sources mentioned in the text; grey shading = Brooks Range; star outlines = lakes mentioned in the text.

In 2013 sediment cores were collected from two sites at Cascade Lake using a percussion-piston coring system (long cores) and Aquatic Instruments universal corer (surface cores). Cores were split and described at the National Lacustrine Core Facility (LacCore) repository at the University of Minnesota, Twin Cities, and archive halves are





125 housed there. The 5.2-m-long composite sedimentary sequence, CASC-4A/2D, described by Steen et al. (this volume) is the focus of the age-depth model reported here, which extends down to the boundary with an underlying diamicton.

2.1 Previous geochronological data

2.1.1 Radiometric data

- 130 Radiometric data from Cascade Lake are detailed in full in Steen et al. (this volume) and summarised here in Table 1. Eleven AMS ¹⁴C samples analysed at the University of California-Irvine AMS Facility are reported. Samples consisted of terrestrial plant macrofossils, insect parts, resting eggs, and aquatic vegetation as available. The oldest sample analysed was from 350 cm composite depth, dating to ~15 cal ka BP. Six ²¹⁰Pb
- 135 measurements were made from the uppermost sediment at Cascade Lake, but as equilibrium (~142 yr BP) is reached within the top 4 cm of the cores these ages are not discussed in the context of the Holocene age models.
- Table 1: Radiometric ages from Cascade Lake (from Steen et al., this volume.). Ages are reported to the nearest whole year (^{210}Pb) or five years (^{14}C) * = ^{14}C ages rejected as outliers; † = samples from surface core CASC-4B, all other samples are140from CASC-4A.

(a) ²¹⁰Pb CRS ages

Composite depth (cm)	Age (yr)	Error (yr)
0-0.5	23	1
0.5-1	48	1
1-1.5	67	2
1.5-2.25	83	2
2.25-3	112	4
3-3.5	143	7
(b) 14C agas		

(b) ¹⁴C ages

Composite depth (cm)	Sample ID (UCIAMS #)	Age (¹⁴ C yr)	Error (¹⁴ C yr)	Material
2.6-4.6†	147384	170	30	Resting eggs, mixed aquatic fragments
5.5-7.5	134422*	1765	20	Insect remains, twigs, leaves, bryophyte, eggs
11-13†	147383	785	45	Leaf fragments, resting eggs, mixed aquatic fragments
30.5-32.5	131742	2825	25	Insect remains, moss fragments, resting eggs, fine unidentified pieces
85.75-87.75	128095	4160	120	Insect remains, twigs, leaves, bryophyte, resting eggs
138-140	131743	5085	20	Insect remains, moss fragments, resting eggs, fine unidentified pieces
197-199	131744	6485	25	Insect remains, moss fragments, resting eggs, fine unidentified pieces
233.5-235.5	134423	8270	35	Insect remains, twigs, leaves, resting eggs, fine unidentified pieces
245-248	128096*	13200	450	Insect remains, aquatic vegetation, twigs, resting eggs
303-304	131745	9875	35	Insect remains, moss fragments, resting eggs, fine unidentified pieces
348.5-351	137726	12690	150	Insect fragments, twig, leaf fragments





2.1.2 PSV ages

145 A composite inclination record and associated age model for Cascade Lake (Steen et al., this volume) was produced using inclination age control points (tie-points) matched to two geometric field models (CALS10k.1b, Korte et al., 2011; pfm9k.1b, Nilsson et al., 2014) and a palaeomagnetic record from nearby Burial Lake (~ 200 km west along the Brooks Range; Dorfman, 2013). PSV scenario 1 (PSV-1) was produced using 14 inclination tie-

150 points in total (Table 2) and successfully extends the age model for Cascade Lake back to ~21 ka.

Table 2: The PSV-1 inclination age model data, with chronological tie-points calculated for Cascade Lake with Burial Lake, CALS10k1b and pfm9k1b. Reported ages are rounded to the nearest five years. See Steen et al. (this volume) for full details.

Tie Point	Composite depth (cm)	Burial Lake Age (cal yr BP)	Burial Lake Age 95% CI (cal yr BP)	CALS10k1b Age (yr BP)	+/-	pfm9k1b Age (yr BP)	+/-
I1	60	2270	1790-2765	2065	500	1955	500
12	80	2755	2440-3045	2945	500	2635	500
13	155	4810	4285-5345	4145	500	4195	500
I4	177	7275	7140-7415	5705	500	5435	500
15	189	-	-	6165	500	6305	500
16	203	-	-	6585	500	7145	500
I7	228	9880	9470-10280	7345	500	7655	500
18	246	-	-	8185	500	8345	500
19	284	11935	11440-12430	9425	500	-	-
I10	357	15455	14745-16155	-	-	-	-
I11	419	17055	16660-17430	-	-	-	-
I12	427	17415	16860-17960	-	-	-	-
I13	454	18130	17360-18930	-	-	-	-
I14	509	20520	19370-21630	-	-	-	-

- 155 The ages of tie-points from the geometric field models are based on a database of 75 selected sedimentary palaeomagnetic records from the SED12k data compilation (Donadini et al., 2010; used by CALS10k.1b, Korte et al., 2011). The database was further parsed to exclude bulk ¹⁴C samples, archaeomagnetic data with large temporal uncertainties, and palaeomagnetic behaviour incompatible with the majority of records during the Holocene
- 160 (pfm9k.1b, Nilsson et al., 2014). Both models have reported estimated temporal resolutions of \pm 500 a. Burial Lake tie-point ages and errors are derived from the ¹⁴C age model of the sediment cores (Dorfman, 2013), which is based on terrestrial macrofossils and shows remarkably linear sediment accumulation over ~ 17 ka cal BP.

2.3 Cryptotephra analysis

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Cryptotephra analyses are reported here from the past 4 ka, as a large number of the most-well-known, dated, and widely distributed tephra in Alaska were erupted during this time period (Davies et al., 2016). This is also the interval when the ¹⁴C ages in Cascade Lake





cores appear to be too old relative to the expected ages of the PSV features and therefore where tephra have significant potential to validate and improve a final age-depth model.

- 170 No visible tephra were located in cores from Cascade Lake (in fact, no visible beds are known north of the Brooks Range); targeted cryptotephra analyses were undertaken using contiguous 1-cm-thick subsamples from 1.42 m composite depth to the surface. Standard methods (e.g. Blockley et al., 2005) were used to produce glass shard concentration profiles throughout the two core sections and the heavy liquid, Lithium Heteropolytungstate (LST),
- 175 was used for density separations. Glass shards for geochemical analysis were re-extracted from peaks in shard concentration using heavy liquid separation and samples were mounted in an epoxy puck and polished to expose glass surfaces before being carbon coated prior to electron probe microanalysis (EPMA). New data are reported here from glass shards analysed on a JEOL 8900 Superprobe at the University of Alberta by wavelength dispersive X-ray
- 180 spectroscopy (WDS) following established protocols (e.g. Jensen et al., 2008, 2019).

A standard suite of ten elements (Si, Ti, Al, Fe, Mn, Mg, Ca, Na, K, Cl) was measured using a 5 μ m beam with 15 keV accelerating voltage and 6 nA beam current. This focussed beam (usually 10 μ m is utilised) can result in Na loss in more sensitive glasses (e.g. Jensen et al., 2019; Foo et al., 2020). However, where intensity data loss does occur, it has

- 185 been shown that empirical corrections can be applied if the data demonstrate linear variance over time (Nielsen and Sigurdsson, 1981). Here Na, and if necessary, Si, were corrected for Time Dependent Intensity (TDI) loss (or gain) using a self-calibrated correction with Probe for EPMA software (Donovan et al., 2015).
- Two secondary standards of known composition were run concurrently with all tephra samples: ID 3506, a Lipari rhyolite obsidian, and a reference sample of Old Crow tephra, a well-characterised, secondarily hydrated tephra bed (e.g. Kuehn et al., 2011). All results were normalised to 100% and are presented as weight percent (wt%) oxides. New major-element geochemical data and associated standard measurements, as well data points for relevant reference material (analysed concurrently, where possible), are reported in the Supplementary
- 195 Information (Tables S1, S2).

2.4 Bayesian age modelling

Three steps are detailed here for identifying and resolving problematic chronometer offsets using the data from Steen et al. (this volume) and new cryptotephra correlated ages. Firstly, ages that were obviously out of stratigraphic sequence were rejected previously by

200 Steen et al. (this volume). Secondly, OxCal's Poisson process model (P_Sequence, Bronk



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Ramsey, 2008) was used to construct independent models for each chronometer. These were then visually compared to detect offsets between the dating methods. This is more effective than using statistical techniques as a first approach as they can be biased by datasets with high numbers of dates and tight distributions. Here, cryptotephra isochrons were used as independent checks for the other chronological methods, e.g. to identify ¹⁴C outliers.

Finally, the resulting chronological data were combined in one composite P_Sequence model (OxCal v4.4; Bronk Ramsey, 2009). This set-up allows variable accumulation rates; here the k parameter (deposition events defined as increments per unit length, controlling model rigidity and resolution) was set as variable rather than fixed to increase model

210 flexibility (Bronk Ramsey, 2013). General (Student's t) outlier analysis was used to identify any remaining anomalous ages in the parsed dataset. All ages were given the prior probability of 5% of ages being incorrect; if an age needs to be shifted substantially (by more than two standard deviations) to fit the resulting age-depth model it was identified as an outlier and downweighed in the process (Blockley et al., 2007).

215 **3 Cryptotephra data**

Glass shards were present in 75% of the samples analysed here. The composite shard concentration profile for the 1.42 m of counted samples is shown in Fig. 2. Twenty-eight peaks were chosen for geochemical analysis based on the relative abundances of shards counted at those depths. For each sample, geochemical analyses were performed on single grains, but 15 of the peaks chosen resulted in fewer than five shards exposed on the EPMA

puck surface. This is likely due to the relatively low concentrations of glass present overall.

Of the remaining 13 samples, five have dominant unique geochemical populations (i.e. single eruptions are strongly represented), six have multiple identifiable trends/populations (representing an amalgamation of shards from multiple eruptions) and two

225 have sparse shards with no discernible geochemical trends. Table 3 and Fig. 3 show the samples analysed, the average major element EPMA data for identified geochemical populations and any geochemical correlations to known eruptions with associated chronological data or similarities to known volcanic sources. Normalised single point major element EPMA datasets and associated standard analyses are provided in Tables S1 and S2.







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Figure 2: Cascade Lake core CASC-4A/2D multi-method chronological controls. (a) The composite depths of radiometric ages (¹⁴C and ²¹⁰Pb; Table 1), PSV-1 tie-points from three models (Table 2) and correlated cryptotephra ages (Table 4). The shaded grey area shows the depth interval of core sampled for cryptotephra analysis (expanded in panel b). (b) Glass shard concentration counts produced down to 145 cm, and the composite depths of analysed glass peaks. Circles = <5 points analysed; triangles =>5 points analysed; filled grey triangles have correlated ages that are used in the age-depth model.

3.1 Unique glass populations

Five samples contained glass shards that show dominant unimodal rhyolitic populations based on between 10 and 38 geochemical analyses. These are interpreted as primary tephra-fall events relating to contemporaneous eruptions (i.e. they show no evidence

of secondary reworking). Four of these five samples can be used as isochrons as they correlate to reference material from known and dated eruptions (University of Alberta reference collection samples, Fig. 3; details provided in Tables 3 and S1). Key information regarding these eruptions and the tephra deposits are summarised in Table 4. Samples are discussed here individually from oldest to youngest and age estimates are given as two sigma calibrate to ge ranges unless otherwise stated.



Table 3: Average major element geochemical data for identifiable populations of analysed tephra samples and suggested correlations. (###) = standard deviation; FeOt = total iron oxide as FeO: H_2O_4 = water by difference. (a) Samples used here as tie-points; (b) Reference material analysed at the University of Alberta, for full details regarding the original sample details please see listed references. (c) Samples with multiple populations or too few points to use as tie-points. Only groups of 3 or more analyses are shown here – for full details see Table SI.

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Correlation	Omolo (OB)	Opaia (Or)		Mt. Churchill –	White River Ash	(northern lobe,	WRAn)			ruppert	IImprove	UIIKIIOWII	A winkohole CEE II	AIIIAKUIAK UFE II
u	10		10		12		22		36		12		11	
H_2Od	3.90	(1.31)	2.23	(1.58)	2.25	(1.89)	2.24	(1.71)	0.48	(1.24)	0.86	(1.18)	0.54	(0.93)
Total	100.00	(0.00)	100.00	(0.00)	100.00	(0.00)	100.00	(0.00)	100.00	(0.00)	100.00	(0.00)	100.00	(0.00)
C	0.12	(0.02)	0.34	(0.03)	0.30	(0.04)	0.31	(0.04)	0.25	(0.03)	0.24	(0.02)	0.20	(0.02)
K_2O	3.83	(0.22)	3.35	(0.25)	3.61	(0.19)	3.49	(0.25)	16.1	(60.0)	2.81	(0.08)	2.91	(0.12)
Na_2O	4.39	(0.23)	4.07	(0.27)	3.65	(0.16)	3.84	(0.30)	4.81	(0.26)	4.33	(0.23)	5.53	(0.23)
CaO	0.77	(0.04)	1.57	(0.13)	0.91	(0.18)	1.21	(0.37)	2.18	(0.14)	2.06	(0.18)	1.64	(0.04)
$0^{5}M$	0.12	(0.02)	0.34	(0.08)	0.15	(0.04)	0.24	(0.11)	0.46	(0.06)	0.49	(0.09)	0.52	(0.03)
OuM	80.08	(0.03)	90'0	(0.01)	0.04	(0.02)	0.05	(0.02)	0.11	(0.02)	0.08	(0.02)	0.13	(0.02)
FeOt	0.60	(0.09)	1.49	(0.20)	1.06	(0.10)	1.26	(0.26)	1.96	(0.15)	1.89	(0.15)	2.34	(0.07)
Al_2O_3	13.27	(0.12)	14.12	(0.32)	12.43	(0.42)	13.20	(0.94)	13.91	(0.23)	13.72	(0.46)	15.19	(0.15)
TiO_2	0.11	(0.03)	0.22	(0.06)	0.16	(0.05)	0.19	(0.06)	0.30	(0.05)	0.40	(0.03)	0.48	(0.03)
SiO_2	76.73	(0.26)	74.52	(0.58)	77.76	(0.69)	76.29	(1.77)	74.16	(0.63)	74.04	(0.95)	71.10	(0.29)
Popn			а		q		all		1		-		a	
Sample #	CL-37		CL-48						CL-74		CL-96		CL-105	

Reference material analyses from the University of Alberta (q

Site	Sample ID	SiO ₂	TiO ₂	Al ₂ O ₃	FeOt	0uM	MgO	CaO	Na ₂ O	K_2O	CI	Total	H_2O_d	u	Correlation	Reference sample details
Duke River	11011	74.32	0.21	14.18	1.54	0.06	0.32	1.67	4.11	3.26	0.33	100.00	2.40	55	White River Ash	(LUUC) 405401
Fan, YT	UA 1044	(0.63)	(0.05)	(0.33)	(0.15)	(0.03)	(0.05)	(0.16)	(0.18)	(0.13)	(0.04)	(0.00)	(1.15)		(northern lobe)	
	UT 1480,	73.95	0.21	14.38	1.50	0.06	0.35	1.77	4.29	3.23	0.34	100.00	2.82	31		
Sixtymile	Sample 16	(2.06)	(0.07)	(0.99)	(0.31)	(0.03)	(0.14)	(0.49)	(0.31)	(0.19)	(0.04)	(0.00)	(0.78)		White River Ash	Preece et al.
KIVET AFEA,	UT 1482,	73.65	0.23	14.50	1.59	0.06	0.38	1.83	4.26	3.25	0.34	100.00	3.06	34	(northern lobe)	(2014), this paper
11	Sample 17	(1.80)	(0.06)	(0.87)	(0.29)	(0.02)	(0.12)	(0.46)	(0.26)	(0.19)	(0.05)	(0.00)	(0.88)			
	11A 12002	59.13	1.40	16.45	7.50	0.22	2.97	6.06	4.61	1.57	0.13	100.00	2.30	17	и -111	
Zagoskin	UA 10024	(0.97)	(0.07)	(0.16)	(0.64)	(0.03)	(0.22)	(0.40)	(0.26)	(0.11)	(0.02)	(0.00)	(1.07)		Aniakcnak CFE II	Ager (2003);
Lake, AK	11071 11	71.07	0.50	15.19	2.55	0.14	0.51	1.78	5.07	3.05	0.19	100.00	2.48	32	(anuesue,	Davies et al.
	UA 10020	(0.52)	(0.05)	(0.28)	(0.2)	(0.03)	(0.08)	(0.19)	(0.29)	(0.13)	(0.02)	(0.00)	(1.56)		111youacite)	(0107)
Ruppert	L33C VII	74.08	0.30	13.96	2.00	0.10	0.46	2.18	4.80	1.93	0.22	100.00	0.53	17	1	Monteath et al.
Lake, AK	1007 VO	(0.39)	(0.05)	(0.16)	(0.12)	(0.03)	(0.04)	(0.10)	(0.14)	(0.10)	(0.02)	(0.00)	(06.0)		ruppert	(2017)
Southern	7066 VII	76.65	0.12	13.31	0.69	0.11	0.12	0.77	4.24	3.91	0.11	100.00	2.65	30	Omole (Illinee III)	Andrews et al.
Kamchatka	0075 VO	(0.20)	(0.04)	(0.12)	(0.03)	(0.03)	(0.02)	(0.03)	(0.11)	(0.0)	(0.02)	(0.00)	(0.71)		Opara (Filase III)	(2018)







Table 3: c)	cont. Other s	samples a	ınalysed											
Sample	Popn	Si02	Ti02	AI203	FeOt	MnO	MgO	CaO	Na2O	K20	C	Total	H2Od	u
CL-0	a	71.02	0.48	14.90	2.36	0.16	0.50	1.67	5.64	3.09	0.23	100.00	1.46	7
		(0.29)	(0.03)	(0.15)	(0.08)	(0.03)	(0.02)	(0.05)	(0.26)	(0.08)	(0.03)	(0.00)	(1.26)	
	p	74.42	0.16	13.99	1.28	0.05	0.32	1.57	4.63	3.33	0.33	100.00	3.12	3
		(1.46)	(0.04)	(0.76)	(0.28)	(0.02)	(0.12)	(0.29)	(0.13)	(0.12)	(0.01)	(0.00)	(1.59)	
	c	74.68	0.32	13.68	2.02	0.09	0.46	2.22	4.43	1.94	0.21	100.00	1.78	6
		(0.84)	(0.04)	(0.67)	(0.16)	(0.02)	(0.04)	(0.19)	(0.34)	(0.05)	(0.06)	(0.00)	(0.62)	
	q	75.63	0.26	13.51	1.22	0.05	0.31	1.30	4.54	3.05	0.15	100.00	2.32	6
		(1.35)	(0.10)	(0.44)	(0.41)	(0.01)	(0.10)	(0.38)	(0.45)	(0.19)	(0.07)	(00.0)	(1.05)	
	e	76.91	0.33	12.27	1.57	0.03	0.26	1.16	4.33	2.97	0.21	100.00	1.61	3
		(0.45)	(0.04)	(0.09)	(0.11)	(0.01)	(0.02)	(0.02)	(0.33)	(0.01)	(0.01)	(0.00)	(0.48)	
CL-2	а	71.00	0.49	15.00	2.48	0.14	0.52	1.68	5.42	3.11	0.22	100.00	1.35	14
		(0.59)	(0.10)	(0.23)	(0.34)	(0.03)	(0.10)	(0.20)	(0.16)	(0.14)	(0.03)	(0.00)	(0.78)	
	q	73.73	0.21	14.46	1.51	0.04	0.37	1.77	4.52	3.13	0.33	100.00	2.81	5
		(0.35)	(0.02)	(0.23)	(0.05)	(0.03)	(0.03)	(0.10)	(0.12)	(0.08)	(0.02)	(0.00)	(1.14)	
	c	74.17	0.32	13.75	1.94	0.12	0.45	2.20	4.82	2.03	0.26	100.00	1.14	4
		(0.45)	(0.03)	(0.16)	(0.08)	(0.02)	(0.01)	(0.18)	(0.23)	(0.04)	(0.01)	(0.00)	(0.74)	
	q	75.86	0.25	13.18	1.56	0.07	0.31	1.66	4.96	2.01	0.17	100.00	2.04	4
		(0.27)	(0.04)	(0.14)	(0.05)	(0.03)	(0.04)	(0.11)	(0.14)	(0.03)	(0.02)	(0.00)	(0.28)	
	e	76.90	0.26	12.63	1.19	0.07	0.22	1.05	4.20	3.35	0.18	100.00	3.07	5
		(1.24)	(0.16)	(0.35)	(0.52)	(0.02)	(0.12)	(0.38)	(0.23)	(0.34)	(0.11)	(0.00)	(0.93)	
CL-4	а	66.61	0.77	15.68	4.45	0.13	1.19	3.50	4.88	2.63	0.20	100.00	1.79	4
		(1.64)	(0.26)	(0.22)	(1.07)	(0.05)	(0.14)	(0.44)	(0.3)	(0.12)	(0.11)	(0.00)	(0.79)	
	p	70.86	0.49	15.00	2.47	0.14	0.51	1.69	5.50	3.17	0.22	100.00	1.09	15
		(0.86)	(0.04)	(0.17)	(0.35)	(0.03)	(0.08)	(0.18)	(0.36)	(0.3)	(0.02)	(0.00)	(0.91)	
	c	76.71	0.12	13.26	0.61	0.12	0.09	0.72	4.25	4.07	0.10	100.00	2.78	3
		(0.16)	(0.04)	(0.02)	(0.17)	(0.03)	(0.06)	(0.03)	(0.18)	(0.36)	(0.04)	(0.00)	(0.86)	
CL-7	а	70.93	0.48	15.03	2.36	0.13	0.59	1.88	5.40	3.03	0.20	100.00	1.04	5
		(0.19)	(0.02)	(0.12)	(0.09)	(0.04)	(0.13)	(0.38)	(0.45)	(0.20)	(0.03)	(0.00)	(0.15)	
	q	75.83	0.23	13.31	1.55	0.07	0.33	1.67	4.82	2.06	0.16	100.00	1.63	6
		(0.61)	(0.05)	(0.39)	(0.04)	(0.03)	(0.08)	(0.24)	(0.23)	(0.1)	(0.04)	(0.00)	(0.62)	
CL-31		77.92	0.13	12.22	1.06	0.06	0.15	0.85	3.84	3.53	0.31	100.00	1.97	6
		(0.7)	(0.02)	(0.55)	(0.1)	(0.03)	(0.04)	(0.23)	(0.34)	(0.26)	(0.01)	(0.00)	(1.16)	
CL-61	a	71.23	0.43	15.05	2.39	0.15	0.44	1.57	5.50	3.09	0.20	100.00	-0.28	3
		(0.80)	(0.04)	(0.28)	(0.07)	(0.02)	(0.05)	(0.13)	(0.56)	(0.19)	(0.02)	(0.00)	(0.28)	
	q	77.70	0.21	12.72	1.23	0.05	0.34	2.05	4.01	1.62	0.10	100.00	0.44	4
		(0.33)	(0.02)	(0.20)	(0.07)	(0.03)	(0.01)	(0.05)	(0.16)	(0.09)	(0.02)	(0.00)	(1.40)	
CL-105	q	71.81	0.51	14.75	2.32	0.10	0.41	1.43	4.85	3.69	0.17	100.00	0.37	4
		(0.55)	(0.09)	(0.03)	(0.22)	(0.02)	(0.05)	(0.13)	(0.22)	(0.04)	(0.01)	(0.00)	(0.14)	





255 **Table 4:** Cascade Lake cryptotephra and their suggested correlative eruptions. Age estimates for the core depth of the cryptotephra from Steen et al. (this issue) are compared with published ages for the listed eruptions. Bayesian modelled ages for both Aniakchak CFE II and Opala are updated here using OxCal and IntCal20.

Sample (Lab #)	Suggest	ed correlation	Cascade Lake age estimates (95% range)			Age estimate fo	or correlated erupt	ion
	Tephra	Source volcano	¹⁴ C (cal yr BP)	PSV-1 (yr BP)	95% range (cal yr BP)	Method	Deposit type	Age estimate reference(s)
CL-37 (UA3721)	OP	Opala, Kamchatka	3180- 2300	1720- 990	1395-1305	Calibrated ¹⁴ C (IntCal20)	Visible tephra (Kamchatka)	Braitseva et al. (1995); updated here using IntCal20
CL-48 (UA3730)	WRAn	Mt. Churchill, Alaska	3895- 2935	2110- 1360	1689-1540	Calibrated ¹⁴ C (IntCal20)	Visible tephra (Alaska, Yukon)	Reuther et al. (2020)
CL-74 (UA3733)	Ruppert	Unknown (likely Alaska)	4920- 4145	2795- 2280	2800-2130	Calibrated ¹⁴ C (IntCal20)	Distal cryptotephra (four bogs - Newfoundland, Canada; Maine, Michigan, New York, USA)	Jensen et al. (submitted)
CL-96 (UA3735)	-	Unknown	5345- 4840	3355- 2600	-	-	-	-
CL-105a (UA3736)	CFE II	Aniakchak, Alaska	5460- 5030	3525- 2675	3590-3545	Calibrated ¹⁴ C (IntCal20) & ice core	Visible tephra (Alaska) and cryptotephra (Alaska, USA; Newfoundland, <u>Canada</u>)	Davies et al. (2016); updated here using IntCal20
					$\frac{3572 \pm 8}{(-20 \pm 5)}$	GICC05 (with correction)	Distal cryptotephra (NGRIP, Greenland)	Vinther et al. (2006); Adolphi & Muscheler (2016); Pearce et al. (2017)







Figure 3: Geochemical biplots showing major element data for the five unique populations of cryptotephra glass identified from Cascade Lake sediment, and data for reference material where relevant. Points for CL-105b are also plotted, for reference. See Table 3 for sample details and Table S1 for point data.

260 3.1.1 CL-105 (Aniakchak Caldera Forming Eruption II)

CL-105, a peak concentration of 12 shards/gram, is a geochemical match for the rhyodacite population of the widespread Late Holocene caldera forming eruption of Aniakchak (CFE II) (Fig. 3; Bacon et al., 2014; Neal et al., 2001; Riehle et al., 1987). Tephra from this eruption have been found visibly across southern and western Alaska, and as

^{cryptotephra in the Bering Sea, Yukon, Newfoundland and Greenland (Davies, 2018; Denton and Pearce, 2008; Pearce et al., 2017, 2004; Ponomareva et al., 2018; Pyne-O'Donnell et al., 2012). A second population of four points was also identified in this sample (CL-105b, Table 3c), however it is unclear if these represent a separate event or alkali loss from the main population.}





- 270 Chronologically, Aniakchak CFE II has been dated with radiocarbon from sequences with visible tephra and distal lakes and peat bogs with correlated cryptotephra, as well as with a precise ice-core model age estimate from distal cryptotephra identified in Greenland ice cores. The latter is supported using geochemically correlated glass shards as well as sulphate peaks and tree ring perturbations recorded in this interval (Coulter et al., 2012;
- McAneney and Baillie, 2019; Pearce et al., 2004). Glass shards correlated to the eruption in two NGRIP intervals have overlapping associated ice-core modelled ages of 3594-3589 yr BP (1641-1639 BCE QUB-1198, 1644-1643 BCE QUB 1201; Coulter et al., 2012; Vinther et al., 2006). When a correction factor of -19 ± 3 a (Adolphi and Muscheler, 2016) is applied to the GICC05 chronology, the resulting NGRIP modelled age for the eruption is

 $280 \qquad 3572 \pm 4 \text{ cal yr BP} \text{ (Pearce et al., 2017)}.$

Here we report updated modelled eruption ages produced using the Tau_Boundary function in OxCal v.4.4 with IntCal20 (following Davies et al., 2016; Fig. S1, see Table S4 for details). The ice-core chronology age discussed above is only compatible with published ¹⁴C ages if two of the three ¹⁴C ages that underlie the tephra in an exposed peat section in

- 285 northwest Alaska (Blackford et al., 2014) are removed as outliers. This is unexpected because the peat section is one of the most precisely dated terrestrial sequences for Aniakchak CFE II, with six samples analysed at 0.5 cm increments over 3 cm immediately surrounding the tephra. While there are no obvious reasons for disregarding these two ages, beyond the disagreement with the ages from the ice cores, in this instance it seems pertinent to do so.
- Modelled Tau_Boundary estimates for the eruption age are: a) 3545-3425 cal yr BP when all ¹⁴C dates are included, b) 3610-3450 cal yr BP with two ¹⁴C dates removed, and c) 3590-3545 cal yr BP including all but two ¹⁴C dates and the NGRIP ice core chronology age (Fig. S1). At Cascade Lake, using either the ice core chronology age estimate of 3572 ± 4 cal yr BP (Adolphi and Muscheler, 2016; Pearce et al., 2017) or the Tau Boundary model age (c,
- 295 above) for Aniakchak CFE II shows that while neither estimated age for this depth from Steen et al. (this volume) overlaps here, the PSV-1 age model is substantially closer than the ¹⁴C age model (Table 4).





3.1.2 CL-96 (unknown)

CL-96 represents a small peak of only four shards/gram but yielded 10 analytical
points that have relatively high values for wt% TiO₂, FeO and CaO (Table 3a). These analyses are similar to CL-74 for many major elements, but have substantially higher wt.% K₂O (2.81 wt.% average vs. 1.91 wt.%, respectively). The shards are likely from a source in Alaska and the Aleutian Arc and are similar to published average analyses for glass from the Katmai volcanic cluster (Fierstein, 2007) but cannot be directly correlated here to a particular vent or eruption. Therefore, there are no associated age estimates that can be used here to compare with other Cascade Lake chronometers.

3.1.3 CL-74 (Ruppert tephra)

CL-74 has a shard concentration peak of 10 shards/gram but a disproportionately high number of analyses (38) when compared to other samples. This rhyolitic population of platy
and cuspate shards has distinctly low wt.% K₂O values (~2.0%) compared to other tephra from Alaska and is a geochemical match for the Ruppert tephra. This tephra was first identified in Newfoundland (NDN-230; Pyne-O'Donnell et al., 2012) and tentatively correlated to Augustine G, although this is now known to be incorrect (Blockley et al., 2015;

Monteath et al., 2017). While it is geochemically similar to glass from Mt. Augustine, no

315 proximal correlative is currently known. The tephra was later found in, and subsequently named after, Ruppert Lake, directly south of Cascade Lake on the southern slope of the Brooks Range (Monteath et al., 2017) and has also been identified in peatlands in the Yukon (Davies, 2018) and eastern USA (Jensen et al., submitted).

Chronologically, Ruppert Lake's ¹⁴C age model shows evidence of old carbon
 contamination (Monteath et al., 2017) but Jensen et al. (submitted) produced a modelled two-sigma age of 2800-2130 cal yr BP using ¹⁴C ages from four distal peat bogs (located in Newfoundland, Canada; Maine, Michigan and New York, USA). This agrees well with Steen et al.'s (this volume) PSV-1 age estimate for this depth (Table 4).

3.1.4 CL-48 (White River Ash, northern lobe)

325 CL-48 is the largest glass concentration peak of the pre-19th century sequence, with 36 shards/gram. These pumaceous rhyolitic shards are geochemically similar to the White





River Ash, which comprises two Late Holocene eruptions from Mt. Churchill (Lerbekmo, 2008; Preece et al., 2014). Major element glass geochemical data for these eruptions are very similar (with substantial overlap) but given the broad range of wt.% SiO₂ values and bimodal

- 330 geochemistry of CL-48 shards, it likely correlates with the older northern-focused eruption (WRAn). The tephra from this eruption is more geochemically diverse than that of the younger eastern lobe (Davies et al., 2019) and is preserved as a visible bed in sediment deposits north of the vent in Alaska and the Yukon. Reference geochemical data from three WRAn samples in the Yukon (Jensen, 2007; Preece et al., 2014) are plotted in Fig. 3 to
- 335 demonstrate the observed variability; distal correlatives trend towards higher wt.% SiO₂ values compared to proximal samples (Davies et al., 2019).

WRAn has a recently updated modelled two-sigma ¹⁴C age of 1689-1560 cal yr BP (Reuther et al., 2020). This is slightly younger than previous published estimates (e.g. 1805-1605 cal yr BP, Davies et al., 2016) as the new ages and modelling methods reported by

340 Reuther et al. (2020) better constrain the eruption, which occurred at a time when there is a fluctuation in the ¹⁴C calibration curve. This age is in good agreement with Steen et al.'s (this volume) PSV-1 age estimate for this depth (Table 4).

3.1.5 CL-37 (OP tephra)

- CL-37 is the second largest pre-19th century peak, with 28 shards/gram. This rhyolite,
 is distinctive from published analyses of glass from Alaska, with notably low wt.% FeOt (average 0.60%) and CaO (average 0.77%). This characteristic geochemical signature has been observed in some volcanic glasses from Kamchatka (e.g. Portnyagin et al., 2020). CL-37 is shown here to correlate with the Late Holocene caldera forming eruption of Opala (OP), Kamchatka (e.g., Andrews et al., 2018; Braitseva et al., 1995, 1997; Kyle et al., 2011;
- 350 Melekestsev et al., 1992; Plunkett et al., 2015). CL-37 is the first ultra-distal correlation of glass from this eruption outside of Kamchatka.

Here we report an updated modelled eruption age for OP of 1395-1305 cal yr BP (Fig. S2). This was produced using the Tau_Boundary function in OxCal v4.4 with IntCal20 following the methodology of Davies et al. (2016) with ¹⁴C ages reported in (Braitseva et al.,

355 1995) (Table S4). This is in good agreement with previous published ages for the eruption and with Steen et al.'s (this volume) PSV-1 age estimate for this depth (Table 4).





3.2 Multimodal/mixed glass populations

Glass shards from six of the remaining analysed shard-frequency peaks have mixed or multimodal geochemical data and two have scattered results with no discernible trend. Higher

- 360 levels of background shards are present from 35 cm to the surface, and the geochemical 'noise' is also particularly evident in the youngest samples, with all peaks analysed in the past millennium showing either multimodal or scattered data. Geochemical biplots for samples with multiple populations of only a few shards (CL-0, -2, -31, -61) are shown in full in Fig. S3.
- CL-61 is the only analysed mixed sample that pre-dates the past millennium, located between the Ruppert (CL-74, 2800-2130 cal yr BP) and WRAn (CL-48, 1689-1560 cal yr BP) tephras. It contains a few shards that are similar to the rhyodacite from Aniakchak volcano and also Augustine tephra (Fortin et al., 2019; Waitt and Begét, 2009), but while these volcanoes have known activity at this time (e.g. Bacon et al., 2014; Waitt and Begét,

370 2009) there are not enough analyses available for a definite correlation.

Of the six mixed samples, only two – CL-4 and CL-7 – have populations that can be identified as dominant from the analyses presented here. Rhyodacitic and dacitic glass shards from these samples overlap geochemically with reference data for Aniakchak (Davies et al., 2016) and are interpreted as strong evidence of eruptive activity at Aniakchak, given both the

- 375 number of shards and the proportion of analyses that they represent. CL-7 also has six points that are geochemically similar to an Early Holocene eruption, KO (~8410-8455 cal yr BP; Braitseva et al., 1997) from Kamchatka, but this does not correlate to any known eruptions from Kamchatka in the timeframe of this deposit. While these are the three most coherent geochemical populations observed in these mixed samples, they are not deemed useful here
- 380 for chronostratigraphic applications (discussed further in Sect. 5.1.1).

An alternative approach for considering these mixed data is to parse by geochemical trend rather than by individual sample. Given the high levels of background shards it is possible that the chosen shard concentration peaks do not relate directly to primary tephra-fall. This is particularly likely where multiple eruptive events are closely spaced and overlap.

385 As each sample might contain shards from multiple eruptions these data can be seen as recording eruptive activity in a broader period, instead of discrete eruptions or accurately dated events.





Using this source-based classification, it is possible to identify eight geochemical groups, illustrated in Fig. 4, for the six mixed samples from the past ~1000 years. Five of these eight geochemical groups correlate with reference glass data for volcanic sources in Alaska (Aniakchak, Mt. Churchill, Redoubt, Augustine, Novarupta-Katmai). These volcanoes all have known eruptions or suspected eruptive activity over this time period (Alaska Volcano Observatory, 2016).



395 Figure 4: Geochemical biplots showing mixed-glass samples from Cascade Lake. Bounding shapes represent simplified geochemical fields for potential source volcanoes (to aid visualisation). For the full glass geochemical-data ranges associated with these volcanic sources see, e.g., Bolton et al. (2020), Davies et al. (2016), Fortin et al. (2019), Zander et al. (2018). Three populations with unknown sources are also shown using the same bounding line and fill. All single point analysis data are listed in Table S1.

400 4 Bayesian age modelling

Step one of our chronometer comparison (see Sect. 2.4) considered if the individual ages fit their expected stratigraphic order. Steen et al. (this volume) noted that two ¹⁴C ages (5.5-7.5 cm and 245-248 cm) were out of sequence as they are anomalously old compared to their surrounding ages. They were therefore excluded from further consideration.





- 405 For step two of our comparison, an initial overlay of the individually modelled chronometers (Fig. 5) showed that there are substantial offsets between ¹⁴C and PSV-1 models above 175 cm, as noted by Steen et al. (this volume). As outlined in Sect. 3.1 and Table 4, the four available cryptotephra correlated ages agree well with PSV-1 tie-points (Fig. 5b) and three further ¹⁴C ages (32.5-30.5 cm, 87.75-85.75 cm and 140-138 cm) are
- 410 therefore also removed as outliers. From 180-290 cm there is also a noticeable divergence between the PSV data model tie-points used from geomagnetic field models and the Burial Lake record (Fig. 5a).



Figure 5: Cascade Lake core CASC-4A/2D multi-method chronometer comparison of downcore age models based on PSV-1 tie-points (light grey shading) and radiocarbon ages (dark grey shading). 2 sigma uncertainties are plotted for all samples; where bars are not visible the uncertainty is smaller than the symbol (values in Table S3). Correlated tephra ages are overlain at their identified depths and show good agreement with the PSV-1 model. (a) Whole model down to 520 cm. Note disagreement between the geomagnetic field model and Burial Lake tie-points from 284-177 cm. PSV-1 model is extrapolated from 520-509 cm (from the base of the unit to the lowest dated sample); (b) enlarged 145 cm section, highlighted with the grey shaded box in panel a, showing cryptotephra correlated ages and the substantial offsets between ¹⁴C and PSV-1 age models.





For step three, a composite P_Sequence model was produced using the PSV-1 tiepoint ages, the four cryptotephra correlated ages and the six remaining ¹⁴C ages (details for OxCal input are given in File S1). This age-depth model was run with a Student's t-test outlier model, which identified four ages with strong likelihoods of being outliers (posterior

- 425 values of 68-100; Fig. S4). These include two further ¹⁴C ages (199-197 cm and 235.5-233.5 cm) and two PSV tie-points from Burial Lake (177 cm and 228 cm). The 284 cm composite depth tie-point from Burial Lake was also removed as it failed the chi-squared test when combined with the tie-point from the pfm9k1b field model and was significantly older than the model results for that depth. These five ages are not included in the final version of the
- 430 age-depth model presented here, as their removal improved the model agreement and reduced the associated uncertainties (Fig. S5).

Figure 6 shows the final age model, which uses 14 PSV-1 tie points, four cryptotephra correlated ages and four ¹⁴C ages to cover 509 cm of core. Ages are extrapolated from 509 cm, the composite depth of the lowest PSV-1 tie-point, to 520 cm, a unit boundary with underlying diamicton. Hence, a well-resolved age model was produced using a combination of ages from three independent chronometers and Bayesian statistical techniques.

5 Discussion

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The data reported here have implications for cryptotephra records in northwestern North America and for Arctic sedimentary sequences and age models through the successful 440 application of PSV dating over the last ~ 21 ka.







Figure 6: Age-depth plot showing the final Bayesian age model for Cascade Lake composite core CASC-4A/2D. Shaded areas show the 1 sigma (68.2%) and 2 sigma (95.4%) confidence ranges. Filled symbols are included in the model and white symbols are identified as outliers. 2 sigma errors are included for all ages; where they are not visible the error is smaller than the symbol used. Full details and values can be found in Table S3.

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5.1 Cryptotephra in Arctic Alaska

This study demonstrates that identifiable concentrations of volcanic glass reach the north flank of the Brooks Range and can be used as chronostratigraphic tools where clear evidence of primary tephra-fall is preserved. In particular, this is the first report of ultra-distal





450 glass from the Late Holocene eruption of Opala, Kamchatka, as well as an unknown tephra, CL-96, likely from a source in the Alaska Peninsula or Aleutian Arc. Ruppert tephra and Aniakchak CFE II are both documented on the southern slope of the Brooks Range (Monteath et al., 2017), and their distributions are expanded here across this large topographic barrier. This is also the first distal identification of WRAn this far to the northwest of Mt. Churchill.

While the cryptotephra profile here only covers the Late Holocene, it highlights eruptive events that are both locally important and widespread and provides possibilities for correlating proxy data within North America and across the Pacific in Kamchatka. Our focus was specifically on the past ~4 ka as there are several widespread, well-dated and

460 geochemically characterised tephra within Alaska during this time period. From 12-4 ka, there is a paucity of well-dated regional tephra that are documented and fully characterised, but it is possible that new tephra from other regions may be identified, as here with OP.

Compared to the cryptotephra stratigraphies published in Monteath et al. (2017) from Ruppert Lake and Woody Bog Pond, located ~150 km south of Cascade Lake on the southern

- 465 slope of the Brooks Range, large differences can be seen in both the number of primary tephra preserved and the overall shard presence and concentrations. Reported glass abundances at the southern sites are at least an order of magnitude higher than those from Cascade Lake (100s 1000s vs 10s shards/gram or less). This likely relates in part to the topographic barrier presented by the Brooks Range, causing increased rain-out of shards
- 470 being transported from the south (e.g. in north trending plumes from Aniakchak CFE II) and deposition of shards before they reach the northern slope. Other factors may include lake size and bathymetry, catchment size, local topography and hydrology. Cascade Lake is an order of magnitude larger and deeper than the southern sites and hence has a larger surface area (~1 km² vs 0.04 and 0.01 km²) but its catchment area is not proportionally larger (~10 km² vs <4</p>
- 475 km^2) and it has no current inflow.

There are common issues affecting cryptotephra research in Alaska that still apply at this distal, Arctic site. The shard concentration profile reported for Cascade Lake is affected by closely spaced eruptions from multiple sources combined with relatively low sediment accumulation rates, causing geochemically variability within individual samples. The

480 presence of glass in the majority of samples analysed shows a level of background deposition



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that must be considered when interpreting data from identified shard concentration peaks. This is particularly important here as the signal:noise ratio between the peaks that have been correlated with known eruptions and the (fairly consistent) background shard concentration is relatively high. This is mostly due to the low concentrations of shards in the identified peaks, compared to other cryptotephra records in the area

5.1.1 Multi-modal samples and historical activity

The issue of 'clear evidence of primary tephra-fall' being preserved is one that affects all cryptotephra records. Low numbers of shard analyses cannot be interpreted as conclusive evidence of an eruption, especially if multiple geochemical populations or trends are

- 490 observed. This appears to only be a problem for certain parts of the Cascade Lake tephrostratigraphic record; there are discernible changes in shard concentrations and samples from the younger portion of the core contains multiple geochemical populations/trends. For example, samples analysed from 30-0 cm have multiple geochemical populations, which are not frequently seen below this. However, this view may be biased by the relatively higher
- 495 number of samples with more than five analyses in this period. Also, the overall shardconcentration profile over the top 15 cm of the core has higher average and peak shard concentration values than the rest of the analysed sediment. These differences could be the result of a myriad of regional (e.g. eruption style, plume altitude, wind direction and strength, shard characteristics) and local (e.g. fallout on snow, sediment accumulation, hydrology,
- 500 bioturbation) factors that affect the distribution, deposition, reworking, and ultimately preservation of shards. A succinct summary for these factors relating to cryptotephra in peatlands is given in Watson et al., (2015), and is largely applicable for lake sediments.

Beyond the five clearly defined cryptotephra samples, we present evidence here of volcanic activity using glass that is geochemically similar to reference data for Mt.

- 505 Augustine, Redoubt, Aniakchak, Mt. Churchill, Novarupta-Katmai (e.g. Bolton et al., 2020) and further possible sources in Kamchatka and Alaska. Focusing on the modern period, this is interpreted as evidence for trace amounts of glass reaching the north flank of the Brooks Range from known eruptions, but without the resolution to interpret individual eruptive events. These shards are unlikely to represent significant reworking from the surrounding
- 510 landscape, or within the lake sediment itself, as there is little ash in the area. This supposition





is supported by the record of known eruptions in the past millennium, including Novarupta-Katmai 1912, six eruptions from Redoubt and 13 from Augustine (Alaska Volcano Observatory, 2016).

- Furthermore, sedimentation rates calculated from the age-model data using OxCal
 v4.4 (Table S3) show that there is a significant decrease, by ~50%, for the depth interval of 12-4 cm (~1840-1250 CE) compared to the Holocene average values (0.015 vs 0.029 cm a⁻¹). This period, coinciding with the Little Ice Age, is therefore expected to show increased background shard concentrations and multi-modal data from 1-cm-resolution samples as each centimetre represents ~67 years of accumulation compared to ~25-40 years as seen here over
- 520 the Holocene. A higher resolution record for this time period may help to address some of the issues detailed here.

For Mt. Churchill there is published evidence for an eruption in the last 500 years: the Lena tephra is dated to 310-285 cal yr BP (Payne et al., 2008). It forms a visible bed in Yukon Territory (Preece et al., 2014) where it sits on top of ~10 cm of peat accumulation

525 above the WRAe. It is possible that shards from CL-0 and -2 relate to these events, although their age is younger than expected. There has not been any observed modern eruptive activity at Mt. Churchill.

There is published evidence for proximal activity at Aniakchak between 560 to 70 yr BP (Neal et al., 2001), but only a small proportion of the associated whole rock geochemical

- 530 data have a rhyodacitic composition similar to the mid-Holocene CFE II eruption (Bacon et al., 2014). Distal tephra preserved in sediment from lakes in the Akhlun Mountains, southwest Alaska, however, have similar glass geochemistry and have been dated at around 400 yr BP (Kaufman et al., 2012). As our age model places the Cascade Lake samples between 350-100 cal yr BP, this currently precludes correlation with these known events.
- 535 This age range is associated with a relatively high uncertainty due to decreased sedimentation rates, so it is possible the chronology does not negate these correlations, but an alternative correlation with a younger eruption from Aniakchak (that has not yet been identified distally) cannot be ruled out. The large number of analyses that geochemically correlate with Aniakchak (47, including 4 dacitic points) over four samples (CL-0, 2, 4 and 7) are therefore
- 540 interpreted here as representing as at least one eruptive event from Aniakchak in the last ~400 years.





5.2 Cascade Lake age models

It is not uncommon for ages produced by multiple chronometers to diverge over part or all of a sediment sequence. Individual chronometers have their own inherent strengths and

- 545 weaknesses, and their different physical properties can be affected by a number of different processes, which in turn affect the preserved and eventually measured signal (e.g. Davies et al., 2018). This is somewhat disheartening as using multiple techniques should provide a check for bias and inaccurate data, but additional independent data can be used to reconcile observed offsets, as shown here.
- 550 Once any obvious outliers have been addressed (i.e. steps one and two from Sect. 2.4), it is not always easy to resolve any remaining disagreements between chronometers. For example, from 303-175 cm in Cascade Lake cores there is a divergence between PSV-1 tiepoints from geomagnetic field models, from Burial Lake and ¹⁴C ages. Logically, the geomagnetic field models incorporate data from multiple regional palaeomagnetic records,
- 555 which should give a valuable, albeit spatially smoothed, resulting record for the area. Their reliability at any given coordinate, however, will depend on the amount and quality of data that is in close proximity. A single, nearby well-dated PSV record (here, Burial Lake) could arguably be more relevant than a field model that incorporates multiple datasets. The use of terrestrial macrofossils for radiocarbon dating at Burial Lake and their consistency over the
- 560 sedimentary sequence suggests they are not affected by, for example, old carbon effects. But, if accurate, the Burial Lake tie-point ages are up to 2000 years older than the other methods for the same composite depths. Outlier analysis performed within OxCal v4.4 was used to assess the ages and statistically identify remaining outliers here (two ¹⁴C ages and three Burial Lake PSV tie-points) in order to resolve this divergence.
- 565 The combination of all three chronometers using Bayesian modelling techniques is therefore shown to result in a refined dataset that produces a reliable age model for the past ~21 ka. This demonstrates the importance of independent chronological validation from marker horizons – here, Late Holocene cryptotephra, which provide additional data in a key period – and the power of Bayesian statistics for age modelling. Furthermore, the
- 570 identification of periods of offset and anomalous or biased ages can allow further investigation of the potential causes, such as mechanical (e.g. mobilisation or redeposition) or chemical (e.g. alteration or degradation) processes affecting the analysed sample material.





Data from Cascade Lake show that PSV-1 provides reliable and accurate tie-points in the Late Holocene that are in agreement with four cryptotephra correlated ages. Comparison of these data across the whole core shows that at least six ¹⁴C ages are too old, including two initially identified as out of sequence (likely old carbon contamination). However, while the 'best ages' produced by PSV-1 are in good agreement with the final age-depth model, the associated uncertainty produced by the geomagnetic field models (± 500 years) is broad compared to other methods that can be applied to this time period.

580 The more commonly applied method of ¹⁴C dating can have lower associated errors but is restricted at some Arctic sites by a lack of suitable material. Where macrofossils are available, they may be affected by old carbon contamination or the redeposition of older material. Cascade Lake's location in limestone terrain likely resulted in a hard-water effect, shown by the ¹⁴C ages reported here. Only four of the eleven analysed samples were included

585 in the final age-depth model and the identified outliers were variably 500-500 ars too old compared to median modelled ages. As mentioned in other studies the use of either terrestrial material or the humic fraction of sediment is recommended, especially when in limestone terrane (Lowe and Walker, 2000). Nonetheless, this study demonstrates that using multiple independent chronometers with Bayesian age modelling techniques can produce accurate and

590 reliable age-depth models for Arctic lake sediments.

6 Conclusions

This research demonstrates the potential for dating Arctic lake sediments in Alaska using PSV tie-points and cryptotephra correlations. The advantages of tephrochronology include the longer period of time over which it can be applied, the level of precision

595 associated with known tephra ages and their potential for independently validating other chronometers. We suggest here that the most robust age models can be produced by using a combination of chronostratigraphic techniques in a Bayesian statistical model. While cryptotephra are best defined regionally for the Late Holocene, it is possible that other welldated cryptotephra from Alaska (e.g. the Early Holocene caldera forming eruptions from

600 Fisher, Stelling et al., 2005; the late Pleistocene Tephra D, Davies et al., 2016) and ultradistal sources (e.g. Kamchatka, Japan) could be identified in northern regions.





Data availability

The major element geochemistry data and associated metadata for individual tephra grains will be made available publicly though the Alaska Volcano Observatory Geochemical

605 Database (Cameron et al., 2019; <u>http://avo.alaska.edu/geochem</u>), part of the larger Geologic Database of Information on Volcanoes in Alaska (GeoDIVA). The Bayesian age-depth model generated in this study, including the underlying radiocarbon ages, lead ages and palaeomagnetic secular variation data are available as supplements to both this paper and Steen et al. (this volume).

610 Supplement information

File S1: OxCal age-depth model input.

Figure S1: Bayesian Tau_Boundary probability density function plots derived using OxCal v4.4 and IntCal20 for the age of Aniakchak CFE II tephra with: all ¹⁴C dates are included (grey right-hand distribution); two ¹⁴C dates removed (green central distribution); and all but two ¹⁴C dates and the NGRIP ice core chronology age (Pearce et al., 2017) (blue left-hand bar). See Table S4 for the ages used for this model.

Figure S2: Bayesian Tau_Boundary probability density function plots derived using OxCal v4.4 and IntCal20 for the age of OP tephra, Opala, Kamchatka. See Table S4 for the ages used for this model.

Figure S3: Major element glass geochemical biplots showing wt.% SiO₂ vs K₂O and FeO vs CaO for samples with multiple geochemical populations. (a) CL-0 and CL-2; (b) CL-4 and CL-7; (c) CL-31 and CL-61; (d) CL-0, -2, -4 and -7 plotted both by sample and by geochemical correlation with a source volcano or region.

Figure S4: OxCal age-depth plot output for the initial Bayesian model for Cascade Lake (v1). Students'-t outlier analysis results are shown. Four ages have more than 50% chance of being an outlier. BL-284 is also excluded as it has an outlier posterior value of 49 and it fails the chi² when combined with pfm9k1b-284.

Figure S5: OxCal age-depth output for the final Bayesian model for Cascade Lake (v2). Five outliers from the previous model (v1) were removed and the students'-t outlier analysis results shown good agreement.

Table S1: Single point major element glass geochemical data for Cascade Lake samples and reference material.

Table S2: Secondary standard data (ID 3506 and Old Crow) for EPMA glass analyses of Cascade Lake samples and reference material.

 Table S3: Final OxCal age model output for 0-520 cm of CASC13-4A/2D, Cascade Lake.

630 Table S4: Ages for tephras reviewed within this study, listed by associated tephra.

Author contribution

LJD carried out the research, helped conceptualise the study, and wrote the manuscript. DSK and BJLJ helped conceptualise and fund the study and revised the manuscript. DSK provided the samples. BJLJ carried out some of the analyses.





635 Competing interests

The authors declare that they have no conflict of interest.

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