Supporting Information for

High-resolution Holocene record from Torfdalsvatn, north Iceland, reveals natural and anthropogenic impacts on terrestrial and aquatic environments

David J. Harning^{1,2}, Christopher R. Florian^{1,2,3}, Áslaug Geirsdóttir², Thor Thordarson², Gifford H. Miller¹, Yarrow Axford⁴, Sædís Ólafsdóttir⁵

¹ Institute of Arctic and Alpine Research, University of Colorado, Boulder, CO

² Faculty of Earth Sciences, University of Iceland, Reykjavík, Iceland

³ National Ecological Observatory Network, Battelle, Boulder, CO

⁴ Department of Earth and Planetary Sciences, Northwestern University, Evanston, IL

⁵ Reykjavík Energy, Bæjarháls 1, 110 Reykavík

Please see the Supporting Data (SD) for accompanying tables (SDT#) and figures (SDF#) referred to in this text.

SDT1: Tephra compositional data from sediment cores 2012NC (this study), 2012BC (Harning et al., 2024) and 2004NC (this study).

SDT2: Number and frequency of eruptive events in Torfdalsvatn as recorded by tephra layer composition.

SDT3: Bulk geochemical and algal pigment proxy data.

1. Tephra layers descriptions

Hekla 1766 (12-TORF-1A-15): At 15 cm depth in 2012NC is a 0.1-cm thick layer of greyish black fine to medium ash, with diverse tephra grains in their attribute and chemical composition. The characteristic grain types are icelandite and rhyolite pumices and shards with minor abundance of basaltic grains (SDT1). Of the 86 grains analyzed, 76 grains (88%) have composition conforming with origin from the Hekla volcanic system (SDT1 and SDF1). This grain population ranges from alkali basalt (n=5, 6%) to icelandite (n=38, 44%) to rhyolite (n=33, 38%), where the latter two compositions dominate the population.

The intermediate Hekla population defines a tight compositional cluster, and on some elemental plots defines two distinct clusters that best match tephra composition from the Hekla 1766 eruption (SDF1). Other icelandite historic Hekla tephra layers, such as Hekla 1206, 1510, 1597, 1845 and 1947, can be ruled out as a source based on their southerly tephra dispersal (Thorarinsson, 1958). Others can be eliminated based on their chemical composition, namely the basaltic icelandite tephras from the Hekla 1970, 1980, 1991 and 2000 events and the dacite of Hekla 1300 and 1158 (SDF1). The tephra from the Hekla 1693 eruption has similar dispersal and composition (e.g., Thorarinsson, 1967; Janebo et al., 2016). However, the 12-TORF-1A-15 icelandite component exhibits a better fit to Hekla 1766 (SDF1). This is one of the larger intermediate Hekla eruption in historical time (VEI 4) that featured an explosive sub-Plinian phase that lasted for 5 to 6 hours on 5 April 1766 CE (Thorarinsson, 1967) with a northward tephra dispersal in the direction of Torfdalsvatn (Janebo et al., 2016).

The rhyolite component, not present in the reference data set obtained from proximal to medial lapilli size Hekla 1766 tephra samples (Harning et al., 2018, 2019), is comprised of two distinct compositional populations. The more abundant low silica population has composition that resembles Hekla 1104 tephra deposits. The high silica population is a minor component and has composition similar to Hekla 4 tephra deposits (SDF1). It is difficult to reconcile that the rhyolitic component is derived from local reworking of Hekla 4 and Hekla 1104 Hekla tephra layers via erosion of the soil cover around Torfdalsvatn, because 1) these compositional groups are in different proportions (0.15 vs 0.85) while the thickness of the layers is roughly the same in local soil sections (Möckel et al., 2021), and 2) of the pristine nature of the grains. It is more likely that this component originated at the source vents. The distance from Hekla to Torfdalsvatn is just over 230 km and the thickness of the 12-TORF-1A-15 tephra layer in the lake is 0.1 cm, matching well with the mapped isopach thickness (Janebo et al., 2016). Consequently, it is logical to assume that tephra from only the most intense initial explosive phase reached Torfdalsvatn. In this regard, it is important to note that 1) the eruption's climax is reached at the very beginning and then intensity drops off rapidly (e.g., Thordarson and Larsen, 2007), and 2) the magma erupted in silicic and intermediate Hekla eruptions are typically compositionally zoned, spanning the felsic realm with the most silicic component erupted first (e.g., Sigmarsson et al., 1992; Sverrisdóttir, 2007; Meara et al., 2020). Alternatively, it may represent Hekla 1104 and Hekla 4 tephra incorporated into the erupting mixture as the magma was opening its path towards the surface at the beginning of the eruption. In any case, we interpret this tephra to be a previously undocumented rhyolitic component of the Hekla 1766 eruption.

The remaining ten grains exhibit tholeiite basalt compositions consistent with the Grímsvötn (n=8, 9%) and possibly Askja volcanic systems (n=2, 2%; SDF1) and may represent tephra produced by eruptions at these volcanic centers in this timeframe.

Hekla 1300 (12-TORF-1A-66): At 66 cm depth in 2012NC is a 0.1-cm thick, greyish white tephra of fine to medium ash. Tephra grains feature diverse attributes and chemical compositions, although the characteristic grain types are rhyolite pumices, with minor abundances of intermediate and basaltic grains (SDT1). Of the 69 grains analyzed, 43 grains (62%) are silicic, where 40 (58%) have Hekla affinities; 38 (55%) are low-silica rhyolite and two are high-silica rhyolite (SDF2). The low silica component falls within the rhyolite compositional fields for Hekla 1104 and Hekla 1300 and the high silica components matches well with the high-silica tail of the Hekla 1300 field (SDF2). The high-silica rhyolite has Hekla 4-like composition. Three rhyolite grains have a mildly alkalic compositions with affinities towards the Katla system, except they feature much higher TiO₂ content (SDF2). Components of Hekla 1300 dacite (n=3), icelandite (n=3) and alkali basalt (n=3) are also present. The remaining 17 grains are of tholeiite basalt compositions, where 13 grains have Grímsvötn (n=13) and 4 grains have Veiðivötn-Bárðarbunga system compositions (SDF2). As the western side of the 0.1 cm isopach of the Hekla 1300 tephra layer is just south of Torfdalsvatn (Thorarinsson, 1967; Janebo et al., 2016), it is likely that only the most evolved tephra from the most intense part of the initial explosive phase from this eruption reached the lake. Hence, the 12-TORF-1A-66 tephra layer is taken here to represent the Hekla 1300 event. In addition, the tholeiite basalt component may represent tephra from roughly concurrent eruptions within the Grímsvötn and Veiðivötn-Bárðarbunga volcanic systems, as several such events are known to have taken place within \pm 70 years of 1300 CE (e.g., Óladóttir et al., 2011).

Hekla 1104 (04-TORF-01, A-1, 90-92, 102-103; 12-TORF-2A-1N-01A-80,5, 84.4): This is a greyish white tephra, which is present in 2004NC over 15.4 cm interval (88.1 to 103.5 cm depth) as three distinct and separate layers/wedges connected by mm-thick, irregular light-colored stringers (Fig. S1). In 2012NC it is present as two distinct layers at depths of 80.5 cm (0.1 cm) and 84.5 cm (0.7 cm). This tephra layer is found in a previously published record from Torfdalsvatn (Alsos et al., 2021), where the stratigraphy is also discontinuous with stringers below and above the main tephra layer horizon (Bender, 2020). The chemical composition of these tephra horizons is identical in all instances, even in the minor components (STD1 and SDF3). Hence, we take this to represent a single layer, which was subsequently deformed to produce the observed features (see main text Section 4.2.3 for Discussion). As a result, we do not explicitly include this tephra layer in the age model.

This horizon is dominantly comprised of rhyolite tephra grains, with minor icelandite and alkalic basalt tephra grains that are mixed with juvenile crystals and minor lithics (SPF3). Of the 222 grains analyzed, 168 (74%) are of rhyolite pumices that not only have affinities to the Hekla volcanic system but also match the composition of the rhyolitic component of Hekla 1104 (SPF3). A total of 14 grains (6%) feature intermediate and 21 grains (9%) have alkali basalt composition consistent with the Hekla volcanic system (SPF3). These compositional results, along with the mapped dispersal thickness (Thorarinsson, 1967; Janebo et al., 2016), strongly suggest that this tephra represents fallout from the largest historical Hekla eruption, Hekla 1104 (VEI 4-5, e.g., Larsen et al., 1999). The residual sulfur in the degassed rhyolite tephra population (SDT1) indicates extended degassing upon eruption, consistent with the notion that Hekla 1104 was a dry magmatic Plinian eruption (e.g., Janebo et al., 2016). Conversely, the residual sulfur in the degassed alkali basalt component of the tephra (SDT1) indicates about 60% degassing during the eruption, suggesting involvement of external water in the explosive activity that produced the basalt tephra (e.g., Thordarson et al., 2001). The remaining 22 grains are tholeiitic basalt of the Grímsvötn (n=10) and Veiðivötn-Barðárbunga (n=12) volcanic systems (SPF3).

C-990 (12-TORF-1A-104, mix of tephra layers): The sample at 104 cm depth in 2012NC is a cryptotephra and has a modeled age of 990 ± 80 cal a BP (960 CE). Of the 46 grains analyzed, 12 (28%) are tholeiitic basalt with composition indicating origin within the Veiðivötn-Barðárbunga volcanic system, whereas 15 (37%) are tholeiite basalt of the Grímsvötn volcanic system (SDF4). In addition, there are 5 (11%) alkalic basalt grains of the Katla volcanic system, matching best with the Eldgjá 934-9 CE compositional field. Grains of rhyolite composition are 10 (24%) and are identical to that of Hekla 1104 (SDT1 and SDF4). This grain population represents an evenly weighted mixture of tephra layers from four different volcanic systems. Hence, we interpret this to be a mixture of tephra layers from the period around 960 CE. As this horizon includes Hekla 1104 grains, it is possible that the mixing event that produced it is related to the event that broke up the Hekla 1104 tephra layer.

C-1180 (12-TORF-1B-9, mix of tephra layers): The sample at 125 cm depth in 2012NC is a cryptotephra and has a modeled age of 1180 ± 70 cal a BP (770 CE). Of the 37 grains analyzed, 20 (54%) exhibit alkalic basalt compositions consistent with the Katla volcanic system, whereas 15 (41%) exhibit tholeiitic basalt compositions consistent with Grímsvötn volcanic system (SDT1 and SDF5). In addition, there are two rhyolitic pumice grains with composition matching the Hekla 4 marker tephra layer, but likely reflect reworked material due to their low abundance. The dominant basaltic composition suggests that this horizon is comprised of tephra fallout from two separate eruptions originating from the Grímsvötn and Katla volcanic systems around 1180 cal a BP. Similar to C-990, it is possible that it was disturbed during the unknown event that broke up the Hekla 1104 tephra layer.

Katla 1220 (12-TORF-1B-15): A 1-cm thick, black layer of fine to medium ash with diffuse upper and lower contacts is located at 131 cm depth in 2012NC and has a modeled age of 1220 ± 50 cal a BP (730 CE). Of the 55 grains analyzed, 49 (89%) exhibit alkalic basalt composition consistent with the Katla volcanic system (SDF6). The remaining 6 grains reflect minor contamination from other systems including Grímsvötn/Kverkfjöll (*n*=3) and Veiðivötn-Bárðarbunga (*n*=2) due to their low abundance (SDT1 and SDF6).

Katla 1270 (12-TORF-1B-26.5): A 2-cm thick, black layer of fine to medium ash is located at 142.5 cm depth in 2012NC and has a modeled age of 1270 ± 40 cal a BP (680 CE). Of the 33 grains analyzed, 29 (88%) exhibit alkalic basalt composition consistent with the Katla volcanic system (SDT1 and SDF7). The remaining 4 grains reflect minor contamination from other systems including Grímsvötn (*n*=3) and Hekla (*n*=1).

C-1850 (12-TORF-1B-86, mix of Katla and Grímsvötn layers): The sample at 202 cm depth in 2012NC is a cryptotephra and has a modeled age of 1850 ± 50 cal a BP. Of the 36 grains analyzed, 20 (56%) exhibit alkalic basalt compositions of the Katla volcanic system and 10 (28%) exhibit tholeiitic basalt compositions consistent with the Grímsvötn volcanic system (SDT1 and SDF8). The remaining grains reveal dacitic to rhyolitic composition that fall into the domains of the Hekla or the Katla volcanic system (SD1 and SDF8). The dominance of basalt tephra grains and lack of a clear stratigraphical horizon suggests that this horizon may be a disturbed mixture of tephra fallout from separate Katla and Grímsvötn eruptions around 1850 cal a BP.

Katla/Grímsvötn 1990 (12-TORF-1B-103): A 0.4-cm thick, black layer of fine to medium ash is located at 219 cm depth in 2012NC and has a modeled age of 1990 ± 140 cal a BP. Of the 32 grains, 21 (66%) are alkalic basalt (SDF9) and one is dacite (SDT1 and SDF9). Their composition is consistent with origin within the Katla volcanic system. The remaining 11 (34%) grains are tholeiite basalt indicating origin at Grímsvötn (n=8) and Veiðivötn-Barðárbunga (n=3) volcanic systems (SDT1 and SDF9). The composition of this layer suggests concurrent tephra fall

from two to three Katla and Grímsvötn (and possibly Bárðarbunga-Veiðivötn) eruptions around 1990 cal a BP.

C-2320 (12-TORF-1B-141, mix of tephra layers): The sample at 257 cm depth in 2012NC is a cryptotephra and has a modeled age of 2320 ± 70 cal a BP. Of the 39 grains analyzed, 17 (44%) are tholeiite basalt originating at the Veiðivötn-Barðárbunga volcanic system, whereas seven (18%) are consistent with the Grímsvötn volcanic system (SDF10). In addition, seven (18%) alkali basalt grains from the Katla volcanic system and three (8%) dacitic grains plus two (5%) rhyolitic grains, both from the Hekla volcanic system (SDT1 and SDF10). The cryptotephra nature of this horizon and dominance of basaltic tephra from Veiðivötn-Barðárbunga, Grímsvötn and Katla suggest it is a disturbed mixture of tephra fall from three closely spaced eruptions around 2320 cal a BP.

Hekla C? (12-TORF01, A-2-47.5): A distinct 0.5-cm thick, grayish brown tephra horizon situated at 197.2 m on the 2004NC core (anticipated at ~340 cm in 2012NC but not identified) and has a modeled age of 2800 ± 80 cal a BP. It features rhyolite, dacite, icelandite and alkali basalt and tholeiite basalt grains. Of the 19 grains analyzed, 12 (63%) have composition with distinct Hekla affinities (SDT1 and SDF11), spanning the range from alkali basalt to rhyolite but dominated by icelandite grains (SDF11). It is possible that this tephra horizon represents the eastern edge of the Hekla C tephra fall (2800 cal a BP, Larsen et al., 2020), although origin via reworking of the underlying Hekla 3 tephra cannot be ruled out, especially since the icelandite component in our samples show stronger affinity with the intermediate component of Hekla 3 rather than Hekla C (SDF11). The remaining seven grains are tholeiite basalt with composition indicating origin within the Grímsvötn volcanic system (SDF11). This may imply that the Hekla C tephra fall coincided with deposition from an explosive eruption at the Grímsvötn volcano.

Hekla 3 (04-TORFA2, 53.5): A 2-cm thick, grayish white tephra is located at 202.2 cm depth in 2004NC and anticipated at ~349 cm in 2012NC but not identified (SDT1 and SDF12). Of the 47 measured tephra grains, 9 (19%) are rhyolite, 30 (64%) have dacite to icelandite composition and two (4%) alkali basalt tephra grains all of which exhibit affinity with the Hekla volcanic system (SDT1 and SDF11). In terms of chemical composition, these components fit well with the compositional trend defined by the Hekla 3 tephra layer (SDF11). Along with its stratigraphic position, these results verify that this is the Hekla 3 marker layer (Larsen and Thorarinsson, 1977; Meara et al., 2020), which has a calibrated C¹⁴ age of 3060 ± 30 cal a BP (Dugmore et al., 1995) and identified in a previous record from Torfdalsvatn (Alsos et al., 2021). The residual sulfur in the degassed alkali basalt tephra population (SDT1) indicates that the magma lost >70% of its volatiles upon eruption, consistent with the notion that Hekla 3 was a dry magmatic Plinian eruption (e.g., Larsen and Thorarinsson, 1977). The remaining six (13%) tephra grains are tholeiite basalt of the Grímsvötn volcanic system and their residual sulfur is indicative of phreatomagmatic origin (SDT1 and SDF11).

Hekla 4 (12-TORF-2A-118): A 1.3-cm thick, grayish white tephra is located at 391 cm depth in 2012NC and at 226.8 cm depth in 2004NC, both as salt and pepper tephra horizons (i.e., light and dark grains). Of the 86 grains analyzed, 81 (94%) are rhyolite (n=68), icelandite (n=2) and alkali basalt (n=11) with composition of the Hekla volcanic system (SDT1 and SDF12). The stratigraphic position and the diagnostic high-silica rhyolite composition verifies this as the Hekla 4 marker tephra (Larsen and Thorarinsson, 1977; Meara et al., 2020) that has a calibrated C¹⁴ age of 4260 ± 10 cal a BP in Icelandic and British Isle peat sequences (Dugmore et al., 1995) and identified in a previous record from Torfdalsvatn (Alsos et al., 2021). The five (6%) remaining tholeiite basalt tephra grains have composition that is consistent with the Grímsvötn volcanic

system. Humic acid extracted from bulk sediments in 2004NC at depth of 227.5 cm immediately below Hekla 4 produced an age of 4030 ± 50 cal a BP (Axford et al., 2007) consistent with previously published Hekla 4 ages. However, due to some irregularities and possible deformation observed in the layer in both the 2004NC core and from a previously published record (Bender, 2020), we do not include this tephra layer in the age model.

Hekla 4270 (12-TORF-2B-03): The sample at 397 cm depth in 2012NC is a cryptotephra and has a modeled age of 4270 ± 180 cal a BP. Of the 37 tephra grains analyzed, 29 (78%) are evolved alkalic basalt (MgO range 4.4 to 5.76 wt. %). While the biplots cannot effectively distinguish between Katla or Hekla origin, the overall variability seems to favor the Hekla volcanic system (SDF13). Moreover, the two rhyolite tephra grains also have composition consistent with the Hekla volcanic system (SDF13, Meara et al., 2020). The remaining six tholeiite basalt grains have composition suggestive of origin from within the Grímsvötn volcanic system (SDF13).

Hekla 5100 (04-TORFA2, 112): A 0.8-cm thick, light grey tephra is located at 261.5cm depth in 2004NC and anticipated at ~450 cm in 2012NC core, but not detected during sampling (SDT1 and SDF12). It has a modeled age of ~5100 cal a BP. A strongly bimodal composition is reflected in the 28 analyzed grains, where 16 (57%) are rhyolite and 12 (43%) are alkali basalt. Both components have clear affinity to the Hekla volcanic system, the basalt features the classic Hekla compositional variability, while the silicic part exhibits resemblance to Hekla 4 but is distinctly different from Hekla 5 in its composition (SDT1 and SDF14). The residual sulfur in the degassed rhyolite tephra population (SDT1) is consistent with full degassing upon eruption, thus, implying a dry magmatic explosive eruption. The residual sulfur in the degassed alkali basalt component of this tephra (SDT1) indicates 65-70% degassing upon eruption, also indicating a magmatic eruption or at best a very minor involvement of external water. This is a previously unidentified bimodal Hekla tephra layer and has the potential of becoming a useful marker tephra in the Middle Holocene.

Askja 5700 (04-TORF-A2, 143.5): A 0.1-cm thick, dark colored tephra is located at 293.1 cm depth in 2004NC, equivalent to ~505 cm in 2012NC (SDT1 and SDF12) and has a modeled age ~5700 cal a BP. This is a tholeiitic basalt tephra of uniform composition (n=19; SDT1 and SDF15) and is one of two olivine tholeiite tephra layers identified in the Torfdalsvatn record. Its composition indicates origin from within the Bárðarbunga-Veiðivötn-Askja-Krafla (VAK) sector of the Northern Volcanic Zone and has relatively high sulfur content, suggesting that degassing upon eruption was arrested prematurely via quenching with external water.

Kverkfjöll/Katla 5850 (12-TORF-2B-118): A 0.1-cm thick, black layer of fine ash is located at 512 cm depth in 2012NC (SDT1 and SDF16) and has a modeled age of 5850 \pm 200 cal a BP. Of the 14 grains analyzed, 10 (71%) feature a tholeiitic basalt composition indicative of origin within the Kverkfjöll volcanic system (SDF16). Several Kverkfjöll tephra layers are identified in soil sections around the Vatnajökull ice cap in the time interval between 6500 and 5500 cal a BP and dated via sediment accumulation rates (Óladóttir et al., 2011). Although one of these is likely the source event for the basalt component of this layer, direct correlation is not currently possible due to multiple correlations possibilities and the uncertainty of soil section tephra age estimates. Four remaining tephra grains are rhyolitic and have composition indicating origin within the Katla volcanic system (Larsen et al., 2001). The stratigraphical position and age of the Holocene SILK (silicic Katla) tephra layer series is well-established and the closest layers in age to TORF-2B-118 are SILK-A1 (~5000 cal a BP) and SILK-A7 (~6200 cal a BP) (Larsen et al., 2001). In addition, another SILK tephra layer (6750 cal a BP) has been identified in the Hvítárvatn lake sediment record (Jóhannsdóttir, 2007). Of these, correlation with the SILK-A7

seems most plausible, however, correlation is not currently possible due to the uncertainty of soil section tephra age estimates. Alternatively, TORF-2B-118 may reflect a previously unidentified Middle Holocene SILK layer.

Katla 6500 (12-TORF-2C-8/04TORF-A3-20.5): A 0.4 to 1.1-cm thick, black layer of medium to coarse ash is located at 557 cm depth in 2012NC and at 318.7 cm depth in 2004NC, and has a modeled age of 6490 ± 130 cal a BP. All 64 grains analyzed are alkalic basalt that generally occupy the compositional fields of the Katla system on bi-elemental plots (SDT1 and SDF17). Residual sulfur content of this tephra is low and indicative of a dry magmatic eruption (see main text Section 5.1.3). The youngest tephra layer described by Björck et al. (1992), labelled Tv-5, has similar composition and thickness to the 12-TORF-2C-8. The same can be said about the tephra layer, termed "Katla S" (SDF17), in lake sediment from Barðalækjartjörn, ~75 km south of Torfdalsvatn (Eddudóttir et al., 2016). Katla S has been ¹⁴C dated using adjacent Betula (6661 to 6501 cal a BP) and Salix twigs (6679 to 6529 cal a BP, Eddudóttir et al., 2015, 2016). Given the overlap of ages derived for Tv-5 in Torfdalsvatn and Katla S in Barðalækjartjörn, in addition to compositional similarity, we suggest these tephra layers were generated from the same Katla eruption. We note that another Katla tephra layer, termed Katla EG, has recently been described with an age of 6240 cal a BP in the lakes Bæjarvötn and Haukadalsvatn (Fig. 1), as well as on the southeast Greenland shelf (Jennings et al., 2014; Harning et al., 2018, 2019). However, small but distinct compositional differences (SDF17) and the modeled age difference indicate that these were generated by tephra fall from two separate eruptions at Katla.

Grímsvötn/Katla 8500 (04-TORF-01-A-3-54.5): A 0.1 to 0.2-cm thick, black layer of fine to medium ash is located at 353 cm depth in 2004NC and has a modeled age of ~8500 cal a BP. Of the 19 grains analyzed, 11 (58%) are tholeiite basalt with distinct Grímsvötn volcanic system affinities and their residual sulfur values are indicative of arrested degassing due to interaction with external water, consistent with origin from a subglacial eruption (SDT1 and SDF18). The remaining 8 (42%) tephra grains exhibit alkali basalt composition that sit within the Hekla basalt compositional field and at the boundary of the Katla basalt field (SDT1 and SDF18). However, the residual sulfur content of this tephra makes Katla origin more viable (see main text Section 5.1.3).

Grímsvötn? 9260 (12-TORF-2D-34): A 0.3-cm thick, black layer of fine to medium ash is located at 730 cm depth in 2012NC (Table 1) and has a modeled age of 9260 ± 300 cal a BP. This layer features pristine and vesicular, basaltic sideromelane grains and a lesser amount of poorly to non-vesicular black basalt grains and minor grey microcrystalline grains. All 19 analyzed tephra grains are tholeiite basalt and the composition of 18 (95%) grains, although variable and dispersed, shows some affinity with the Grímsvötn volcanic system (SDT1 and SDF19).

G10ka Series 9410 to 10400 (12-TORF-2D-38 to -85 and 04-Torf01-A3-105 to -140): The G10ka Series in 2012NC core is represented by sequence of five closely spaced, black tephra layers of fine to medium ash in the depth interval of 734-749 cm and a 26 cm-thick black tephra horizon, extending from 756 to 782 cm depth. The five layers range in thickness from 0.5 to 1.1 cm, each separated by 3-4 cm of lake sediments, while the 26 cm-thick horizon is comprised of alternating beds of very fine gray and coarse black ash (SDF-A). In core 2004NC, it is represented by a 0.3 cm thick tephra layer at 438.3 cm depth and an overlying 26.7 cm thick horizon at depth of 403.9 to 430.6 cm, comprised of mm-dm thick black and brownish black sub-horizons of tephra separated by mm to cm thick packages of organic sediment. Like 2012NC, the 2004NC tephra unit is distinctly bedded with alternating beds of very fine gray and coarse black ash (SDF-A). The modeled age range represented by the G10ka Series horizon in 2012NC is 9410 to 10400 cal a BP.

This estimate is underpinned by two humic acid dates from 2004NC at 432 cm depth (in between the G10ka Series 26.7 cm-thick horizon and 0.3 cm layer at 438.3 cm depth) and at 369 cm depth (i.e., 34.9 cm above the G10ka Series tephra), giving calibrated ages of 10240 ± 10 cal a BP and 9510 ± 20 cal a BP, respectively (SDF-A, Axford et al., 2007). Furthermore, based on chemical composition, total thickness and rhythmic bedding, this unit is correlated with the Tv-4 tephra as originally identified in Torfdalsvatn by Björck et al. (1992). The G10ka Series tephra horizons as well as individual layers were sampled every cm in the 26-cm thick horizon in core 2012NC. In total 46 samples were collected and 44 were analyzed for their chemical composition and described and examined for their physical components (SDT1 and SDF-A).

Of the 672 grains analyzed from these horizons, 660 (98 %) are tholeiite basalt of remarkably uniform composition with distinct affiliation to the Grímsvötn volcanic system, thus confirming a G10ka Series origin (SDT1 and SDF20-21). The remaining 12 grains are tholeiite basalt (n=2) from the Bárðarbunga-Veiðivötn system and alkali basalt (n=10) from Hekla and/or Katla volcanic systems. Via analysis of the appearance, components, and chemical composition, we have identified 12 separate tephra fall events within the G10ka Series tephra in Torfdalsvatn. In addition, the residual sulfur in the degassed basalt tephra, as measured in a select subpopulation of the samples (SDT1), shows that these Grímsvötn magmas released between 35-50% of the volatiles upon eruption, indicating that these tephra falls were produced by phreatomagmatic eruptions (e.g., Thordarson et al., 1996). These 13 events are briefly described below in chronological order from oldest to youngest.

Samples 12-TORF-2D-85 and -84, representing the lowest 2 cm of the 26 cm-thick G10ka Series tephra horizon at 780 to 782 cm depth in core 2012NC, are comprised of medium to coarse ash containing distinguishing sideromelane grains, including achneliths and fragments of golden pumice. This along with a distinctly broader compositional range compared to the tephra above (i.e., 12-TORF-2D-83 and -79; SDF21) as well as more abundant diatom mud clumps and round clastic sediment grains in 12-TORF-2D-84, are taken to indicate that this is the first tephra fall event recorded in Torfdalsvatn (i.e., G10ka Series event #1). In terms of componentry and chemical composition, the 12-TORF-2D-85 and -84 level correlates with the basal layer of the G10ka series in the 2004NC core (i.e., sample 04-TORF-A3, 140; SDF21). The level represented by samples 12-TORF-2D-83 and -82 (778 to 780 cm depth) is also medium to coarse ash and contains relatively high abundance of silvery grey microcrystalline and black opaque tephra grains and is devoid of diatom mud. This evidence, along with significantly tighter compositional range (SDF21), are taken to indicate a new tephra fall event (i.e., G10ka Series event #2). The level represented by samples TORF-2D-81 to -78 (i.e., 778 to 775 cm depth) is made up of very fine to fine ash. In the lower 2 cm it contains a significant portion of open-framework frothy (i.e., reticulite-like = >90% vesicularity) sideromelane grains featuring very delicate protrusions. This grain type feature less strongly in the upper 2 cm of this level, which is typified by upward increase in the diatom mud component along with appearance of diatoms. Although this level has similar composition 12-TORF-2D-83 and -82 (SDF21), the abrupt shift to a more dominant sideromelane clast populations at 778 cm-depth along with increased sediment contribution at 777-775 cm, supports the notion that this interval represents a separate tephra fall (i.e., G10ka Series event #3). The level represented by 12-TORF-2D-77 to -73 at 775 to 770 cm depth, has a lower part comprised of medium ash (TORF-2D-77 and -76) and an upper part of very fine to fine ash (12-TORF-2D-75 to -73). The medium ash in the lowest 2 cm is devoid of diatom mud and contains reticulite-like grains in moderate abundance, indicating it represents the onset of a new tephra fall. The overlying 3 cm of very fine to fine ash are as follows: 12-TORF-2D-75 characterized by poorly

vesicular black glass and grey microlite-rich grains, containing relatively high abundance of diatom mud and may represent the tapering out phase of G10ka Series event #4. TORF-2D-74 is, again, typified by golden pumice and reticulite-like grains, although mixed in with some diatom mud. 12-TORF-2D-73 is typified by abundant fines that are a mixture of diatom mud and granulated highly vesicular grains. Hence, samples 12-TORF-2D-74 to -73 (i.e., 771 to 769 cm depth) is taken to represent tephra fall G10ka Series event #5 in the Torfdalsvatn record. In this context, it is noteworthy that 12-TORF-2D-78 and -77 correlate, in terms of chemical composition and physical characteristics, well with samples 04TORF-A3, 130 and 131 from the base of the 26.7 cm-thick tephra horizon in the 2004NC core. The significance of this correlation is the C¹⁴ age determination of 10240 ± 10 cal a BP in core 2004NC (Axford et al., 2007), thus providing an internal age control for the 2012NC horizon (SDF-A). G10ka Series event #6 corresponds to the level represented by samples 12-TORF-2D-72 to -68 is comprised of fine ash with a slight overall fining upwards and contain abundant golden pumice fragments and reticulite-like grains, whereas diatom mud is absent. G10ka Series vent #7 is represented by samples TORF-2D-67 to -63, which range from black medium ash (TORF-2D-67) to grey fine ash (TORF-2D-66 to -63). TORF-2D-67 is clean tephra comprised of golden pumice fragments and reticulate-like sideromelane glass grains in addition to black opaque, greyish black microcrystalline grains, and a minor plagioclase component. In contrast, TORF-2D-66 to -63 also feature white diatom mud, which may represent the tail-end of the fallout as well as redeposition of the tephra after reworking at the land surface. G10ka Series event #8 of the G10ka Series record in Torfdalsvatn is cm represented by samples 12-TORF-2D-62 to -60 from the top 3 cm (depth of 759-756 cm) of the 26 cm -thick tephra horizon in core 2012NC. It is comprised of fine to medium ash with similar grain morphology and composition as the underlying G10ka Series event #7 deposit. However, G10ka Series event #8 is devoid of white diatom mud, indicating pristine tephra fall.

G10ka Series events #9-13 are present in core 2012NC as a series of five 0.5 to 1.1 cmthick, closely spaced Grímsvötn system tephra layer of fine to medium ash. These layers are in the depth interval of 749-734 cm depth and are separated by 3-4 cm of lake sediments. These five layers correlate with the upper part of the 26.7 cm-thick horizon in 2004NC core (SDF-A and SDT1). Their modeled age spans the range of 9410 to 9960 cal a BP and overlap significantly with the published age range of ~10,625 \pm 50 to 9590 \pm 315 cal a BP for the G10ka Series elsewhere (Óladóttir et al., 2020).

Hekla 10550 (12-TORF-2D-106): A 0.3-cm thick, black layer of fine to medium ash is located at 802 cm depth in 2012NC (SDT1) and has a modeled age of 10550 ± 150 cal a BP. Of the 28 grains analyzed, 22 (79%) are alkali basalt from the Hekla volcanic system (Fig. 11). The remaining six grains are tholeiite basalt, four from the Grímsvötn volcanic system and two from an unknown source. This layer is taken to represent a basaltic tephra fallout from the Hekla volcanic system with a minor contribution from a Grímsvötn system eruption that took place at a similar time. Note that Hekla 10550 is older than the two andesitic Hekla layers identified in Bæjarvötn (~9600 and 9400 cal a BP, Harning et al., 2018).

I-THOL-I? (TORF-2D-108): A 0.3-cm thick, black layer of fine to medium ash is located at 804 cm depth in 2012NC (SDT1) and has a modeled age of 10560 ± 150 cal a BP. Of 30 tephra grains analyzed, 25 are rather primitive tholeiite basalt (MgO = 8.4 ± 0.26 wt %) that appears be derived from the VAK sector of the Northern Volcanic Zone (NVZ) and most likely from within the Askja volcanic system, possibly the Gígöldur eruption. The remaining five grains are also tholeiite basalt but from the Grímsvötn volcanic system (Fig. 11). This layer is a basaltic tephra fallout distinctly primitive composition from an unknown explosive eruption within the NVZ, with

an indication of contribution from an explosive eruption at the Grímsvötn system of similar age. It has similar chemical attributes to the Tv-3 tephra previously identified in Torfdalsvatn (Björck et al., 1992) and in the lake Bæjarvötn (Harning et al., 2018) support a correlation to the I-THOL-I marker tephra layer of the North Atlantic Ash Zone I (Kvamme et al., 1989). We also note here that Tv-3 likely correlates to the KOL1-2269 tephra identified in marine sediment core MD99-2269 on the North Iceland Shelf (10570 \pm 106 cal a BP, Kristjánsdóttir et al., 2007).

Kverkfjöll 10630 (12-TORF-2D-116.5): A 0.3-cm thick, black layer of fine to medium ash is located at 812.5 cm depth in 2012NC (Table 1 and SDT1) and has a modeled age of 10630 \pm 150 cal a BP. Of the 35 grains analyzed, 33 grains define three distinctive compositional groups particularly in terms of TiO₂, FeO and MgO wt%. Seven of the 33 grains are alkali basalt from the Katla volcanic system, six tholeiite basalt from the Bárðarbunga-Veiðivötn system and 17 grains are tholeiite basalt with composition that makes the Kverkfjöll the most likely source system (). We take this layer to represent basaltic tephra fallout from Kverkfjöll volcanic system eruption. The minor components suggest that tephra fall from explosive eruptions within the Katla and Bárðarbunga-Veiðivötn systems took place at the similar time.

Please see Harning et al. (2024) for detailed descriptions of the oldest four tephra layers from sediment core 2012BC: Katla 11,170, Katla 11,295, Katla 11,315 (Tv-2), and Hekla 11,390 (Tv-1).

2. Tephra stratigraphy implications

Excluding the 26 cm-thick horizon within the G10ka Series, the average tephra horizon thickness is 0.7 ± 0.5 cm (total range <0.1 to 2 cm), implying that the source eruptions were of subplinian to Plinian/phreatoplinian intensities (Thordarson and Larsen, 2007; Thordarson and Höskuldsson, 2008), activity that is taken to typify explosive eruptions producing magmas of silicic and intermediate compositions (e.g., Walker, 1973). Yet over 65% (>80% if we include the 26 cm thick G10ka horizon) of the events recorded in the Holocene sediment record from Torfdalsvatn are mafic (i.e., basaltic), including two of primitive magma composition (7.42 \pm 0.14 and 8.39 \pm 0.26 wt. % MgO, respectively). Furthermore, the thicknesses of individual basalt layers within G10ka Series range from 0.5 to 1.1 cm and up to 3 cm if we include data from the 26 cm thick G10ka series horizon. In addition, the number of Hekla and Katla basalt tephra events in the record are close to that of Hekla and Katla silicic to intermediate events. Collectively, this demonstrates not only that basaltic explosive events can be as powerful and widely dispersed as their silicic and intermediate counterparts in Iceland, but they also have the capacity to be valuable as stratigraphic markers in distal sediment records. Finally, Grímsvötn, Hekla and Katla are the most common sources, consistent with them being three of the most active volcanic systems in Iceland (Thordarson and Larsen, 2007; Thordarson and Höskuldsson, 2008). However, the high frequency of tephra fall from Grímsvötn is principally a consequence of the G10ka Series eruption episode in the Early Holocene, between ~10400 and 9400 cal a BP.

2.1. G10ka Series Tephra

The Saksunarvatn Ash has been one of the most widely applied tephra markers sourced from Iceland in Northern Hemisphere tephra stratigraphies (Mangerud et al., 1986; Merkt et al., 1993; Björck et al., 1992; Grönvold et al., 1995; Birks et al., 1996). However, evidence in Iceland and abroad demonstrates that as many as seven compositionally indistinguishable Saksunarvatn-like tephra layers (i.e., G10ka Series tephra) were deposited between 10400 and 9900 cal a BP, with

no more than five reported in superposition (Jóhannsdóttir, 2007; Jennings et al., 2014; Kristjánsdóttir et al., 2017; Harning et al., 2018b, 2019; Wastegård et al., 2018; Timms et al., 2019; Óladóttir et al., 2020). Although Greenland ice core records date one layer of the G10ka Series tephra to $10,300 \pm 90$ cal a BP (Rasmussen et al., 2006), ¹⁴C-based age estimates on the G10ka Series elsewhere range from $10,625 \pm 50$ to 9590 ± 315 cal a BP (Óladóttir et al., 2020), which supports successive Grímsvötn eruptions rather than a single event.

In Torfdalsvatn, we correlate 13 layers to the G10ka Series tephra based on Grímsvötn composition (Fig. S2) and modeled ages (10,400 to 9410 cal a BP) that overlap with ¹⁴C-based age estimates elsewhere (10,625 \pm 50 to 9590 \pm 315 cal a BP, Óladóttir et al., 2020). Of these 13 total layers, the 26 cm thick unit in 2012NC reflects eight separate tephra layers based on distinct variations in grain size, sedimentology and morphology. The remaining five G10ka Series tephra layers are located immediately above the 26 cm thick unit and are each separated by organic sediment. Although Björck et al. (1992) attribute their 22 cm thick "Saksunarvatn Ash" to a single event, the authors do describe a rhythmic tephra unit, which was later proposed to be partially comprised of reworked tephra (upper 10 cm) from catchment erosion based on the incorporation of pollen (Rundgren, 1998). However, in addition to the detailed grain morphology and compositions, each of the high-resolution samples (every cm) retains delicate protrusions that would not survive aeolian or fluvial erosion, supporting the interpretation that the entire 26 cm thick sequence is comprised of multiple, primary tephra deposits. In the lake Bæjarvötn, NW Iceland (Fig. 1), a similar conclusion is also supported by the presence of at least two G10ka Series tephra layers that are separated by tephra from the Veiðivötn-Bárðarbunga volcanic system (Harning et al., 2018). Despite our high-resolution sampling strategy throughout the 26 cm thick unit in Torfdalsvatn, we find no evidence that tephra from Veiðivötn-Bárðarbunga tephra or elsewhere is interbedded. However, the lowermost sample tephra layer in Torfdalsvatn (TORF-2D-53) does feature a more clustered composition compared to the subsequent G10ka Series layers (Fig. 7), which may aid in future correlations. Given the lack of firmer age control on individal layers as well as the compositional indistinctions on the subsequent G10ka Series layers, it remains difficult to correlate Torfdalsvatn sequence to other sites in Iceland where multiple G10ka Series layers have been identified, such as southern (Geirsdóttir et al., 2022) western (Jóhannsdóttir, 2007; Harning et al., 2019) and northwestern lake sediments (Harning et al., 2018). In addition, it still remains unclear how the G10ka Series tephra layers found in Iceland correlate to the Saksunarvatn Ash layers found in Greenland and the Faroe Islands (e.g., Óladóttir et al., 2020), but it is the focus of ongoing research.

2.2. Middle and Late Holocene tephra stratigraphy

Given the presumed distribution of several key marker tephra layers over Torfdalsvatn, it is surprising that they were not found in our records. These marker tephra include the widely distributed and rhyolitic Hekla 5 tephra (~7075 cal a BP, Meara et al., 2020), a local but distinctly intermediate Katla tephra layer found in Barðalækjartjörn (Fig. 1A main text), termed HUN (5530 \pm 60 cal a BP, Eddudóttir et al., 2016), and the trachydacitic to rhyolitic Snæfellsjökull-1 tephra (1820 \pm 90 cal a BP, Larsen et al., 2002) that has been found across western and northern Iceland (Larsen et al., 2002; Gudmundsdóttir et al., 2012, 2018; Harning et al., 2018, 2019). Although these tephra layers were not present in our Torfdalsvatn sediment cores, some such as Hekla 5 have been found in nearby Barðalækjartjörn (Eddudóttir et al., 2016). Given the known distribution, distinct compositional characteristics, and relatively firm age control of these tephra

layers, it is conceivable that these could become useful marker layers in north Iceland with additional sediment records from nearby lakes.

Notable Middle Holocene tephra layers in Torfdalsvatn derive from the Katla, Kverkfjöll, Askja and Hekla volcanic systems (Table 2, main text). One of the thickest is the Katla 6500 tephra layer (1.1 cm thick, TORF-2C-6), which correlates with the previously termed Tv-5 tephra in prior Torfdalsvatn studies (Fig. S3B, Björck et al., 1992). In lake sediment from Barðalækjartjörn, ~75 km south of Torfdalsvatn (Fig. 1A, main text), a compositionally similar tephra layer has been termed Katla S (Fig. S3B, Eddudóttir et al., 2016), which was ¹⁴C-dated using adjacent Betula (6661 to 6501 cal a BP) and Salix twigs (6679 to 6529 cal a BP, Eddudóttir et al., 2015, 2016). Given the overlap of ages derived for Katla 6500 in Torfdalsvatn and Katla S in Barðalækjartjörn, in addition to compositional similarity, we suggest these tephra layers were generated from the same Katla eruption. Additional distinct tephra layers in the Middle Holocene include the olivine tholeiitic basalt Askja 5700, the bimodal Hekla 5100, and a rhyolitic Hekla layer (Hekla 4270, TORF-2B-03) that precedes the Hekla 4 marker tephra layer (TORF-2A-118, Fig. S3). Given the lack of current correlations in other nearby Icelandic sediment records, these layers likely reflect the product of narrow dispersals from their respective sources.

The two uppermost Late Holocene Hekla tephra layers (Hekla 1300 and 1766) are wellknown to feature dacitic or andesitic compositions (Thórarinsson, 1967). However, in addition to the dacitic and andesitic tephra grains, each of these tephra layers also contained substantial proportions of high-SiO₂ and low-SiO₂ rhyolitic glass consistent with Hekla 4 and Hekla 3/Hekla 1104 compositions, respectively (TORF-1A-66 and TORF-1A-80.5, respectively, Fig. S1). It is difficult to reconcile that these are derived from local reworking of Hekla 4 and Hekla 3/Hekla 1104 tephra via erosion of the soil cover around Torfdalsvatn because: (1) the proportion of high and low-SiO₂ rhyolitic grains are substantially different, whereas Hekla 4 and 3 are close to each other in local soil profiles and have roughly the same thickness (Möckel et al., 2021) and (2) the pristine nature of these grains that suggests no post-depositional reworking. For Hekla 1300, since Torfdalsvatn is at its western margin of dispersal (Thórarinsson, 1967; Janebo et al., 2016), it is possible that the most intense part of the initial phase was directed more to the north resulting in the dominantly rhyolitic tephra that we observe (Fig. S1). For Hekla 1766, although the initial rhyolitic phase was about 2-3 times less intense than that of Hekla 1300 (Janebo et al., 2016), our data suggests that Torfdalsvatn also saw tephra fall from this initial explosive phase resulting in its rhyolitic as well as intermediate character (Fig. S1). However, it is also possible that the Hekla 1104-like rhyolitic components reflect material incorporated from units within the Hekla volcanic system during the latter two volcanic eruptions. On the other hand, it is unlikely that the Hekla 4like component could have been incorporated in a similar fashion as the Hekla 4 eruption and its high-SiO₂ magma predates the presumed construction of the modern Hekla edifice (~3000 cal a BP, e.g., Jóhannesson and Einarsson, 1992; Larsen et al., 2020). Although more work is needed to better establish the origins and dispersal limits for the rhyolitic components of Hekla 1300 and 1766 that we observe in Torfdalsvatn, our datasets suggest that rhyolitic tephra from Hekla were generated more frequently during the last millennium than currently established.

3. Residual sulfur content as indication for phreatomagmatic eruptions

In this study, the residual sulfur content was measured in a selected suite of samples (bold font, Table 2, main text) to assess whether the events that produced those tephra layers involved interaction with external water upon eruption. Evidence of such interaction is an indicator of wet

vent environment and thus a proxy for eruptions from within glaciers or through standing body of water (lake or the sea).

During the Early Holocene, recent syntheses show that the Icelandic Ice Sheet retreated from a shallow marine shelf position at ~12,000 cal a BP to the central highlands, when catastrophic drainage of previously ice dammed lakes occurred, between 10,700 and 10,400 cal a BP (Geirsdóttir et al., 2022). The Early Holocene Hekla 11390 basalt tephra has residual sulfur contents showing that the magma lost >70% of its volatiles upon eruption, indicating that this tephra was produced by a dry magmatic eruption (e.g., Thordarson et al., 1996, 2001). This is consistent with a rapid northeastward retreat of the Icelandic Ice Sheet from its position at the Búði moraine complex southwest of Hekla (e.g., Geirsdóttir et al., 2022). Around the same time, the Early Holocene Katla 11,315 and 11,170 tephra layers are typified by residual sulfur indicating about 55% loss of their volatiles upon eruption, suggesting that these tephra layers were produced by wet phreatomagmatic eruptions (e.g., Thordarson et al., 2001). While this may indicate that the Katla volcano was either still covered by the retreating Icelandic Ice Sheet or its caldera was filled with a lake when the eruptions occurred, recent glacier modeling and reconstructions suggest ice sheet cover was more likely (Anderson et al., 2019; Geirsdóttir et al., 2022).

The G10ka Series (10400 to 9410 cal a BP) has residual sulfur showing that these Grímsvötn magmas released between 35-50% of the volatiles upon eruption, implying that these tephra falls were produced by wet phreatomagmatic eruptions (e.g., Thordarson et al., 1996) and most likely from vents within the Vatnajökull glacier during the Early Holocene. These observations are consistent with a recent Holocene glacier model that simulates the presence of Vatnajökull's northern sector, which overlies the Grímsvötn volcanic system's central volcanos, at this time (Anderson et al., 2019). The Grímsvötn/Katla 8500 tephra layer is comprised of tephra fall from two eruptions, a tholeiite basalt from the Grímsvötn volcanic system and alkali basalt from either the Katla or Hekla system. The Grímsvötn portion has residual sulfur values indicative of arrested degassing due to interaction with external water, consistent with origin from a subglacial volcanic eruption. This is also consistent with the glacial model that simulates the persistence of Vatnajökull's northern sector to ~8000 cal a BP, although the timing also depends on glacial isostatic adjustment and precipitation changes, neither of which are well-constrained (Anderson et al., 2019). The alkali basalt portion of the 8500 cal a BP tephra layer has a composition that on bi-element plots sits within the Hekla basalt compositional field and at the boundary of the Katla basalt field but has residual sulfur contents indicative of arrested degassing via interaction with external water. This makes Katla origin a more viable eruption source given that the Katla central volcano likely had some ice cover at this time, albeit smaller than today (Anderson et al., 2019), while the Hekla volcanic system was likely ice-free.

During the Holocene Thermal Maximum (HTM, ~7900 to 5500 cal a BP, Geirsdóttir et al., 2020), warmer than present summer temperatures (Harning et al., 2020) resulted in the significant retreat or disappearance of residual ice caps in Iceland (Larsen et al., 2012; Harning et al., 2016; Geirsdóttir et al., 2019). However, discrepancies exist regarding the HTM persistence of some, including Mýrdalsjökull, the ice cap that today covers Katla. For instance, HTM ice cap presence has been suggested based on sulfur content of other Katla tephra layers over the last 8400 years (Óladóttir et al., 2007). On the other hand, surface mass balance–equilibrium line altitude glacier models suggests the persistence of HTM ice is highly unlikely (Anderson et al., 2019). Sulfur analysis of the Katla 6500 tephra layer features a low and tightly clustered residual sulfur content indicating degree of degassing exceeding 70%, which is typical for dry magmatic basalt eruptions (e.g., Thordarson et al., 2003). Hence, while we cannot draw conclusions about the entirety of the

HTM, the Katla 6500 tephra sulfur content as well as several other Katla tephra layers' sulfur content in this time frame (Óladóttir et al., 2007) indicate that Katla's caldera was at least periodically free of ice and water during the HTM.

Askja 5700 has composition that is consistent with origin within the Bárðarbunga-Veiðivötn-Askja-Krafla (VAK) sector of the Northern Volcanic Zone. For magma of this composition (SDT1), un-degassed values are typically between 1100-1300 ppm S and fully degassed values between 300-400 ppm S (e.g., Thordarson et al., 2003). The measured residual sulfur in this tephra is ~800 ppm on average (SDT1) and thus suggestive of quenching-induced arrest of degassing due to interaction with external water (e.g., Thordarson et al., 1996, 2001). At the time of the eruption the Krafla volcanic system is ice-free and its main phreatomagmatic constructs (i.e., the ~10000 cal a BP Lúdent and ~2500 cal a BP Hverfjall tuff cones) are either much too old or young to be considered likely sources for this tephra layer (Sæmundsson, 1991). Of these three systems, the only one likely to have featured a glacier at this time is the Veiðivötn-Bárðarbunga volcanic system (Anderson et al., 2019). In the pre-Hekla 4 time, the caldera of the Askja volcano did not contain the lavas that fill it to the brim today. It also featured a major episode of phreatomagmatic volcanism during this time that produced the NE- and SW tuff cone complexes (Hartley et al., 2013, 2016). Hence, it is reasonable that the Askja central volcano featured a caldera lake at this time. Although the layer's major element composition is such that our bi-element discrimination plots do not clearly discriminate between the two systems, the overall trend does lean towards Askja origin (SDF15). In addition, the glass composition of the tephra from the NE- and SW tuff cone complexes (Hartley, 2012; Hartley et al., 2013) exhibits similarities to this tephra layer. Hence, we propose that this olivine tholeiite tephra was produced by phreatomagmatic eruption at the Askja central volcano at about 5700 years ago.

Hekla 1104 reveals residual sulfur contents in the degassed rhyolite tephra population (see SDT1) that is indicative of extended degassing upon eruption and consistent with the notion that Hekla 1104 is a dry, magmatic Plinian eruption (e.g., Janebo et al., 2016). Conversely, the residual sulfur in the degassed alkali basalt component of the tephra (SDT1) indicates about 60% degassing upon eruption, suggesting involvement of external water in the explosive activity that produced the basalt tephra (e.g., Thordarson et al., 2001). These contrasting results indicate either that 1) the basalt component is from an older tephra formation that was produced by phreatomagmatic eruption and accidentally incorporated into the silicic tephra of Hekla 1104 during the eruption or 2) it originated from a separate, but simultaneously active, Hekla system vent where the basaltic magma interacted with external water.



Figure S1: Late Holocene Hekla rhyolite tephra layer compositions from core 2012NC in comparison to known Hekla eruptions. See Supporting Data for source references.



Figure S2: G10ka Series compositions from core 2012 NC in comparison to volcanic source fields. See Supporting Data for source references.



Figure S3: Notable Middle Holocene tephra layers compositions from core 2012NC in comparison to fields of volcanic sources and known eruptions. See Supporting Data for source references.

References

Alsos, I. G., Lammers, Y., Kjellman, S. E., Merkel, M. K. F., Bender, E. M., Rouillard, A., Erlendsson, E., Guðmundsdóttir, E. R., Benediktsson, I. Ö., Farnsworth, W. F., Brynjólfsson, S., Gísladóttir, G., Eddudóttir, S. D., and Schomacker A.: Ancient sedimentary DNA shows rapid post-glacial colonisation of Iceland followed by relatively stable vegetation until the Norse settlement (Landnám) AD 870. Quaternary Science Reviews, 259, 106903, 2021.

Anderson, L. S., Geirsdóttir, Á., Flowers, G. E., Wickert, A. D., Aðalgeirsdóttir, G. Th., and Thorsteinsson, T.: Controls on the lifespans of Icelandic ice caps. Earth and Planetary Science Letters, 527, 115780, 2019.

Axford, Y., Miller, G. H., Geirsdóttir, Á., and Langdon, P.: Holocene temperature history of northern Iceland inferred from subfossil midges. Quat. Sci. Rev., 26, 3344-3358, 2007.

Bender, E. M.: Late Quaternary tephra stratigraphy and paleoenvironmental reconstruction based on lake sediments from North and Northeast Iceland. MS thesis, UiT The Arctic University of Norway, 2020.

Birks, H. H., Gulliksen, S., Haflidason, H., Mangerud, J., and Possnert, G.: New radio-carbon dates from the Vedde ash and Saksunarvatn ash western Norway. Quaternary Research, 127, 119-127, 1996.

Björck, S., Ingólfsson, Ó., Haflidason, H., Hallsdóttir, M., and Anderson, N. J.: Lake Torfadalsvatn: a high resolution record of the North Atlantic ash zone I and the last glacial-interglacial environmental changes in Iceland. Boreas, 21, 15-22, 1992.

Dugmore, A. J., Cook, G. T., Shore, J. S., Newton, A. J., Edwards, K. J., and Larsen, G.: Radiocarbon dating tephra layers from Britain and Iceland. Radiocarbon, 37, 379-388, 1995.

Eddudóttir, S. D., Erlendsson, E., and Gísladóttir, G.: Life on the periphery is tough: Vegetation in Northwest Iceland and its responses to early-Holocene warmth and later climate fluctuations. Holocene, 25, 1437-1453, 2015.

Eddudóttir, S. D., Erlendsson, E., Tinganelli, L., and Gísladóttir, G.: Climate change and human impact in a sensitive ecosystem: the Holocene environment of the Northwest Icelandic highland margin. Boreas, 45, 715-728, 2016.

Geirsdóttir, Á., Harning, D. J., Miller, G. H., Andrews, J. T., Zhong, Y., and Caseldine, C.: Holocene history of landscape instability in Iceland: Can we deconvolve the impacts of climate, volcanism and human activity? Quaternary Science Reviews, 249, 106633, 2020.

Geirsdóttir, Á., Miller, G. H., Andrews, J. T., Harning, D. J., Anderson, L. S., Florian, C., Larsen, D. J., and Thordarson, T.: The onset of Neoglaciation in Iceland and the 4.2 ka event. Climate of the Past, 15, 25-40, 2019.

Geirsdóttir, Á., Miller, G. H., Harning, D. J., Hannesdóttir, H., Thordarson, T., and Jónsdóttir, I.: Evidence for recurrent outburst floods and active volcanism in Icelandic lacustrine settings during dynamic Younger Dryas-Early Holocene deglaciation. Journal of Quaternary Research, 37, 1006-1023, 2022.

Gudmundsdóttir, E. R., Larsen, G., and Eiríksson, J.: Tephra stratigraphy on the North Icelandic Shelf: extending tephrochronology into marine sediments off North Iceland, Boreas, 41, 719-734, 2012.

Gudmundsdóttir, E. R., Schomacker, A., Brynjólfsson, S., Ingólfsson, Ó., and Larsen, N. K.: Holocene tephrostratigraphy in Vestfirðir, NW, Iceland. Journal of Quaternary Science, 33, 827-839, 2018.

Grönvold, K., Óksarsson, N., Johnsen, S.J., Clausen, H. B., Hammer, C. U., Bond, C., and Bard, E.: Ash layers from Iceland in the Greenland GRIP ice core correlated with oceanic and land sediments. Earth and Planetary Science Letters, 135, 149-155, 1995.

Harning, D. J., Curtin, L., Geirsdóttir, Á., D'Andrea, W. J., Miller, G. H., and Sepúlveda, J.: Lipid biomarkers quantify Holocene summer temperature and ice cap sensitivity in Icelandic lakes. Geophysical Research Letters, 47, e2019GL085728, 2020.

Harning, D. J., Geirsdóttir, Á., Miller, G. H., and Zalzal, K.: Early Holocene deglaciation of Drangajökull, Vestfirðir, Iceland. Quaternary Science Reviews, 153, 192-198, 2016.

Harning, D. J., Thordarson, T., Geirsdóttir, Á., Miller, G. H., and Florian, C. R.: Repeated Early Holocene eruptions of Katla, Iceland, limit the temporal resolution of the Vedde Ash, Bulletin of Volcanology, 86, 2, 2024.

Harning, D. J., Thordarson, T., Geirsdóttir, Á., Miller, G. H., and Ólafsdóttir, S.: Marker tephra in Haukadalsvatn lake sediment: A key to the Holocene tephra stratigraphy of Northwest Iceland. Quat. Sci. Rev., 219, 154-170, 2019.

Harning, D. J., Thordarson, T., Geirsdóttir, Á., and Zalzal, K.: Provenance, stratigraphy and chronology of Holocene tephra from Vestfirðir, Iceland. Quat. Geochronol., 46, 59-76, 2018.

Hartley, M. E.: Postglacial volcanism and magmatism on the Askja volcanic system, North Iceland. PhD thesis, University of Edinburgh, 2012.

Hartley, M. E., Thordarson, T., and de Joux, A.: Postglacial eruptive history of the Askja region, North Iceland. Bulletin of Volcanology, 78, 2016.

Hartley, M. E., and Thordarson, T.: The 1874-76 volcano-tectonic episode at Askja, North Iceland: lateral flow revisited. Geochemistry, Geophysics, Geosystems, 14, 2286-2309, 2013.

Janebo, M. H., Thordarson, T., Houghton, B. F., Bonadonna, C., Larsen, G., and Carey, R. J.: Dispersal of key subplinian-Plinian tephras from Hekla volcano, Iceland: implications for eruption source parameters. Bull. Volcanol., 78, 1-16, 2016.

Jennings, A. E., Thordarson, T., Zalzal, K., Stoner, J., Hayward, C., Geirsdóttir, Á., and Miller, G. H.: Holocene tephra from Iceland and Alaska in SE Greenland Shelf Sediments. In: Austin, W.E.N., Abbott, P.M., Davies, S.M., Pearce, N.J.G., and Wastegård, S. (eds) *Marine Tephrochronology*. Geological Society, London, Special Publications, 398, 2014.

Jóhannesson, H., and Einarsson, S.: Hekla, fjall med fortíd. Náttúrufrædingurinn, 61, 177–191, 1992.

Jóhannsdóttir, G. E.: Mid-Holocene to late glacial tephrochronology in west Iceland as revealed in three lacustrine environments. MS thesis, University of Iceland, 2007.

Kristjánsdóttir, G. B., Stoner, J. S., Jennings, A. E., Andrews, J. T., and Grönvold, K.: Geochemistry of Holocene cryptotephras from the North Iceland Shelf (MD99-2269): intercalibration with radiocarbon and palaeomagnetic chronostratigraphies. Holocene 17, 155-176, 2007.

Kvamme, T., Mangerud, J., Furnes, H., and Ruddiman, W. F.: Geochemistry of Pleistocene ash zones in cores from the North Atlantic. Norsk Geologisk Tidsskrift, 69, 251–272, 1989.

Larsen, D. J., Miller, G. H., Geirsdóttir, Á., and Ólafsdóttir, S.: Non-linear Holocene climate evolution in the North Atlantic: a high-resolution, multi-proxy record of glacier activity and environmental change from Hvítárvatn, central Iceland. Quaternary Science Reviews, 39, 14-25, 2012.

Larsen, G., Dugmore, A. J., and Newton, A. J.: Geochemistry of historical-age silicic tephras in Iceland. Holocene, 9, 463-471, 1999.

Larsen, G., Eiríksson, J., Knudsen, K. L., and Heinemeier, J.: Correlation of late Holocene terrestrial and marine tephra markers, north Iceland: implications for reservoir age changes. Polar Research, 21, 283-290, 2002.

Larsen, G., Newton, A. J., Dugmore, A. J., Vilmundardóttir, E. G.: Geochemistry, dispersal, volumes and chronology of Holocene silicic tephra layers from the Katla volcanic system, Iceland. J. Quat. Sci., 16, 119-132, 2001.

Larsen, G., Róbertsdóttir, B. G., Óladóttir, B. A., and Eiríksson, J.: A shift in eruption mode of Hekla volcano, Iceland, 3000 years ago: two-coloured Hekla tephra series, characteristics, dispersal and age. J. Quat. Sci., 35, 143-154, 2020.

Larsen, G., and Thórarinsson, S.: H-4 and other acid Hekla tephra layers. Jökull, 27, 27-46, 1977.

Mangerud, J., Furnes, H., and Jóhansen, J.: A 9000-year-old ash bed on the Faroe Islands. Quaternary Research, 26, 262-265, 1986.

Meara, R. H., Thordarson, T., Pearce, N. J. G., Hayward, C., and Larsen, G.: A catalogue of major and trace element data for Icelandic Holocene Silicic tephra layers. J. Quat. Sci., 35, 122-142, 2020.

Merkt, J., Müller, H., Knabe, W., Müller, P., and Weiser, T.: The early Holocene Saksunarvatn tephra found in lake sediments in NW Germany. Boreas, 22, 93-100, 1993.

Möckel, S. C., Erlendsson, E., Prater, I., and Gísladóttir, G.: Tephra deposits and carbon dynamics in peatlands of a volcanic region – lessons from the Hekla 4 eruption. Land Degrad. Dev., 32, 654-669, 2021.

Óladóttir, B. A., Larsen, G., and Sigmarsson, O.: Holocene volcanic activity at Grímsvötn, Bárdarbunga and Kverkfjöll subglacial centres beneath Vatnajökull, Iceland. Bull. Volcanol., 73, 1187-1208, 2011.

Óladóttir, B. A., Thordarson, T., Geirsdóttir, Á., Jóhannsdóttir, G. E., and Mangerud, J.: The Saksunarvatn Ash and the G10ka series tephra. Review and current state of knowledge. Quat. Geochron., 56, 101041, 2020.

Óladóttir, B. A., Thordarson, T., Larsen, G., and Sigmarsson, O.: Survival of the Mýrdalsjökull ice cap through the Holocene thermal maximum: evidence from sulfur contents in Katla tephra layers (Iceland) from the last ~8400 years. Ann. Glaciol., 45, 183-188, 2007.

Rasmussen, S. O., Andersen, K. K., Svensson, A. M., Steffensen, J. P., Vinther, B. M., Clausen, H. B., Siggaard-Andersen, M.-L., Johnsen, S. J., Larsen, L. B., Dahl-Jensen, D., Bigler, M., Röthlisberger, R., Fischer, H., Goto-Azuma, K., Hansson, M. E., and Ruth, U.: A new Greenland ice core chronology for the last glacial termination. Journal of Geophysical Research, 111, 1-16, 2006.

Rundgren, M.: Early Holocene vegetation of northern Iceland: pollen and plant macrofossil evidence from the Skagi peninsula. The Holocene, 5, 553-564, 1998.

Sæmundsson, K.: Jarðfræði Kröflukerfisins (Geology of the Krafla volcanic system). In: Gardarsson, A. and Á. Einarsson eds. *Náttúra Mývatns*, Hið íslenska náttúrufræðifélag, Reykjavík, 25–95, 1991.

Sigmarsson, O., Condomines, M., and Fourcade, S.: A detailed Th, Sr and O isotope study of Hekla: differentiation processes in an Icelandic Volcano. Contrib. to Mineral. Petrol., 112, 20–34, 1992.

Sverrisdóttir, G.: Hybrid magma generation preceding Plinian silicic eruptions at Hekla, Iceland: evidence from mineralogy and chemistry of two zoned deposits. Geological Magazine, 144, 643–659, 2007.

Thorarinsson, S.: The Öræfajökull eruption of 1362. Acta Nat. Islandica II, 1–99, 1958.

Thorarinsson, S.: The eruptions of Hekla in historical times: a tephrochronological study. In: Einarsson, T., Kjartansson, G., Thorarinsson, S. (eds) *The eruption of Hekla 1947-1948*. Societas Scientiarum Islandica, Reykjavik, 1-177, 1967.

Thordarson, T., and Höskuldsson, Á.: Postglacial volcanism in Iceland. Jökull, 58, 197-228, 2008.

Thordarson, T., Miller, D. J., Larsen, G., Self, S., and Sigurdsson, H.: New estimates of sulfur degassing and atmospheric mass-loading by the 934a.d. Eldgjá eruption, Iceland. J. Volcanol. Geotherm. Res., 108, 33–54, 2001.

Thordarson, T., and Larsen, G.: Volcanism in Iceland in historical time: volcano types, eruption styles and eruptive history. J. Geodyn., 43, 118–152, 2007.

Thordarson, T., Self, S., Miller, D. J., Larsen, G., Vilmundardóttir, E.G.: Sulphur release from flood lava eruptions in the Veidivötn, Grímsvötn and Katla volcanic systems, Iceland. In: C. Oppenheimer, D.M. Pyle, and J. Barclay (Editors), *Volcanic Degassing*. Geol. Soc. London, Spec. Publ. **213**, 103-121, 2003.

Thordarson, T., Self, S., Óskarsson, N., and Hulsebosch, T.: Sulphur, chlorine, and fluorine degassing and atmospheric loading by the 1783-1784 AD Laki (Skaftár Fires) eruption in Iceland. Bull. Volcanol., 58, 205-225, 1996.

Thordarson, T., Miller, D. J., Larsen, G., Self, S., and Sigurdsson, H.: New estimates of sulfur degassing and atmospheric mass-loading by the 934a.d. Eldgjá eruption, Iceland. Journal of Volcanology and Geothermal Research, 108, 33–54, 2001.

Timms, R. G. O., Matthews, I. P., Lowe, J. J., Palmer, A. P., Weston, D. J., MacLeod, A., Blockley, S. P. E.: Establishing tephrostratigraphic frameworks to air the study of abrupt climatic and glacial transitions: a case study of the Last Glacial-Interglacial Transition in the British Isles (c. 16-8 ka BP). Earth-Science Reviews, 192, 34-64, 2019.

Walker, G. L.: Explosive volcanic eruptions—a new classification scheme. Geol Rundsch, 62, 431–446, 1973.

Wastegård, S., Gudmundsdóttir, E. R., Lind, E. M., Timms, R. G. O., Björck, S., Hannon, G. E., Olsen, J., and Rundgren, M.: Towards a Holocene tephrochronology for the Faroe Island, North Atlantic, Quaternary Science Reviews, 195, 195-214, 2018.