Item S1: (MS Word doc). Detailed methods for major, minor, and trace-element analysis:

 Electron-microprobe analysis (EMA)

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**SUPPLEMENTAL MATERIAL, sections A through E**

**A: DETAILED METHODS FOR MAJOR, MINOR AND TRACE-ELEMENT ANALYSIS**

**Electron-microprobe analysis (EMA)**

Major-element composition of glass shards was determined at the Department of Earth Sciences, University of Toronto, Canada, the U. S. Geological Survey at Menlo Park, California, U.S.A., and the Earth Sciences Department at the University of Utah, Salt Lake City, U.S.A. (sections B, C, D).

At the University of Toronto, the bulk tephra samples were sieved to obtain the 0.25 – 0.125 mm size fraction and then ultrasonically cleaned for 20 minutes, dried, and sieved again to concentrate the coarsest material. Volcanic glass shards were separated from minerals and lithic fragments, and the platy and bubble-wall glass shards were separated from pumiceous glass shards using a Frantz isodynamic magnetic separator. The fraction containing the platy and bubble-wall glass was then used for compositional analysis and age determination. Major-element concentrations in the glass were determined using a JEOL JXA-8230 Electron Probe Micro-Analyzer with 5 wavelength-dispersive X-ray spectrometers. Operating conditions were 15kV accelerating voltage, 6 nA current, and 10 µm beam diameter. The MPI-DING ATHO-G reference glass (Jochum and others, 2006) was used to monitor accuracy of the glass analyses.

At the U.S. Geological Survey Tephrochronology Laboratory, volcanic glass shards were separated from field samples collected for electron-microprobe (EMA) by disaggregating or crushing, and wet-sieved using nylon screens and plastic screen holders with openings of 45, 80, and 150 mm. Samples were further separated using a magnetic separator into magnetic, non-magnetic, and intermediate fractions, and the last was separated further using a mixture of a heavy liquid (methylene iodide) and acetone to float the volcanic glass from other components. This last separate was used for both the EMA and the INAA analyses (see below).

Electron-microprobe analyses were determined on three successive instruments: ARL, SEQM, and JEOL8900 microprobes, over a time span of ~1970 to ~2020 (section C). With the JEOL8900, we use a 15-kV accelerating voltage, 10.05-nA current, and 10 µm beam diameter. Acquisition time is 10 seconds for Na, and 20 seconds for the remaining elements: Si, Al, Fe, Mg, Mn, Ca, Ti, and K. We use GSC (Corning Glass Standard) and An40 (anorthite) as standards, and RLS-132, a homogenous obsidian from La Puebla, Mexico, as an internal standard. The ZAF reduction program is used to obtain oxide concentrations for the elements analyzed. Replicate analyses of internal standards and type tephra samples over the past ~50 years indicate that analyses are reproducible and precise despite differences in instrumentation over time. Precision and accuracy of analyses for each oxide are indicated by replicate analyses of RLS-132 as well as of specific widespread tephra layers (SECTION C).

EMA and LA ICP-MS makes it possible to determine the composition of individual glass shards and, thus, obtain a measure of the homogeneity or heterogeneity of the glass-shard population of a sample. Sodium, and to a lesser degree, potassium concentrations can be quite variable between samples of the same tephra layer, a consequence of both the problem of sodium analysis as well as differential leaching or enrichment of the mobile alkalis in volcanic glass under different natural storage conditions. For these reasons, both the alkalis that were analyzed, sodium and potassium, appear to have errors that are greater than expected for the concentrations in which they are present in volcanic glass shards (Fig. 5). See Sarna-Wojcicki, 1971, 1976, 2000; Sarna-Wojcicki and others 1984, 2005, 2011, for additional information on sampling, analytical procedures and problems, and data interpretation.

**Instrumental neutron activation analysis (INAA)**

For INAA, sized fractions of separated glass shards prepared for EMA were additionally chemically cleaned with 10% HCl, then briefly etched with 8% HF, and rinsed in an ultrasonic bath in deionized water, if not done initially. Abundances of thirty-three major, minor and trace elements, including 11 rare-earth elements (REE), were determined on ~0.5 - 1 g samples of separate, chemically cleaned glass shards by INAA at the USGS-Denver laboratory. In this procedure, two irradiations are performed. The samples are first irradiated in the USGS-TRIGA reactor at a neutron flux of 2.5x1010 n/cm2/sec for thirty minutes followed by a long irradiation at a flux of 2.5x1012 n/cm2/sec for six hours. The absorption of neutrons causes these elements to emit characteristic gamma-rays which also decay at unique rates. Gamma-ray radiation emitted by a radionuclide is converted to an electrical signal by a semiconductor detector. The electrical signal is analyzed by a multichannel analyzer. A gamma spectrum is produced and the intensities of each gamma-ray photopeak are measured. The counts accumulated for each elemental photopeak are corrected for background, elemental interferences and decay, and the count rates are directly proportional to the concentration. Counting is performed immediately following the short irradiation, whereas three sequential counts at 7, 14, and 65 days are made after the longer irradiation utilizing coaxial and planar germanium detectors. The abundances of elements that produce very short-lived radionuclides (e.g., K, Dy and Mn) are determined in the first irradiation. Radionuclides with half-lives on the order of hours (e.g., Na, La, and Sm) are measured in the early counts, and long-lived radionuclides (e.g., Cs, Zr, Gd and Zn) in the later counts. Both precision and accuracy for most of the elements determined range from 2 to 5 %, including the REEs: La, Ce, Sm, Eu, Tb, Yb, Lu; and Hf and Ta. Only five elements have precision errors greater than 10% (Tm, W, Sb and Au) based on one sigma counting statistics [errors] (i.e., the standard deviation of the measurements). Accuracy is based on [the] analysis of USGS standard reference materials, including BHVO-1. A summary of the INAA procedure used at the USGS is given in Budahn and Wandless (2002).

The elements we found most useful in correlation of silicic tephra layers are the incompatible trace elements, and several minor and major elements. In order of increasing atomic number these are: the lighter metallic elements Sc, Mn, Fe, Rb, Cs, Ba, the rare earth elements (REE) La, Ce, Nd, Sm, Eu, Tb, Dy, Yb, Lu, and the heavy metals Hf, Ta, Th, and U. The usefulness of specific elements for correlation varies to some extent among volcanic source areas, their tectonic settings, the major-element compositions of the parent rocks from which magma is derived, the compositions of the magmas from which the tephra layers were generated, and the temperatures of eruption. These factors determine the abundances of the elements/oxides in the volcanic glass at and above detection levels, and the presence or absence of these elements in crystalline phases associated with the volcanic glass. The presence of microphenocrysts in glass shards containing elements of interest, such as Sr, Zn, Fe, Ba, U, Th, and Zr, can introduce scatter, and thus lower precision, in chemical analysis of volcanic glass by INAA.

**Laser-ablation inductively-coupled plasma mass spectrometry (LA-ICP-MS)**

Trace, minor and major elements were analyzed using the LA-ICP-MSA system at the Department of Earth Sciences, University of Toronto following methods outlined below. We used a New Wave UP-213 (solid state frequency quintupled Nd-YAG) laser ablation microscope connected to a VG PQ-Excell quadrupole ICP-MS. Helium was used as the transfer gas at a rate of 1000 cc/min and the plasma was tuned to achieve oxide production of less than 2% monitoring ThO+/Th+ (masses 248/232) and U+/Th+ (masses 238/232) as close as possible to unity. Samples were analyzed for 80 seconds of which the first 20 seconds consisted of background. The sample cell was flushed for a minimum of 60 seconds between each analysis. Glass shards from each sample were analyzed in batches with NIST 610 being analyzed at the beginning and end of each batch. ATHO-G was also analyzed in each batch as a reference standard. Laser conditions for NIST 610 analyses were 10 Hz repetition rate with a 55 µm spot in lines approximately 250 µm long. Glass shards and ATHO-G were analyzed using a 25 µm spot as lines, 150 µm long for ATHO-G and 120 – 200 µm long for glass shards, depending on the size of the shard, with the same 10 Hz repetition rate and a scan speed of 10 µm/sec. Fluence values for ATHO-G and glass shards ranged from 9.0 to 9.30 J/cm2 within each run and for NIST 610 the fluence was 6.5 J/cm2. The resultant time resolved data were reduced offline using the Glitter version 4.4.4 software package (MacQuarie Research Ltd.). Elements were quantified using NIST 610 (Pearce and others 1997) and ablation yields were corrected using SiO2 values (anhydrous) obtained by electron microprobe analysis. This procedure gave an average composition for each glass shard with fractionation effects minimized, if present, by the shallow penetration of the laser.

**Isothermal-plateau fission-track method of age determination**

The glass-fission-track method used to determine the age of tephra beds in Montana – beds that are correlated to the IHT – is the zeta calibration method, which is based on analysis of age standards (Hurford and Green, 1983; Wagner and Van den Haute, 1992). These determinations were carried out at the University of Toronto. The age standard used is the Moldavite tektite glass (Lhenice locality) with a 40Ar/39Ar age of 14.808 ± 0.021 Ma (2σ) (Schmieder and others 2018). The population-subtraction variant was used (Westgate, 2015), and correction for partial track fading was carried out using the isothermal plateau method (Westgate, 1989) or the diameter correction method (Sandhu and Westgate, 1995). Zeta value is 311 ± 4 based on 7 irradiations at the McMaster Nuclear Reactor, Hamilton, Ontario, using the NIST SRM 612 glass dosimeter and the Moldavite glass. Area was determined with an eyepiece graticule or estimated using the point-counting technique (Naeser and others, 1982). The Walcott Tuff, (6.27 ±0.05, Morgan and McIntosh, 2005; also see Watts and others, 2011) was used as an internal age standard to monitor the accuracy of the neutron fluence estimate.

Four glass-fission-track (ft) ages were determined on IHT samples in Montana. Glass shards in these samples were thick enough to give sufficient surface area for meaningful age determinations. An attempt to determine the glass-ft age of IHT at site 8 in Nebraska failed because the glass shards were too thin, the site being 1400 km east of the source vent. Two independent methods were used to correct for partial track fading and estimation of the glass surface area (Table 5). All 4 age estimates are statistically the same, the weighted mean age and error being 11.49 ± 0.44 Ma (1σ).

**Correlation Methods**

Correlation of tephra layers can be accomplished by several graphical and statistical methods (Sarna-Wojcicki, 2000; Lowe, 2011). In this study, the reported EMA data are normalized to 100 percent (water-free basis) and compared to over 7000 other analyses in the U.S. Geological Survey tephra database using the similarity coefficient of Borchardt and others (1972). Similarity coefficients were calculated using SiO2, Al2O3, Fe2O3, CaO, and TiO2. Samples were analyzed by INAA for 34 major, minor, and trace elements. Of these, Sc, Mn, Fe, Ba, Rb, Sr, Zr, Cs, La, Ce, Nd, Sm, Eu, Tb, Dy, Yb, Lu, Hf, Ta, Th, and U were found in sufficient concentration and consistency for correlation. For this study, Ba, Rb, Cs, La, Ce, Nd, Sm, Eu, Tb, Dy, Yb, Lu, and Hf were used in this study because of the high precision of measurement and consistent results in replicate analyses. Scatter and low precision of some elements (i.e., Ba, Sr, and Zr) may be the result of concentration in small phenocrysts and microlites that have escaped our separation procedures. Samples of glass shards were also analyzed by LA-ICP-MS for 24 elements. Several samples were analyzed by both techniques, and good agreement was obtained in most instances. Previous studies have shown that solution inductively-coupled mass spectrometry (S-ICP-MS and INAA analyses) of some of the same volcanic glass samples yield very similar results (Knott and others, 2007). In this study, tephra layers were correlated on the basis of similarity in compositions (similarity coefficient), relative stratigraphic position to other tephra layers or homotaxial sequence (Huxley, 1862), independent dating and other methods such as biostratigraphy. Graphically, rare-earth elements were normalized using the values from Boynton (1992).

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