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Assessing New and Old Methods in Paleomagnetic Paleothermometry: A Test Case at Mt. St. Helens, USA

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Assessing New and Old Methods in Paleomagnetic Paleothermometry: A Test Case at Mt. St. Helens, USA

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Key Points:
- Large overestimates in deposit temperature result from multidomain (MD) juvenile material; smaller overestimates from MD lithic material
- Small to moderate overestimates may result from thermally activated reordering in titanomagnetite during deposit cooling
- Stratigraphic variations in Curie temperatures arising from thermally activated reordering are modeled in terms of deposit temperature

Supporting Information:
- Supporting Information S1

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Abstract Paleomagnetic data can be used to estimate deposit temperatures (T_{dep}) of pyroclastic density currents (PDCs) by finding the laboratory temperature at which a PDC-associated thermal remanence is removed. Paleomagnetic paleothermometry assumes that (1) blocking (T_b) and unblocking (T_{ub}) temperatures are equivalent, and (2) the blocking spectrum remains constant through time. The first assumption fails for multidomain (MD) grains, and recent evidence shows that the second is violated in many titanomagnetites, where T_c is a strong function of thermal history. Here we assess the extent to which the standard paleomagnetic method may be biased by a changing T_b spectrum, and we explore a new magnetic technique that instead exploits these changes. Using samples from the 1980 PDCs at Mt. St. Helens, we find that standard methods on oriented lithic clasts provide a T_{dep} range that overlaps with measured temperatures, but is systematically slightly higher. By contrast, juvenile pumice give T_{dep_min} estimates that greatly exceed lithic estimates and measured temperatures. We attribute this overestimate to (1) depth-dependent variations in T_c and T_{ub} resulting from thermally activated crystal-chemical reordering and (2) MD titanomagnetite where $T_{ub} > T_b$. Stratigraphic variations in T_c are interpreted in terms of T_{dep}, giving results mostly consistent with measured temperatures and with the lower end of estimates from lithic clasts. This new method allows us to evaluate temporal and spatial variations in T_{dep} that would not have been possible using standard paleomagnetic techniques in these lithic-poor deposits. It also provides information on deposits not accessible by surface temperature probes.

1. Introduction

Pyroclastic density currents (PDCs) are among the most of dangerous of volcanic hazards. Mixtures of hot gas and pyroclastic material, PDCs rapidly travel downslope and can devastate anything in their path. Assessing the temperature of PDCs allows for a better understanding of the volcanic processes at play and aids in assessing future hazards. Perhaps the only way of determining PDC paleotemperatures over a wide temperature range is the use of paleomagnetic techniques (e.g., Aramaki & Akimoto, 1957; Bardot & McClelland, 2000; Hoblitt & Kellogg, 1979; Kent et al., 1981; Paterson et al., 2010).

The traditional approach is to extract remobilized lithic clasts incorporated into the PDC. The lithic material typically carries a magnetization that predates the eruption, and some or all of the magnetization may be overprinted when the clast is reheated as part of the PDC. By thermally demagnetizing the clasts and evaluating the temperature at which this overprint is removed, we can infer the temperature to which the clast was reheated. The temperature interval over which this technique is effective depends on the temperature spectrum over which the magnetization is removed, or unblocked, which in turn depends on magnetic mineral composition and grain size distribution. The maximum temperature that can be constrained by this method is often 580°C, the Curie temperature (T_c) of pure magnetite, although (titano)hematite (if present) may unblock up to 675°C. The method has been compared to direct temperature measurements in at least two locations: the 1980 Mt. St. Helens PDCs (Erwin, 2001; Paterson et al., 2010) and the 1982 eruption of El Chichón (Sulpizio et al., 2008). In both cases, the paleomagnetic method reasonably reproduced measured temperatures.

The technique is most effective when two clear directional components of magnetization are present in the clasts. A component removed at laboratory temperatures below the PDC deposit temperature (T_{dep}) should

be parallel to the expected field direction at the time of the eruption. The high-temperature component ($T > T_{dep}$) should be randomly oriented and equates to the remaining pre-PDC magnetization.

It has been suggested that single-component magnetizations can also be used to partially constrain T_{dep} (Hoblitt & Kellogg, 1979). In this case, if the magnetization is parallel to the expected field, T_{dep} is greater than the maximum unblocking temperature ($T_{ub\text{-}max}$). If the magnetization is not parallel to the expected field, the clast cannot have been reheated above the minimum unblocking temperature ($T_{ub\text{-}min}$). When applied to lithic fragments, which likely had a pre-existing magnetization, single-component magnetizations should typically produce reliable results. However, when applied to juvenile material (pumice, ash, scoria), which is not expected to have a pre-eruption magnetization, it has a history of mixed results.

Kent et al. (1981) examined lithic fragments, as well as ash and pumaceous matrix from the AD79 Vesuvius eruption. While the lithics showed two-component behavior indicating a T_{dep} between 350°C and 400°C, the matrix had a single component with unblocking up to 550°C. This high-temperature unblocking was interpreted to be not of thermal origin. Zlotnicki et al. (1984) (Guadeloupe, French West Indies); McClelland and Thomas (1990) and McClelland and Druitt (1989) (Santorini, Greece); Donoghue et al. (1999) (Ruapehu, New Zealand); and McClelland et al. (2004) (Taupo, New Zealand) all found that nearly all pumice clasts had a single component remanence that unblocked all the way to the maximum T_c. McClelland and Thomas (1990) and McClelland and Druitt (1989) additionally found that this $T_{ub\text{-}max}$ was consistently greater than T_{dep} estimated from multicomponent lithic fragments, sometimes by hundreds of degrees. All of these studies concluded that the high unblocking temperatures represented a chemical remanence acquired post-deposition. Erwin (2001) reported extensively on MSH deposits, sampling both lithic and pumice material. However, results were not reported by clast type, so the relative success of the pumice is unknown. Paterson et al. (2010) examined both pumice and lithic clasts from the 12 June and 24 July 1980, eruptions at Mt. St. Helens, USA, and the 1993 eruption at Láscar, Chile. In both cases, $T_{dep} > T_c$, and all pumice and all but one lithic had a single component magnetization that unblocks all the way to T_c. Finally, Rader et al. (2015) sampled both lithic and juvenile scoria clasts from the 2006 eruption at Tungurahua and the 1977 eruption at Cotopaxi, both in Ecuador. They found that the juvenile material consistently gave significantly higher temperatures (>590°C) and interpreted the deposits as having heterogeneous temperature distributions.

This paleomagnetic method of temperature determination requires that the blocking (T_b) and unblocking temperatures are equivalent and that the blocking spectrum remains constant through time. In other words, a magnetization acquired at $T \leq T_b$ should be completely removed by laboratory heating to exactly $T_{ub} = T_b$. As noted above, a remanence which is not thermal in origin, such as a chemical remanent magnetization (CRM), does not meet these requirements. A magnetization carried by coarse, multidomain (MD) grains also does not meet the requirement that $T_{ub} = T_b$, and an MD thermal overprint acquired at $T < T_{dep}$ may not completely demagnetize until T_c (where T_c is always $\geq T_{ub}$).

Another complication related to blocking temperatures comes from observations that T_c of common titanomagnetites ($Fe_{3-x}Ti_xO_4$, $0 \leq x \leq 1$) is a strong function of prior thermal history (Bowles et al., 2013; Jackson & Bowles, 2014). Natural or laboratory annealing at moderate temperatures (~300–450°C) can cause T_c to increase by >100°C, and as T_c varies, the blocking spectrum must also change. If Curie and blocking temperatures are modified during heating and cooling of the deposit materials, we may reasonably expect the resulting T_{dep} estimates to be affected.

In this study, we report on experiments designed to assess the amount of uncertainty or bias in T_{dep} that might be expected when the remanence is carried by MD titanomagnetite that undergoes thermally activated crystal-chemical reordering. We also explore another magnetic method of constraining T_{dep} that may be useful especially when lithic clasts are not present. This method exploits the variability of T_c that arises from thermally activated reordering within the titanomagnetite and can be used on unoriented pumice clasts and/or ash matrix. We selected Mt. St. Helens (MSH) as the sampling location because direct emplacement temperature measurements were made following the 18 May 1980, eruptions (Banks & Hoblitt, 1996).

2. Summary of Titanomagnetite Reordering Effects

The reordering phenomenon is strongest in titanomagnetites of intermediate composition ($x \sim 0.2 - 0.4$), which are common in volcanic materials. The process is thought to arise from some form of thermally

activated cation or vacancy reordering within the crystal lattice. The equilibrium degree of order is temperature dependent, and at high temperatures, a lesser degree of order is associated with a lower T_c; at low temperatures, a higher degree of order is associated with a higher T_c. The rate of reordering is also temperature dependent, with faster reordering at higher temperatures.

If a sample is cooled rapidly from magmatic temperatures, it will become continuously more ordered until it reaches a rate-dependent closure temperature, T_{close}, where the reordering process is too slow to remain in equilibrium and the order state at T_{close} is locked in. Rapid cooling from any temperature $T > T_{close}$ will result in the same T_c ($T_{c\text{-}close}$). By measuring susceptibility as a function of temperature, $\chi(T)$, $T_{c\text{-}close}$ is identified on the cooling leg of this experiment, because the sample is rapidly cooled from 600°C $> T_{close}$. The exact temperature is composition dependent, but the titanomagnetites of the 18 May PDCs all have similar composition and $T_{c\text{-}close} \sim 375$°C.

Applying these concepts to a PDC where $T_{dep} < T_{close}$, the juvenile material will cool rapidly during transit, from magmatic temperatures through T_{close}, and will be deposited at T_{dep} with a degree of order lower than the equilibrium degree. Material at the surface of the flow will continue to cool rapidly to ambient temperatures and this low degree of order will be locked in, with $T_c = T_{c\text{-}close}$. Samples deeper in the flow, however, will remain hot for some time and will slowly evolve toward the equilibrium state at T_{dep}, with a higher T_c. This can lead to an increase in T_c with depth, as previously observed in one MSH PDC deposit (Jackson & Bowles, 2014). As temperature continues to decrease, reordering becomes so slow as to be insignificant and the final state is frozen in. We can calculate the T_c differential (ΔT_c) that results from spending time at elevated temperatures $T < T_{close}$ by calculating the difference between T_c measuring on warming and T_c measured on cooling during the $\chi(T)$ experiment.

See Bowles et al. (2013), Jackson and Bowles (2014), and Jackson and Bowles (2018) for a more detailed discussion of the reordering process.

3. Geologic Background and Sampling

On 18 May 1980, at 8:32 a.m., a landslide weakened and collapsed the northern slope of Mt. St. Helens resulting in a violent lateral blast as the volcano erupted. An additional five phases in the Plinian eruption sequence occurred over the next 10 h, resulting in multiple ash flows and PDCs (Christiansen & Peterson, 1981; Criswell, 1987). Five distinct PDC units have been identified, ranging in average thickness from 4 to 7 m, with variable amounts of ash, crystals, pumice, and lithics, although the majority are composed of nonwelded pumice and ash with minor lithics (Brand et al., 2014). Subsequent smaller eruptions on 25 May, 12 June, 22 July, 7 August, and 16 October all left additional PDC deposits of nonwelded dacitic pumice and ash (Christiansen & Peterson, 1981).

Temperature profile measurements of all the 1980 PDC deposits were made by inserting temperature probes into the deposits in the days and weeks following the eruptions (Banks & Hoblitt, 1996). The measured deposit temperatures for the 18 May PDCs ranged from 297°C to 418°C. Temperatures in the region sampled for the present study are shown in Figure 1. The later PDCs were progressively hotter than the 18 May PDCs.

The majority of samples were collected in 2014 from the 18 May PDCs (denoted MSH14), but one deposit (MSH10-02) was sampled in 2010 (Figure 1). This latter deposit is closer to the vent and likely represents a later event. Whenever possible, we targeted deposits with intact surfaces of capping airfall ash in order to precisely measure depth within a flow. All sample depths are reported as depth below this airfall. Unoriented matrix (ash, pumice, and lithic lapilli) was collected below these ash layers every 5–10 cm down to ~1.0–1.5 m in depth. Most deposits were considerably thicker than this, meaning we only sampled the uppermost meter. In two cases the lowermost 0.5–1.0 m of the overlying deposit was sampled. Lithic lapilli found in the matrix were <1 cm diameter and pumice clasts were <2 cm diameter, but at MSH14-01 and MSH14-08, pumice clasts were up to ~10 cm. In two cases (MSH10-02, MSH14-04), oriented 1 in. diameter cores were taken from larger pumice using a battery-powered drill and oriented using a magnetic or sun compass. Oriented cores were also taken from a single layer of concentrated large (up to 30 cm diameter) lithic blocks (MSH14-05). Several oriented lithic and pumice clasts (<~10 cm diameter) from sites MSH10-02, MSH14-02, and MSH14-06 were brought back to the University of Wisconsin-Milwaukee, where they

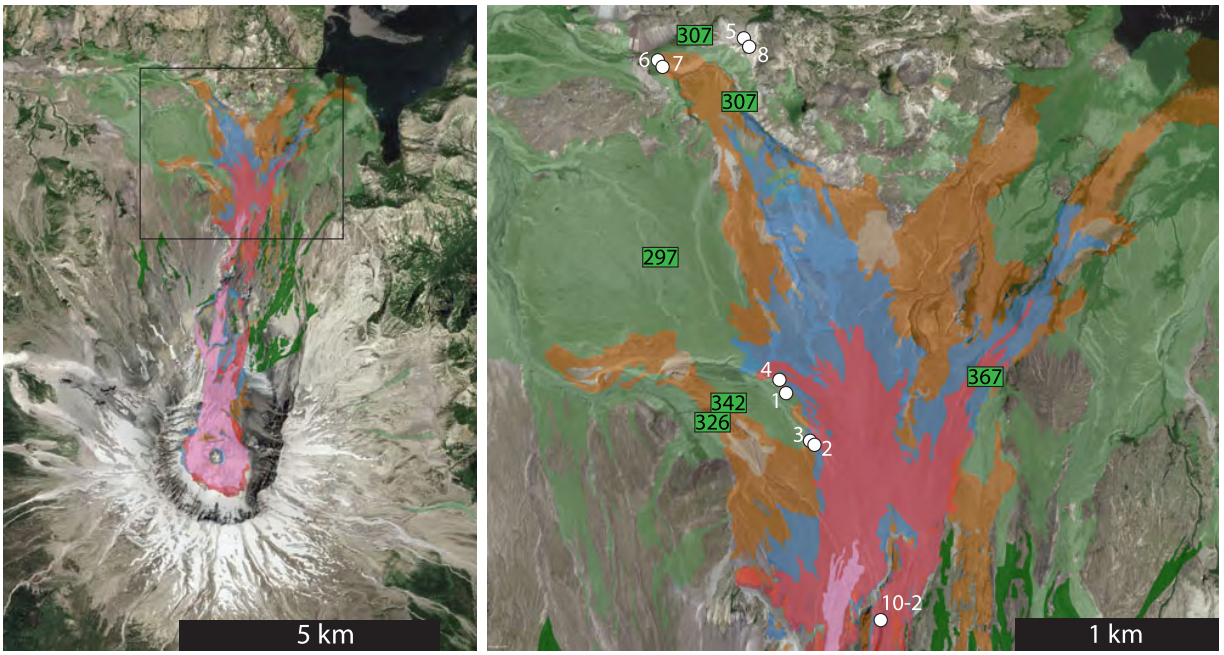

Figure 1. Sample location map. Sampling locations shown by white circles. Sites sampled in 2014 (MSH14) are indicated by a single number. Site sampled in 2010 (MSH10) is 10-2. 18 May 1980 deposit temperatures measured by Banks and Hoblitt (1996) shown in green boxes. Extent of PDCs (Kuntz et al., 1990) shown by colored shading: 18 May (green), 12 June (tan), 22 July (blue), 7 August (red), and 16 October (pink). Base imagery from GoogleEarth.

were either cored or cut into cubes. Site coordinates, deposit descriptions, and materials collected are given in supporting information Table S1. Additional deposit descriptions and photographs can be found in Gerzich (2016).

Site MSH14-08 is within several meters of and equivalent to site MSH12-08 of Jackson and Bowles (2014), where T_c was found to systematically increase with depth from ~375°C at the top to nearly 500°C at 1.6 m depth. The pumice from this site also has a relatively simple magnetic mineralogy: predominantly single-phase, multidomain, homogeneous titanomagnetite ($Fe_{2.55}Ti_{0.26}Mg_{0.10}Al_{0.08}O_4$; Bowles et al., 2013, 2015; Jackson & Bowles, 2014). This simplicity allows experiments to isolate the magnetic mineralogy responsible for the time-dependent and temperature-dependent magnetic properties. The ash matrix from this site, as well as pumice from some other MSH locations, is known to also contain oxy-exsolved titanomagnetite with ilmenite lamellae (Jackson & Bowles, 2014). The homogeneous and oxy-exsolved titanomagnetites are interpreted to be primary magmatic constituents, and the fraction of oxy-exsolved grains may be linked to deposit temperature. Some matrix samples have a more complicated magnetic mineralogy with three or more Curie temperatures, and Kuntz et al. (1981) additionally identified iron-titanium oxides with complex intergrowths and altered margins that were interpreted as xenocrysts or xenolith fragments.

4. Laboratory Methods

Unoriented pumice samples from MSH14-08 and MSH14-01 were cut into 1 in. cubes and were thermally demagnetized in an ASC Model TD-48SC in nitrogen atmosphere. Oriented lithic and pumice samples were thermally demagnetized in air. Because so few lithic clasts large enough to orient were found, 29 small unoriented lithic lapilli fragments from sites MSH14-01, MSH14-02, MSH14-03, MSH14-04, MSH14-06, and MSH14-08 were separated from the matrix and immobilized in glass tubes for thermal demagnetization in air. Room-temperature susceptibility was measured on a Bartington MS2 susceptibility bridge following each heat treatment in order to test for possible changes in the magnetic mineralogy during heating. All remanence measurements were made on a Molspin Minispin spinner magnetometer housed in the shielded room at the University of Wisconsin-Milwaukee (UWM). The exceptions to these protocols are: (1) the small, unoriented lithics were measured on a 2G Enterprises superconducting rock magnetometer (SRM) with DC SQUIDs; and (2) the oriented pumice samples from MSH10-02 were heated in air in a

Schoenstedt thermal demagnetizer and measured on a 2G Enterprises SRM with RF SQUIDs in the shielded room at the Institute for Rock Magnetism, University of Minnesota. Directional analysis of remanence data was carried out using the PmagPy Pmag GUI (Tauxe et al., 2016).

χ(T) was measured on an Agico MFK Kappabridge susceptibility bridge with CS4 furnace attachment under argon atmosphere at UWM. Curie temperatures were determined by finding the (negative) peak in the first derivative of χ(T) (Petrovský & Kapička, 2006).

Hysteresis was measured on pumice and lithic clasts using a Princeton Measurements vibrating sample magnetometer at the Institute for Rock Magnetism in maximum applied fields of $\pm$1 T. Coercivity of remanence was measured from a backfield isothermal remanent magnetization acquisition following saturation in a 1 T field.

5. Results

5.1. Hysteresis Data

Hysteresis results (supporting information Figure S1) show that average sample domain state ranges from near single-domain (SD) to MD, with lithic samples closer to the SD end-member and pumice samples closer to the MD end-member. MD states in the pumice are consistent with previous observations by optical microscopy, which showed common grain sizes of a few hundred micrometers (Kuntz et al., 1981; Jackson & Bowles, 2014). Migration toward the SD end-member in the pumice is associated with an increasing fraction of oxy-exsolved grains.

5.2. χ(T) Results

Thermomagnetic data from both ash matrix and pumice clasts are shown in Figure 2. In most cases, the pumice data reflect a simpler magnetic mineralogy with a single dominant T_c between ~375°C and ~475°C when measured on warming. When measured on cooling T_c is consistently close to 375°C. We interpret this T_c to be associated with the homogeneous titanomagnetite that experiences thermally activated reordering, and T_c measured on cooling is ~$T_{c\text{-}close}$. The exceptions to this single-phase mineralogy are pumice from MSH14-07 (Figure 5j) and MSH14-04 (supporting information Figure S2). At these two sites, there may be several prominent, discrete Curie temperatures on both warming and cooling.

Data from the ash matrix are typically more complex. In addition to the T_c that we infer to be associated with homogeneous titanomagnetite, there are also one or more additional phases with higher Curie temperature that do not exhibit the same systematic behavior. We interpret these as related to an oxy-exsolved titanomagnetite and/or a higher fraction of xenocrysts or very fine lithic lapilli which have more heterogeneity in magnetic mineralogy.

Sites MSH14-02 (Figures 2c and 2d), MSH14-06 (Figures 2g and 2h), and MSH14-08 (Figure 2l) all show a systematic increase with depth in the T_c associated with homogeneous titanomagnetite. This is consistent with previous observations at MSH12-08 (Jackson & Bowles, 2014). In contrast, data from MSH14-01 (Figures 2a and 2b), MSH14-03 (Figures 2e and 2f), and MSH10-02 (not shown) are nearly constant with depth, showing no systematic increase. Data from MSH14-07 (Figures 2i and 3j) and MSH14-04 (supporting information Figure S2) are extremely variable with depth.

To show the depth-dependent trends in T_c more clearly, the derived homogeneous titanomagnetite Curie temperatures have been plotted versus depth below the capping ash layer (Figure 3), in addition to the difference between T_c measured on warming and T_c measured on cooling (ΔT_c).

5.3. Thermal Demagnetization Data

For all the oriented lithics, samples showed two types of demagnetization behavior (Figure 4): (1) univectoral decay where most unblocking occurred at relatively high temperatures (>400–500°C) and the resulting principal component is randomly oriented with respect to the 1980 field; and (2) two-component decay where a low-temperature (low-T) component is close to the 1980 field direction and the high-temperature (high-T) component is randomly oriented (Figure 5). In one sample (MSH14-05-a1, Figure 4g), three components were present, and the lowest-temperature component was parallel to the 1980 field. In some cases, the fraction of magnetization removed at low T_{ub} is small, and the direction is therefore poorly defined; nevertheless, it is consistently close to the 1980 direction. For one clast (MSH14-06-b1), the low-temperature

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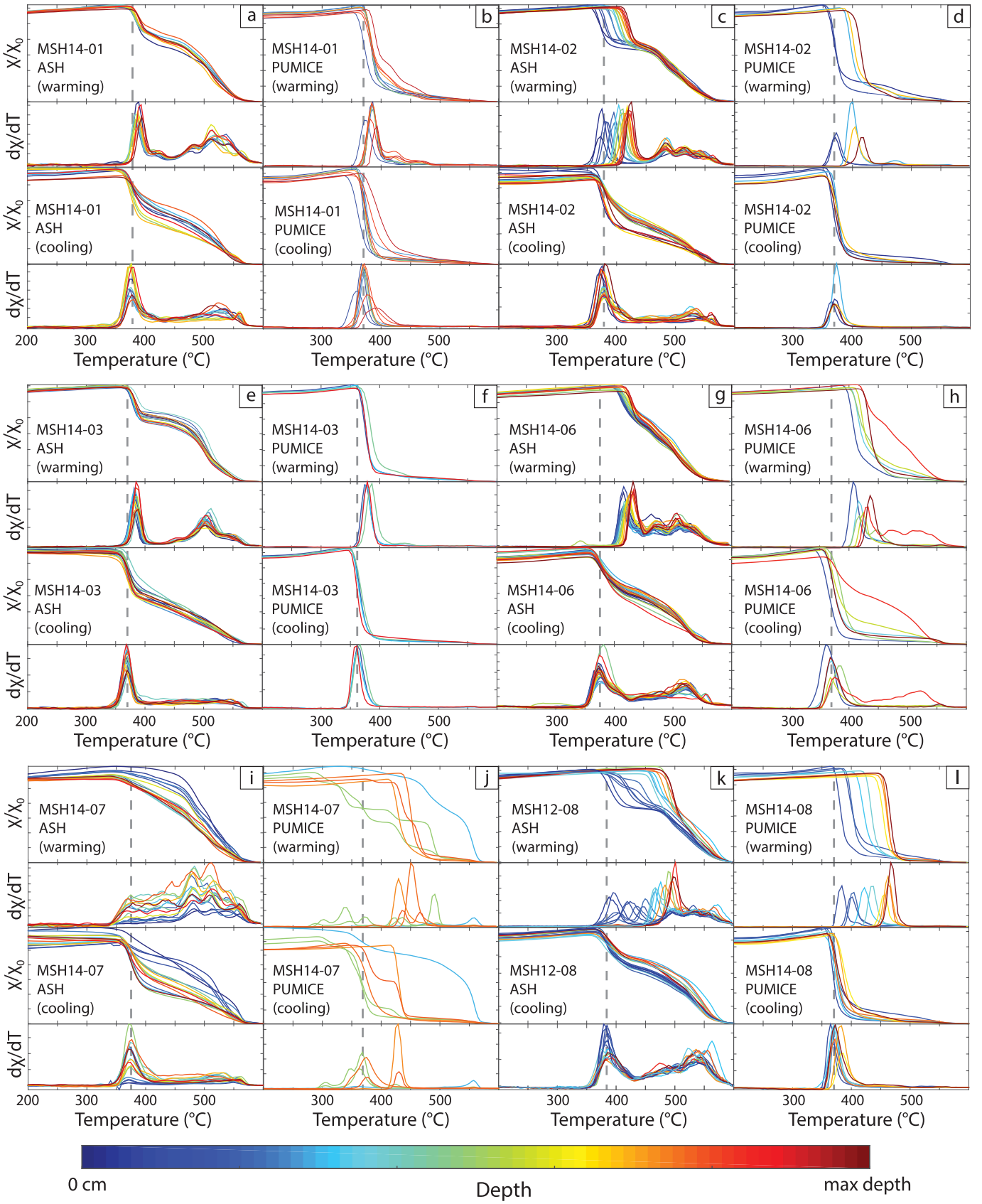

Figure 2. (continued)

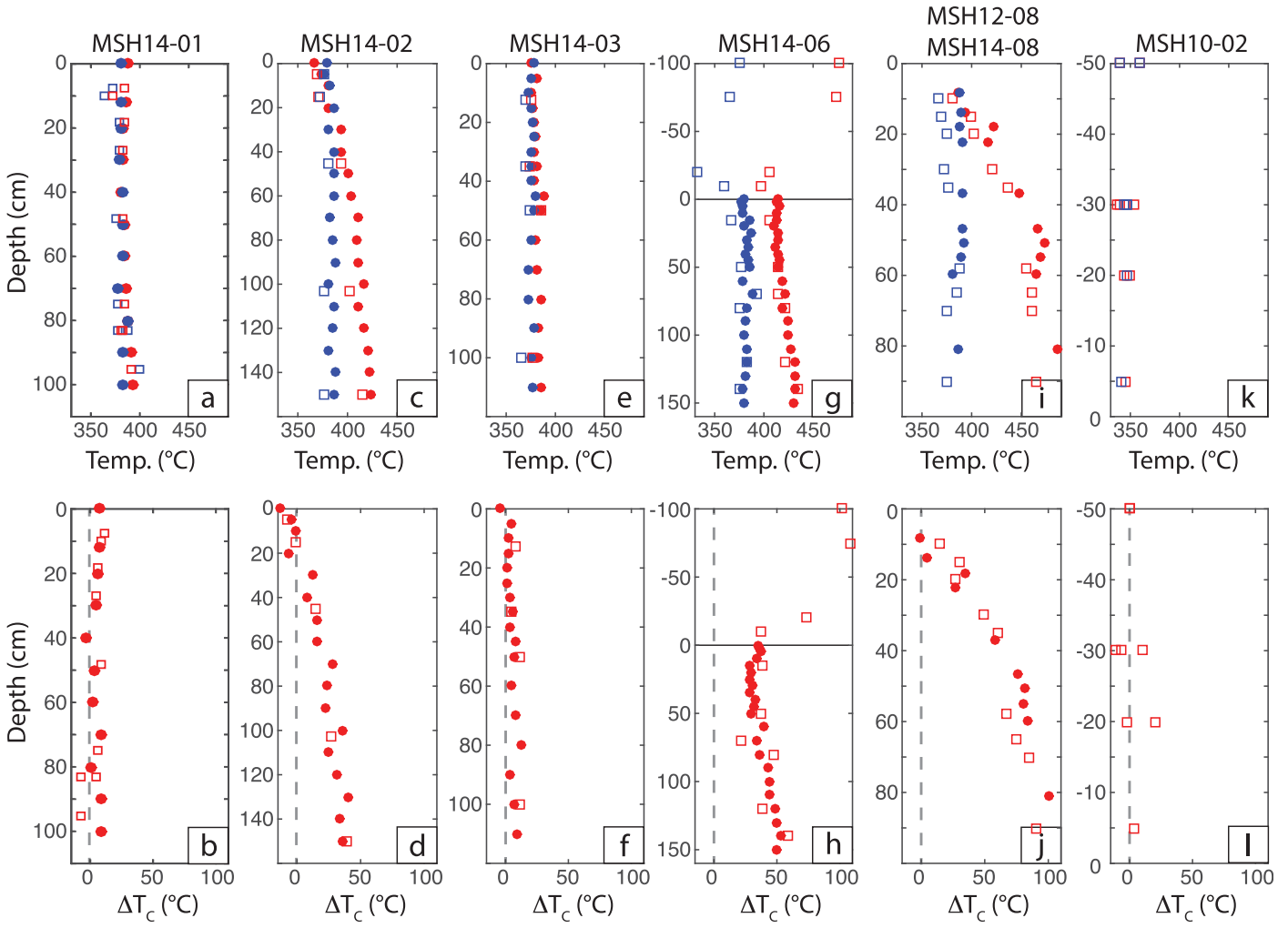

Figure 3. Depth-dependence of Curie temperature for homogeneous titanomagnetite. Top row is T_c measured on warming (red) and cooling (blue) for both ash/matrix (solid circles) and pumice (open squares). Bottom row is the difference between the warming and cooling T_c. (i,j) Ash data are from MSH12-08 (Jackson & Bowles, 2014); pumice data are from MSH14-08. Negative depths indicate height above the unit boundary or reference horizon.

component was not parallel to the 1980 field, and we suspect that the clast was misoriented in the field or during subsampling in the lab. A second clast from the same site has a low-T component with the 1980 direction.

The temperature break between the low-T and high-T components is rarely sharp, and we list this temperature interval in supporting information Table S2, along with the directions of the two components and the Curie temperature(s). The selection of this interval is admittedly somewhat subjective, but we take $T_{dep\text{-}min}$ as the last temperature that is clearly part of the low-T trend and $T_{dep\text{-}max}$ as the first temperature clearly part of the high-T trend. The range of uncertainty defined in this way corresponds to the curved interval in the demagnetization plots in which the two overlapping components unblock simultaneously at differing rates. Where only one component is present and is not parallel to the 1980 field, we take $T_{dep\text{-}max}$ as the

Figure 2. Thermomagnetic data. Data are grouped by site and by material (ash/matrix or pumice). Data measured on warming are shown above data measured on cooling. $d\chi/dT$ is the first derivative of $\chi(T)$. Color represents depth within each deposit. Dark blue is at the surface, and dark red is the deepest sample from that site. Sites MSH14-02, MSH14-06, and MSH14-08 show a consistent increase in T_c with depth. Dashed vertical line shows the approximate location of the dominant T_c measured on cooling for each site. Ash data from site 8 (MSH12-08) are from Jackson and Bowles (2014). Data from MSH14-04 are extremely variable and are given in supporting information Figure S2.

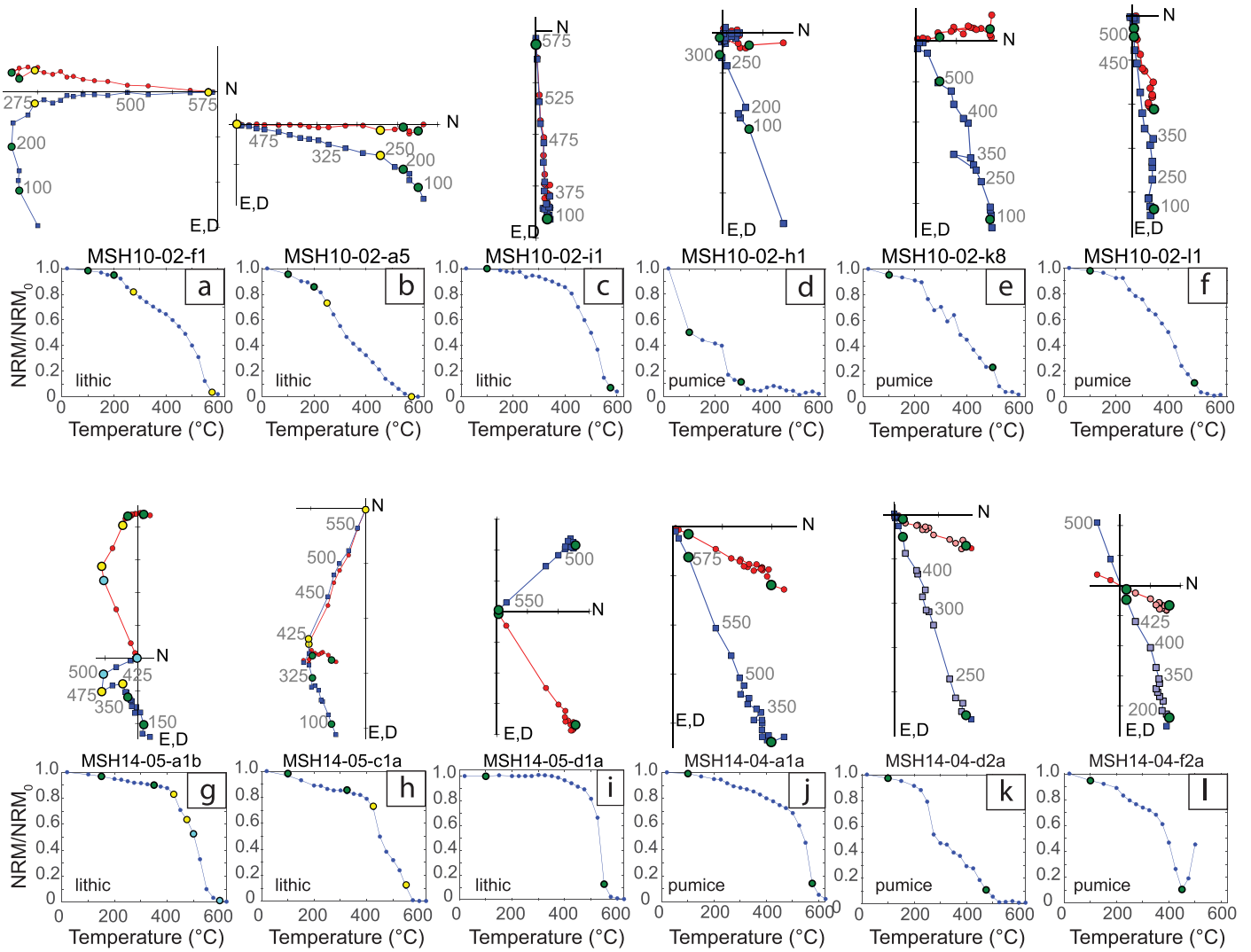

Figure 4. Examples of NRM demagnetization behavior of oriented samples. Vertical component in vector endpoint diagrams shown in blue squares, horizontal component in red circles. Starting and ending temperatures of directional components indicated by colored dots.

lowest unblocking temperature; if T_{dep} was greater than this, some of the unblocking spectrum would be overprinted.

For the small, unoriented lithics we only interpret specimens that clearly had two or more directional components. In the case of a single component, the ability to identify the 1980 field direction is required to know whether a clast was emplaced at low temperatures (random direction) or high temperatures (1980 direction). Eleven of the 29 clasts had two or more components (supporting information Table S2).

All pumice samples except one had a single directional component, and the oriented samples all had a direction close to the 1980 field (Figures 4 and 5). $T_{ub\text{-}max}$ in this case is an estimate of $T_{dep\text{-}min}$ (Hoblitt & Kellogg, 1979). We take the temperature at which less than ~5% of the NRM remains as $T_{ub\text{-}max}$ (supporting information Table S2). Five of 12 pumice samples from MSH14-04 show evidence for self-reversed behavior (e.g., Figure 4l). In this case, we take $T_{dep\text{-}min}$ as the last temperature before the net direction reverses.

Pumice samples from MSH14-01 and MSH14-08 also show depth-dependent unblocking behavior consistent with the T_c variations. At MSH14-08, the maximum unblocking temperature increases with depth (Figure 6b), just as T_c increases with depth (Figure 3i). We interpret this as remanence held predominantly by homogeneous titanomagnetite that has undergone reordering during cooling, elevating the blocking spectrum. At MSH14-01, where T_c is constant with depth (Figure 3a), there is no systematic depth-dependent

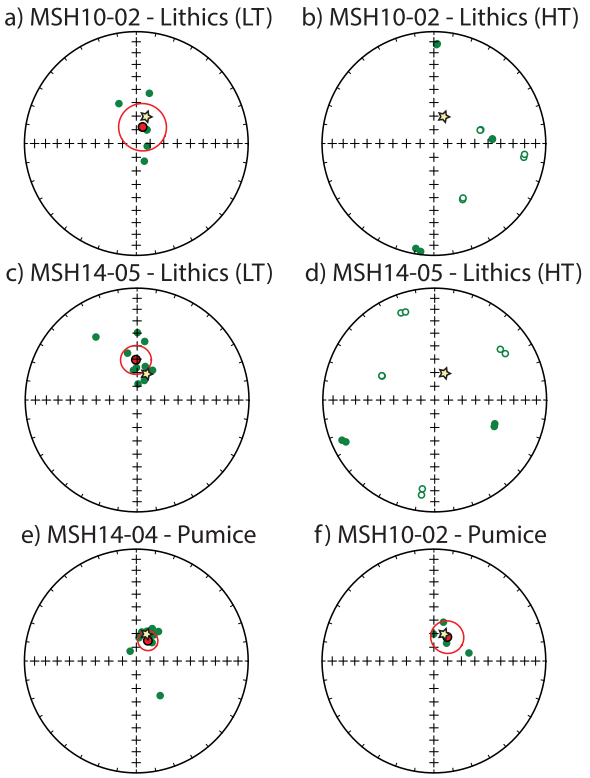

Figure 5. Principal component directions by site and clast type. Solid (open) symbols are lower (upper) hemisphere. LT, low-temperature component. HT, high-temperature component. All pumice samples only have a single component. Red dot and circle are Fisher mean and α_{95} uncertainty ellipse. Yellow star is 1980 field direction at the site.

trend in unblocking (Figure 6a). All samples undergo significant unblocking between about 150°C and 250°C, and we interpret this as the remanence held by homogeneous titanomagnetite. The remaining 30–50% is not fully removed until about 400–500°C, and this fraction is likely held by oxy-exsolved magnetite or other compositions of titanomagnetite. The thermomagnetic data for these samples (Figure 2b) show evidence for one or more Curie temperatures above the dominant T_c at ~380°C, as well as a small fraction of the susceptibility with a T_c near 550°C.

6. Discussion

6.1. Thermal Overprint Estimates of T_{dep}

Different authors have taken different approaches to final interpretation of the clast data, taking into account within-deposit variability in clast sizes, sources, and initial temperatures. Bardot (2000) and Paterson et al. (2010) suggest that the minimum estimate from any one location represents the equilibration temperature of the deposit (i.e., hot clasts cool down and cold clasts heat up until the deposit as a whole begins to cool). Cioni et al. (2004) and others (Di Vito et al., 2009; Zanella et al., 2007) take a different approach, using the temperature overlap between different clasts as a range that excludes both superheated clasts and colder, thermally unequilibrated clasts. Rather than assigning specific deposit temperatures, we prefer here to look at how estimates derived from different methods and materials vary and discuss possible errors and biases not likely to be related to peak clast temperature.

Figure 7 shows T_{dep} estimates derived from different methods and materials. Starting with the thermal demagnetization methods, the oriented lithic clasts (blue triangles) likely to be from the 18 May PDCs (MSH14-05, MSH14-06) all provide T_{dep} estimates that overlap with the measured 18 May deposit temperatures. When considering the full range of direct temperature measurements (297–418°C) there is little evidence of bias toward the high or low temperature end of the spectrum. However, excluding a single measurement made very close to the vent (418°C) the maximum measured temperature is 367°C (Figure 1), and most $T_{dep\text{-}max}$ estimates are significantly higher than 367°C. This could mean simply that hot clasts were derived close to the vent and retained their heat even as colder air and materials mixed in as the PDC moved downslope. Alternatively, the assumption that $T_b = T_{ub}$ may be violated, and this is discussed below.

The unoriented lithic clasts (purple stars) mostly overlap with measured temperatures. At site MSH14-06, however, the estimated T_{dep} range is significantly higher and is also higher than the range obtained from the oriented lithics. One of these clasts has very little unblocking at $T < 450$°C, so it is possible that the clast underwent a higher-temperature thermal event prior to being incorporated into the PDC, which in turn was not hot enough to produce a significant overprint in a clast with high unblocking temperatures. However, the second clast has continuous unblocking between room temperature and ~550°C, and there is no clear lower-T overprint other than a (presumably) viscous component removed by 125°C. This highlights the disadvantage of using unoriented clasts, where the timing of the acquisition of low-T component is unclear.

The estimates from the oriented lithics sharply contrast with the estimates from the pumice (red triangles). The minimum T_{dep} derived from pumice clasts is consistently higher than both the lithic estimates and the measured temperatures, sometimes by hundreds of degrees. Pumice from sites MSH14-04 and MSH10-02 are both oriented and the single component is consistent with magnetization in the 1980 field. MSH14-08 and MSH14-01 pumice samples are unoriented, but previous work on MSH14-08 has demonstrated that the single-component magnetization is parallel to the 1980 field (Bowles et al., 2015), and there is no reason to suspect that the same would not be true for MSH14-01. Clearly, the interpretation of $T_{ub\text{-}max}$ as an estimate of $T_{dep\text{-}min}$ is not appropriate here.

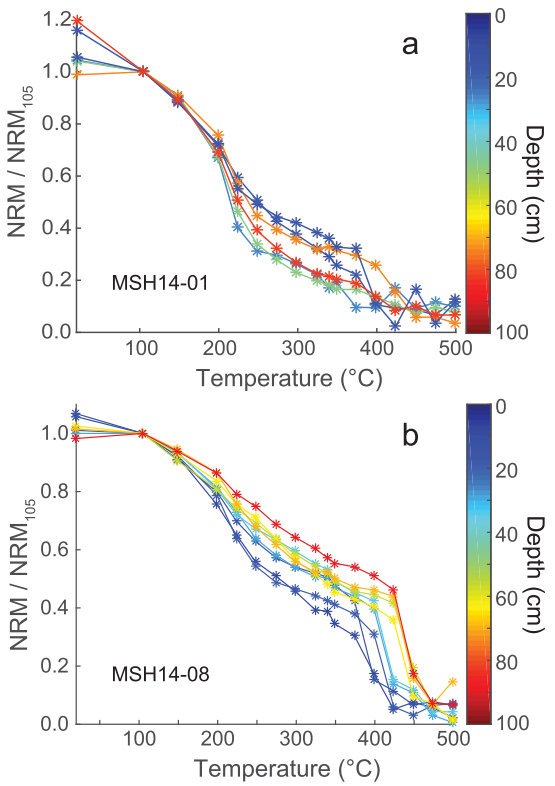

Figure 6. Thermal demagnetization of NRM of pumice clasts from sites MSH14-01 and MSH14-08. Data normalized to the 105°C step after removal of a small overprint. Color corresponds to depth in the flow with blues at the surface and reds at depth. Samples from MSH14-08 show a systematic increase in blocking temperature with depth, which is interpreted to arise from "hot" emplacement and variation in cooling rate with depth. Samples from MSH14-01 do not show a similar increase, and this deposit is assumed to have been emplaced below the temperature at which reordering is significant.

As noted above (section 1), similar high-unblocking data from juvenile material has been interpreted as either actual high-temperature deposition or as remanence of nonthermal origin. Rader et al. (2015) interpret the high unblocking temperatures in juvenile scoria in terms of hot scoria mixed in with cooler matrix material. There is some evidence for pumice blocks at MSH that also were not in equilibrium with the matrix at the time of deposition. Banks and Hoblitt (1996) find that for the 12 June PDCs, temperatures of pumice blocks encountered during profiling are up to ~50–75°C above *or below* the temperature of the matrix. This does not explain the $T_{dep\text{-}min}$ estimates that are consistently much higher than estimates from other materials or methods. Both sites MSH10-02 and MSH14-04 were discolored (pink and orange) as if they had been oxidized or hydrothermally altered, in which case they could carry a CRM. However, the remaining two sites (MSH14-01 and MSH14-08) showed no evidence of this. While that does not exclude a CRM, there are at least two other possibilities that might contribute to the anomalously high unblocking temperatures in the pumice.

The presence of MD grains and/or thermally activated reordering in the titanomagnetite would violate our assumption of an unchanging blocking spectrum where $T_b = T_{ub}$. These violations are more likely to affect T_{dep} estimates from pumice, but should also be considered in relation to the lithics. In MD grains, $T_{ub\text{-}max} > T_b$, and a remanence acquired at $T < T_{dep}$ will not be fully removed by laboratory heating to T_{dep} and may continue unblocking up to T_c (Bol'shakov & Shcherbakova, 1979; Xu & Dunlop, 1994). In the case where at least two magnetic components are present (as in the lithics), a T_{dep} estimate can still be made, but with greater uncertainty. The blocking spectrum of the low-T component will overlap into the high-T component (Dunlop et al., 1997), and the directional break between them will not be sharp (as seen in most samples here). This leads to a wider temperature window between the two components, where T_{dep} is (in principle) the lower bound. This means that the $T_{dep\text{-}min}$ estimate from each lithic clast is likely to be closer to the true clast temperature and may explain the temperature range that seems biased too high at sites MSH14-05 and MSH14-06.

For MD samples with a single directional component (as in the pumice), it is impossible to obtain an accurate T_{dep} estimate because there is no directional change at T_{dep} that arises from a pre-existing high-T component. This alone likely explains most of the overestimates found in the pumice in this study, and possibly in some previous studies. Although the pumice here have large titanomagnetite grain sizes near the MD end-member, that may not be true for all pumice everywhere, and it is likely not true for the scoria that Rader et al. (2015) describe as SD. Nevertheless, it is clearly preferable to rely on nonjuvenile material whenever possible precisely because the pre-PDC magnetization aids significantly in the interpretation. If juvenile material must be used (because temperature heterogeneities are suspected or due to lack of other materials), it is advisable to determine domain state. McClelland et al. (2004) also recommend against using pumice, suggesting that it is more likely to carry a CRM, the presence of which may sometimes be inferred through a careful examination of the magnetic mineralogy.

An MD contribution may be the biggest source of error or uncertainty, but another violation of our assumptions comes from modification of the blocking spectrum via reordering in the titanomagnetite during heating and cooling. The increase in blocking temperature with depth in MSH14-08 pumice strongly suggests that the blocking spectrum is affected by thermal history. If the deposit temperature is hot enough to activate the reordering process, T_c and T_b will slowly evolve toward their equilibrium state. The difference in maximum unblocking between the top of the deposit and 100 cm depth is ~50°C, which explains a small portion of the overestimate.

The effects of reordering on the lithics are more difficult to assess. It is possible that the same increase in T_b we observe in the pumice during cooling also takes place in the lithics. χ(T) data from the lithic clasts (supporting information Figure S2) show that some (but not all) clasts have clear evidence of a large ΔT_c suggesting a highly ordered state. However, the magnitude of ΔT_c suggests that the clast would have to be annealed for a much longer time than it likely took this flow to cool. There is also no correlation between the magnitude of ΔT_c (if it exists) and the T_{dep} estimate from the specimen. In this study, where T_{dep} is $<\sim$400°C, it is likely that any elevation in T_c and T_{ub} happened primarily at some point earlier in the clast's history and should not affect the T_{dep} estimate. If, however, $T_{dep} > T_{close}$, the titanomagnetite may revert to its more disordered state on heating above T_{dep}, and the final order degree, T_c, and T_b will reflect the cooling history in the PDC.

Our results can be compared to those of Erwin (2001), who also sampled the 18 May MSH deposits (along with other 1980 deposits). Pumice and lithic results are not distinguished, but in general they find that most that individual clast temperature ranges are close to but always higher than measured temperatures. They interpret interclast variability to arise from thermal heterogeneity between casts, and after accounting for this they still find a bias toward higher temperatures of $\sim$34°C. This is very similar to our own observation, but they interpret the bias to arise from a cooling-rate dependence on blocking temperatures. This type of blocking temperature bias is distinct from bias arising from multidomain grains or reordering (as we suggest) and assumes a single-domain mineralogy, which may or may not be correct for these deposits. Clearly, there are a variety of processes that can result in increased blocking temperatures, and all of them will lead to some degree of overestimation in T_{dep} when applying the standard method of NRM thermal demagnetization.

6.2. Independent T_{dep} Estimates From T_c Variations

Because lithic clasts were sparse, we place meaningful temperature constraints on the deposits by using T_c variations in the matrix material (pumice lapilli and bulk ash matrix). As demonstrated in Jackson and Bowles (2014) and discussed above, Curie temperatures at sites MSH12-08 and MSH14-08 systematically increase with depth in the deposit (Figure 3i) and this is shown to be linked to a thermally activated reordering phenomenon. Because the rate of reordering is time-dependent and temperature-dependent, it is possible to place temperature constraints on the deposits by comparing observed variations in T_c with predicted variations based on laboratory isothermal annealing experiments (Jackson & Bowles, 2014; supporting information Figure 3). Qualitatively, sites MSH14-01, MSH14-03, and MSH10-02 have small-to-zero ΔT_c at all depths, suggesting that they were emplaced at relatively low temperatures compared to sites MSH14-02, MSH14-06, and MSH14-08, which have T_c profiles that increase significantly with depth.

Quantitatively, we can model stratigraphic T_c variations by combining a conductive cooling model with the isothermal laboratory experiments which tell us how T_c changes with time and temperature. A similar approach to modeling T_c variations with time and temperature was outlined in Bowles and Jackson (2016). Here we combine that approach with a simple half-space conductive cooling model (Turcotte & Schubert, 2014). We assume the PDC is emplaced at a uniform temperature, maintains a fixed upper boundary condition of T = 10°C, and has a thermal diffusivity of 2.5×10^{-7} m^2 s^{-1}, the average found by Ryan et al. (1990) for the 18 May directed blast deposits. The model assumes Curie temperatures are initially uniform and represent the relatively disordered (quenched) state; the initial T_{c0} is taken to be the lower of T_c measured on cooling during the χ(T) experiments or T_c at the top of the deposit. For all of the 18 May deposits, this temperature is relatively uniform, ranging between 366°C and 382°C. For site MSH10-02, T_{c0} is considerably lower (343°C) reflecting a different titanomagnetite composition in this later deposit. Because the time-temperature dependence of T_c is composition dependent, we use the same isothermal anneal data (supporting information Figure 3) for all of the 18 May deposits; these data are generated using samples from MSH12-08. We use a different set of anneal data based on samples from MSH10-02 for that site (Bowles et al., 2013; Jackson & Bowles, 2014).

Resulting T_c profiles are modeled for T_{dep} in 5°C increments, and the best fit deposit temperature is found by minimizing the residual sum of squares. We use the site 8 pumice and ash data from the 2012 sampling season (MSH12-08) because it is more densely spaced and extends to greater depths (Jackson & Bowles, 2014). Only ash data are used for sites MSH14-02 and MSH14-03 because few pumice were available. Only pumice data were available for MSH10-02.

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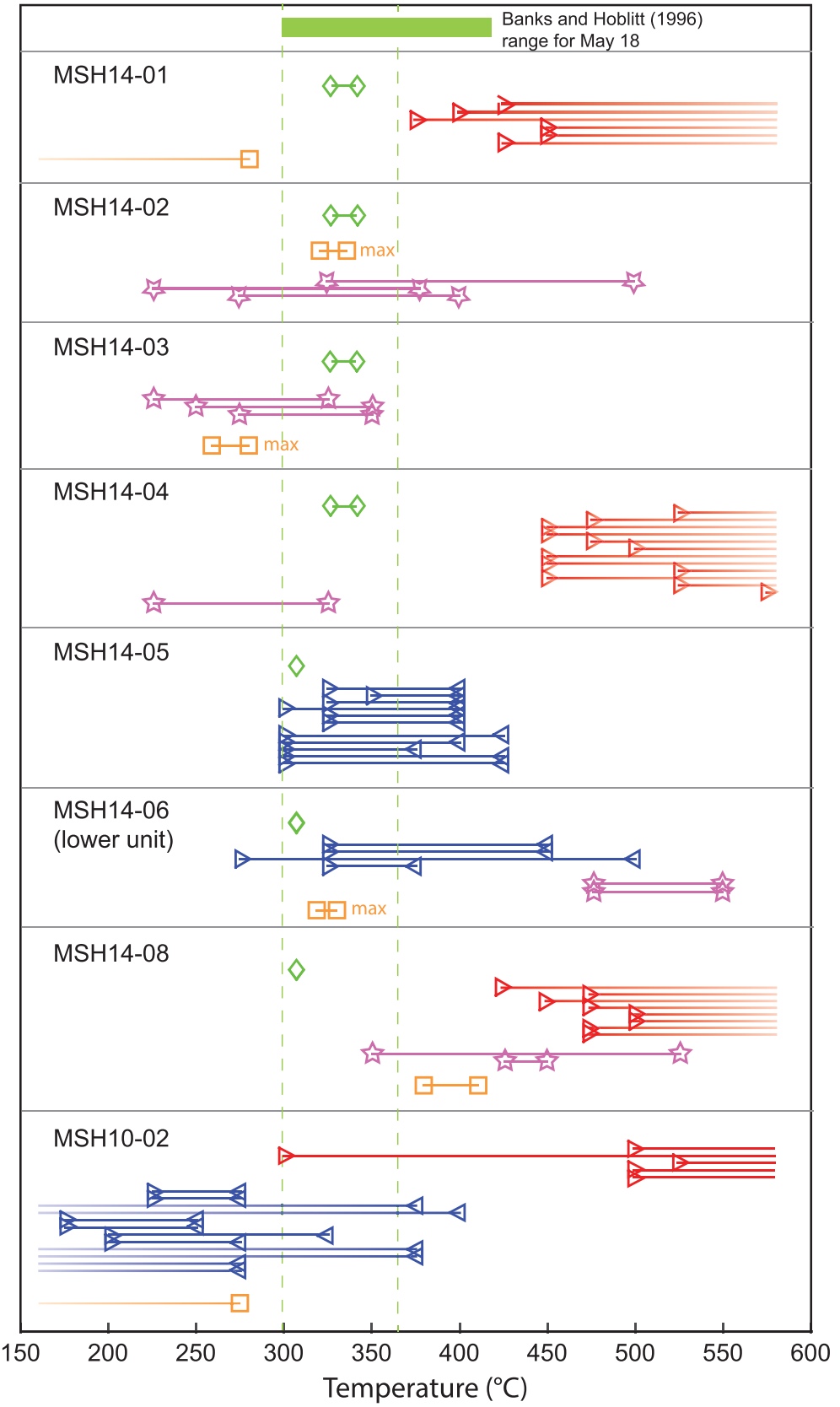

Figure 7. (continued)

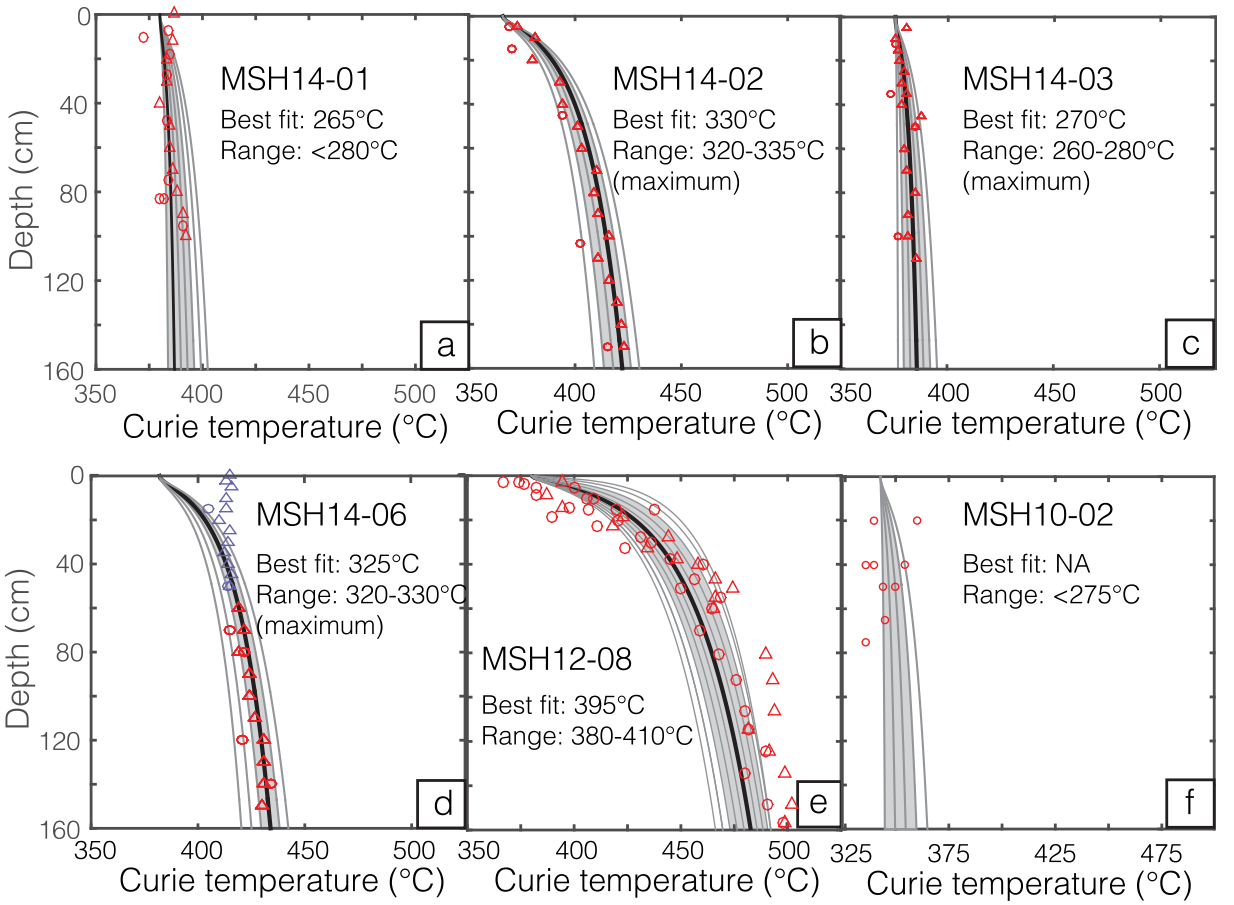

Figure 8. T_c profile modeling results. Red symbols represent measured T_c data, circles for pumice and triangles for ash. Purple symbols in Figure 8d MSH14-06 were not used in fitting. Solid lines represent modeled T_c profiles for different T_{dep} values in 5°C increments. Black line is best fit. Shaded range represents plausible range of acceptable T_{dep} (see text).

Modeled T_{dep} results are shown in Figure 8. The best fit model is shown by the bold line, and a range of plausible models is given by the shaded region. This range is admittedly subjective and was chosen to encompass most of the data. Site MSH12-08 is the most straightforward to interpret because the deposit was not overlain by additional deposits and was most likely to cool under our assumed boundary conditions. However, the fit to these data is the least satisfactory. The best fit T_{dep} is 395°C, but the model overestimates T_c at the top of the deposit and underestimates it at depths greater than ~80 cm. It is possible that the conductive cooling assumptions were violated here; a contribution from convective cooling—perhaps following a rain storm(s)—could produce a similar profile. Jackson and Bowles (2014) also found that the cooling times predicted by a simple conductive cooling model for this site did not reproduce the expected relationship between T_c and time that was observed in isothermal annealing experiments.

Sites MSH14-01, MSH14-02, MSH14-03, and MSH14-06 were all overlain by other 18 May deposits, so we treat these T_{dep} estimates as maxima because the sampled units may have been thermally insulated by the overlying units. However, ΔT_c remains near zero at the deposit top for all sites except MSH14-06 (Figure 3h).

Figure 7. Summary plot showing T_{dep} estimates derived from various methods and materials. Green bar at top shows range of direct temperature measurements (Banks & Hoblitt, 1996). Vertical dashed lines show temperature range measured in the vicinity of sampled deposits. Green diamonds show range of temperatures measured nearest individual sites (see Figure 1). Blue (red) lines and triangles are from thermal demagnetization of lithic (pumice) clasts. Purple stars are from thermal demagnetization of small, unoriented lithic clasts. Orange squares are modeled ranges derived from stratigraphic variation in T_c. In all cases, a fading line with no terminating symbol means that the estimate is either a minimum or a maximum. For sites MSH10-02, MSH10-03, and MSH10-06, the given range is a maximum (see text).

ΔT_c near zero implies that the materials were rapidly cooled, preserving their far-from-equilibrium disordered state and low T_c. The kinetics of the reordering process are prohibitively slow at temperatures less than about 275°C. This means that the materials must be reheated to >275°C to reorder on the relevant timescales. This reordering would be accompanied by an increase in T_c which we do not see. Therefore, the overlying deposit was not hot enough to elevate the surface of the underlying deposit to $T > 275$°C.

By contrast, the two units sampled at MSH14-06 appear to have formed a compound cooling unit. ΔT_c is ~30°C at the surface of the lower unit and increases to ~50°C at 150 cm. Above the unit boundary, ΔT_c is much larger, up to ~100°C, consistent with a relatively high T_{dep}. Because we do not know the total thickness of the upper unit, it is not possible to model the two units together. However, excluding data at depths <50 cm in the lower unit, the remaining data are well fit to the simple cooling model defined above, and the resulting T_{dep} represents a maximum at ~325°C. Further, assuming the two units were emplaced in relatively rapid succession (within a few hours), the overlying unit must have been hotter. If it was the same or lower temperature, ΔT_c of the lower unit would be about the same as ΔT_c of the upper unit.

The top of MSH10-02 has been removed by erosion, so true depths are not known. However, because ΔT_c is essentially zero at all depths, a relatively cool T_{dep} is inferred. As noted above, T_c data from MSH14-07 and MSH14-04 are extremely variable with depth and were therefore not modeled.

Comparing these independent T_{dep} results with our other estimates (Figure 7), we see decent agreement between the standard lithic estimates and the ΔT_c-derived estimates. As predicted above, the T_c-based estimates coincide most closely with the low end of the range derived from the lithics.

6.3. Geological Interpretation

In the discussion below, we draw primarily on the ΔT_c estimates of T_{dep} because they are available for the most sites. Linking all the 18 May deposits in a single stratigraphy is difficult, because the character of each pyroclastic episode changes with distance from the vent and is influenced by topography. Unit boundaries may be diffuse, and in some places units are duplicated (Brand et al., 2014). What is clear is that temperature varies between the different episodes, from <275°C to >380°C. Banks and Hoblitt (1996) found evidence for temperature variability between multiple flow units in the 12 June, 22 July, 7 August, and 17 October PDCs, but not in the 18 May PDC. They attributed this temperature variability to variations in eruptive vigor. Our data demonstrate that the same phenomenon is found in the 18 May deposits and was perhaps not noted because the temperature probes did not penetrate deep enough into the more voluminous 18 May deposits.

There are two locations where direct stratigraphic variations in T_{dep} are observed: site MSH14-02 overlies MSH14-03; and at MSH14-06 we sampled two separate units. In both cases, the overlying unit was hotter, which may suggest increasing temperature over the course of the day. If the lowest temperatures are confined to the earlier flows, these temperatures would not have been sampled by Banks and Hoblitt (1996) and explain their slightly higher temperature range.

We also have some evidence for temperature variability within a single PDC episode/unit. MSH14-01 and MSH14-02 are located ~300 m apart and are both overlain by a 35–50 cm thick ash layer, followed by a pink, block-rich unit. We interpret these to be the same unit, yet their temperature varies by at least ~40°C. This is not particularly surprising, as a wide variety of factors may contribute to observed significant lateral and vertical facies changes within the 18 May PDC deposits (Brand et al., 2016, 2014). The dynamics of the flow may be affected by slope, surface topography, current velocity, particle concentration and size, and internal pore pressure. Erosion of the substrate leads to the incorporation of cold material, and flow over irregular topography (Zanella et al., 2007) or convergence of two flows (Brand et al., 2014) can lead to the incorporation of cool air, both of which will lead to thermal heterogeneity in the flow.

Site MSH10-02 has T_{dep} estimates in the range of ~200–275°C based on both the oriented lithics and ΔT_c data. This is considerably lower than most of the other sites that are attributed to the 18 May PDCs. As noted above, this site is much closer to the vent and is likely associated with a later PDC. We note that the Curie temperatures (measured on cooling) are ~25°C lower than at any of the other sites. This suggests a different composition of titanomagnetite and is consistent with a different eruptive event. However, all of the later events were significantly hotter than the 18 May event, so these low emplacement temperatures are difficult to explain, unless this deposit represents an unusually cool pulse not detected by Banks and

Hoblitt (1996). Alternatively, this deposit is anomalously lithic rich, and the incorporation of a large volume of cold lithic material may have locally depressed T_{dep}, as noted above.

6.4. General Application of New Method

The use of pumice or juvenile matrix variations in T_c to model deposit temperatures should be generally applicable, with some caveats. It may be especially useful in lithic-poor deposits or to supplement data from thermal demagnetization of lithics. Because incorporated lithics can be thermally heterogeneous, use of matrix ash may provide a more direct assessment of the deposit equilibrium temperature. The technique works best when the matrix has a relatively simple magnetic mineralogy, and one of the components must be homogeneous intermediate-composition titanomagnetite. Fortunately, this is a very common composition in andesitic, dacitic, rhyolitic, and some basaltic eruptive materials (Ghiorso & Evans, 2008). Because the exact time-temperature evolution of T_c is composition dependent (reflecting variable amounts of cation substitution and vacancies; Jackson & Bowles, 2018), application to a new site would require calibration of a new titanomagnetite composition. This involves isothermal annealing experiments, similar to those reported on in Jackson and Bowles (2014). While not overly laborious, it does require a considerable amount of time.

The technique provides the most information in the approximate temperature interval $275°C > T > T_{close}$, where T_{close} is approximately 450–500°C. 275°C is the approximate temperature at which reordering begins to take place at the relevant timescales. If $T_{dep} < 275°C$, the modeling will provide an approximate upper bound. If $T_{dep} > T_{close}$, the model assumptions are violated. The model assumes that once deposited at $T < T_{close}$, materials are in a disequilibrium state and then slowly move toward equilibrium during cooling. If $T_{dep} > T_{close}$, the cooling deposit starts out in an equilibrium state and the modeling will fail. Data from a 2010 PDC from Soufrière Hills, Montserrat, shows a strong and systematic change in T_c with depth (Bowles et al., 2014). However, all independent information suggests these deposits were emplaced at $T > 500°C$, and the modeling totally fails to produce a reasonable fit to the data. This failure is important, because it means that in such situations the method will produce a recognizable null result rather than an erroneous estimate for T_{dep}.

Of the studies that have previously worked with pumice and found that it failed to produce reliable results based on remanence data, McClelland et al. (2004) show thermomagnetic data demonstrating that the Taupo pumice have relatively simple magnetic mineralogy including intermediate-composition titanomagnetite. Paterson et al. (2010) report T_cs mostly between 425°C and 500°C for pumice from the 1993 Lascar PDCs, and Donoghue et al. (1999) report a T_c for Ruapehu pumice clasts of approximately 300°C, all of which are consistent with intermediate-composition titanomagnetite. Juvenile material with the requisite mineralogy is therefore not uncommon.

7. Summary and Conclusions

Of the different methods and materials we have examined, it is clear that thermal demagnetization of the pumice clasts provides biased results, much higher than other methods. We interpret this as due primarily to the multidomain nature of the magnetic mineralogy and we recommend avoiding juvenile material unless it can be shown that the remanence is not carried by an MD component. Lower estimated temperature ranges are obtained from the lithic clasts, but the upper end of these ranges is typically significantly higher than the direct measurements as well as the estimates derived from Curie temperature variations (ΔT_c). If the T_{dep} range between the low-T and high-T components is due in whole or in part to an MD remanence, then the lower end of this range will closer to the true T_{dep} because $T_{ub} > T_b$. Interpreted in this light, we find good agreement between measured emplacement temperatures, thermal demagnetization of lithics, and estimates from ΔT_c variations. With the exception of MSH14-06, the unoriented lithics also broadly agree with these other estimates, but we do not recommend reliance only on unoriented lithics because of the inability to identify the nature of the low-T component.

Thermally activated reordering in titanomagnetite during slow cooling of the deposits may in principle lead to overestimation of T_{dep}. In the lithic clasts, this is only likely to be important if $T_{dep} > T_{close}$ (~450–500°C). In juvenile material, we find here a bias of up to ~50°C, but in principle it could be significantly greater.

The development of a new method of assessing PDC deposit temperature by evaluating stratigraphic variations in titanomagnetite Curie temperatures enables us to assess T_{dep} in lithic-poor flows. In this study, it allows for a more detailed assessment of temperature variations between and within deposits than would have been achieved only by using lithic clasts.

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