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Key Points:

- Illustrates in detail the weakness of pTRM checks to detect alteration in Thellier-series paleointensity experiments
- Uses comprehensive stepwise after-heating rock magnetic property changes to study thermophysicochemical alteration
- Proposes a RESET method and CAL check to monitor overall thermal alteration of specimens

Supporting Information:

- Supporting Information S1
- Data Set S1

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RESET: A Method to Monitor Thermoremanent Alteration in Thellier-Series Paleointensity Experiments

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Abstract Accurately estimating the paleomagnetic field intensity recorded in terrestrial and planetary materials is the key to understanding dynamo processes. Thellier-series stepwise-heating methods with partial thermoremanent magnetization (pTRM) checks for alteration are considered the most reliable technique even though pTRM checks are shown to be incapable of detecting the entirety of thermal alteration. We utilize a recently developed multidomain-correction experiment (or RESET) to monitor thermally induced magnetic property alteration that may happen to the very specimen used in Thellier-series experiments. We also use rock magnetic property changes to track the physicochemical alteration of fresh companion specimens of Galapagos lavas that were previously used for paleointensity determinations. Our results show that laboratory heating induced thermal alteration of a typical Galapagos lava sample starts to occur at around 500°C. It escaped detection by pTRM checks but was caught by our RESET method.

Plain Language Summary To date, there has been no way to detect the entirety of thermal alterations that may happen during laboratory heating to specimens used in Thellier-series paleointensity experiments. This study develops an experimental approach to address this issue.

1. Introduction

The strength of ancient magnetic fields (paleointensity) recorded in rocks provides key information on the evolution of Earth and planetary dynamo processes (Weiss et al., 2008), interactions between the magnetosphere and solar wind (Tarduno et al., 2010), and even the formation of the solar system (Wang et al., 2017). Accurate paleointensity estimates may also enable us to infer when the inner core began to form (Biggin et al., 2015; Bono et al., 2019), to study the interaction between the magnetic field and other Earth systems (Knudsen & Riisager, 2009), and to establish the likelihood of an approaching geomagnetic polarity reversal (Wang et al., 2015).

Various experimental techniques have been developed to retrieve paleointensities recorded by various rocks (Tauxe & Yamazaki, 2007). The classic Thellier-Thellier double-heating experiment (Thellier & Thellier, 1959) and its variants like the Coe (Coe, 1967b), Aitken (Aitken et al., 1988), IZZI (Tauxe & Staudigel, 2004), and BZF (Wang & Kent, 2013) protocols (Thellier-series experiments henceforth) are considered the most reliable methods and are widely used to estimate absolute paleointensities from igneous rocks. All these protocols involve comparisons of the natural remanent magnetization (NRM), expected to include a thermoremanent magnetization (TRM) component that records the paleointensity, and a TRM reproduced on the same specimen in a known laboratory-applied field. This is usually done in a sequence of thermal treatment steps from room temperature to the Curie temperature whereby the NRM is demagnetized by cooling samples in a zero-field environment at the same temperature steps as the acquisition of a partial TRM (pTRM) by cooling samples in the presence of a known weak laboratory-applied field. Each paired demagnetization-acquisition step provides an independent estimate of paleointensity by the law of additivity of pTRM (Thellier, 1938).

A major limitation on obtaining reliable paleointensity is the alteration due to physical (magnetic domain state) and chemical (formation and/or destruction of ferromagnetic minerals) changes induced during laboratory heating. The main procedure to detect alteration is the pTRM check (Thellier & Thellier, 1959), which is given after a particular heating step at a previous lower temperature. If the pTRM acquired in a pTRM-check step is different than the previously acquired pTRM at the same lower temperature, alteration

must have occurred (Thellier & Thellier, 1959), and only those steps up to the previous lower temperature are deemed capable of retaining the original TRM spectrum to provide reliable estimates of paleointensity. However, the lack of a discrepancy between a pTRM check and the original pTRM acquisition does not guarantee the absence of alteration. This is because a pTRM check is not capable of detecting thermal alteration of grains with higher blocking temperatures than the pTRM-check temperature.

Although this ambiguity of the pTRM check was realized soon (Coe, 1967a) after the development of the Thellier-Thellier technique with pTRM checks (Thellier & Thellier, 1959), pTRM checks are still heavily relied upon as a sufficient (and usually the only) indicator of thermal alteration for Thellier-series paleointensity experiments. For example, 23 out of 25 paleointensity studies from 2003 to 2014 (Table S1) that had been analyzed for a paleointensity data quality factor, QPI (Biggin & Paterson, 2014), employed a Thellier-series protocol, and 22 of those conducted pTRM checks. Nonetheless, the known weakness of the pTRM check has not received wide attention, and for example, only one (Qin et al., 2011) of these 25 studies discussed this issue.

Even less well acknowledged is that a failed pTRM check may actually be due to multidomain (MD) effects rather than alteration, as demonstrated by Wang and Kent (2013). For MD grain-bearing igneous samples, pTRM checks will tend to fail simply because of MD tails (i.e., lack of reciprocity) and thus will be even less indicative of thermal alteration. This means we cannot be sure that a sample has thermally altered even if there are discrepancies between pTRM checks and previous pTRM acquisitions. The reason is that both the pTRM check and the original pTRM acquisition are calculated by subtracting the remanence measurements of zero-field steps from those of in-field steps (or subtraction of two opposite in-field steps for the Thellier-Thellier protocol). The effects of MD tails (Riisager & Riisager, 2001) are automatically included in the calculation and can contribute to the discrepancy between pTRM checks and previous pTRM acquisitions, even without thermal alteration. The resultant tendency to accept data up to only lower temperatures will bias the estimated paleointensity to anomalously higher values due to the typical concave-up Arai diagram for MD samples (Levi, 1977; Smirnov et al., 2017).

There is thus a clear need to determine the onset of magnetic alteration independent of the ambiguous pTRM check. In practice, Thellier-series experiments are often accompanied by thermomagnetic (e.g., susceptibility versus temperature, χ - T , and/or saturation magnetization versus temperature, J_s - T) heating and cooling curves (as performed in 23 of the 25 studies in Table S1), as well as other rock magnetic measurements (e.g., hysteresis loops and/or first-order reversal curves, FORCs) measured on selected sister specimens before and after heating. These may provide key information on the possibility, extent, and nature of thermal alteration. However, perhaps due to the intensive effort needed, systematic comparison of fresh-and-heated measurements is not usually carried out; for example, only 4 of the 25 studies in Table S1 conducted such measurements (Calvo-Rathert et al., 2013; Mochizuki et al., 2013; Mochizuki et al., 2011; Qin et al., 2011). Moreover, such measurements, when done, are typically on only a few selected sister specimens and are thus unlikely to be broadly representative of the alteration experienced by the tens or even hundreds of specimens that are often used in Thellier-series paleointensity studies. Even with universal rock magnetic characterization of sister specimens, there is still no guarantee that even closely paired sister specimens will behave exactly the same when heated (Coe et al., 2004; de Groot et al., 2014). A method that is able to pinpoint thermally induced magnetic alterations (particularly thermoremanent capacity changes) in the very specimens that are used in Thellier-series paleointensity experiments is thus crucial for us to obtain accurate paleointensities.

In the next section, we illustrate the ambiguity of the pTRM check in detecting thermal alteration in Thellier-series experiments by using a plausible theoretical TRM spectrum for stable single-domain (SSD) grains. In Section 3, we present illustrative before-and-after heating rock magnetic data from two typical Galapagos lavas that were used in a paleointensity study (Wang & Kent, 2013) to show that magnetic alteration occurred but was not detected by pTRM checks. Lastly, in Section 4, we propose how the “Repeated Thellier-Series Experiment” (RESET; formerly MD-correction technique) (Wang & Kent, 2013) can be used to verify whether or not thermal alterations have occurred in the very specimens undergoing Thellier-series experiments. A previously known benefit is that the RESET also detects and corrects for MD effects, which can be distinguished from alteration and allow unbiased paleointensity estimates to be retrieved from a thermally stable sample bearing MD magnetic carriers.

2. Ambiguities of the pTRM Check

Thellier-series experiments are designed to estimate paleointensities of SSD-bearing igneous rocks (or other materials that cooled from high temperatures in the geomagnetic field, like baked contacts and pottery) and are theoretically supported (strictly only for uniaxial SSD particles, like in the Tiva Canyon Tuff (Jackson et al., 2006)) by the thermal relaxation theory of TRM (Néel, 1949) whereby a population of strictly SSD grains is expected to satisfy Thellier's three laws of pTRM (Thellier, 1938): 1. reciprocity, 2. independence, and 3. additivity. If the pTRM blocking temperature spectrum of a sample remains essentially the same during heating cycles, these ideal properties allow accurate estimation of absolute paleointensity by comparing the NRM to a laboratory-applied TRM (via reciprocity). In a stepwise Thellier-series experiment, the NRM remaining after thermal demagnetization is plotted against the pTRM acquired for each temperature step (Arai diagram; Nagata et al., 1963), which allows multiple estimates of paleointensity to be obtained (via independence and additivity). For a sample with a TRM dominated by SSD grains that remain unaltered, the Arai diagram should be a straight line with a slope representing the ratio of the paleointensity to the intensity of the known laboratory-applied field (Figures 1a and 1c).

The original Thellier-Thellier protocol consists of two heating steps at each temperature with the laboratory field applied in opposite directions, from which the remaining NRM and the acquired pTRM are calculated and used to make successive estimates of paleointensity (Thellier & Thellier, 1959). The Coe variant (zero-field heating followed by in-field heating at each temperature step) (Coe, 1967b), the Aitken variant (in-field and then zero-field) (Aitken et al., 1988), the IZZI variant (alternating between the Aitken and Coe methods for every other temperature step) (Tauxe & Staudigel, 2004), and the BZF variant (back-field, and zero-field, and then forward-field heating treatments for each temperature step) (Wang & Kent, 2013) all include an NRM demagnetization step for each heating temperature, which allows the remaining NRM to be directly measured. These protocols are found to be mostly interchangeable with the original Thellier-Thellier method in their net results for estimating paleointensities (Wang & Kent, 2013). A pTRM check can be added to any of the Thellier-series protocols, usually after a zero-field heating (NRM demagnetization) step.

Here, we consider a theoretical demonstration (Figure 1g) of detailed heating steps of the Coe protocol applied to an igneous sample with an ideal SSD magnetite population that satisfies Thellier's three laws of pTRM. After the initial room temperature (denoted as 0°C for convenience) NRM measurement, we use 100°C increments to 600°C for the Thellier-series experiments and plot the pTRMs from each temperature interval to obtain the TRM spectrum of the sample. We assume the sample is thermophysicochemically stable below 300°C but then starts to alter above 300°C (Figure 1g). The alteration produces an increase in the TRM spectrum, which, in practical terms, can be caused by the formation of new ferromagnetic particles from alterations of silicates or preexisting iron oxides (Zhao et al., 2014). This kind of alteration can result in an increase of the sample's pTRM carrying capacity and will cause a shallowing of the Arai plot in the 300°C–600°C interval (Figures 1b and 1d).

The NRM (blue bars in Figure 1g; see Table S2 for values) is the original TRM acquired in the paleomagnetic field. From 0°C to 600°C, the incremental NRM demagnetization plotted against the pTRM acquisition is the Arai diagram (Figures 1a and 1b) to which pTRM checks are added after demagnetization steps from 200°C to 600°C (Figure 1g). No alteration occurs below 300°C, but then alterations are imposed to occur during the first heating (NRM demagnetization) steps at 400°C, 500°C, and 600°C (steps 10, 13, and 16 in Figure 1g, respectively). Thus, the back-to 100°C and 200°C pTRM checks (steps 5 and 8 in Figure 1g, respectively) show no change in the TRM spectrum, in agreement with the absence of imposed alteration, but this does not guarantee the ability of pTRM checks to detect thermal alterations when they actually happen. For example, after the 400°C zero-field heating, a pTRM check is performed by in-field heating to 300°C (step 11 in Figure 1g) that can be compared with the previously applied pTRM from 300°C (step 9 in Figure 1g). In our theoretical demonstration, alteration is imposed to occur in the zero-field heating to 400°C (dashed box in step 10 in Figure 1g), which causes an increase of the pTRM carrying capacity in the 300°C–400°C interval. However, the 400°C back to 300°C pTRM check shows no change in the Arai plot (Figure 1b) and thus would erroneously indicate the absence of thermal alteration from 300°C to 400°C. This is because the pTRM checks only apply pTRMs to grains with blocking temperatures up to the pTRM-check temperature, which is the previous heating temperature (i.e., to 300°C in this case), but not to grains with blocking temperatures higher than that temperature (i.e., above 300°C in this case). Thus, even if those

Figure 1. (a) Circles and black line are Arai plot of an ideal SSD sample without thermal alterations (see Table S2 for NRM and pTRM values). Green triangles are pTRM checks with dashed green lines tracing their temperatures. (b) Circles and dashed black line are Arai plot of a SSD sample with thermal alterations (increased TRM capacity) from 300°C to 600°C. Note the shallowing of the slope in the 300°C–600°C interval. (c) NRM remaining and calculated pTRM acquisition plotted against temperature of the thermally unaltered sample shown in (a). (d) Dashed red line shows the calculated pTRM acquisition of the thermally altered sample shown in (b). (e) Circles and pink line are MD-corrected Arai plot of the SSD sample with thermal alterations from 300°C to 600°C. Dashed line shows the Arai plot of the ideal SSD sample without thermal alterations for reference. Note the shallowing of the slope of the pink line in the 300°C–600°C interval. (f) Circles and black line are the repeated Arai plot from the RESET of the SSD sample with thermal alterations from 300°C to 600°C that already happened in the original round of Thellier-series experiment. Green triangles are pTRM checks with dashed green lines tracing their temperatures. Red line indicates the tTRM alteration check. Dashed black line shows the original Arai plot of the ideal SSD sample without thermal alterations for reference. (g) Experimental steps for the Coe protocol shown on the modeled TRM spectrum. Numbers in the circles indicate heating steps in the experimental treatment sequence. Blue bars indicate original NRM. White bars indicate thermally demagnetized NRM. Red bars indicate laboratory-applied pTRMs in the pTRM acquisition steps. Green bars indicate laboratory-applied pTRMs in the pTRM-check steps to previous lower temperatures. Dashed bars indicate the increase of TRM recording capabilities due to thermal alteration. Arrows indicate the pTRM-check paths from the current temperatures to the previous temperatures. (h) Experimental steps for the repeated Coe protocol for the RESET procedure shown on the modeled TRM spectrum. The laboratory-applied tTRM (cyan bars) is equivalent to a traditional NRM here. Numbers, colors, and arrows are the same as in Figure 1g.

grains with blocking temperatures above 300°C (between 300°C and 400°C in our demonstration) experience changes in TRM carrying capacity due to thermal alteration, the pTRM check will still indicate the absence of any differences (Figures 1b and 1g).

Continuing the theoretical demonstration, a pTRM acquisition step is next applied at 400°C (step 12 in Figure 1g). This is the first time in our virtual experiment that the altered TRM spectrum carries a pTRM, and it would be useful if pTRM checks could detect the altered TRM spectrum in later steps. However, even though further thermal alteration occurs during the zero-field heating to 500°C, the 500°C back to 400°C pTRM check only imparts a pTRM to the magnetic grains with blocking temperatures up to 400°C (step 14 in Figure 1g), which is the same as the pTRM previously applied at 400°C. Thus, the 500°C back to 400°C pTRM check is also incapable of detecting thermal alterations that occurred in step 10. In fact, even when thermal alterations are imposed in every temperature interval from 300°C to 600°C, pTRM checks continue to indicate the absence of any magnetic alteration. This weakness is caused by the inherent nature of the pTRM check that only applies pTRMs to the grains with blocking temperatures below the pTRM-check temperature. Thus, ferromagnetic grains with blocking temperatures higher than that temperature are basically invisible to pTRM checks. There is only a sign of the thermal alteration in the Arai diagram wherein the 300°C–600°C interval would have a shallower NRM-pTRM slope (Figure 1b), compared to that of the 0°C–300°C interval, due to the increase of pTRM carrying capacity caused by thermal alterations (Figure 1d) as a result of changes in the TRM spectrum (Figure 1g). However, other than being produced by alterations in SSD samples, concave-up Arai diagrams with shallower NRM-pTRM slope at higher temperature intervals can also be caused by behaviors of larger MD carriers (Levi, 1977; Smirnov et al., 2017; Xu & Dunlop, 2004). Thus curved Arai diagrams cannot be used to definitively identify thermal alterations that may happen to non-SD samples, which commonly exist in nature.

We also applied the above theoretical demonstration of thermal alteration of the TRM spectrum in the Aitken (Aitken et al., 1988) (Figure S1), Thellier-Thellier (Thellier & Thellier, 1959) (Figure S2), IZZI (Tauxe & Staudigel, 2004) (Figure S3), and the BZF (Wang & Kent, 2013) (Figure S4) protocols and found that pTRM checks are incapable of detecting the overall thermal alterations in any of these protocols due to the fact that grains with blocking temperatures higher than the pTRM-check temperatures are not carrying TRM. And pTRM checks to a previous lower temperature are always done after the first heating treatment (either in-field or zero-field) of each temperature step, so that any thermal alteration that occurred during the first heating treatment to each successive temperature to the grains with blocking temperatures higher than the pTRM-check temperatures will always be undetected, no matter which protocol is used. Accordingly, we conclude that for samples bearing SSD grains, the absence of a discrepancy between pTRM checks and previous pTRM acquisitions is not sufficient to warrant the absence of thermophysicochemical alteration during heating. For example, see the case of Galapagos lava specimen GA84.6c (Figure 2 and Figure 10 of Wang & Kent, 2013).

It would seem appropriate for the converse that a discrepancy between the pTRM check and the previous pTRM acquisition is sufficient to conclude that the sample has indeed experienced thermal alteration, and hence data from above the pTRM-check temperature should be excluded from qualified paleointensity results. But this would only apply for samples assuredly dominated by SSD magnetic carriers. For MD-bearing igneous samples, pTRM checks will tend to fail simply because of MD tails (i.e., lack of reciprocity) and will thus be even less indicative of general thermal alterations. Examples of such a case are illustrated by paleointensity data from Galapagos lavas (e.g., GA79.5c in Figure 2, details in Wang & Kent (2013)).

3. Rock Magnetic Results for Galapagos Lavas

Rock magnetic properties of sister specimens are sometimes used to gauge thermal alterations in parallel to paleointensity experiments (e.g., Bowles et al., 2011; Qin et al., 2011; Smirnov & Tarduno, 2003). To provide independent constraints on the efficacy of pTRM checks in Thellier-series experiments, we conducted comprehensive rock magnetic measurements on several Galapagos lavas, which were previously used for a (heated in air) RESET paleointensity study (Wang & Kent, 2013). Here, we focus on one sample (GA79.5) that yielded a qualified RESET paleointensity result despite failed pTRM checks attributed to MD effects, and another sample (GA84.6) that failed to provide a satisfactory RESET paleointensity estimate

Figure 2. (a)–(d) RESET paleointensity results for GA79.5c (a and b) and GA84.6c (c and d) from the original and repeated Thellier-BZF experiments, calculated using Thellier protocol according to the highest quality control factors (reproduced under rights granted to authors from a previous study by the same authors (Wang & Kent, 2013)). (a), (c) Arai diagrams of the original (thick black line) and repeated (thin black line) Thellier-BZF experiments, with circles indicating NRM to 375°C and solid squares indicating 400°C–575°C. Orange triangles show the pTRM checks, with orange dashed lines indicating their temperatures. Red lines represent pTRM gains in the original versus those in the repeated Thellier-BZF experiments (i.e., the tTRM check), with light blue dashed lines as the 1:1 reference. Green dashed lines are the theoretical linear prediction of Arai diagrams for SSD grains for repeated Thellier-BZF experiments. (b), (d) Arai diagrams of the original (thick black line) Thellier-BZF experiments with pTRM checks (orange triangles). Pink lines represent the original NRM unblocking remaining from the original BZF experiment versus the laboratory-applied tTRM unblocking from the repeated BZF experiment, with blue dashed lines representing the linear regression for the 400°C to 575°C temperature interval. Figures are modified from Figures 9e, 9f, 10g, and 10h of Wang & Kent (2013). (e) Day plot of GA79.5y (blue) and GA84.6y (red) measured at room temperature after each heating step (temperatures as labeled). Percentages next to crosses on dashed curve are modeled volumes of MD contribution from the SSD-MD mixing curve #3 (Dunlop, 2002). (f–s) high-resolution FORC diagrams (field increments of 0.6 mT; number of curves = 513; measurement time ~10 h; smoothing factor of 6) for GA79.5y (f–l) and GA84.6y (m–s) in the original fresh state and after 200°C, 400°C, 450°C, 500°C, 550°C, and 600°C heating treatment steps. FORCs were processed using FORCinel v3.06 (Harrison & Feinberg, 2008).

even though pTRM checks passed. The following rock magnetic measurements aim to provide insights into this issue.

Wang and Kent (2013) and this study used fresh ~20 mg chips cut very closely from the paleointensity specimens (see Figure S5 for the sample cutting sketch) for rock magnetic measurements. We understand that the rock magnetic sister specimens may not completely represent the properties of the paleointensity specimens because a lava flow is not perfectly homogeneous even at the specimen level (e.g., a Hawaiian lava showed in (de Groot et al., 2014)). However, our sampling technique helps to ensure that the rock magnetic chips resemble the properties of the paleointensity specimens as closely as possible.

Previous thermomagnetic (J_s - T) curves (heating in air) and hysteresis measurements of chip specimens GA79.5s and GA84.6s (in Figure 3 of Wang & Kent, 2013) indicate that GA79.5s experienced less thermal alteration than GA84.6s. The J_s - T curves of both of these specimens show reversible heating and cooling curves, indicating little change in their magnetic mineralogical states.

To further track possible thermal alterations step-by-step as the samples are being heated in the Thellier-series experiments, we measured hysteresis properties and high-resolution FORCs (Egli et al., 2010; Wang et al., 2013) for fresh specimens GA79.5y and GA84.6y (~20 mg chips) at room temperature after they were heated in air to successively higher temperature steps. Hysteresis parameters (M_r/M_s (saturation remanence to saturation magnetization) versus B_{cr}/B_c (remanent coercivity to coercivity)) of specimens GA79.5y and GA84.6y after each heating step (Figure 2e) plotted on a Day plot (Day et al., 1977) show that both specimens alter to more SSD-like behavior (higher M_r/M_s and lower B_{cr}/B_c), with GA84.6y becoming somewhat more SSD-like than GA79.5y. High-resolution FORC diagrams (Figures 2f–2s) clearly show that GA79.5y stayed relatively stable all the way up to at least 550°C (if not 600°C), whereas GA84.6y started to alter around 500°C, with a shift of the central ridge and an increase of the peak of coercivity likely due to reduction of effective magnetic grain size (increases of microcoercivity and TRM carrying efficiency). Thus, our rock magnetic results of GA84.6y (Figures 2e and 2m–2s) and previous RESET paleointensity results of GA84.6c (Figure 2c) support the inferences from our theoretical demonstrations in Section 2 by showing the incapability of pTRM checks to consistently detect the entirety of thermal alterations in Thellier-series experiments. On the contrary, GA79.5 is a revealing example of a sample that failed the pTRM check (Figure 2a) but showed insignificant evidence of magnetic alteration with heating (at least up to 550°C) by independent rock magnetic characterizations (Figures 2e and 2f–2l).

4. Monitoring Thermal Alteration Using the RESET Method

The RESET procedure that Wang and Kent (2013) developed to correct for the concave-up Arai plot caused by MD effects is conducted by repeating the Thellier-series experiment after giving the same sample a laboratory-applied total TRM (tTRM) from the Curie temperature (Figure 1h) after the original round of Thellier-series experiments. The corrected Arai diagram is then plotted with the original NRM remaining (unblocking) versus the tTRM loss (unblocking) (pink lines in Figures 1e, 2b, and 2d); to the degree that both the original NRM and the laboratory tTRM are carried by the same population of unaltered magnetic grains (which can be SSD, MD, or some mixture), the plot will be linear and provide a valid paleointensity value.

Using the RESET procedure, Wang and Kent (2013) also plotted the pTRM acquisition in the original Thellier-series experiment versus that of the repeated experiment, which provides a so-called “tTRM check” that measures alteration of the TRM spectrum (red lines in Figures 1f, 2a, and 2c). The tTRM check is a more powerful (yet still not infallible) approach to unveiling alteration than the pTRM check because it compares the TRM spectrum acquired from the original Thellier-series experiments to the TRM spectrum acquired from the repeated Thellier-series experiments after heating the sample to the Curie temperature, which potentially can cause more changes in the TRM carrying capabilities than heating just to the next one or two successive temperature steps as in pTRM checks. However, the tTRM check is still incapable of detecting thermal alteration if grains with blocking temperatures that are lower than each pTRM acquisition temperature do not alter after being heated to the Curie temperature. In this case, the pTRM acquisition from the repeated Thellier-series experiment (steps 3, 6, 9, 12, 15, and 18 in Figure 1h) will be the same as the original experiment (steps 3, 6, 9, 12, 15, and 18 in Figure 1g), yielding a falsely passed tTRM check

(linear red line in Figure 1f). Thus, neither the pTRM nor the tTRM check is capable of reliably detecting the overall thermal alteration in Thellier-series experiments.

Nevertheless, since the corrected Arai diagram plots NRM unblocking versus tTRM unblocking, MD effects cannot contribute to its curvature. After excluding secondary magnetization overprints, such as viscous remanent magnetization (VRM), the only factor that is able to affect the linearity of the corrected Arai plot is the alteration of the TRM spectrum. Thus, departure from linearity of the corrected Arai plot can be used to detect the overall thermal alteration during Thellier-series experiments, except in the implausible situation that alteration multiplies all parts of the TRM spectrum by a constant. We name this test the “corrected Arai linearity check” (CAL check). For example, after excluding the VRM bearing low-temperature interval, specimen GA79.5c (GA84.6c) with a linear (curved) corrected Arai plot shows the absence (presence) of such alteration (Figures 2b and 2d), as suggested by our comprehensive rock magnetic measurements (Figures 2e and 2f–2s).

To quantitatively evaluate the CAL check, we can simply use the coefficient of determination (r^2) to gauge the linearity of the selected temperature interval of the corrected Arai plot (CAL values), which is the square of its linear regression correlation coefficient (P-Int-R) (Wang & Kent, 2013). For example, specimen GA79.5c and GA84.6c have CAL values (and corresponding P-Int-R values shown in brackets) of 0.9986 (0.9993) and 0.9031 (0.9503), respectively. Critical minimum CAL values (P-Int-R values) of 0.9801 (0.9900) (Wang & Kent, 2013), 0.9604 (0.9800) (Wang et al., 2015), or similar values can be applied to select qualified RESET paleointensity results.

In the history of paleointensity studies, many samples with concave-up Arai plots and passed pTRM checks (similar to the original Arai plot of GA84.6c in Figure 2c) are attributed to MD effects (e.g., specimen hw108b1 in Figure 2i of Cromwell et al., 2015). However, to generate a factor-of-two shallowing of the high-temperature interval of the Arai plot, the average magnetite grain size needs to be larger than 30 μm , with M_r/M_s ratios less than 0.05 (Smirnov et al., 2017). Coarse grain sizes like this are not commonly observed in lavas that are used for paleointensity studies. Thus, our results suggest that many of the previously observed concave-up Arai plots may be due to thermal alterations that are not detected by pTRM checks rather than by MD effects. This could be tested by using our RESET method to see if the tTRM checks and, especially, the CAL checks yield significant departures from linearity.

5. Conclusions

Alteration of the original TRM carrying capacity in Thellier-series experiments is a decisive factor in determining the reliability of the paleointensity record. To date, the pTRM check is virtually the only indicator for laboratory-induced thermal alteration embedded in Thellier-series paleointensity experiments (Paterson et al., 2014), the results of which are relied upon no matter which set of data qualification criteria is used (e.g., ThellierTool-A and ThellierTool-B (Leonhardt et al., 2004); PICRIT-03 (Kissel & Laj, 2004); PQI (Biggin & Paterson, 2014), CCRIT (Cromwell et al., 2015; Tauxe et al., 2016)). However, satisfying a pTRM check does not guarantee the absence of thermal alteration, which raises significant questions about the overall quality of the paleointensity results in the current databases. Conversely, failed pTRM checks in many cases may be attributed to MD behaviors rather than alteration effects, which, if not distinguished, may also bias paleointensity estimates inadvertently derived from MD samples. Our illustration of the weakness of the pTRM check strongly suggests it should be treated more judiciously in paleointensity studies.

We propose a new method that utilizes a recently developed RESET procedure with tTRM checks and the newly advanced CAL checks to monitor the overall thermal alteration that may have happened to the very specimen used in Thellier-series experiments. Our rock magnetic results reinforce the effectiveness of the CAL check, which we suggest is practically the only theoretically sound method to reveal the entirety of thermal alterations in Thellier-series experiments.

We suggest that the RESET procedure can be routinely carried out not only to differentiate MD behaviors from alteration effects but also to pinpoint possible thermal alteration in SSD-bearing samples. To facilitate universal application in future paleointensity studies on SSD samples, it is also possible to use a pTRM-based RESET experiment (truncated at a moderate temperature instead of the Curie temperature), which

leaves the original pTRM between the truncation temperature and the Curie temperature intact and can be left out in the paleointensity analysis. Using pTRM-based RESET instead of tTRM-based RESET procedure allows the maximum heating temperature to be lowered from the Curie temperature so that thermal alteration can be reduced, which could lead to more accurate paleointensity estimates.

And conveniently, even previously thermally demagnetized samples can be used with the RESET procedure to acquire corrected Arai plots to estimate paleointensities by just thermally demagnetizing laboratory imparted total TRMs. Although pTRM checks and tTRM checks cannot be added in this sequence, accurate paleointensities can still be achieved since the automatically built-in CAL checks can be used to detect thermal alteration. Such an abbreviated experimental protocol could help make it practical to obtain sufficient data for routine evaluation of site-level consistency in paleointensity, in parallel to what is customarily done for paleomagnetic directions and which could ideally be combined as full-vector representations of the paleofield.

Data Availability Statement

Data presented in this study can be downloaded from <http://doi.org/10.5281/zenodo.4432110>.

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