

COMMENT

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This article is a comment on Brandt et al. (2020), <https://doi.org/10.1029/2018JB016968>.

Key Points:

- The Late Pennsylvanian glacial rhythmites of the Mafra Formation have a complex rock-magnetic signature
- These rocks have been overprinted during the Jurassic-Cretaceous in the context of widespread remagnetizations in South America
- The nature of the magnetization recorded by these rocks precludes their use for the evaluation of paleosecular variation models

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Comment on "New Late Pennsylvanian Paleomagnetic Results From Paraná Basin (Southern Brazil): Is the Recent Giant Gaussian Process Model Valid for the Kiaman Superchron?" by Brandt et al.

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Abstract The late Pennsylvanian glacial rhythmites of the Mafra Formation (Itararé Group) from the Paraná Basin of Southern Brazil, have a complex rock-magnetic signature. Rock-magnetic tests imply that both hematite and magnetite in varying grain sizes are responsible for the magnetic remanence. Thermal and alternating field (AF) demagnetization of the natural remanence reveal different behaviors that are attributed to a mixture of remanence-carrying magnetic minerals and grain sizes, implying a series of magnetic overprints. A great circle analysis of the distribution characteristic directions reported suggests that these rocks may not be entirely immune from magnetic overprints acquired during the Jurassic-Cretaceous in the context of widespread remagnetizations in South America. The nature of the magnetization recorded by these rocks warrants caution in their use for the evaluation of paleosecular variation models.

1. Comment

Brandt et al. (2019) present a study conducted on the Itararé Group glacial rhythmites of the Carboniferous (late Pennsylvanian) Mafra Formation from the Paraná Basin of Southern Brazil. They use paleomagnetic and rock-magnetic evidence to support a primary origin of the magnetization and in turn use this to question the validity of the TK03 paleosecular variation (PSV) model (Tauxe & Kent, 2004), when applied to the Paleozoic. However, the evidence presented for primary magnetizations is scant, and the study neglects a growing body of evidence that testifies to widespread secondary magnetizations affecting Paleozoic to early Mesozoic rocks in South America (e.g., Bilardello et al., 2018; Font et al., 2011, 2012; Henry et al., 2017). The failure to properly consider the extent and consequences of remagnetization in these rocks undercuts the authors' conclusions about paleofield behavior and validity of PSV models for the Paleozoic.

Brandt and coworkers claim that single domain (SD) magnetite grains are the only magnetic carriers observed in the rocks and are solely responsible for the magnetic remanence. However, from the evidence presented, it is questionable that the rocks contain a single magnetic mineralogy. While stable SD (SSD) grains have longer relaxation times and are thus stable remanence recorders that do not acquire viscous overprints (e.g., Butler, 1992), they are not immune from secondary magnetizations of different origin (e.g., thermal). Moreover, whether SD grains are dominant in the studied rocks or not, they have not been directly associated to the paleomagnetic characteristic remanence isolated. Other magnetic minerals and/or grain size fractions, whether primary but occurring in minor quantities and/or of secondary origin (e.g., thermochemical precipitation), may be responsible for the remanence. The paper lacks a clear discussion linking the inferred magnetic minerals to the isolated paleomagnetic directions, and unfortunately, the rock-magnetic experiments conducted are insufficient to support the claims presented in the article, and a more detailed rock-magnetic characterization is required.

This comment addresses the rock-magnetic results and paleomagnetic data separately, detailing how the evidence presented is not clear-cut and the conclusions reached debatable. The comment also highlights inaccuracies that do not justify the conclusion that the TK03 model of PSV may be invalid for the Paleozoic.

1.1. Low-Temperature Magnetometry

The study attributes behavior of laboratory remanences (FC-ZFC curves) measured as a function of temperature to single domain magnetite grains, presenting data from one specimen only. The inference is based on

the observation that the field-cooled (FC) curve presented (remanence measured upon warming after cooling the specimen in an applied field) has stronger magnetization than the zero field-cooled (ZFC) curve (remanence measured upon warming after cooling the specimen in a zero field and applying a saturation isothermal remanence (SIRM) at low temperature). This results from an anisotropy induced by cooling through the Verwey transition (T_V) in a strong field, which orients the monoclinic easy axes as they form (parallel to one of the three symmetrically equivalent 100 axes of the cubic phase) preferentially along the cooling field (Carter-Stiglitz et al., 2002, 2004 and references therein). Conversely, multi domain (MD) grains become more strongly magnetized by an SIRM, resulting in stronger magnetizations when compared to a FC remanence, due to the different orientation distributions of monoclinic twin boundaries in the two states (Carter-Stiglitz et al., 2006; Kasama et al., 2013; Kostrov & Fabian, 2008). However, although FC remanence higher than ZFC is indicative of the presence of SD grains in some quantity, it cannot exclude the occurrence of MD grains in the same specimens. Importantly, because SD grains have higher remanence ratios (M_{RS}/M_S) than MD grains, they will dominate the remanence even if present in smaller quantities and regardless of their origin (e.g. Smirnov, 2009). In fact, the separation of the FC and ZFC curves shown by Brandt et al. (2019) is actually fairly small compared to specimens truly dominated by SD grains (e.g., Moskowitz et al., 2008; Pan et al., 2005), indicating that a significant fraction of the remanence is carried by larger grains. There is only a small fraction of the low-T remanence lost at T_V for both the FC and ZFC remanences; most of the loss is distributed over the range from 20 to 300 K, as is typical of the unblocking of nanophases. There is also a separation of FC and ZFC that persists above T_V , typical of oxidized magnetite (e.g., Kostrov, 2003) or oxyhydroxides such as goethite (e.g., Guyodo et al., 2003). Moreover, the remanence acquired after imparting an SIRM at room temperature and measured while cooling to low temperatures and back (RTSIRM curves) has a distinctive curvature. This “humpiness” has been shown to be the hallmark of maghemitization (Özdemir & Dunlop, 2010), implying some oxidation of the sample. This evidence for oxidation does not necessarily affect the fidelity of the remanent directions recorded but directly implies some alteration of the mineralogy, whether pre-, syn-, or post-depositional.

1.2. Hysteresis Data

The hysteresis loop shown is never described. However, the figure reported shows a loop that is wasp wasted, implying the presence of grains with different coercivities, whether from one or multiple magnetic minerals (e.g., Tauxe et al., 1996). This evidence further demonstrates that SD magnetite grains alone cannot be solely responsible for the overall magnetization of the specimen.

The first-order reversal curve (FORC) diagram shows B_C values extending from 0 to 100 mT. Positive distributions are shown throughout the background of the diagram and likely result from measurement drift that has not been sufficiently accounted for. Performing more efficient smoothing, for example, utilizing the VARIFORC protocol of Egli (2013), and showing the corresponding signal/noise envelope would have been ideal for such data. Unfortunately, the FORC was acquired utilizing the irregular grid protocol of Zhao et al. (2015), who explicitly state, “Although the proposed irregular measurement protocol is not as efficient at suppressing noise as recently developed postprocessing techniques (e.g., VARIFORC), it enables efficient high-resolution analysis for relatively strongly magnetized samples where measurement noise is not detrimental to FORC distribution estimation.” Given the amount of measurement noise present, the experiment could have been more informative utilizing a classic protocol allowing for more efficient smoothing.

Inspecting the diagram closely, it is apparent that the distribution of coercivity extends beyond the range of B_C of the diagram (100 mT). Switching fields greater than 100 mT are rare for magnetite and have only been observed in elongated grains and chains (e.g., Egli et al., 2010; Roberts et al., 2000), suggesting the presence of a higher coercivity phase like hematite. Focusing on the main distribution of coercivity, the strongest peak occurs at very low coercivities with some “smearing” along B_U , suggesting a low coercivity phase like MD magnetite, in addition to a higher coercivity phase responsible for the high-coercivity distribution along B_C identified by the authors. A positive distribution is also present along the negative B_U axis: this is not consistent with the expected behavior of stable uniaxial SD grains (e.g., Egli et al., 2010; Newell, 2005) and is more indicative of relaxation of the magnetization after the application of the reversal fields, therefore implying a superparamagnetic (SP, or “unstable SD” grains) contribution to the magnetization of the sample (Lanci & Kent, 2018; Pike et al., 2000; Roberts et al., 2000). The presence of SP grains in a sedimentary rock could reflect in situ growth of new magnetic grains and thus secondary magnetizations. If this is the case,

then it would not be unreasonable to envision interacting SD magnetite grains on top of the primary mineralogy. The resolution of the FORC, however, does not allow discriminating among these possibilities.

It must also be pointed out that the dominant distribution observed along the B_C axis represents a small percentage of the total FORC area and that the saturation field of 300 mT utilized for this experiment is not sufficient to fully activate, let alone saturate, a high-coercivity phase like hematite. The FORC experiment appears to be designed to highlight the contribution of magnetite over any other magnetic phase present. Similarly, the majority of the induced FORC (iFORC) area shown seems to be largely affected by measurement noise. This observation is confirmed by the FORCs shown in the inset, which demonstrate measurement drift and nonclosure of the outer loop. It is thus questionable whether the rather poorly defined three regions (negative-positive-negative) identified are actually representative of the SD population.

While it is probable that the specimens do contain SD magnetite particles, the rock-magnetic evidence presented suggests that pseudo single domain (PSD) and MD grains may be contributing a substantial fraction of the bulk remanence together with a high-coercivity mineral. Moreover, if SD grains are present, these could just as easily be interacting SD grains and thus point to a secondary origin. Simple rock-magnetic tests such as isothermal and anhysteretic remanent magnetization acquisition and subsequent alternating field (AF) demagnetization experiments (e.g., Cisowski, 1981; Henkel, 1964) and unmixing of coercivity spectra (e.g., Maxbauer et al., 2016) would have provided useful evidence on the coercivity distribution and the nature of the remanence. Likewise, extending the experiments already performed by acquiring AF susceptibility data as a function of frequency and temperature (e.g. Till et al., 2011) would shed light on the domain state of the grains present.

1.3. Paleomagnetism and Shape Analysis of the Distribution of Directions and VGPs

Thermal demagnetization of the natural remanent magnetization (NRM) for specimen DDL078B2B is shown to hold remanence up to 670°C, and the characteristic remanent magnetization (ChRM) was isolated up to that temperature (cf. Brandt et al.'s (2019) figure 2 and supplementary data), which is close to hematite's Néel temperature and far past magnetite's 580°C Curie temperature. The labels on the orthogonal demagnetization and stereonet plots in Figure 2, however, state 580°C as the upper limit of the ChRM. While most of the remanence is lost by ~580°C, the observation of remanence held up to 670°C confirms that high-coercivity/high unblocking temperature hematite is present in the samples, as suggested by the FORC data, with a magnetization direction subparallel to that of magnetite and thus contributing to the ChRM.

The high-coercivity phase contribution cannot be fully removed with AF demagnetization, resulting in the great circle patterns observed on a stereonet, as well as the difference between the thermal and AF mean directions. This behavior further implies that the directions carried by the harder phase and the magnetite are not perfectly aligned, with at least one of the two phases being either secondary or overprinted. Additionally, the magnetite may also exist in different grain sizes, displaying a range of coercivities and thus recording different components of magnetization. The great circle distributions obtained through AF demagnetization, as convincingly shown in the study, have very similar orientation to the plane containing the secondary (North and up) and ChRM (SE and down) directions isolated through thermal demagnetization up to 600°C, further demonstrating overlapping remanence components. Given that AFs do not fully demagnetize the high-coercivity component and that the ChRM isolated through thermal demagnetization is likely "contaminated" by the same component, the similar orientation between these two sets of planes is reassuring. The mean ChRM and secondary directions can be fitted with a great circle that also includes the early Cretaceous direction expected for the area, as calculated from the mean paleopole of Somoza and Zaffarana (2008) ($D = 355.4^\circ$, $I = -40.2^\circ$, Figure 1a). The extensive magmatic events throughout the Paraná basin at this time (e.g., Ernesto et al., 1999) provide a likely mechanism for the overprint observed.

The ChRM directions isolated by thermal demagnetization, however, have a different distribution orientation than those derived through AF demagnetization. As informed by the thermal demagnetization temperature intervals over which the directions have been isolated (typically 350–600°C but also up to 670°C), these reasonably represent the distribution of hematite, and the more coercive of the magnetite grains presents (i.e., not the grain size fraction responsible for the low temperature direction). However, while the majority of the ChRM directions isolated by Brandt and coworkers through both thermal and AF demagnetization point to the southeast and have high positive inclinations, a few directions have been isolated through both thermal and AF demagnetization that have negative inclinations. Unfortunately, these are not reported.

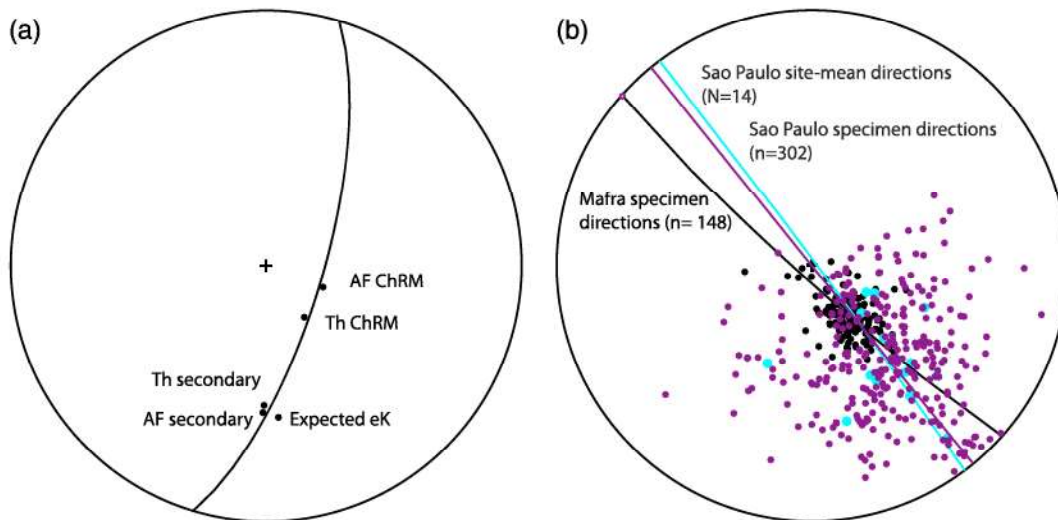


Figure 1. (a) Mean ChRM and secondary directions isolated from the Mafra Fm rhythmites reported by Brandt et al. (2019) from both AF and thermal (Th) demagnetization, and the expected mean early Cretaceous (eK) direction for the area, together with their best fit plane plotted on a lower hemisphere stereonet; (b) paleomagnetic specimen directions from the Itararé Group Mafra Fm reported by Brandt et al. (2019) and best fit plane ($n = 148$, black), together with the mostly south and down specimen directions only isolated by Bilardello et al. (2018) from the Itararé Group rocks from the state of Sao Paulo and their best fit plane ($n = 302$, purple), and their site-mean directions and best fit plane ($N = 14$, cyan and larger symbols), plotted on a lower hemisphere stereonet. Despite the scatter, the Mafra and Sao Paulo distributions have a large overlap and similar orientations regardless of whether the latter are analyzed at the specimen or site-mean level. See text for details.

Brandt et al. (2019) state, “These are spurious directions because they have no consistency with nearby samples” and eliminate them from further analysis. Likewise, they eliminated directions with lower precision (maximum angular deviation (MAD) angles $>15^\circ$) from their analysis. In total, 11% of the total data set was eliminated, and it cannot be entirely ruled out that the different distribution of the thermal data set is due to the selection process.

Bilardello et al. (2018) had isolated discordant ChRM site-mean directions from the Itararé Group in the northern Paraná Basin even within the same drill cores. They attributed this behavior to heterogeneities in grain size, which is characteristic of glacial sediments, with different grain sizes recording different components of magnetization. However, instead of discarding these data, they showed that results that appear discordant based on their directions and lower statistical precision ($\alpha_{95} > 15^\circ$) are actually in agreement when performing distribution analyses in the context of remagnetization circles, defining distinct elongated distributions. Plotting only the ~SE and down directions (both at the specimen, $n = 302$, and site-mean level, $N = 14$) isolated from the Itararé Group rocks by Bilardello et al. (2018) and comparing these to the specimen directions presented in Brandt et al.’s (2019) study ($n = 148$), similar orientations of their distributions are obtained, although the distribution from Sao Paulo has an overall shallower mean and higher overall scatter at both specimen and site-mean level (Figure 1b). The comparison suggests that the distribution of Brandt et al. (2019) could be an artifact of data selection. Conversely, its primary nature needs to be fully demonstrated.

Maintaining the directions that do not point to the Southeast and down, Bilardello et al. (2018) obtained two distinct distributions for the Sao Paulo rocks that are substantially different from the one shown above. These distributions are better evaluated when plotted as (South) virtual geomagnetic poles (VGPs), allowing for a better comparison among results from different localities. In Figure 2a, VGPs and best fit distribution of the Mafra Fm rhythmites are reported, together with those obtained from the Itararé Group rocks from the Rio do Sul and Itu rhythmite exposures (Franco et al., 2012) and the Itararé Group rocks sampled across the state of Sao Paulo (Bilardello et al., 2018). Notably, Franco et al. (2012) had concluded that the magnetic mineralogy of the Rio do Sul and Itu rocks consists of primary magnetite and hematite grains which both acquired their magnetization during early stages of diagenesis and further presumed the Itu rocks to contain additional authigenic magnetite formed during the same time interval. In particular, the Rio do Sul

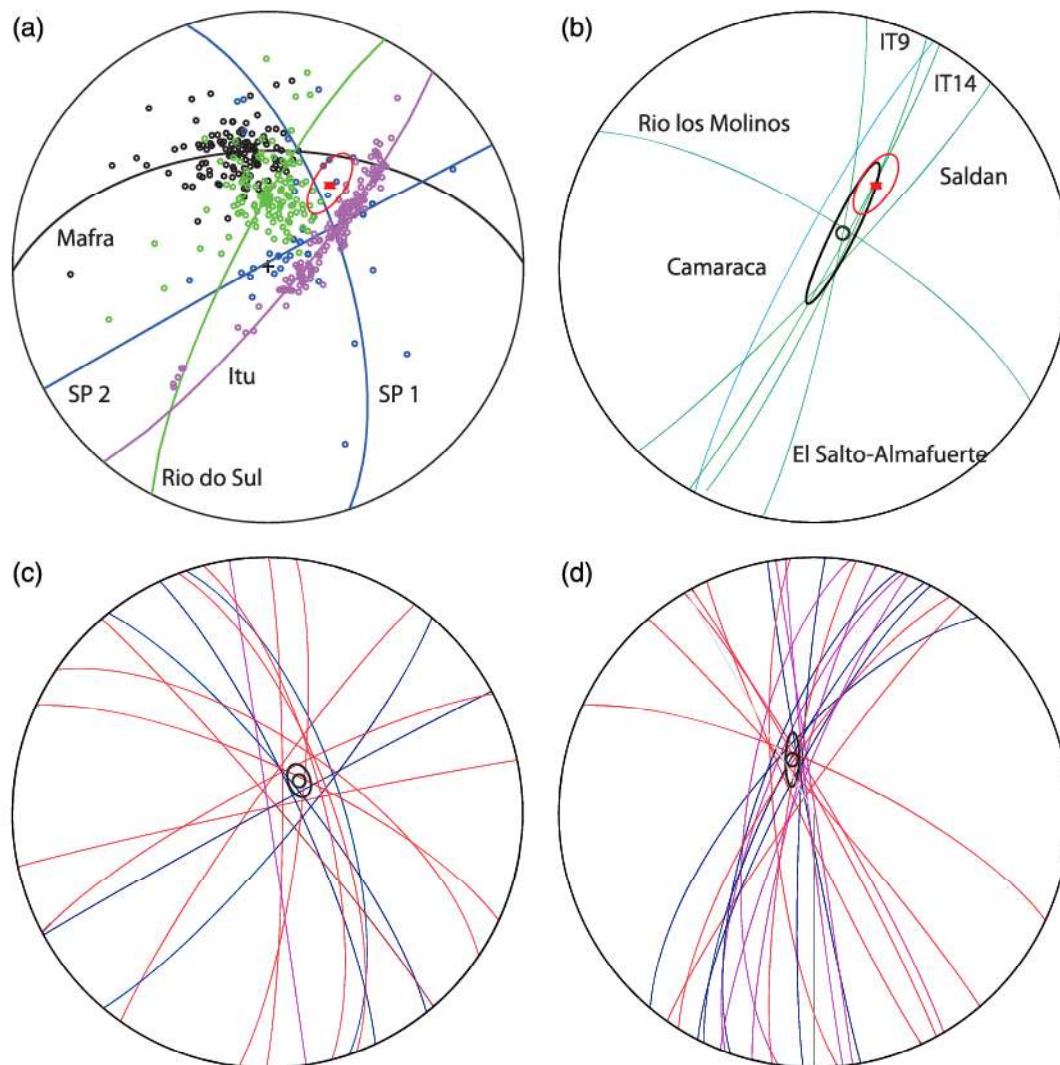


Figure 2. Distributions of south VGPs and best fit planes determined from South American rock formations plotted on upper hemisphere stereonets. (a) Distribution of Itararé Group rocks from the Mafra Fm (in black) from Brandt et al. (2019), Rio do Sul (green) and Itu (purple) from Franco et al. (2012), and across the state of Sao Paulo (SP1 and 2, in blue) from Bilardello et al. (2018). The mean intersection and confidence ellipse is shown in red; (b) mean intersection of the Itararé VGP distributions in (a) and the mean intersection and confidence ellipse (in black) of problematic Jurassic and Cretaceous rock formations from South America, including two sills (IT9 and IT14) that intrude the Itararé in the State of Sao Paulo; (c) Carboniferous (blue), Permian (red), and Triassic (purple) best fit planes through VGPs from different South American formations showing a mean intersection that coincides with the problematic Jurassic and Cretaceous mean paleopoles (black circle and confidence ellipse). (d) Carboniferous, Permian, and Triassic (colors as in panel c) best fit planes through VGPs from different South American formations showing a mean intersection that coincides with the late Cretaceous mean paleopole (black circle and confidence ellipse). Panels b, c, and d modified after Bilardello et al. (2018); see references within for the original data sets.

rhythmites, which have a paleopole in reasonable agreement with that of the Mafra Fm, exhibited unstable thermal demagnetizations behavior, and their ChRM was exclusively determined through AF demagnetization up to 160 mT (up to ~50% of the natural remanence remained, Cf. figure 8 of Franco et al., 2012). These observations raise concerns regarding the nature and timing of the magnetizations carried by these rocks.

Plotted on an upper hemisphere stereonet, the elongations of the VGP distributions tend to intersect in a common area defined by a bootstrap ($N = 10,000$) confidence ellipse (Figure 2a). Moreover, the mean intersection of the great circles fitted through the Itararé Group rocks is within error the same as that obtained from certain Jurassic-Cretaceous igneous rocks associated with the opening of the South Atlantic, which have been considered problematic owing to their elongated distributions, including igneous rocks that intrude the Itararé Group sedimentary rocks (Figure 2b) whose elongation reflects that observed by

Ernesto et al. (1999) across the state of Sao Paulo (see Bilardello et al., 2018). Moreover, the mean intersection is close to that obtained from a compilation of South American Carboniferous to Triassic formation-derived VGP distributions (Figures 2c and 2d), convincingly suggesting that these late Paleozoic and early Mesozoic rocks were affected by the Jurassic-Cretaceous volcanism, resulting in overprinting by thermochemical magnetization events, regardless of their age and location. In truth, based on the distribution and mean pole position, it must be noted that among all the Itararé Group rocks shown, the magnetic remanence directions isolated through thermal demagnetization from the rhythmites of the Mafra Fm appear the least affected by such secondary overprints, yet they are probably not completely immune and the exact nature of their distribution needs to be fully determined. If the distribution reported by Brandt et al. (2019) is indeed an artifact resulting from incomplete cleaning of the remanence and data selection criteria, as suggested in this comment, then it would be reasonable to interpret it as the result of unremoved early Cretaceous overprints. Such overprints would naturally preclude any analysis in the context of PSV and fits to PSV models.

The overprints, however, do not necessarily invalidate the cyclostratigraphic study. Whether the secondary magnetizations are hosted by secondary minerals or constitute a resetting of the primary iron oxides, the response of the original magnetic mineralogy likely still dominates the laboratory remanence and the susceptibility signal from which the cyclostratigraphy is obtained, providing a cyclicity that may still be representative of the time of deposition. Likewise, the anisotropy of anhysteretic remanent magnetization (AARM) fabrics reported are also largely immune from secondary magnetizations because the field settings used to apply the magnetizations (100- μ T DC field over 35- to 100-mT AF demagnetization) primarily isolate SD magnetite and are unaffected by any secondary resetting of the magnetization of detrital grains and/or authigenic grains with coercivities that do not fall within that field range. Li and Kodama (2005) showed that AARM fabrics of remagnetized natural and synthetic samples inherit the primary depositional fabrics and that these can be enhanced by authigenic grains that grow mimetically along the primary fabrics. Such and similar results obtained by Henry et al. (2003) and Borradaile and Lagroix (2000) on different rock types preclude any interpretation of a primary remanence from AARM fabrics alone.

1.4. Validity of the TK03 Model

The study by Brandt et al. (2019) questions the validity of the TK03 model for the Paleozoic and consequently the validity of applying the E/I inclination correction technique, that is based on that model, to Paleozoic rocks. Brandt et al. (2019) state that the E/I method has not been tested for Paleozoic times, although it has been applied by some authors (presumably to rocks of that era) and that the model should be abandoned as a reference for inclination shallowing corrections for Carboniferous rocks. This statement, however, is inaccurate: Bilardello et al. (2011) performed a comparison of the E/I and anisotropy-based inclination correction techniques on the Carboniferous Shepody Formation red beds from North America reported by Bilardello and Kodama (2010). In that study, 104 specimen directions from 19 sites were obtained through thermal demagnetization, while 60 specimen directions were obtained through chemical demagnetization of 16 sites. The site-mean directions isolated through both techniques were statistically undistinguishable, attesting the robustness of both results and their validity for paleomagnetic investigations. After applying an anisotropy-based correction, the uncorrected mean inclination of the 19 site-mean directions obtained through thermal demagnetization changes from 20.4° to 30.1° ($\alpha_{95} = 7.7^\circ$) (Bilardello & Kodama, 2010) and to 30.21° (confidence on the inclination of 8.92°) after propagating the uncertainty from the correction itself (Bilardello et al., 2011). Applying the E/I technique to the 104 individual directions, the mean inclination of 20.7° corrects to 30.0°, with low and high corrections of 21.7° and 37.6° (corresponding to 8.3°–7.6° of uncertainty, respectively). This comparison attests to an excellent agreement between the anisotropy and E/I techniques, including the uncertainties around the corrected inclination. While this constitutes merely one test on the applicability of the E/I technique on late Paleozoic rocks, the more systematic studies of Tauxe et al. (2008) for the Mesozoic and Cenozoic eras and Tauxe and Kodama (2009) for the Proterozoic find the E/I technique and assumption robust, lending more confidence that the Giant Gaussian process-type TK03 model can be considered reliable for times more ancient than five million years, including the Kiaman Superchron.

The negligence of a growing body of evidence for widespread remagnetization in South America has wide consequences. In fact, remagnetizations have led to a wealth of incongruent paleomagnetic data that bear

direct implications in at least two long-standing paleomagnetic problems, the anomalous elongation of certain Jurassic-Cretaceous VGPs (e.g., Geuna & Vizan, 1998) and the Pangea controversy (e.g., Irving, 1977), as well as challenges to PSV models. Clearly, any data-based study that aims at assessing PSV models must ensure data quality, and this article does not meet this criterion. It is well documented that the Paraná Basin was affected by widespread magmatism related to the opening of the South Atlantic (e.g., Abbot & Isley, 2002; Araujo et al., 2000, 2005; Ernesto et al., 1999; Menezes & Travassos, 2000; Milani & Zalán, 1999; Thomaz Filho et al., 2008), providing a likely mechanism for remagnetization. There is a need to embrace a more candid and holistic approach to paleomagnetism and the evaluation of paleomagnetic data.

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