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Supporting Information for

Pinpointing the mechanism of magnetic enhancement in modern soils using high-resolution magnetic field imaging

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Introduction

This Supporting Information file contains text describing the methodology for estimating grain size from ARM acquisition precision and a corresponding figure.

S1. Application of thermoremanent magnetization (TRM) statistical recording limit analysis to anhysteretic remanent magnetization (ARM)

All natural forms of remanent magnetization except for lightning result in only a weak bias of magnetic grain magnetizations towards the ambient magnetic field direction. As a result, a substantial number of independent ferromagnetic grains is necessary to reveal a statistically significant bias. Even more grains are required to quantify the strength and direction of the ambient field with high precision. Berndt et al. (2016) quantified the relationship between the number of ferromagnetic grains and the precision of paleomagnetic recording, represented by the variance in direction and magnitude of the resulting magnetization, assuming a TRM acquisition process on single domain grains. Under these assumptions, the equilibrium probability, p, of a single grain being magnetized in the direction of the bias field is (Eq. 4-5 in Berndt et al. 2016):

$$p(\phi) = \frac{1}{2}[1 + \tanh(x_b \cos\phi)] \tag{1}$$

where ϕ is the angle between the ambient field and the grain easy axis and x_b describes the balance between magnetic alignment energy and randomizing thermal energy:

$$x_b = \frac{\mu_0 V M_s B_0}{k T_B} \tag{2}$$

where μ_0, V, M_s, B_0, k, and T_B are the permeability of free space, grain volume, saturation magnetization at time of grain blocking, ambient field strength, the Boltzmann constant, and the blocking temperature, respectively.

The analysis of Berndt et al. (2016) was derived for thermal, viscous, and chemical remanent magnetizations (TRM, VRM, CRM) only. In these processes, the magnetic remanence carriers reach a thermodynamic equilibrium just before blocking. To relate ARM precision to the number of remanence-carrying grains, the ARM, too, needs to be dominated by particles that reache a thermodynamic equilibrium (Eq. 1) upon remanence acquisition. This is satisfied only if sufficient opportunities are available to each grain for thermally activated processes, described by the energy balance of Eq. 2, to modify the grain magnetization. In TRM, VRM, and CRMs this is always the case since before blocking, magnetic grains are free to change their magnetic moment due to thermally activations. For ARMs, however, due to the applied strong AF field, particles can generally only change their magnetization in one direction: i.e., align with the AF field direction at each time. The frequency of thermally activated grain realignment following classic single domain Néel theory for uniaxial anisotropy is (Dunlop and Ozdemir, 1997, Eq. 8.3):

$$f(B) = f_0 \exp\left[-\frac{B_k M_s V}{kT}\left(1-\frac{B}{B_k}\right)^2\right] \tag{3}$$

where B_k is the microscopic coercivity, B is the ambient field, and T is the ambient temperature, which is room temperature for ARM experiments. The factor $f_0 \approx 10^9$ s^{-1} represents the frequency with which thermal fluctuations result in opportunities to remagnetize the grain and f is the frequency at which such remagnetizations actually occur. For TRM, VRM, and CRMs, the field B is the weak (geomagnetic) ambient field; for ARMs, however,

$$B = B_{pulse} + B_0 \approx B_{pulse} \tag{4}$$

is the sum of the field strength of a single AF pulse B_{pulse} and the ARM bias field B_0,the latter of which is negligible compared to the former. For example, in our experiments a bias field of $B_0 = 300$ μT was used while the AF field may have been $B_{pulse} = 10$ mT for a given AF pulse during a single ARM acquisition. Since in our experiments the AF frequency is 613 Hz, a single pulse would have lasted for $\tau = 816$ μs. For the duration of this pulse, all particles with a coercivity B_k below B_{pulse} align with the field; particles with a coercivity above B_{pulse} may also align with the field due to thermal activations. No particle, however, can anti-align with the field, during this pulse, however, preventing full thermal equilibrium from being reached during a single pulse.

Rearranging eq. (3), one can interpret the ratio f/f_0 as the probability that the grain is remagnetized in a single (10^{-9} s duration) thermal fluctuation. The probability that a grain is not remagnetized during a single thermal fluctuation is therefore $1 - f(B_{pulse})/f_0$, and the probability that a grain is not remagnetized at least once during a single AF pulse of amplitude B_{pulse} and duration τ is therefore

$$p(B_{pulse}) = 1 - \left(1 - f(B_{pulse})/f_0\right)^{\tau f_0} \tag{5}$$

where the product τf_0 can be seen as the number of "thermal activation attempts" during a single AF pulse duration.

When the applied field is equal to or greater than B_k, the probability of remagnetization towards the bias field direction during each pulse (or indeed each thermal fluctuation) is 1, corresponding to deterministic remagnetization of the grain towards the applied field direction and making the field application equivalent to an isothermal remanent magnetization (IRM). As a result, upon all AF pulse applications $\geq B_k$, the magnetization of the grain is not in thermal equilibrium and the statistical reliability/scatter of the direction cannot be used to assess the number of particles.

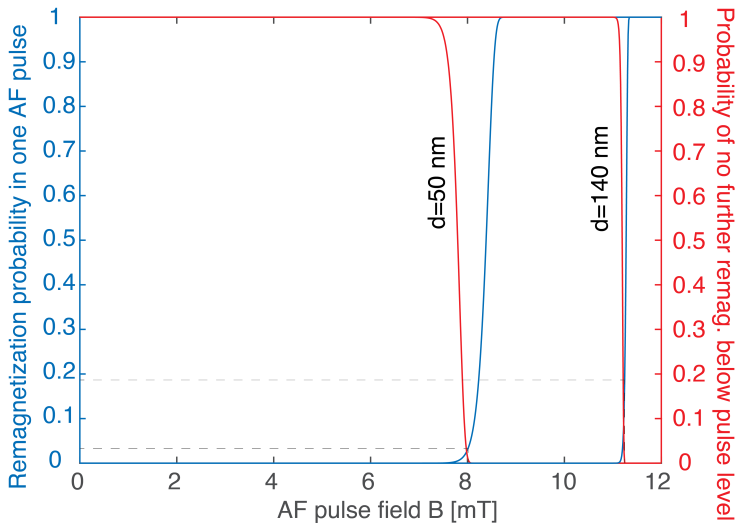

Figure S1. Predicted behavior of magnetite particles upon AF or ARM application. Blue curves indicate the probability of field-assisted thermal remagnetization of a grain with the given diameter during a single AF pulse. Red curves denote the probability that the grain would undergo no further remagnetizations during the AF ramp down after a given AF pulse field level. Dashed gray lines indicate the single-pulse remagnetization probability of a grain that has 99% probability of being remagnetized during the remaining AF ramp down. A low value indicated by the gray line implies that the grain is more likely to reach thermal equilibrium. Assumed grain sizes and $B_k = 12$ mT correspond to the ultrafine magnetite population found in the high precipitation soil. See text for details and additional parameters.

Probabilistic, thermally-assisted remagnetization of the grain according to Eq. 3 may occur during the AF pulses after the field value falls below B_k. In these cases, the grain under a 0 K environment would not be remagnetized, but at laboratory conditions may be remagnetized due to a combination of thermal energy and the pulse field decreasing the energy barrier according to Eq. 3. For AF pulses slightly smaller than B_k, the probability of remagnetization during each pulse remains high, implying that the resulting grain magnetizations are still mostly deterministic (see blue curves in Fig. S1). Conversely, for the pulses where B_{pulse} is significantly smaller than B_k, the outcome becomes less deterministic and the distribution of grain magnetization would converge towards the thermal equilibrium of B_0 in Eqs. 1-2 after infinitely many pulse applications.

Naturally, for very weak AF fields B_{pulse}, the probability of a thermally activated change in magnetization for a particle approaches zero. It would therefore take an infinitely long time and require infinitely many pulses to reach thermal equilibrium. There is, however, an intermediate range where the probability that a particle is remagnetized during one pulse is significantly smaller than 1, yet large enough such that repeated application of AF pulses will reach a thermal equilibrium. For example, if the probability of remagnetization during one pulse is 1%, then the application of 500 repeated pulses leads to a total probability of remagnetization of $(1 - 0.01)^{500} = 99\%$, achieving virtually thermal equilibrium.

One needs to keep in mind, however, that the AF field is continuously being ramped down. Hence, any given grain can only reach thermodynamic equilibrium due to repeated application of successively lower fields. The probability of a grain avoiding remagnetization (i.e., failing to reach thermodynamic equilibrium) after the AF ramped down to zero from a given value B_{pulse} can be computed as the product of the grain avoiding remagnetization at each pulse at and below B_{pulse} (Eq. 5; red curves in Fig. S1).

In summary, two conditions need to be fulfilled for a grain to reach thermodynamic equilibrium. First, it needs to have a high probability of being remagnetized at some point during successive AF pulse applications after B_{pulse} drops well below its microscopic coercivity B_k. Second, the grain must simultaneously have a low probability $p(B_{pulse})$ of being remagnetized during a single AF pulse, since this would otherwise be a deterministic process akin to an IRM. Having these two conditions fulfilled implies that the grain is highly likely to be remagnetized at least once under conditions where the effect of the AF pulse field is non-deterministic and the probability of remagnetization is guided by the balance of magnetic and thermal energies.

Therefore, applying a large number of AF pulses each representing a small probability of thermally remagnetizing the grain can result in a final grain magnetization distribution close to thermal equilibrium. To do so, we apply an AF ramp with very slow decay in peak pulse field strengths. We find that, for our AF frequency of 613 Hz, a ramp down rate of 1 mT s^{-1} results in 99% of 50-140 nm grains being remagnetized after the per-pulse probability of remagnetization has declined to less than 0.2 (Fig. S1). This implies that these ramp parameters are sufficient for magnetite grains associated with the ultrafine ferrimagnetic population to approximate thermal equilibrium, thereby validating the use of recording limit results for TRM on our ARM acquisition data.

References:

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