

## Nanophase magnetism and past global change

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**Abstract** Variations in the concentration of small magnetic particles in certain geological sequences reflect major climatic shifts during ice ages. The resulting magnetoclimatological signal is clear, but the underlying mechanism remains debatable. Nevertheless, spectral analysis of the resulting time series provides strong support for the notion that ice ages are driven by orbital (Milankovitch) forcing.

The geological record is a rich archive of past global change. In recent years it has emerged that some of these changes are encoded magnetically in certain deposits, giving rise to the term magnetoclimatology [1]. Among other things this field of endeavour includes the rapid, nondestructive testing of great thicknesses of strata, particularly by means of magnetic susceptibility which - to first order - measures the amount of iron oxide present. Mineralogically, the most important ingredients are magnetite ( $\text{Fe}_3\text{O}_4$ ), maghemite ( $\gamma\text{-Fe}_2\text{O}_3$ ) and hematite ( $\alpha\text{-Fe}_2\text{O}_3$ ). Even though they typically represent a mass fraction of less than 1%, variation in their amounts acts as a sensitive monitor of environmental change. For example, in many parts of the world thick deposits of loess (windblown silt) retain a record of the waxing and waning of ice ages. During a typical glacial interval windblown silicate grains (a few tens of microns in diameter) might build up a sedimentary layer a metre thick in ten thousand years or so. In the following interglacial period soil-forming processes, or pedogenesis (Greek  $\pi\epsilon\delta\omicron\nu$ , ground), create iron oxide particles (a few tens of nanometres in diameter) that lead to enhanced magnetism. In the celebrated geological outcrops in China the mix of physical, chemical and biological mechanisms that make up the complex process of pedogenesis actually renders these palaeoclimatic changes visible, in the form of alternating bands of light- and dark brown strata (figure 1). Magnetic profiles have been measured from top to bottom of many such sections. It is found that the susceptibility of the pristine airfall material that accumulated during glacial intervals yields a fairly constant background to which is added the interglacial pedogenic magnetism reaching peaks more than an order of magnitude greater (figure 2). In terms of the particle sizes involved, it is observed that the size distribution spans the single-domain (SD) field (less than  $\sim 140$  nm for magnetite) and extends below the superparamagnetic (SP) threshold ( $\sim 30$  nm for magnetite). As the mineral grains pass from SD to SP behaviour their susceptibility discontinuously jumps by more than an order of magnitude, a feature that was first emphasized in connection with the nanoparticles of pure iron found in the dust on the lunar surface [2,3].

The actual mechanism that produces the magnetic enhancement is still a matter of debate. One possibility is that magnetotactic bacteria leave the legacy of their magnetosomes after they die (for excellent examples of relevant microscopy see <http://geography.lancs.ac.uk/cemp/atlas>). However,

these have not been identified in sufficient numbers to quantitatively explain the observations. Recent investigations [4], based on the coercivity spectra of laboratory-induced isothermal remanent magnetizations, suggests that up to two thirds of the remanent magnetism in the fossil soils (palaeosols) resides in slightly oxidised magnetite that was newly created from iron-bearing clay minerals and mafic silicates. Regardless of the mineralogical details underlying the magnetic signal in these Chinese stratigraphic sequences, there is little doubt that it is climatically controlled. This was recognised in an early paper [5] that demonstrated a clear correlation between the magnetic time series and the well-known marine oxygen isotope stages, the canonical framework to which all palaeoclimatic proxies are nowadays referred (<http://www.ngdc.noaa.gov>). This framework essentially reflects the response of sea level to the uptake and release of water as the polar ice caps grow and decay; it provides a truly global yardstick. Furthermore, spectral analysis of magnetic susceptibility time series from continental sediment packages as well as from sediments in very old lakes (such as Lake Baikal, Siberia) yield the frequencies expected from the so-called Milankovitch cycles caused by slight variations in the Earth's orbital parameters [6-8]. This confirms the concept that ice ages are triggered by orbital forcing. But problems remain concerning the relative magnitudes of the eccentricity, obliquity and precessional components.



Figure 1. A geological exposure in China consisting of an alternating sequence of light- and dark brown strata that reflect glacial and interglacial intervals, respectively. During interglacial times warmer and wetter conditions lead to the production of soil (preserved as fossil soils, or palaeosols, in sections like the one shown here). One important result of the many complex processes that take place when soils form is an increase in the amount of iron oxide mineral grains present and a concomitant enhancement of magnetic susceptibility. The person on the right, at the base of the cliff, gives the scale. Photograph taken by the author.

Thick sediment packages on the continents and their counterparts in lakes are not the only magnetic archives of past global change. There are also many important examples from marine sediments. Hesse [9] presents a particularly good record from the Tasman Sea in which he was able to identify bacterial magnetosomes of different morphologies thought to represent distinct species. Depth variations in their relative abundances are interpreted as the response to palaeoenvironmental changes, most probably caused by fluctuations in the concentration of oxygen in the deep water. Another excellent marine example [10] provides a magnetic susceptibility pattern similar to that illustrated in figure 2, with warm intervals yielding values about twice as large as the intervening cold stages.

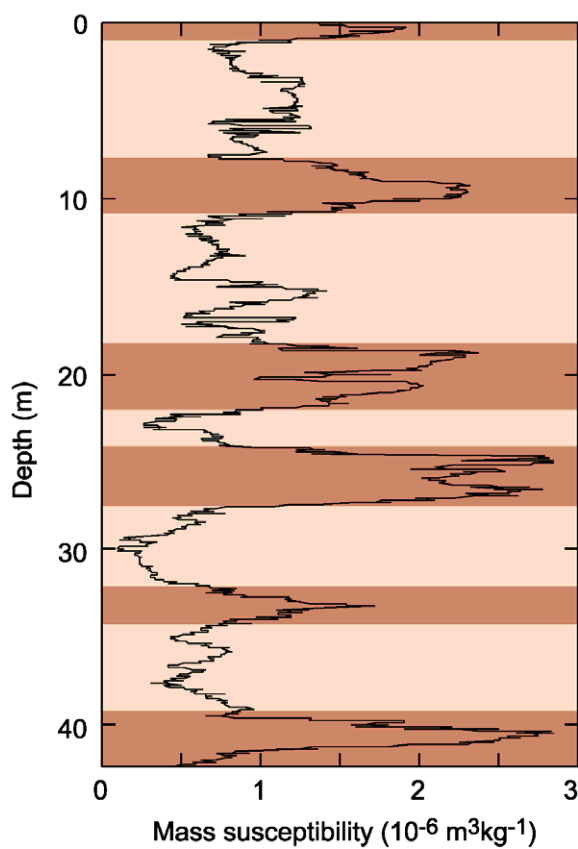


Figure 2. A detailed magnetic susceptibility profile of a stratigraphic sequence like that shown in figure 1. The peak values occur in the interglacial palaeosols (dark brown strata). Some 2000 samples were measured to construct the profile illustrated here, which represents about half a million years. Data kindly provided by Dr. S. Spassov from his doctoral project carried out at ETH Zurich under the supervision of Prof. F. Heller.

Furthermore, electron microscopy reveals the presence of chains of 20- to 200-nm magnetite particles that are so diagnostic of bacterial magnetosomes. Detailed measurements of the size and axial ratios of such grains indicate that they mostly fall in the SD field, a necessary requirement for magnetotaxis to be effective. This is clearly illustrated in figure 3 where the measurements from [10] are plotted with reference to the pseudo-single-domain (PSD)/single-domain (SD)/superparamagnetic (SP) boundaries.

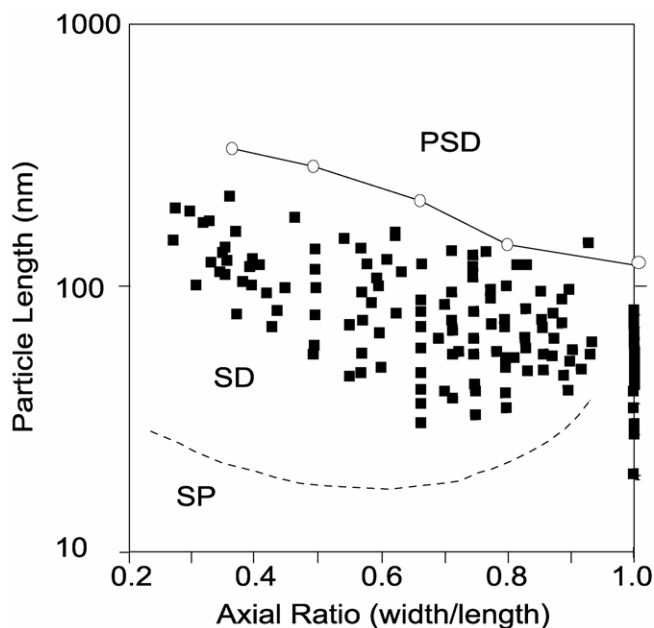


Figure 3. Size and shape distribution of magnetite magnetosomes from marine sediments collected near New Zealand [10]. The PSD/SD boundary is that obtained from a full three-dimensional micro-magnetic calculation [12].

The above discussion indicates that the use of magnetic measurements to monitor climatic change in the geological past has had some noteworthy successes. Nevertheless, in a thorough review of the underlying physics, Rancourt [11] concludes that mineral magnetometry remains underused as a means of characterising environmental samples. Perhaps the synergy resulting from conferences of this kind will help remedy this situation.

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