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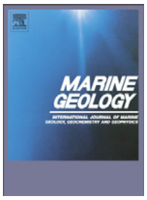

Variations in relative abundances of goethite and hematite in Bengal Fan sediments: Climatic vs. diagenetic signals

Alexandra Abrajevitch *, Rob Van der Voo, David K. Rea

Department of Geological Sciences, University of Michigan, Ann Arbor, Michigan 48109-1005, USA

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A B S T R A C T

Relative abundances of goethite and hematite in marine sediments have been increasingly used in reflectance spectrometry studies to infer the precipitation regime at the source area of detrital sediments. This interpretation is contingent on the assumption that the ratio of these minerals is not modified by diagenetic processes. Our rock-magnetic study of the ~10.5–4.0 Ma interval of the Bengal Fan sedimentary sequence indicates that this is not always the case. We demonstrate that a statistical analysis of isothermal remanent magnetization acquisition curves provides an adequate estimate of the content of goethite and hematite in sediments. We also identified distinctive biogenic and “detrital” components (likely (titano-) magnetite or maghemite) in the studied samples. The sediments of the Bengal Fan contain a significant change in relative abundance of goethite and hematite at ~7 Ma, which is approximately coeval with a postulated climatic turnover in the sediment source region. However, systematic variations in the “detrital” and biogenic components, as well as an increase in the total organic carbon content and the appearance of pyrite concretions at the same stratigraphic level, suggest that this change in magnetic mineral content reflects a change in the degree of diagenetic alteration of the initial detrital assemblage rather than a climatic signal. When assigning climatic interpretations to changes in the relative abundance of goethite and hematite in marine sediments in future studies, the possibility of diagenetic modification should be evaluated. With rock-magnetic methods, alteration of ancient sediments can be detected and the severity of alteration can be estimated based on the presence and grain-size variation of the ferrimagnetic fraction (magnetite/maghemite), which is more sensitive to early reductive dissolution than high coercivity minerals (goethite and hematite).

1. Introduction

The relative abundance of two pedogenic iron oxides goethite (G) and hematite (H) is largely controlled by moisture availability during soil formation (e.g., Yapp, 2001; Cornell and Schwertmann, 2003; Evans and Heller, 2003). Detailed studies of soil sequences showed that the G/G + H ratio does not depend on the composition of parental rocks, but that it is influenced only by climatic factors (e.g. Kampf and Schwertmann, 1983; Tardy and Roquin, 1992; Sangode and Bloemendal, 2004; and references therein). Higher average annual temperatures favor formation of hematite, whereas higher excess moisture and higher organic carbon promote goethite formation. In tropical and subtropical regions where organic carbon content and moisture availability are generally related, the relative abundance of goethite (high G/(G + H)) can be used as an indicator of higher precipitation, whereas low values imply drier/warmer conditions. The high sensitivity of the G/(G + H) ratio to precipitation is demonstrated by its correlation with topography on small (hundred meter) scales; soils with higher concentrations of hematite occur on the drier slopes and grade into goethite-rich soils in the wetter depressions (Curi and Franzmeier, 1984; Santana, 1984; da Motta and Kampf, 1992).

A similar climatic interpretation of the relative abundances of goethite and hematite has been increasingly applied to marine sediments (e.g. Harris and Mix, 1999, 2002; Clift, 2006; Zhang et al., 2007). That is, goethite vs. hematite content in the detrital fraction is thought to reflect the precipitation regime in the source area of the sediments. Marine conditions, however, impose another variable on the ratio of pedogenic minerals. Bacterially-mediated processes of suboxic diagenesis (Froelich et al., 1979) may lead to modification of the initial assemblage of detrital iron oxide grains. In reflectance spectroscopy studies of marine sediments, the possibility of the diagenetic modification of the G/(G + H) ratio was either not considered (Clift, 2006), or was inferred to be non-existent (Harris and Mix, 1999, 2002; Zhang et al., 2007). As strong evidence against dissolution, the above studies invoked an absence of a systematic down-core decline in amplitude of the variability, and the abundance, of the pedogenic iron oxide ratios.

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* Corresponding author. Center for Advanced Marine Research, Kochi University, Kochi 783-8502, Japan. Tel.: +81 090 2827 5424; fax: +81 88 864 6713.
E-mail address: alexabra@umich.edu (A. Abrajevitch).

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To better understand the applicability of the G/(G + H) parameter as a precipitation proxy in studies of marine sediments, we conducted a rock-magnetic study of the Bengal Fan sediments. The Bengal Fan is fed by the Ganges–Brahmaputra river system that carries sediments eroded from the Himalayas (Fig. 1a), including a share of sediments that had been temporarily stored on the Indo-Gangetic plain and that were chemically altered during their residence there. Various proxy records indicate that there was a significant climate change in the Himalaya–Indian Ocean region at ~7–8 Ma. The climatic turnover has been alternatively interpreted as an indication of the onset or marked strengthening of the monsoon (e.g., Quade et al., 1989; Prell et al., 1992; Ding et al., 2001), but also as marking increased aridity in the region (Derry and France-Lanord, 1996; Stern et al., 1997; Gupta et al., 2004). Both options imply a significant change, albeit of opposite sense, in the average amount of precipitation in the Himalayan Foreland. The mode of change in the relative abundances of goethite and hematite in sediments derived from the foreland basin could therefore discriminate between these end members because the G/(G + H) parameter should increase with intensification of the monsoon, and decrease with aridification.

1.1. Background: diagenetic alteration of iron oxides in marine environments

To what degree the initial detrital oxides will be affected by diagenesis depend on availability of reactive organic matter and the presence of terminal electron acceptors for anaerobic respiration. In general, more energy-efficient electron acceptors, such as oxygen, nitrate and manganese oxides are used first, followed by Fe^{3+}-oxides and sulfate (e.g. Curtis, 1983). If the amount of organic matter in the sediments is high enough, such that suboxic diagenesis involves iron or sulfate reduction, then reductive dissolution of detrital ferrimagnetic iron oxides will occur (Karlin and Levi, 1983, 1985; Canfield and Berner, 1987; Karlin, 1990a,b; Leslie et al., 1990a,b; Bloemendal et al., 1992). Fe^{2+} dissociated from detrital oxides during diagenesis is either taken up into pyrite or greigite (Canfield and Berner, 1987; Rowan et al., 2009) or diffuse along concentration gradients to positions in the section where oxygen is available in pore-waters (Colley et al., 1984; Jarvis and Higgs, 1987). There, in the oxidized zone of the sediment, biogenic and chemical precipitation of secondary oxides and hydroxides (including magnetic Fe-compounds) can occur (e.g. Lowrie and Heller, 1982, Karlin et al., 1987).

Diagenetic modification of the iron oxide content of marine sediments has been identified in regions of high organic flux, e.g., the continental shelf and slope (e.g., Richter et al., 1999; Liu et al., 2004; Kawamura et al., 2007; Rowan et al., 2009) or areas of high-productivity (e.g. Tarduno and Wilkison, 1996). In slowly accumulating pelagic sediments, the stage of Fe-oxide reduction is seldom reached (Canfield et al., 1992), except in some specific environmental contexts; for example, in turbidite-dominated abyssal plain sediments. In a study of late Cenozoic turbiditic sediments in the North Atlantic (Robinson et al., 2000), reductive/oxidative processes were found to be important only in organic-rich (>0.5% organic carbon

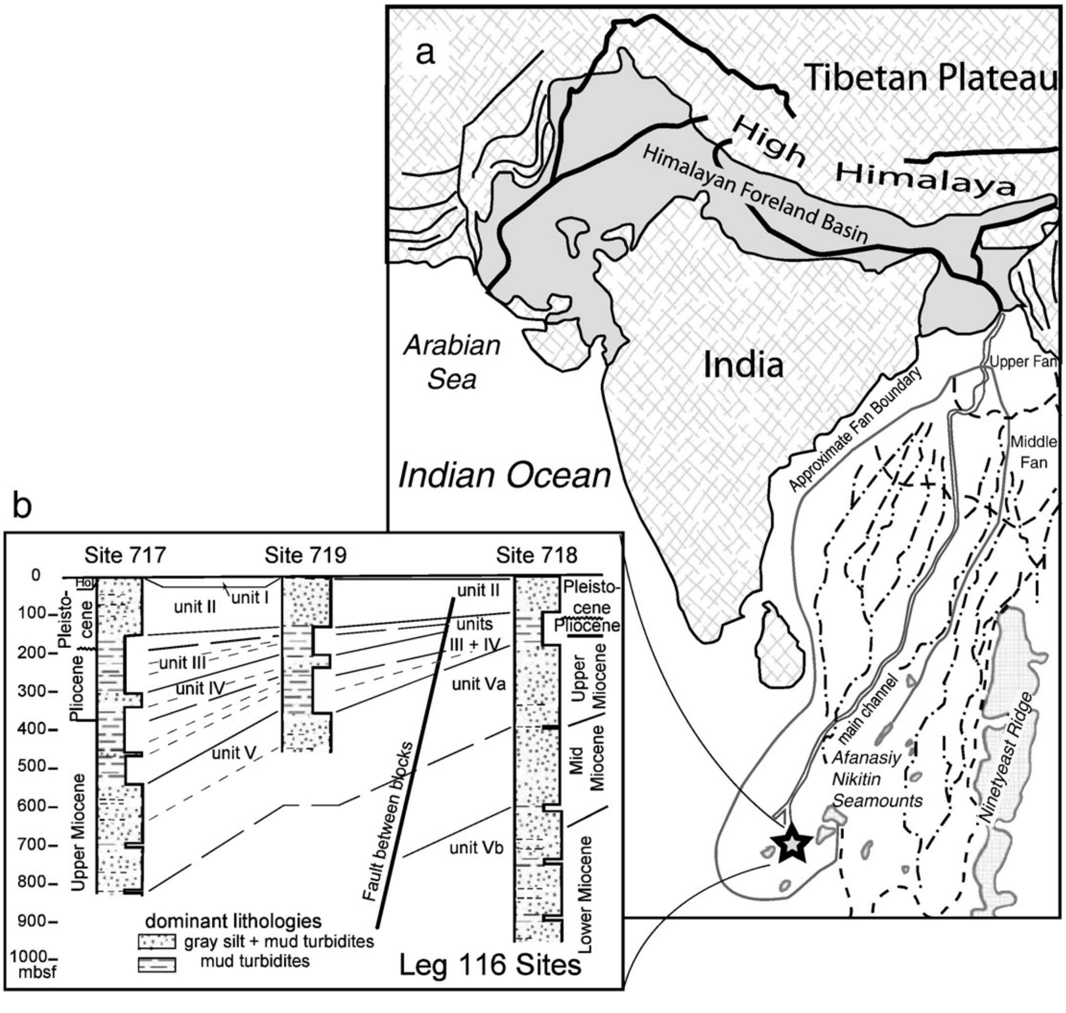

Fig. 1. a) Location map of the Himalayan Foreland and Bengal Fan with ODP Leg 116 drill site locations. Modified after Burbank et al. (1993) and Emmel and Curray (1984). b) Summary of sites 717, 718, 719 (Leg 116), with lithostratigraphic units and correlations. Modified after Stow et al. (1990).