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The MIF-S Record of Sedimentary Rocks: An Indicator of Atmospheric, Thermal, or Biological Evolution of the Earth?

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The presence of mass-independently-fractionated sulfur isotopes (MIF-S) in pre-2.4 Ga sedimentary rocks, but the general absence of MIF-S in post-2.4 Ga sedimentary rocks, has been used by many recent investigators as the most conclusive evidence for the Great Oxidation Event ∼2.4 Ga. This is because the only known mechanism to generate MIF-S signatures in nature has been the dissociation of volcanic SO_2 gas to S^0 and SO_4^{2-} in the atmosphere by UV, and the UV photolysis of SO_2 is prevented when an ozone shield exists and the atmospheric pO$_2$ is $>10^{-6}$ atm (or $>10^{-3}$ atm). However, this simple interpretation that links MIF-S to a simple atmospheric evolution model has been seriously questioned by recent discoveries of: (1) the absence of MIF-S signatures in many Archean sedimentary rocks between 3.46 Ga and 2.7 Ga in age (Watanabe et al., 2005; Peters et al., 2005; Ohmoto et al., 2006; Ono et al., 2006; Mojzsis, 2006); (2) the presence of small, but distinct MIF signatures in recent volcanic ashes (Savariano et al., 2003) and in diagenetic pyrites in a Jurassic ammonite (Ono et al., 2006); (3) the serious discrepancy between the MIF-S signatures in laboratory photochemical reaction products and those in natural samples (Ohmoto et al., 2006); (4) the presence of abnormal geochemical characteristics (e.g., very high organic C contents; submarine hydrothermal signatures) in sedimentary rocks with MIF-S signatures (Ohmoto et al., 2006); and (5) the presence of distinct MIF-S signatures (Δ^{33}S values up to +0.6‰) in the H_2S generated from thermochemical sulfate reduction by amino acids at 150˚C, and the general trend of increasing Δ^{33}S values with decreasing rates of sulfate reduction (Watanabe et al., 2006).

Considering these recent discoveries, we present the following three interpretations for the MIF-S record of sedimentary rocks: (1) the atmospheric O_2 level greatly fluctuated (i.e., “yo-yo atmosphere”) during the Archean era: anoxic before $\sim$3.5 Ga; oxic during most of the following $\sim$800 Ma period ($\sim$3.5-2.7 Ga); and anoxic during the next $\sim$200 Ma period ($\sim$2.7-2.5 Ga); (2) the atmosphere has remained oxic since $\sim$3.8 Ga, and large MIF-S signatures represent times and regions of violent volcanic eruptions that ejected large volumes of SO_2 to the stratosphere; and (3) the atmosphere has remained oxic since $\sim$3.8 Ga, and the large MIF-S signatures were created by thermochemical sulfate reduction in environments where unusually high accumulations of organic matter and extensive hydrothermal activities occurred.