Seawater-sulfate origin for volcanic SO_2 on Earth

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The presence of MIF-S, presumably created by the UV photolysis of volcanic SO_2 in an O_2-poor atmosphere, in some (though not most) Archean-aged sedimentary rocks has been the "definitive evidence" for the "Great Oxidation Event (G.O.E.) at ~2.5 Ga". Yet, apparently contradicting this "evidence", a current paradigmn states that the G.O.E. occurred due to a change from H_2S-dominant submarine volcanism to SO_2-dominant subaerial volcanism, caused by the emergence of large exposed-land-masses ~2,5 Ga ago. In order to constrain the origin of volcanic SO_2 on modern Earth and to estimate the dominant S-bearing volcanic gas on early Earth, I have carried out: (a) thermodynamic and kinetic analyses of reactions in the S-H-O-C-Fe system at T = 0-1,500 °C and P = 1- 2,000 bar; (b) analyses of literature data on the chemical and isotopic compositions ($\delta^{34}S$ and $\Delta^{33}S$) of volcanic gases, -rocks, and submarine hydrothermal deposits (e.g., VMS); and (c) investigations of the geology of Archean terranes.

Based on these investigations, I suggest the following: (1). Emissions of S-bearing gases (H_2S, SO_2, S_8, etc) into the atmosphere by submarine volcanism have been insignificant, because they all become trapped in the oceans as aqueous- and solid species. The atmospheres of ocean-covered planets, such as the pre-3.0-Ga Earth as postulated by a current paradigmn, would have been S-free. (2). Volcanic emissions of SO_2 into the Earth's atmosphere have been carried out almost entirely by subaerial volcanism associated with arc magmas, which have become oxydized and SO_4^{2-}-rich due to the contributions of seawater SO_4^{2-}. If the atmosphere had been anoxic, seawater would have been SO_4^{2-} -poor, and the volcanic gases H_2S-rich and SO_2-poor. (3). Since ~3.5 Ga ago, large exposed-land-masses have existed, and subaerial volcanism has been as important as today. (4). The fO_2 values of mantle-derived igneous rocks, as well as the mineralogy and S isotopic compositions of VMS deposits, of Archean ages suggest that the oxygenation of the atmosphere-ocean-crust-mantle systems and plate tectonic had begun by ~3.5 Ga ago; SO_2 has been the dominant volcanic gas on the oxygenated Earth. (4). The MIF-S in Archean (and younger) sedimentary rocks could not have been created by the UV photolysis of S-bearing gases; MIF-S is not evidence of an anoxic atmosphere.

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