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Episodes of massive felsic volcanism between 3.5 and 3.2 Ga deposited sulfate on early Earth

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The reports of mass-independent sulfur isotope anomalies (MIF-S) in sediments older than 2.45 Ga have been attributed to photolytic reactions involving volcanic SO_2 in an oxygen-poor atmosphere. Photolysis experiments of SO_2 coupled with various UV shielding scenarios provided additional links to the early atmosphere. However, no simple model can reproduce the mismatch in the Δ33S-δ34S relationship between the reference Archaean sulfide array (positive Δ33S-δ34S correlation) and product sulfate (negative Δ33S but positive δ34S). The discrepancy in the temporal and spatial record of sulfur isotope anomalies, with three main sulfate horizons deposited within less than 300 Ma compare to a global distribution of sulfide over more than 1,500 Ma, is also unexplained. Here we report a new Δ33S-δ34S linear trends recovered in two felsic volcanic ash layers of the 3.2 Ga Mapepe Formation in South Africa. This « felsic volcanic array » forms a tight Δ33S-δ34S linear correlation that is best approximated by SO_2 photolysis experiments at deep UV wavelength. The perfect match to the Δ33S-δ34S values of associated sulfate and equivalent felsic volcanoclastic and sulfate horizons of the 3.5 Ga old Dresser Formation, Western Australia, indicates that the exogenic sulfur cycle that produced this array was linked to felsic volcanism and sulfate precipitation. An emerging scenario for the early Earth atmosphere is a continuous photochemical haze that is perturbed between 3.5 and 3.2 Gyr by massive and optically thick volcanic plumes. This volcanic activity coincides in time with a period of massive crust formation resulting from high degrees of melt extraction from the mantle.