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Mass-independent sulfur isotopic compositions in stratospheric volcanic eruptions

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The interpretation of the observed mass-independent sulfur isotopic composition (S-MIF) in Archean rocks remains debated especially in terms of the oxygenation of the early Earth atmosphere. If the starting point of the S-MIF seems to be well-defined and linked to the photo-oxidation of SO_2 by UV light at wavelengths lower than 220 nm, the detail of the chemical steps responsible from its generation remains unknown. Such limitation prevents us a full interpretation and extraction of the information embedded in the Archean records.

It has been showed previously that S-MIF was not limited to Archean times but that modern volcanic sulfate extracted from ice core records possessed this unique sulfur isotopic composition. However, the presence of S-MIF was observed only when the explosion of the volcano had enough energy to inject the SO_2directly in the stratosphere where UV lights are available.

In order to pursue this work and better document this phenomenon, we have extracted and analyzed the two most recent stratospheric eruptions frozen in the snow of Antarctica. The sulfur isotopic composition of the volcanic sulfate from the Agung (March 1963) and Pinatubo (June 1991) eruptions were measured. Contrary to previous volcanic events analyzed, in the present study a time-dependent evolution of the sulfur isotopic composition was performed. It is observed that the sulfur isotopic anomaly of both events changes with time, moving from an initial positive component to a negative value. According to the current understanding of the transformation of SO_2 to sulfuric acid droplets in volcanic plume reaching the stratosphere, it is deduced that the S-MIF is generated on a monthly time basis. Using a 2D transport chemistry

model we demonstrate that this oscillation can be reproduced if one takes into account the microphysics of aerosol related to the condensation of H_2SO_4 gas on preexisting particles size distribution. Preferential sedimentation of larger particles seems to be a possible mechanism to physically separate the aerosols in two reservoirs with opposite MIF anomalies. Alternatively, this study demonstrates that S-MIF can be an excellent proxy of stratospheric volcanoes, opening the way for a robust evaluation of the impact of volcanoes on the natural climate variability.