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Mass-independent oxygen and sulfur isotopic composition of the Pinatubo and Agung volcanic eruptions as recorded in a Dome C snow pit.

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Massive volcanic eruptions are well-known to profoundly modify the atmospheric system by injecting large amount of dusts and gases. Among them, sulfur dioxide is of special interest as its oxidation in the stratosphere leads to the formation of a sulfuric acid layer that changes the radiative properties of the atmosphere. Snow and ice cores provide a unique opportunity to study the chemical and climatic consequence of such brutal events.

We used the mass-independent oxygen and sulfur isotopic composition of volcanic sulfates to characterize their isotopic response and understand the underlying chemistry. An oxygen or sulfur isotopic composition which deviates from the mass-dependent relationships ($\delta^{17}O \approx 0{,}52\ \delta^{18}O$ and $\delta^{33}S \approx 0{,}515\ \delta^{34}S$, $\delta^{36}S \approx 1{,}91\ \delta^{34}S$ for oxygen and sulfur respectively) is termed mass-independent and is quantified by the relations : $\Delta^{33}S = \delta^{33}S - 1000*[(1+\delta^{34}S/1000)^{0,515}-1]$, $\Delta^{36}S = \delta^{36}S - 1000*[(1+\delta^{34}S/1000)^{1,91}-1]$ and $\Delta^{17}O \approx \delta^{17}O - 0{,}52\delta^{18}O$. The oxygen isotopic anomaly in sulfates originates from the incorporation of anomalous oxidative species (i.e. O_3, H_2O_2 or OH radicals) into SO_2during its oxidation in the troposphere or stratosphere. .On the other hand, the sulfur isotopic anomaly is produced by UV photolysis of SO_2 below 310 nm, a photon energy only available in the stratosphere

We studied the two most important stratospheric events of the last 50 years, the Agung (8˚S, 115˚E, March 1963) and the Pinatubo (15˚N, 120˚E, June 1991) eruptions which

injected more than 10 Tg of SO_2into the stratosphere. The volcanic sulfate recorded in a Dome C (75˚S, 123˚E) snow pit allowed a fine sampling time resolution of the sulfur and oxygen isotopic composition, with approximatively one measurement every 6 months subsequent to the eruptions.

The sulfur isotopes results are similar for the 2 volcanic eruptions: we observed a change in sign with time, starting with a positive phase. This oscillation in Δ^{33}S is paralleled by a variation of the δ^{34}S isotopic composition of the volcanic sulfate. The oxygen isotopic anomaly variation is more significant for the Agung eruption than the Pinatubo eruption, as a possible result of the superimposition of the Cerro Hudson (46˚S, 73˚W, August 1991) event with the Pinatubo. The Cerro Hudson was a high tropospheric, low stratospheric eruption that may have formed its sulfate aerosols in a different chemical context. However, in both case, the maximum sulfur isotopic anomaly got ahead of the maximum oxygen isotopic anomaly suggesting that the processes involved in the mass-independent fractionation formation of these two elements do not occur at the same time and space.

As a result of the study of sulfur and oxygen mass-independent fractionation in volcanic sulfates, we show for the first time, the time-dependency of isotopic anomalies and as a consequence, we demonstrate how the sampling step is important in the interpretation of the isotope data. The reproducibility of such isotopic anomaly signatures for two stratospheric events also shows that we have a reliable tracer to assess the impact of volcanic eruption on the atmospheric system.