Modelling the tropospheric and stratospheric sulfur isotopes in a column model for volcanically quiescent periods

Juhi Nagori¹, Narcisa Nechita-Bândă¹, Masumi Shinkai², Sebastian Danielache², Thomas Röckmann¹, and Maarten Krol¹,³

¹Utrecht University, Institute of Marine and Atmospheric Research (IMAU), Physics, Utrecht, Netherlands (j.v.nagori@uu.nl)
²Sophia University, Faculty of Science and Technology Department of Materials and Life Science, Tokyo, Japan
³Wageningen University, Meteorology and Air Quality (MAQ), Environmental Sciences, Wageningen, Netherlands

It is debated how much stratospheric sulfate aerosol (SSA) in volcanically quiescent times is replenished by carbonyl sulfide (COS) oxidation products. The atmospheric COS budget is also currently uncertain, with missing sources and sinks. Isotopic analysis can be used to allocate the missing sources of COS and also to further constrain the relevance of COS to SSA. The measured tropospheric isotopic signature of COS (δ³⁴S) ranges from 10-14 ‰ (Kamezaki et al., 2019; Angert et al., 2019; Hattori et al., 2020; Davidson et al., 2020), whereas SSA δ³⁴S is constrained by only one single measurement at 18 km of 2.6 ‰ (Castleman, 1974). We use an atmospheric column model to constrain the COS isotopic budget and understand the contribution of COS to sulfate. We find that the COS tropospheric signal is determined by the signatures of its precursors (carbon disulfide, CS₂, and dimethyl sulfide, DMS) and fractionation during plant uptake and oxidation. Photolysis of COS is important in the stratosphere; the isotopic signal of COS propagates through sulfur dioxide (SO₂) to sulfate in the stratosphere. The model can reproduce δ³⁴S between 1-5 ‰ in the lower stratosphere, which encapsulates the observations from Castleman (1974).

References