

Atmos. Chem. Phys. Discuss., referee comment RC2
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Comment on acp-2022-68, approach not suitable to obtain new findings

Anonymous Referee #2

Referee comment on "Modelling the atmospheric ^{34}S -sulfur budget in a column model under volcanically quiescent conditions" by Juhi Nagori et al., Atmos. Chem. Phys. Discuss., <https://doi.org/10.5194/acp-2022-68-RC2>, 2022

General comments

The paper tries to use S-isotopes to improve the understanding of stratospheric and tropospheric sulfur budgets. However, the used one-dimensional global average model contains too many simplifications and too much tuning to provide new findings compared to existing literature. It contains, except for better assumptions for the isotopic signature of CS_2 , almost the same flaws as the version submitted in 2020. Another problem is that it ignores existing satellite observations for SO_2 and COS. The paper requires substantial improvements in the text and the model setup including new simulations to be publishable.

The calculation of photolysis rates uses an extremely high spectral resolution, maybe to avoid parameterisations in the Schumann-Runge bands, which play only a minor role, or to account for the redshift in the isotopic bands of COS (Hattori et al, 2011), but ignores Rayleigh scattering which has a large impact on the altitude and wavelength dependent photon fluxes. Replacing a diurnal average of photolysis rates by using a calculation with an average zenith angle screws up the altitude dependence. Errors due to these oversimplifications are much larger than the ones introduced by a coarser spectral resolution, also with consequences for the fractionation by photolysis.

Observations of COS by Montzka et al (2007) show that there are large seasonal and spatial effects on the mixing ratios related to uptake by vegetation and oceanic and anthropogenic sources which cannot be treated with a global average model. A twodimensional model is insufficient here too but this would at least simulate the vertical and meridional transport processes better.

Specific comments, in addition to RC1

Page 2, line 16f: Here papers on the ATAL (Asian Tropopause Aerosol Layer) and SO_2 transport by the summer monsoon might be mentioned but Kjellström (1997) is outdated.

Page 3, line 25: Use "photochemistry" since reaction with $\text{O}(^3\text{P})$ is also important here.

Page 5, line 3ff: It leads to inconsistent results if prescribed oxidants and radicals estimated for midlatitudes are used for tropical processes. (see also page 6, top).
Page 6, line 7ff: These oversimplifications lead to large errors. There are plenty of efficient models available to do it better. From S gases only SO₂ can have an impact on attenuation, and this only in case of a major volcanic eruption.
Page 6, line 20ff: This fine spectral resolution is not justified over the whole range. Most relevant is the "stratospheric window", about 200 to 220nm. Which solar spectra are used (reference)?
Table 2: DMS oxidation should be treated with the individual steps, as done for other species. MSA formation is important.
Table 3: I suppose you mean O(³P) with O. That can be formed also by visible radiation. Where is O(¹D)? For H₂SO₄ photolysis in the visible (Vaida et al, 2003) should not be ignored. It is needed to explain observations.
Page 9, line 7: It is odd to assume a single particle size for aerosol, which is too big for the stratosphere and inconsistent with observations, and to use this for a crude parameterization of sedimentation.
Section 3.1: Here comparisons to MIPAS observations should be included (Höpfner et al., 2013, 2015, Glatthor et al. 2017)
Page 18, line 6f: address also enrichment by the O(³P) reaction.

Technical corrections

The abbreviation "SSA" is used also for "single scattering albedo" and other things in the community, better avoid this.
The reference Hattori et al. 2020 is incomplete.
Text and Figure S2 are inconsistent, there must be something wrong with the x-axis.
To show processes covering 4 orders of magnitude in a linear plot like Figure S7 makes no sense.

Additional references

Glatthor, N., et al. (2017), Global carbonyl sulfide (OCS) measured by MIPAS/Envisat during 2002–2012, *Atmos. Chem. Phys.*, 17, 2631–2652, doi:10.5194/acp-17-2631-2017.
Höpfner, M., et al. (2013), Sulphur dioxide (SO₂) as observed by MIPAS/Envisat: Temporal development and spatial distribution at 15–45 km altitude, *Atmos. Chem. Phys.*, 13, 10,405–10,423, doi:10.5194/acp-13-10405-2013.
Höpfner, M., et al. (2015), Sulfur dioxide (SO₂) from MIPAS in the upper troposphere and lower stratosphere 2002–2012, *Atmos. Chem. Phys.*, 15, 7017–7037, doi:10.5194/acp-15-7017-2015.
Vaida, V., H. G. Kjaergaard, P. E. Hintze, and D. J. Donaldson (2003), Photolysis of sulfuric acid vapor by visible solar radiation, *Science*, 299, 1566–1568.