

We thank the reviewer for their valuable feedback on our paper. We respond here to some of the larger comments the reviewer has detailed in their review.

Our model is a first attempt at fully incorporating the isotope scheme in a 1D model, with all the S-molecules included. This an important step before incorporating the isotope scheme in a 3D model, in which the spatial and regional effects can be better scrutinized. We feel that this effort is not valued enough by this reviewer.

The spectral resolution used in the model is a key feature to model photochemically-induced isotopic effects. In this study the Schumann-Runge bands are not central except when strong contributions from stratospheric SO<sub>2</sub> photolysis by volcanic emissions are considered. The reviewer quite correctly points out that the spectral resolution is needed to calculate isotopic imprint during photolysis created by the red shift in the spectra. The spectral features of the solar flux are also accounted by the spectral resolution implemented in the model and we can explain this better in a future version of the manuscript.

The Rayleigh scattering is a feature that was not considered relevant and it could be added to the opacity term in eq. 4 (in the manuscript). However, there are two main features that render the scattering effect irrelevant at the stratospheric altitudes at which we evaluate photolysis.

First, the effect of Rayleigh scattering on the COS photolysis rate. The absorption cross section of COS at 224—225 nm (peak) is  $3 \times 10^{-19} \text{ cm}^2$ . At the same wavelength the Rayleigh scattering cross section is  $1.515\text{-}1.259 \times 10^{-25} \text{ cm}^2$  (Bucholtz, 1995). For comparison, the cross section of O<sub>2</sub> at the same wavelength is  $0.5 \times 10^{-23} \text{ cm}^2$ . There is a 6 orders of magnitude difference between the Rayleigh and the COS spectrum and 2 orders of magnitude than the O<sub>2</sub> Herzberg continuum (which is accounted for in eq. 4 opacity term). Hence, we do not expect for the Rayleigh scattering to be of any significance to the stratospheric COS photolysis.

Second, any molecule absorbing or scattering with a cross-section several orders of magnitude smaller the absorption cross-section of the molecule in question, could potentially inflict a significant isotopic effect. This only happens if the spectrum (Rayleigh scattering in this case) has a pronounced spectral structure (just like the Schumann-Runge bands). However, the Rayleigh scattering spectrum only shows very small changes over the wavelengths that overlap with the COS absorption cross-sections.

To verify the neglect of Rayleigh scattering on the photolysis, we also looked at the actinic flux in the Tropospheric Ultraviolet and Visible (TUV) radiation model (NCAR, 2022) and especially the difference between the direct flux and diffuse radiation between 200—300 nm between altitudes of 10—40 km. These ranges were looked at since we are concerned about the stratospheric radiation and photolysis of COS, which is significant only above 15 km. Above 40 km no COS is left to consume (See Figure 1 in manuscript). The wavelength range contains the COS absorption peak (224—225 nm). Below 200 nm oxygen and ozone absorption dominate (Molina & Molina, 1986).

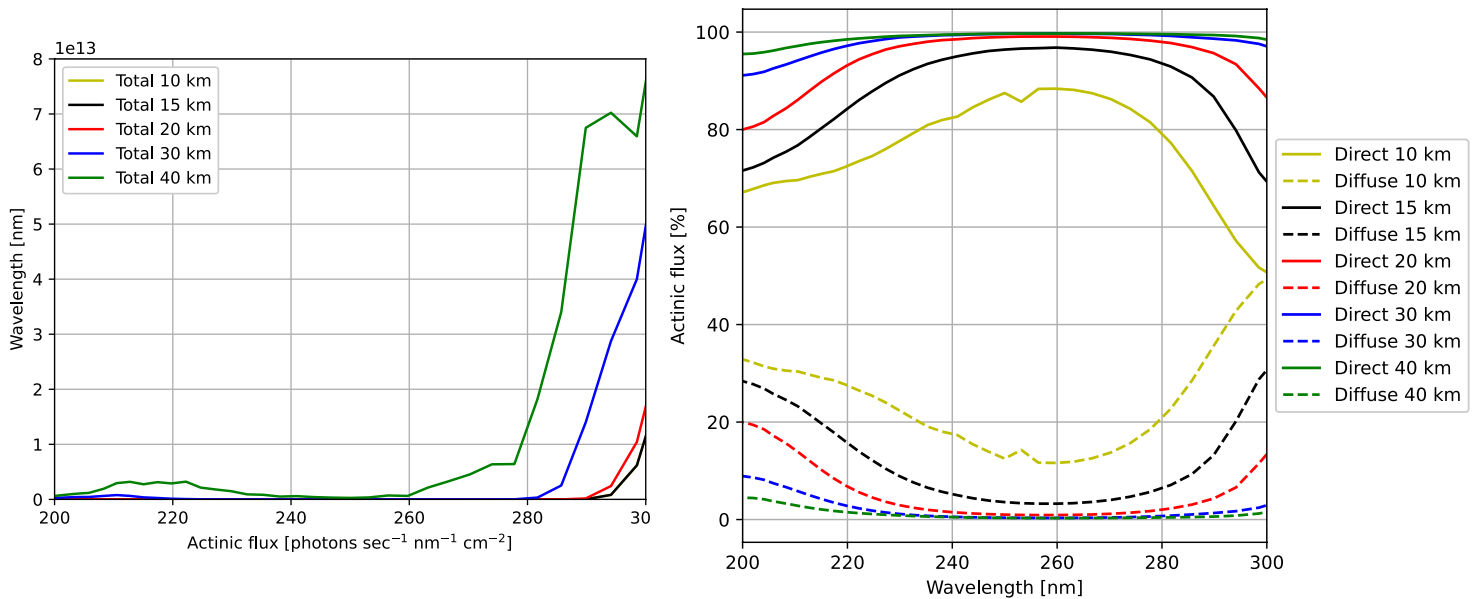


Figure 1: Actinic flux calculated (left) between 10–40 km between 200–300 nm where COS photolysis takes place. (Right) Percentage of this actinic flux that is direct (solid lines) versus diffuse (dashed lines) radiation between 200–300 for 5 different heights shown in different colours.

First, the actinic flux is much higher above 280 nm as most of the absorption takes place at shorter wavelengths. We can see that at 30 km (where COS is efficiently removed) the direct radiation dominates ( $\sim 98\%$ ) the diffuse radiation. This is what led us to not include Rayleigh scattering in our model, as we are mostly concerned with photolysis of COS in the stratosphere as it is considered the major source of SSA formation.

The reviewer also mentions that "Replacing a diurnal average of photolysis rates by using a calculation with an average zenith angle screws up the altitude dependence", yet we are able to reproduce the COS mixing ratio with altitude, though we agree that can better show this by using COS satellite observations.

Thus, in order to further improve the manuscript, we also will add observations that exist for COS mole fractions. For COS profiles, there are satellite observations like the Atmospheric Chemistry Experiment Fourier Transform Spectrometer (ACE-FTS) and the Michelson Interferometer for Passive Atmospheric Sounding (MIPAS). Though the shape of the COS profile looks similar to satellite observations (see: Ma et al., 2021), we would include the MIPAS observations since we are concerned about reproducing the upwelling of COS to the stratosphere in the tropics in order to achieve the  $40 \text{ Gg S yr}^{-1}$  stratospheric loss of COS (Glatthor et al., 2017, Ma et al., 2021). ACE-FTS, operating in solar occultation, has less sensitivity to the tropics.

To conclude, we agree with some of the more substantive remarks of this reviewer. However, the reviewer clearly undervalues some of the novelty of the paper and/or misjudges the

scrutiny we attempted in writing the manuscript. We think that – with the additions we propose above – the paper provides enough interesting material for the scientific community.

### References

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