

The contribution of tropospheric SO₂ to the background stratospheric sulfur budget in the Asian summer monsoon season

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Motivation

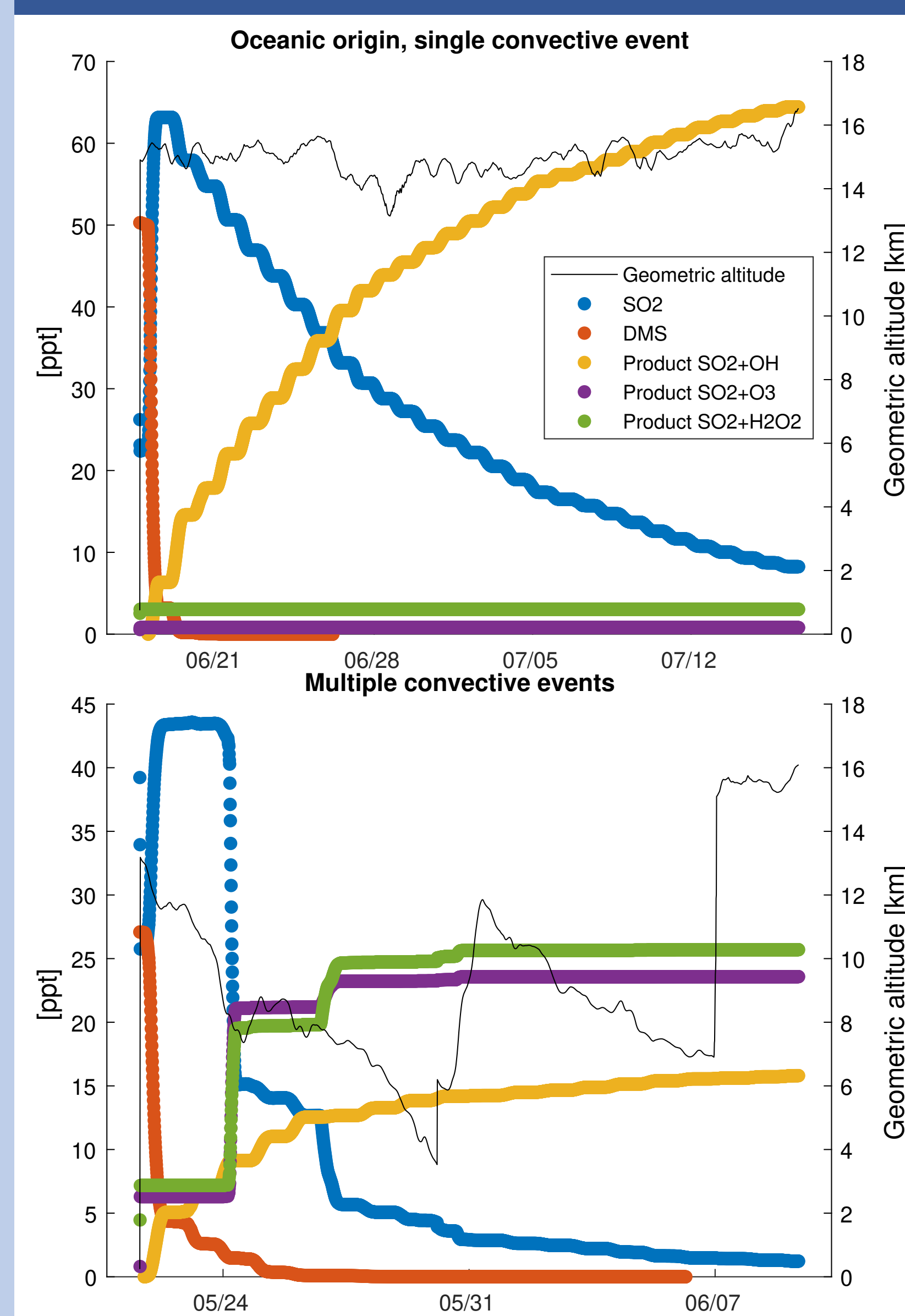
- ▶ The stratospheric aerosol layer is important for the radiative balance of the Earth, plays a significant role in stratospheric chemistry and has been proposed as a target for geo-engineering.
- ▶ In contrast, many processes related to the aerosol layer are not well understood, including the contribution of tropospheric SO₂ to the aerosol layer in volcanically quiescent periods.

Model case study

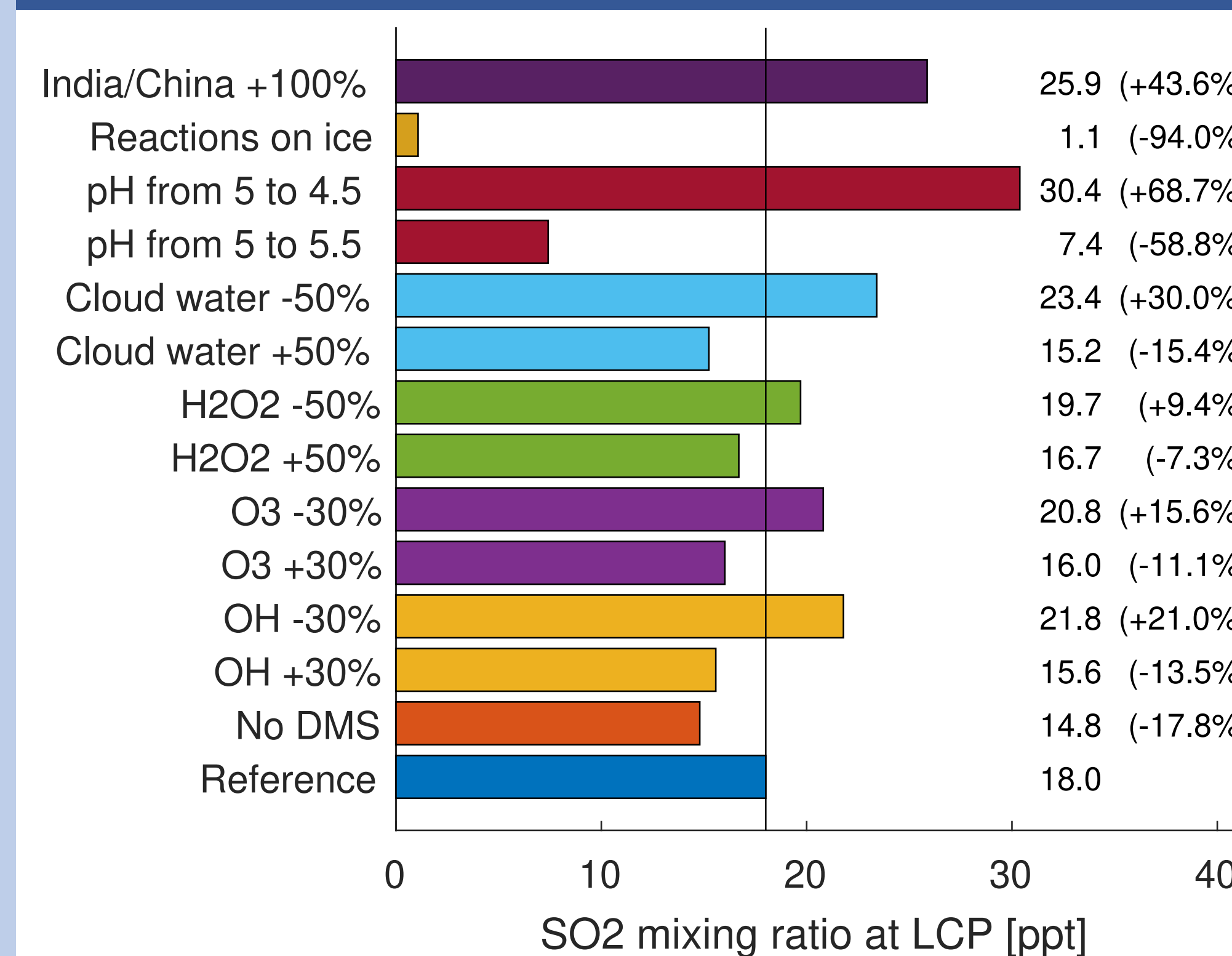
- ▶ Transport and chemistry of SO₂ in the troposphere by running a chemical box model on a high number of trajectories ascending from the boundary layer to the tropopause.
- ▶ The trajectory model includes a detailed treatment of convection for individual trajectories.
- ▶ Case study for the time period May–August 2009 in the monsoon season.

Model

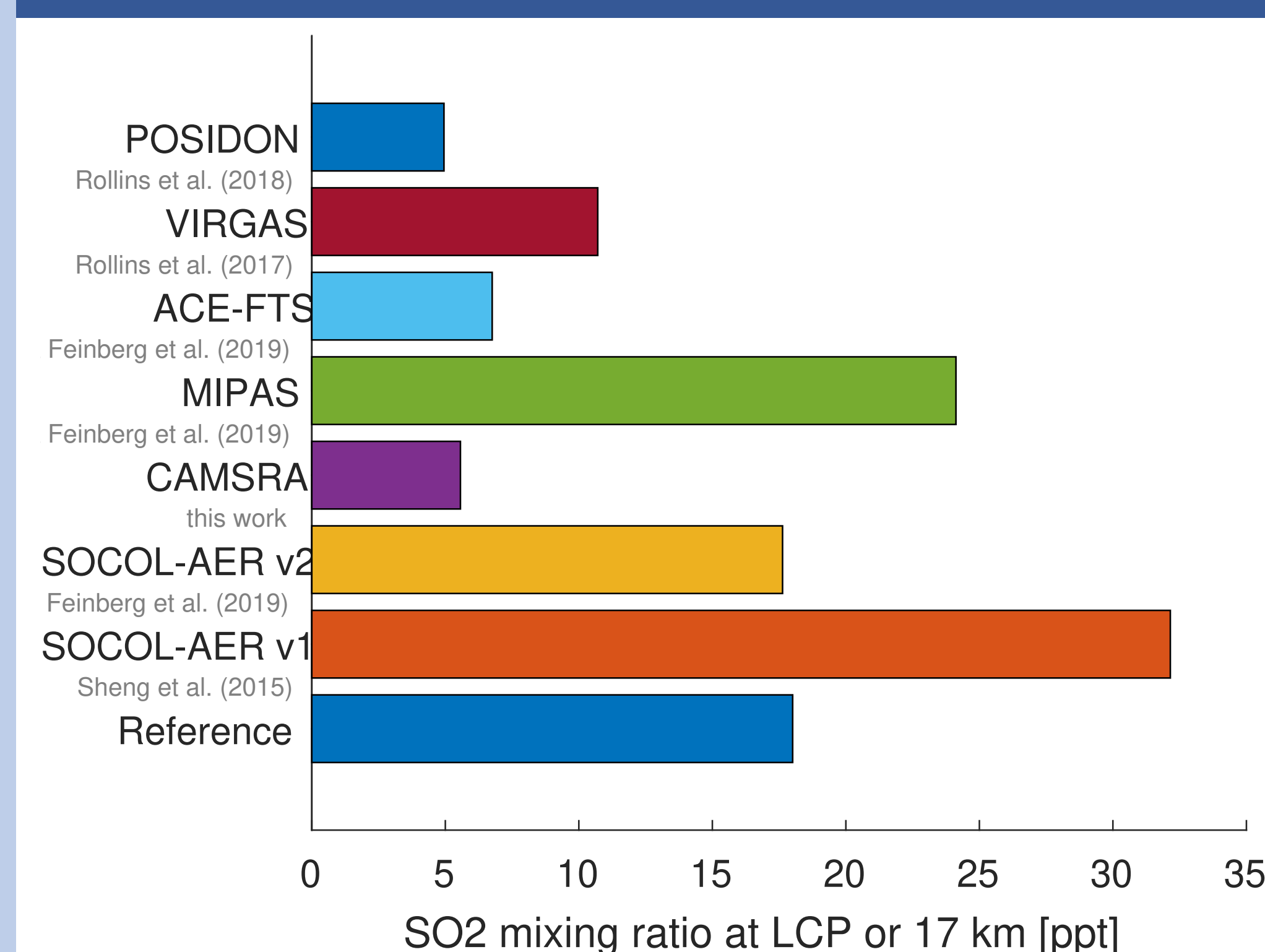
- ▶ Trajectory model of the ATLAS chemistry and transport model.
- ▶ Detailed convection model for individual trajectories developed specifically for this study (Wohltmann et al., 2019)
- ▶ Driven by meteorological data from the ECMWF ERA5 reanalysis (including convective mass fluxes and detrainment).
- ▶ 5580 backward trajectories started at 400 K potential temperature in tropics on regular grid. Start date 31 August 2009, duration 4 months. Parts between Local Cold Point (tropopause) and 80 % surface pressure (boundary layer) used for chemistry.
- ▶ Chemical box model includes only a small set of reactions directly relevant to SO₂ chemistry: SO₂+OH and DMS+OH=SO₂ in the gas phase and S(IV)+O₃ and S(IV)+H₂O₂ in the aqueous phase.
- ▶ Initialization at boundary layer (SO₂, DMS) and fixed background fields of OH, O₃, H₂O₂ from ECMWF global chemical composition reanalysis CAMSRA.
- ▶ Avoids the large computational overhead of a full tropospheric chemistry model and is justified by the small feedback that SO₂ has on OH or O₃. Allows for a high number of sensitivity runs.



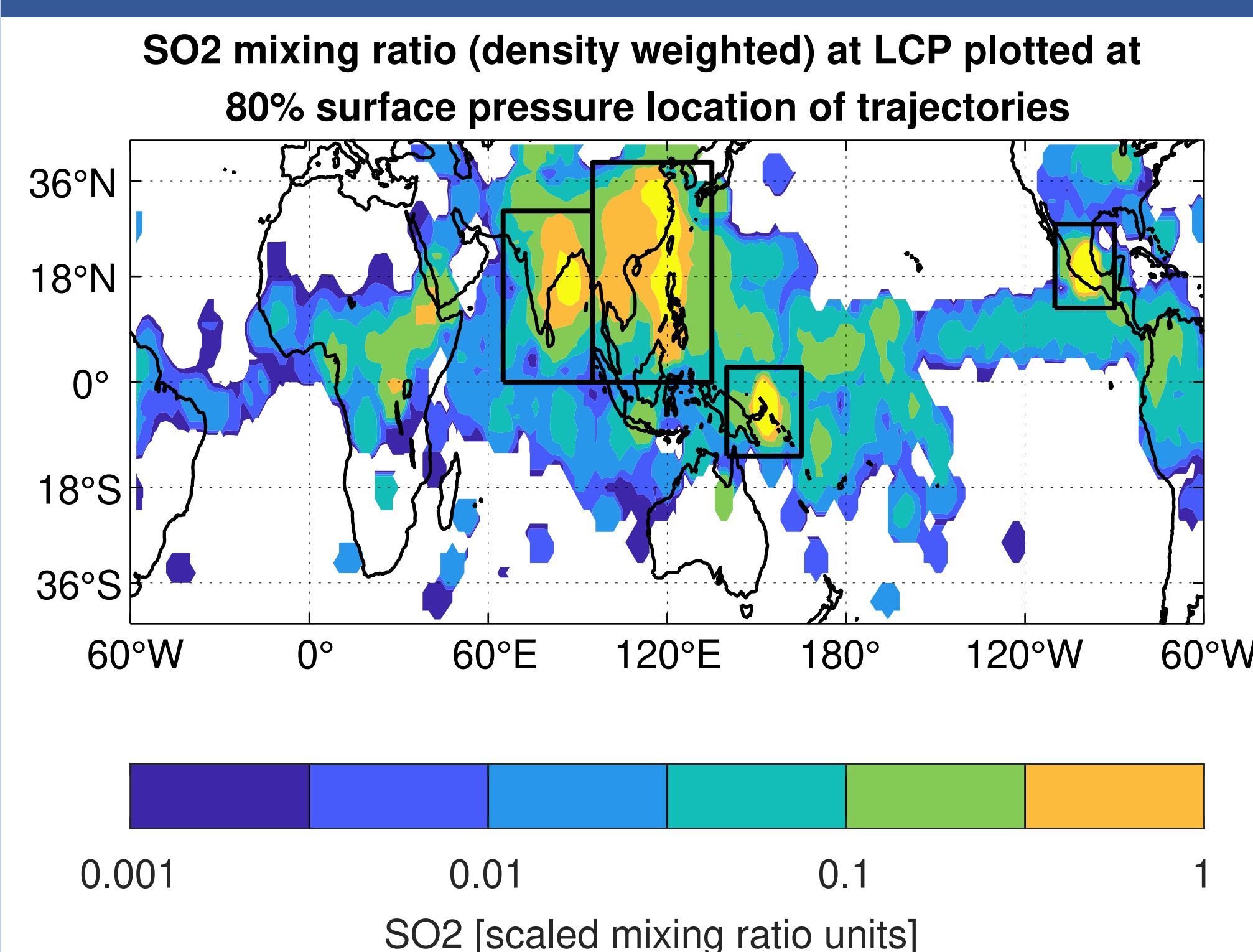
Example trajectories.



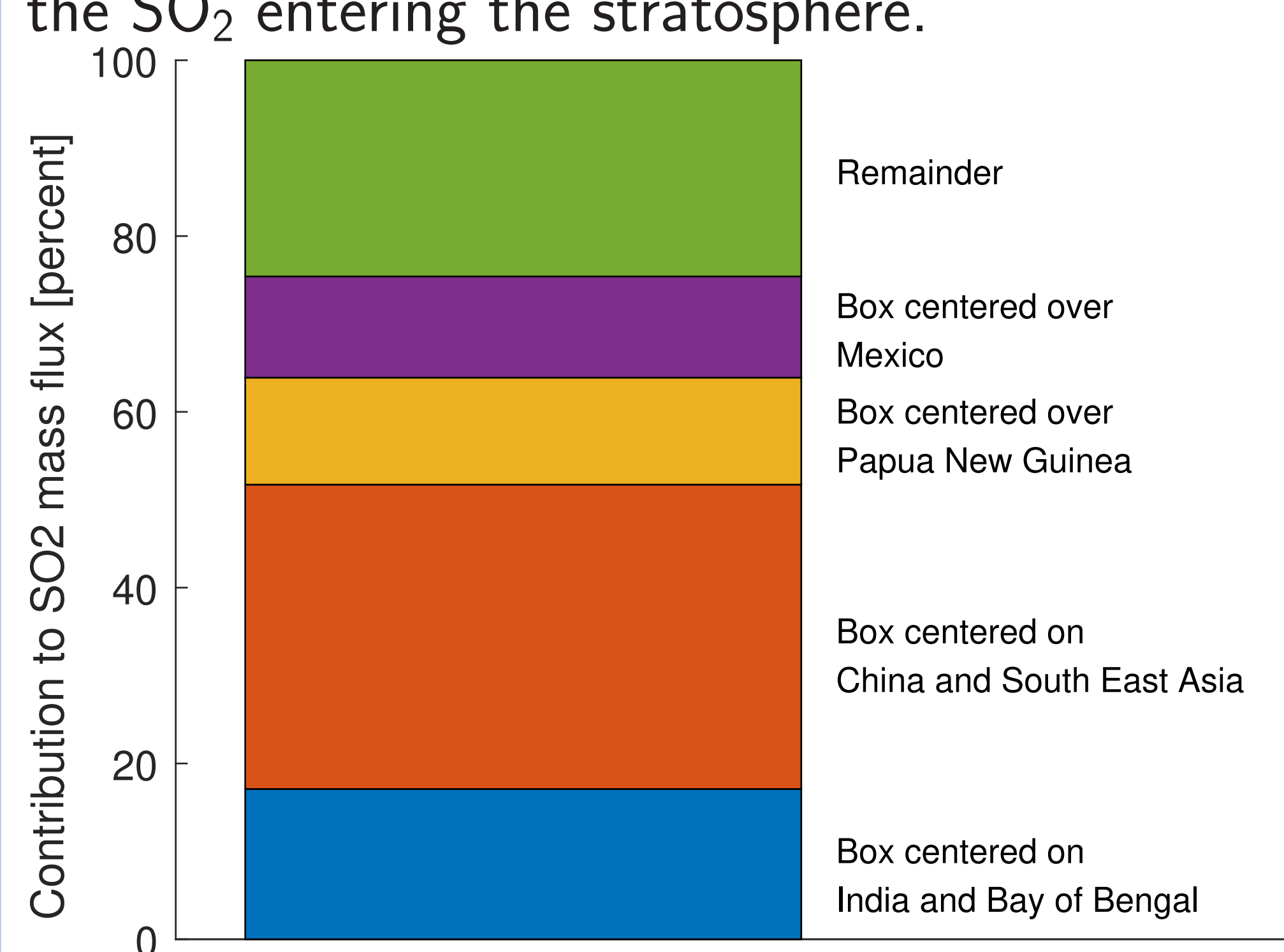
Mean SO₂ volume mixing ratio at the LCP for the reference run and the sensitivity runs. Backward trajectories that do not reach 80 % surface pressure do contribute zero values. Notes: First run increases emissions. Second run includes reactions on ice clouds in addition to water clouds (experimental).



Comparison of SO₂ volume mixing ratios to observations (POSIDON, VIRGAS: in situ, ACE-FTS, MIPAS: satellite) and modelling studies (CAMSRA, SOCOL-AER). References: See Nalapalu et al. below.



SO₂ volume mixing ratios in the reference run at the LCP (tropopause), but plotted at the location of the trajectories at 80 % surface pressure. Weighted by the number of trajectories within a radius of 500 km to give values proportional to percentage contributions (but does not show actual mixing ratios). Gives an impression of the source regions of the SO₂ entering the stratosphere.



Contribution of the SO₂ mass fluxes from the regions indicated by boxes in figure above to the total SO₂ mass flux. China, India: Mainly anthropogenic, Papua, Mexico: Outgassing volcanoes (Bagana, Popocatepetl).

Results

- ▶ About 50 % of the SO₂ entering the stratosphere originates from anthropogenic sources in China, India and South East Asia in our case study.
- ▶ Up to about 25 % of the SO₂ entering the stratosphere originates from permanently outgassing volcanoes.
- ▶ Large uncertainties in the SO₂ entering the stratosphere are associated with cloud pH values and changing emissions in China and India.

Reference

- ▶ Nalapalu et al., 2026, J. Geophys. Res. preprint, doi:10.22541/essoar.15001905/v1
- ▶ Wohltmann et al., 2019, Geosci. Model. Dev., doi:10.5194/gmd-12-4387-2019