Model calculations of the contribution of tropospheric SO_2 to the stratospheric sulfur budget

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Questions:

- How much sulfur from tropospheric sources of SO_2 and DMS (dimethyl sulfide) reaches the stratosphere?
- Where does it come from?
- What is the contribution of the Asian monsoon ("smoke stack" vs. "purifier")?

"Smoke stack":
- Over strong sources of SO_2
- Fast upward transport (convection)

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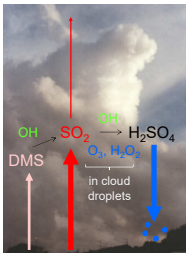
"Purifier":
- Strong precipitation → Wet deposition

Model:

Lagrangian model with SO_2 chemistry

- Backward trajectories (≤ 4 months) starting at 400 K in [30°N, 30°S], ending at 0.8 · p_s (p_s = surface pressure):
- Wind fields and cloud water content ← ERA-Interim
- Convection: trajectories are displaced vertically with a certain probability
  ← ERA-Interim convective entrainment rates, mass flux, and detrainment rates
- Chemistry is run forward in time on the trajectories
- Chemical scheme: cf. figure on right-hand side
- Initial SO_2 and DMS (at 0.8 · p_s) ← GEOS-Chem
- O_3, OH, H_2O_2 ← GEOS-Chem

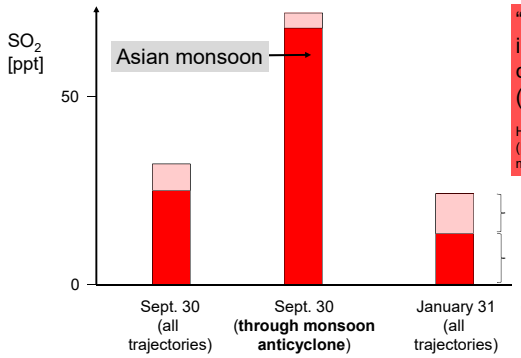


Results:

- Mean SO_2 ≈ 30 ppt at Lagrangian cold points (LCP)
- Mean SO_2 ≈ 70 ppt at Lagrangian cold points of trajectories passing through the monsoon anticyclone
- Larger contribution of anthropogenic sources in northern hemispheric summer
  ← convection closer to sources of pollution
- Significant contribution to SO_2 at LCP by degassing volcanoes in Papua New Guinea (red circle in plots below)

How much SO_2 is in air travelling through the monsoon anticyclon?

SO_2 at Lagrangian cold point of trajectories arriving at 400 K, [30°N, 30°S] on indicated date:



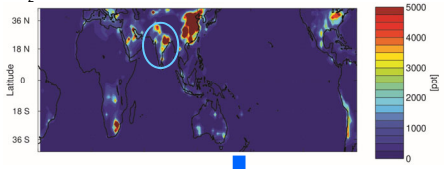
"Smoke stack" effect dominates in model simulations despite counter-acting wet deposition (cf. below).

However, only ≤ 7% of the air in the tropical pipe ([15°S, 15°N], 400 K) travelled through the monsoon anticyclone (Ploeger et al., ACP 2017, Fig. 2).

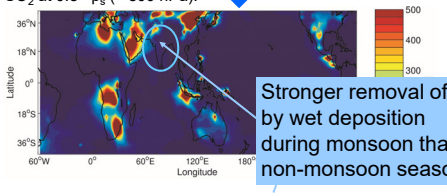
Where does the SO_2 come from?

Sources

SO_2 near the surface:



SO_2 at 0.8 · p_s (≈ 800 hPa):



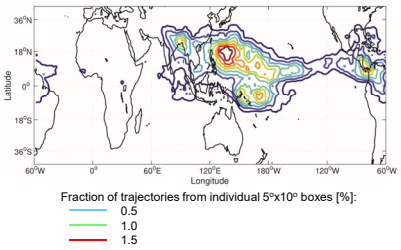
Stronger removal of SO_2 by wet deposition during monsoon than in non-monsoon season

Transport

Northern hemispheric summer

(September 2009)

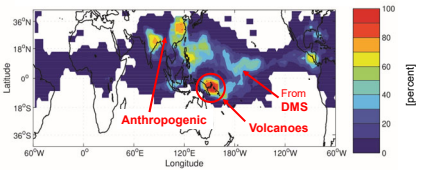
Origin of trajectories at 0.8 · p_s (≈ 800 hPa) (arriving at 400 K, [30°N, 30°S] on September 30):



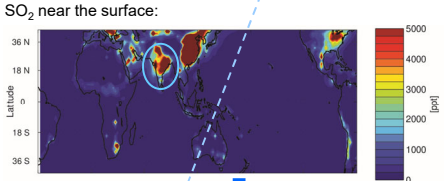
Fraction of trajectories from individual 5°x10° boxes [%]:
0.5
1.0
1.5

SO_2 arriving at LCP

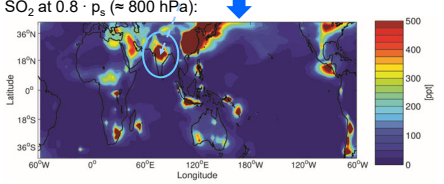
Contribution to mean SO_2 (from SO_2 and DMS sources) arriving at LCPs, plotted at position of trajectories at 0.8 · p_s (percentiles):



SO_2 near the surface:



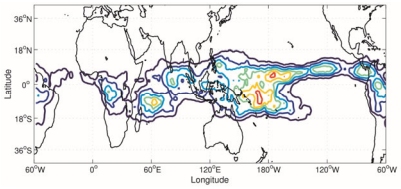
SO_2 at 0.8 · p_s (≈ 800 hPa):



Northern hemispheric winter

(January 2010)

Origin of trajectories at 0.8 · p_s (≈ 800 hPa) (arriving at 400 K, [30°N, 30°S] on January 31):



Fraction of trajectories from individual 5°x10° boxes [%]:
0.5
1.
1.5

Contribution to mean SO_2 (from SO_2 and DMS sources) arriving at LCPs, plotted at position of trajectories at 0.8 · p_s (percentiles):