

Model calculations of the contribution of SO₂ to the stratospheric aerosol layer

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Motivation

Many processes in the stratospheric aerosol layer are not well known

- E.g. Contribution of tropospheric species like COS and SO₂ to stratospheric aerosol layer poorly quantified

Approach

- Chemical box model on backward trajectories
- Examine chemistry of SO_2 and its transport to the stratosphere
- Numerous sensitivity runs to assess range of uncertainty

Model I: Transport

- Backward trajectories from the ATLAS model
- Driven by GEOS-5 analysis data
- Start at 400 K between 30° N/S on 2° x 2° grid
- Start on 31 Jan 2010 back for 4 months
- Only trajectory parts between 800 hPa and the Local Cold Point are used in the chemistry calculations

Model II: Chemistry

Reactions:

- $\text{SO}_2 + \text{OH} + \text{M} \rightarrow \text{Products}$ (gas phase)
uptake of SO_2 into liquid phase is considered
- $\text{DMS} + \text{OH} \rightarrow \text{SO}_2 + \text{Products}$ (gas phase)
Two reaction pathways (addition, abstraction)
- $\text{SO}_2 \cdot \text{H}_2\text{O} + \text{H}_2\text{O}_2 \rightarrow \text{Products}$ (liquid phase)
- $\text{S(IV)} + \text{O}_3 \rightarrow \text{Products}$ (liquid phase)
 $\text{S(IV)} = \text{HSO}_3^- + \text{SO}_2 \cdot \text{H}_2\text{O}$

plus Henry constants for SO_2 , O_3 , H_2O_2 and equilibrium constant between HSO_3^- and $\text{SO}_2 \cdot \text{H}_2\text{O}$

Model III: Chemistry

Initial values for SO_2 and DMS at 800 hPa from GEOS-Chem CTM

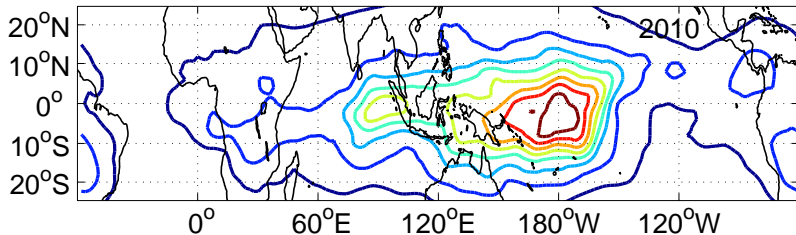
Precalculated background fields taken from the GEOS-Chem CTM
(not interactive):

- OH
- H_2O_2 (only outside cloud)
- O_3

Model IV: Clouds

- Cloud water from GEOS-Chem CTM
- Mixing ratios of SO_2 and H_2O_2 calculated separately inside and outside cloud
- SO_2 inside and outside cloud nudged to mean SO_2 value with a time constant of 1 h
- H_2O_2 inside cloud nudged to outside cloud H_2O_2 climatology from GEOS-Chem with a time constant of 1 h
- Mixing ratios of SO_2 and H_2O_2 are corrected for changing cloud size in mass-conserving way
- Cloud pH is 4.5
- Parameterization for unresolved convection not implemented so far (work in progress)

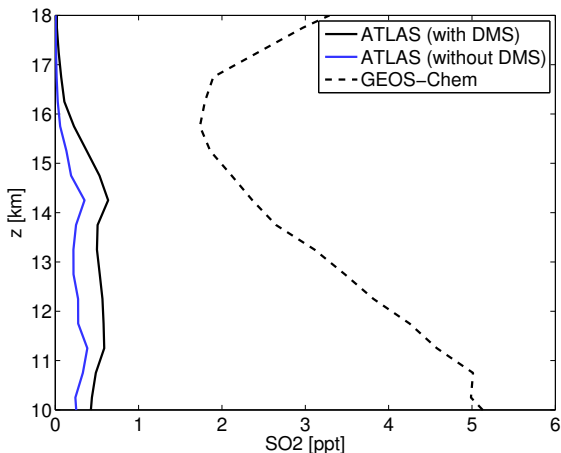
Which airmasses do we see?



Density of all trajectory points between 800 hPa and LCP
(over all altitudes, contours: factor 1–8 relative to uniform contribution)

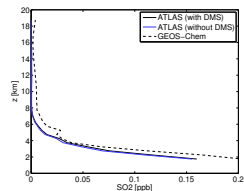
Next plots: Means over all trajectory points (as function of z)

Mean SO₂: Sensitivity to DMS and Comparison with GEOS-Chem

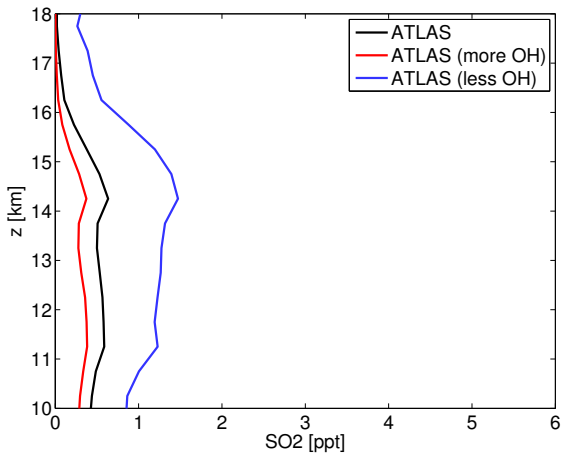


Ref (black)
No DMS (blue)

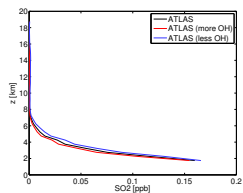
GEOS-Chem
(dashed)



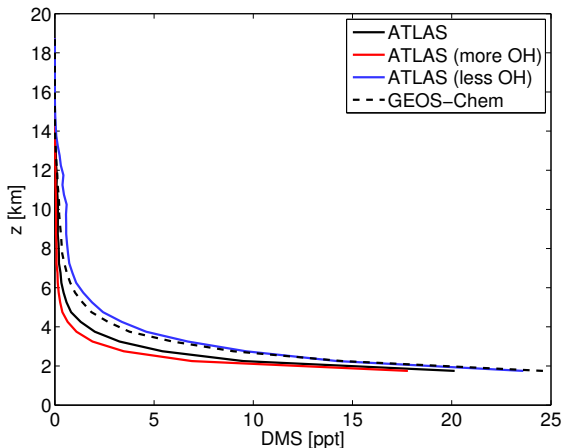
Mean SO₂: Sensitivity to OH



Ref (black)
- 50% OH (blue)
+ 50% OH (red)



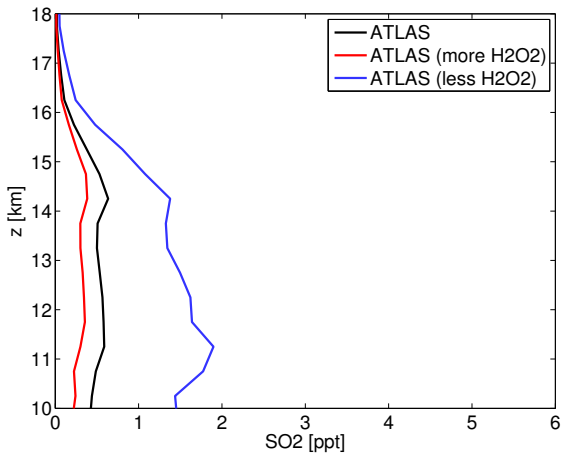
Mean DMS: Sensitivity to OH and Comparison with GEOS-Chem



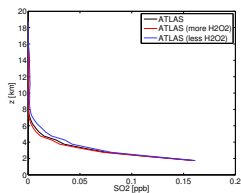
Ref (black)
-50% OH (blue)
+50% OH (red)

GEOS-Chem
(dashed)

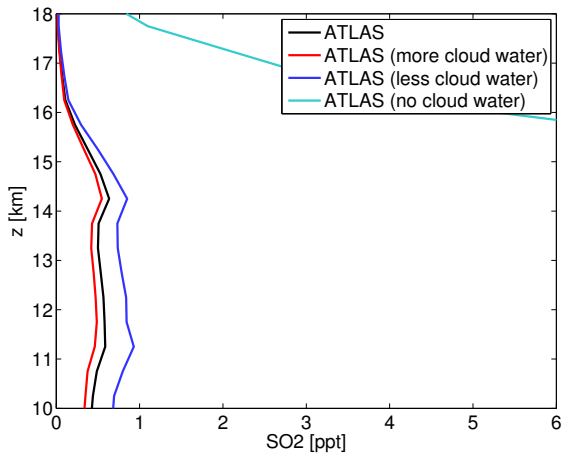
Mean SO₂: Sensitivity to H₂O₂



Ref (black)
- 50% H₂O₂ (blue)
+ 50% H₂O₂ (red)



Mean SO₂: Sensitivity to cloud water

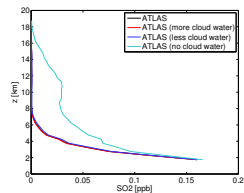


Ref (black)

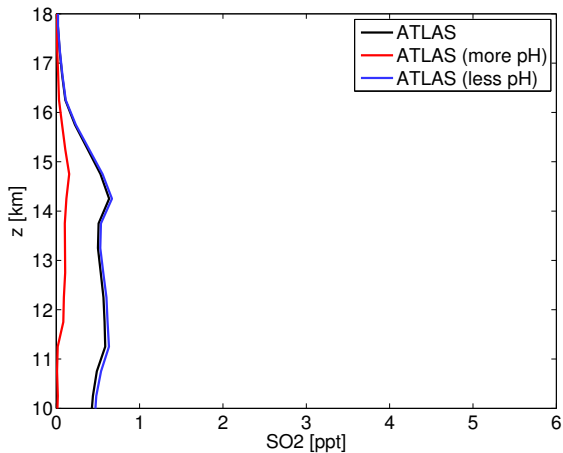
No cld water (cyan)

-50% cld water (blue)

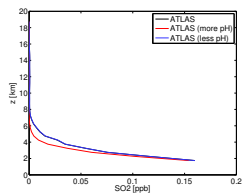
+50% cld water (red)



Mean SO₂: Sensitivity to pH



Ref pH 4.5 (black)
pH 3 (blue)
pH 7 (red)



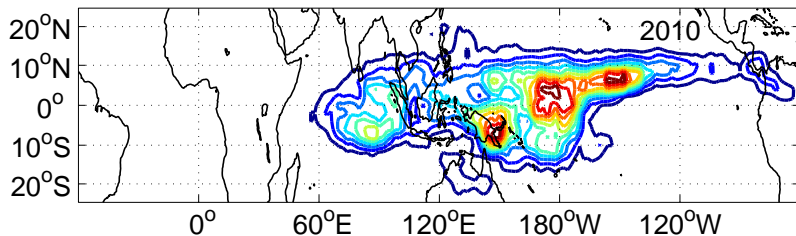
Conclusions

- SO_2 values at tropical tropopause (16–17 km) 1–5 ppt according to our runs.
- Difference between our reference run and full GEOS-Chem CTM due to different transport schemes (Eulerian vs. Lagrangian).
- Modelled SO_2 significantly lower than MIPAS SO_2 background climatology from Höpfner et al. (2015)

Conclusions

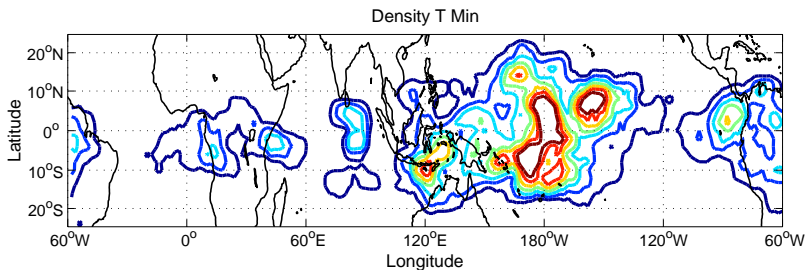
- Large sensitivity at the tropopause in run with 50% of OH reference values. Negative correlation between OH and SO_2 caused by $\text{DMS} + \text{OH}$ (and not by $\text{SO}_2 + \text{OH}$): Less OH \rightarrow less DMS loss in lower troposphere \rightarrow more DMS is transported upward \rightarrow overcompensates for the lower OH values there \rightarrow more SO_2 in the upper troposphere.
- Only if conditions are much drier than assumed by GEOS-Chem, higher SO_2 at tropopause expected since $\text{SO}_2 + \text{H}_2\text{O}_2$ not effective then.

Density at 800 hPa



Density of all trajectory points at 800 hPa
(contours: factor 1–8 relative to uniform contribution)

Density LCP



Density of all trajectory points at LCP
(contours: factor 1–8 relative to uniform contribution)