Model calculations of the contribution of SO_2 to the stratospheric aerosol layer

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Motivation

Stratospheric aerosol layer is important for

- Radiative balance of earth and climate change
- Stratospheric chemistry
- Geo-engineering

Many processes in the stratospheric aerosol layer are not well known

- E.g. Contribution of tropospheric species like COS and SO_2 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:

- $SO_2 + OH + M \rightarrow$ Products (gas phase)
  uptake of SO_2 into liquid phase is considered
- $DMS + OH \rightarrow SO_2 +$ Products (gas phase)
  Two reaction pathways (addition, abstraction)
- $SO_2 \cdot H_2O + H_2O_2 \rightarrow$ Products (liquid phase)
- $S(IV) + O_3 \rightarrow$ Products (liquid phase)
  $S(IV) = HSO_3^- + SO_2 \cdot H_2O$

plus Henry constants for SO_2, O_3, H_2O_2 and equilibrium constant between HSO_3^- and $SO_2 \cdot H_2O$

Model III: Chemistry

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

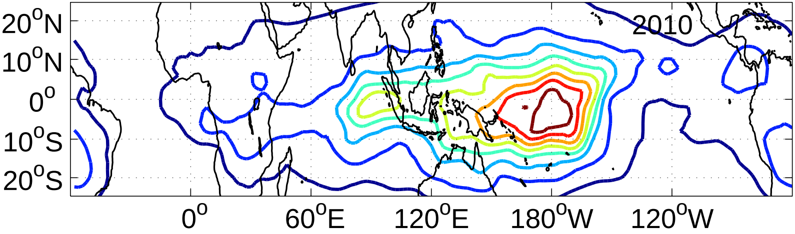
Precalculated background fields taken from the GEOS-Chem CTM (not interactive):

- 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)

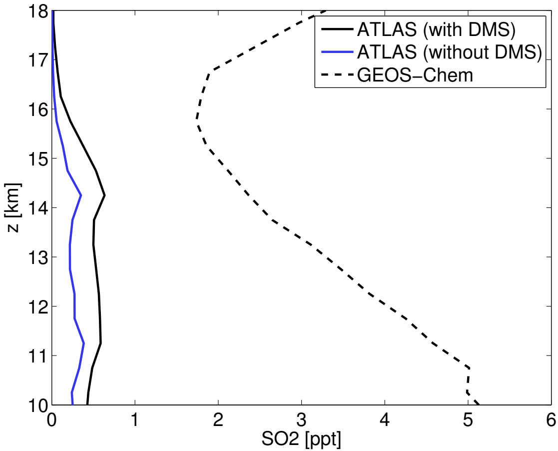
Which airmasses do we see?



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

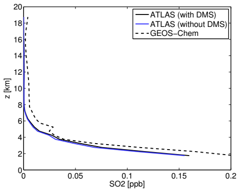
Next plots: Means over all trajectory points (as function of z)

Mean SO_2: Sensitivity to DMS and Comparison with GEOS-Chem

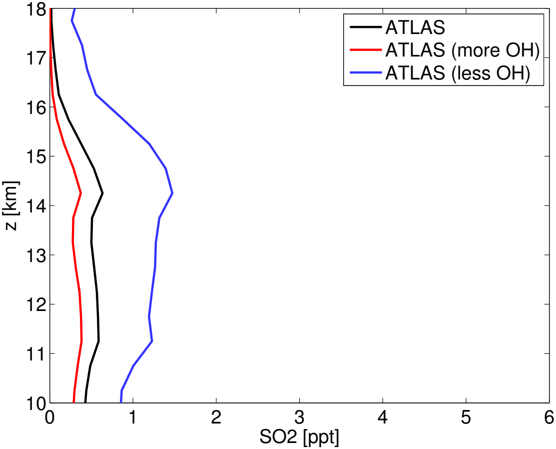


Ref (black)
No DMS (blue)

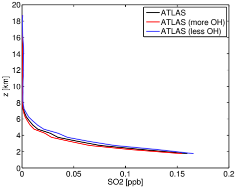
GEOS-Chem (dashed)



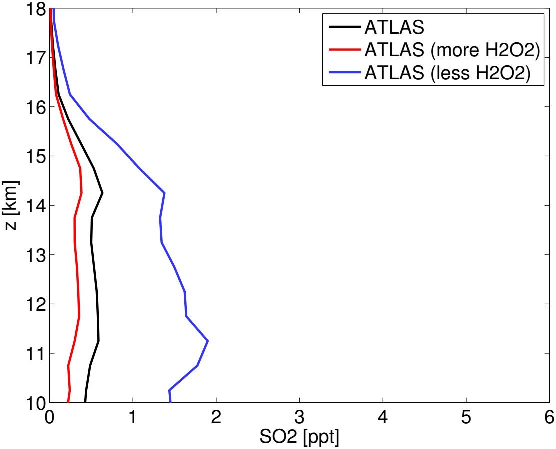
Mean SO_2: Sensitivity to OH



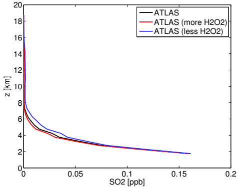
Ref (black)
−50% OH (blue)
+50% OH (red)



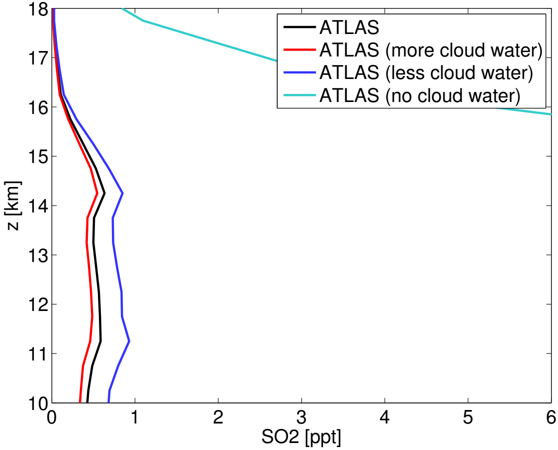
Mean SO_2: Sensitivity to H_2O_2



Ref (black)
-50% H_2O_2 (blue)
$+50\%$ H_2O_2 (red)



Mean SO_2: Sensitivity to cloud water

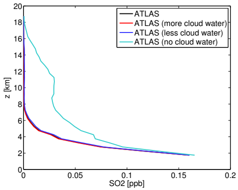


Ref (black)

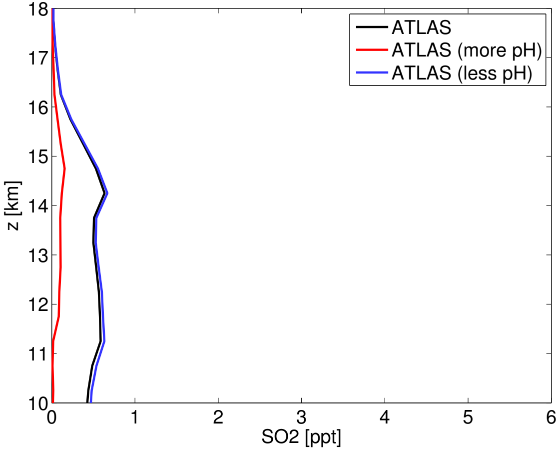
No cld water (cyan)

−50% cld water (blue)

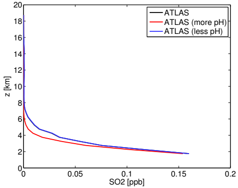
+50% cld water (red)



Mean SO_2: 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 convection (only implemented in GEOS-Chem) and different transport schemes (Eulerian vs. Lagrangian).
- Large sensitivity at the tropopause in run with 50 % of OH reference values. Negative correlation between OH and SO_2 caused by DMS + OH (and not by SO_2 + 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 $SO_2 + H_2O_2$ not effective then.