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 of the stratospheric aerosol layer are not well known

- Contribution of tropospheric species like SO_2 to stratospheric aerosol layer poorly quantified

Approach

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

Model: Transport

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

Model: Gas phase chemistry

Gas phase chemistry:

- Only calculated if air parcel not in convection!
- Every air parcel only small part of time in cloud and reactions sufficiently slow
- Knowledge about OH values in clouds very limited

Reactions:

- $SO_2 + OH + M \rightarrow$ Products
- $DMS + OH \rightarrow SO_2 +$ Products
  Two reaction pathways (addition, abstraction)
- $DMS + NO_3 \rightarrow SO_2 +$ Products

Model: Cloud chemistry

Cloud chemistry:

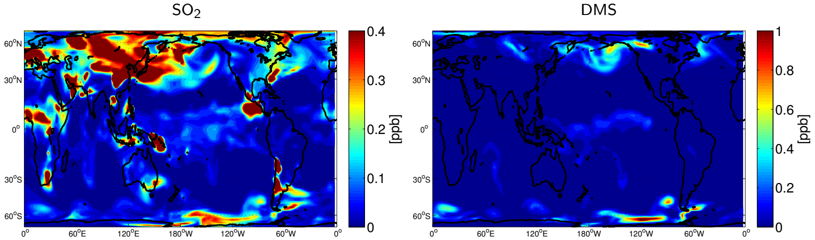
- Only calculated if air parcel in convection!
- Complete washout of products assumed

Reactions:

- $SO_2 \cdot H_2O + H_2O_2 \rightarrow$ Products
- $S(IV) + O_3 \rightarrow$ Products
  $S(IV) = HSO_3^- + SO_2 \cdot H_2O$

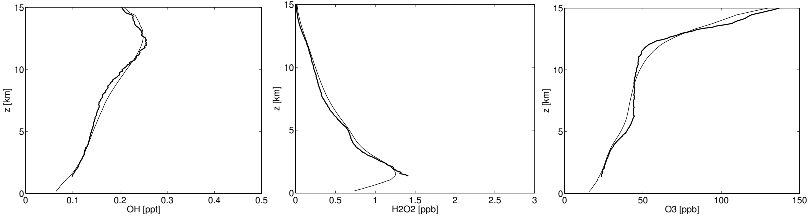
plus Henry constants for SO_2, O_3, H_2O_2 and equilibrium constant between HSO_3^- and $SO_2 \cdot H_2O$

Model: Initialization and boundary conditions



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

Model: Initialization and boundary conditions



Mixing ratios averaged over all trajectories

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

- OH
- H_2O_2 (only outside cloud)
- O_3

Model: Clouds

- Cloud water from ERA Interim
- H_2O_2 runs free inside cloud, reset to H_2O_2 climatology from GEOS-Chem outside cloud
- Cloud pH is 4.5

Model: Convection

Mass of air parcel much larger than mass in convective cell!

Basic idea (statistical approach):

- Throw a dice to determine if air parcel is entrained
- If entrained, move up by vertical updraft velocity for one time step
- If entrained, throw a dice in every time step to determine if parcel is detrained

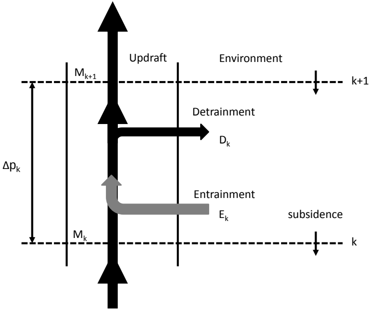
Result averaged over many trajectories is correct

Collins et al., QJRMS, 128, 991 (2002), Forster et al., JAMC, 46, 403 (2007), Rossi et al., GMD, 9, 789 (2016)

Model: Convection

Entrainment probability in layer k

$$\varepsilon_k = \frac{g_0 E_k \Delta t}{\Delta p_k}$$



Detrainment probability

$$\delta_{\mathrm{parcel}} = \frac{\int_{z_{\mathrm{start}}}^{z_{\mathrm{start}}+\Delta z_{\mathrm{conv}}} D' \,\mathrm{d}z}{M_{\mathrm{start}} + \int_{z_{\mathrm{start}}}^{z_{\mathrm{start}}+\Delta z_{\mathrm{conv}}} E' \,\mathrm{d}z}$$

Vertical updraft velocity

$$w_k = \frac{M_k R T_k}{c_k p_k}$$

E entrainment rate, D detrainment rate, M convective mass flux, $\Delta p \sim$ mass of layer, Δt trajectory time step, c convective area fraction

Model: Convection

Mass flux M, detrainment D and entrainment E taken from ERA Interim

Mass balanced by subsiding all air parcels outside convection

$$\Delta p_{\text{subsidence}} = g_0 M_{\text{parcel}} \Delta t$$

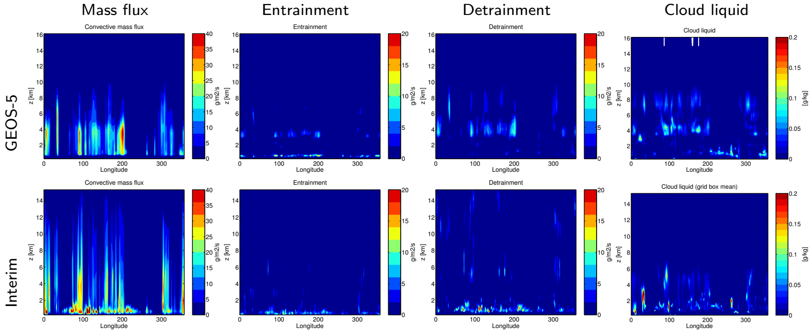
Backward trajectories require some straightforward modifications

$$\varepsilon_k = \frac{g_0 D_k \Delta t}{\Delta p_k}$$

etc.

Convection in the analyses

Curtain plot along equator for arbitrary day (0-16 km)



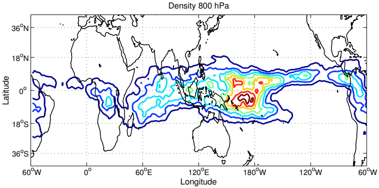
Some fairly large differences between analyses!

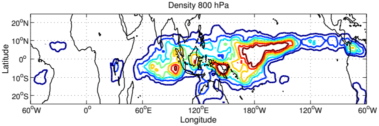
Source region of stratospheric air at 800 hPa

Convection

No convection

Interim



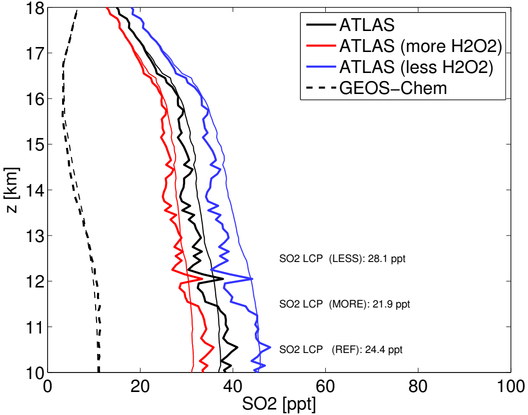


Density of all trajectory points at 800 hPa

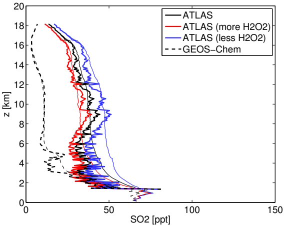
Next plots:

- Means over all trajectory points (as function of z)
- NOTE: NO OBSERVED PROFILES. Air determined to go into stratosphere is only tiny fraction of all air in troposphere
- Thin: Average including interpolated values if trajectory has no value in height bin
- Thick: Without interpolated values

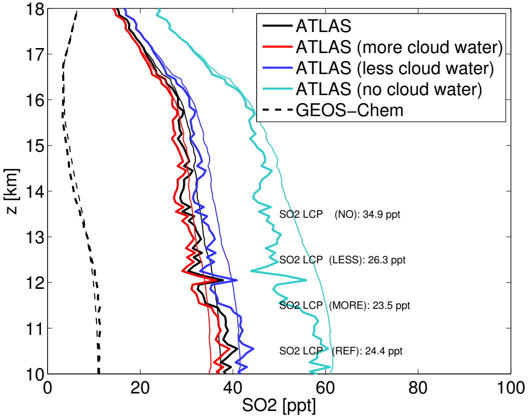
Mean SO_2: Sensitivity to H_2O_2



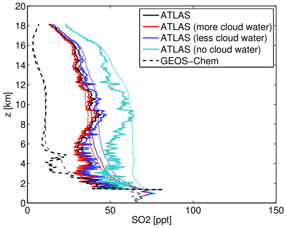
-50% H_2O_2 (blue)
$+50\%$ H_2O_2 (red)



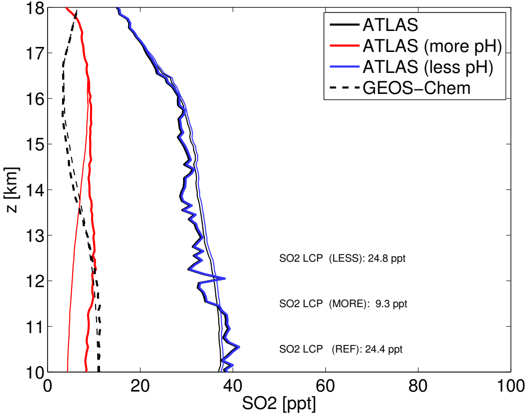
Mean SO_2: Sensitivity to cloud water



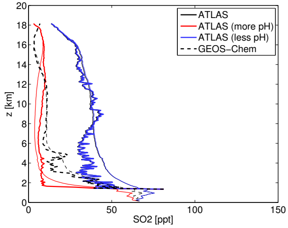
No cld water (cyan)
−50% cld water (blue)
+50% cld water (red)



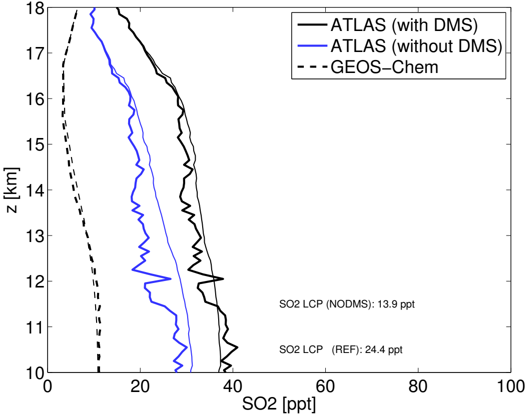
Mean SO_2: Sensitivity to pH

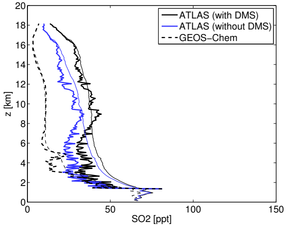


pH 4.5 (black)
pH 3 (blue)
pH 7 (red)

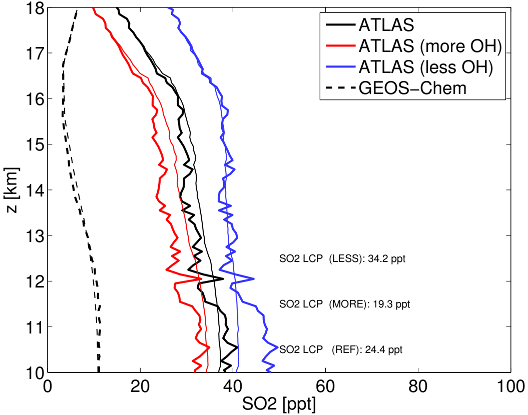


Mean SO_2: Sensitivity to DMS



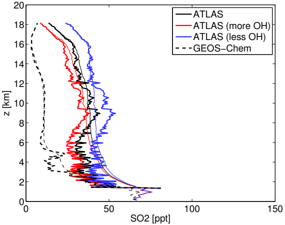


Mean SO_2: Sensitivity to OH

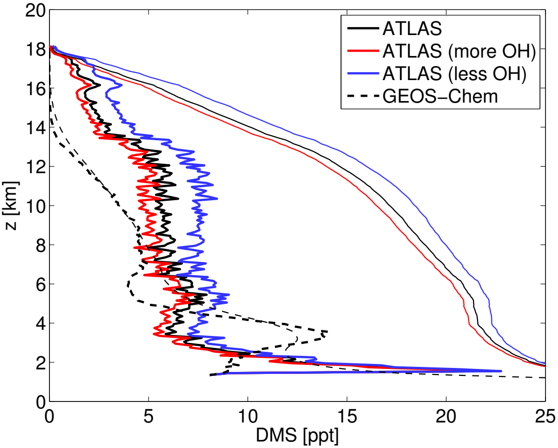


−50% OH (blue)

+50% OH (red)

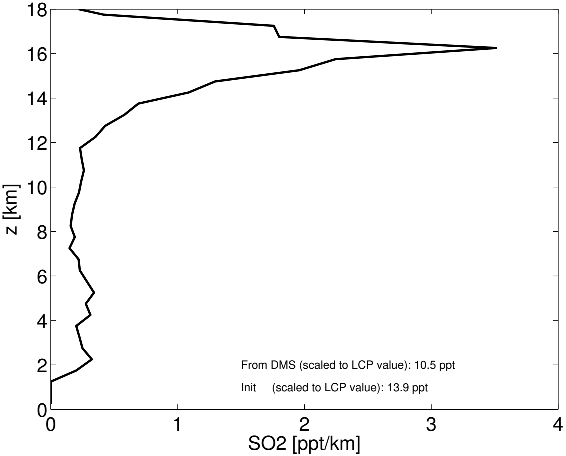


Mean DMS: Sensitivity to OH



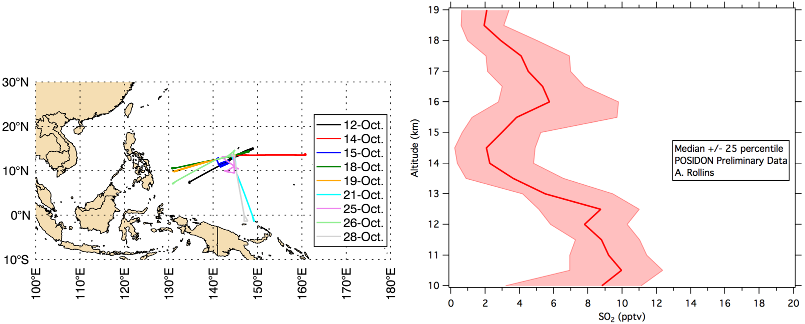
−50% OH (blue)
+50% OH (red)

Altitudes in which SO_2 at LCP was produced by DMS



Comparison to POSIDON measurements

NASA POSIDON campaign WB-57F flights in October 2016 (courtesy of A. Rollins)



Conclusions

- SO_2 values at tropical tropopause (16–17 km) about 10–30 ppt according to our runs.
- Large difference between our reference run and full GEOS-Chem CTM due to different transport schemes (Eulerian vs. Lagrangian) and ???
- Modelled SO_2 compares relatively well to MIPAS SO_2 background climatology from Höpfner et al. (2015) now (with convection)

Conclusions

- Large sensitivity at the tropopause in run with $\pm 50\,\%$ of OH reference values.
  Negative correlation between OH and SO_2 caused by DMS + OH (and also by SO_2 + OH):
  Less OH
  $\rightarrow$ less DMS loss in lower troposphere
  (less SO_2 produced there by DMS is washed out)
  $\rightarrow$ more DMS is transported upward
  $\rightarrow$ overcompensates for the lower OH values there
  $\rightarrow$ more SO_2 production in the upper troposphere
- Only if conditions are much drier than assumed by GEOS-5, higher SO_2 at tropopause expected since $SO_2 + H_2O_2$ not effective then