How does tropospheric OH variability influence the transport of SO_2 from the surface to the stratosphere? – A conceptual study

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Introduction

- 4-7%/year increase in strat. sulfate aerosols (1990-2009)[1]
- No volcanoes → transport of carbonyl sulfide (COS) and sulfur dioxide (SO_2) via tropical tropopause layer (TTL) maintains aerosol layer
- COS → low reactivity and solubility in the troposphere
- SO_2 → lifetime largely determined by the hydroxyl (OH) concentration
- SO_3 → highly soluble, reasonably efficiently removed
- Non-soluble fraction of S compounds determines flux to stratosphere
- Last removal of soluble species at Lagrangrian cold point (LCP)

What we did

- ATLAS[2] model → impact of trop. OH on strat. SO_2 flux
- Back trajectories → start at 400K, 2°x 2° long/lat grid, 30°N to 30°S, 120 days starting on 31 January 2010, must go to 800 hPa (3460)
- Troposphere → vertical winds used for vertical motion
- Upper TTL and strat → full sky heating rates for vertical motion
- Winds and heating rates from ECMWF reanalysis data – ERA Interim
- Latitude/altitude OH fields from GEOS-Chem tropospheric Chemistry Transport Model[3] (Figure 2), provide OH along the trajectories
- Box model → run from 800 hPa to LCP to represent OH oxidation:

$$SO_2 + OH + M \rightarrow HSO_3 + M \qquad (R1)$$

- R1 is the dominant gas-phase reaction
- HSO_3 → reacts with oxygen to produce SO_3
- SO_3 → with water vapour, converted rapidly to sulfuric acid (H_2SO_4)
- H_2SO_4 → uptake into water droplets major source of trop sulfate
- $SO_2/SO_3/HSO_3/H_2SO_4$ gas to particle conversion, i.e. microphysical processes not considered in box model

  → conceptual study at this point

Findings and outlook

- High OH scenarios → 2-4% of air masses contribute >10% of initial SO_2 to strat.
- OH hole → ~20% of air masses deliver >10% of initial SO_2. On average, 3 times more SO_2 reaches stratosphere than under the high OH scenarios
- Southwest and Central Pacific, Southeast Asia, and the Indian Ocean are the main contributing regions to stratospheric SO_2
- Low OH increases mean SO_2 lifetime by 5 days in trop. and TTL
- Temperature dependence of (R1) reaction rate leads to an increase in SO_2 lifetime by ~2 days from low to high temp.
- Outlook → include microphysics as ~50% of strat. sulfur delivered as aerosol

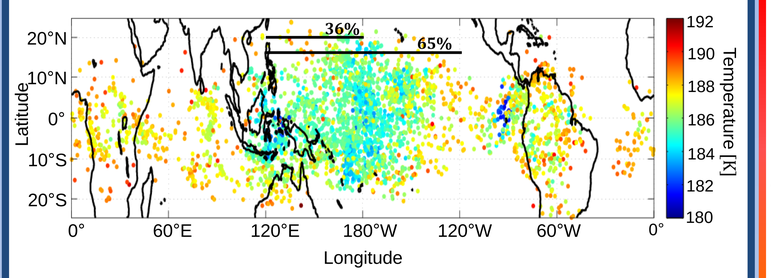

Figure 1: LCP locations and corresponding minimum temperatures. West Pacific is main strat. entrance region for trop. air masses (36%). 2009-2010 El Niño event → 29% of trajectories reach LCP above Central Pacific.

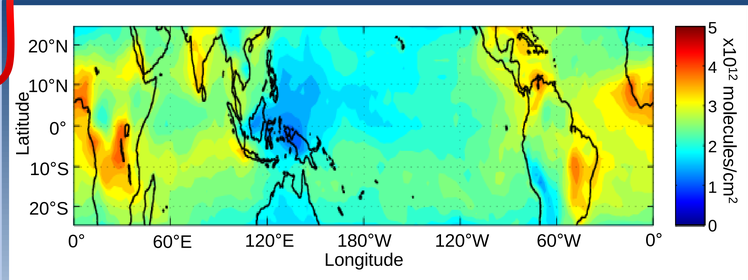

Figure 2: Trop. OH columns from GEOS-Chem (1-15 October 2009). Four scenarios considered:
(i) OH zonal mean → OH 'global' scenario
(ii) OH mean from 125°E-140°E → OH 'hole' scenario
(iii) OH mean from 50°W-50°E → OH 'Atlantic' scenario
(iv) OH mean from 80°W-100°E → OH 'without Pacific'

- Studies of the flux of species often based on zonal mean OH concentrations (i)[4]
- recent study by Rex at al.[5] showed existence of pronounced minimum in OH-concentrations above the West Pacific

  → impact of such variability in trop. OH concentrations on strat. SO_2 flux analysed here

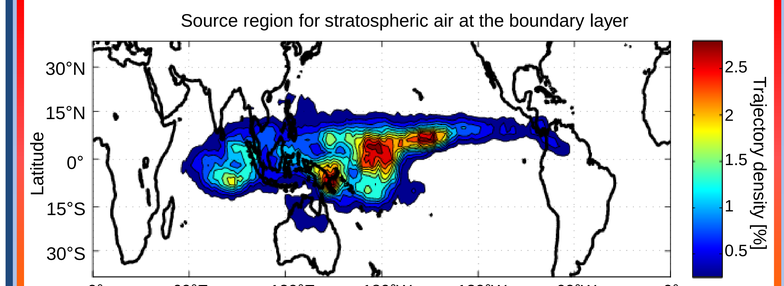

Figure 4: 800 hPa trajectory origins. Biomass burning and small volcanic eruptions in SW Pacific, SE Asia, and Central Pacific can contribute more to strat. sulfur budget than previously thought.

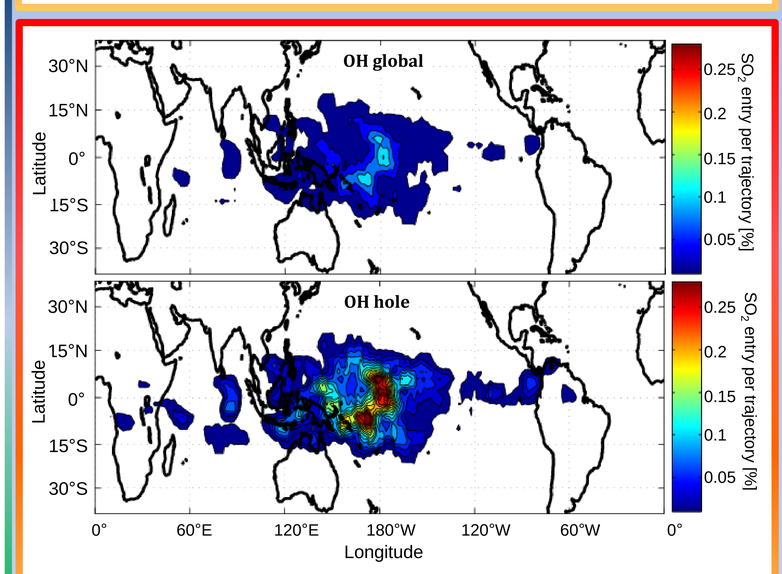

Figure 5: Regional contribution to the total % of SO_2 reaching the LCP. 10°long x 5°lat regions.

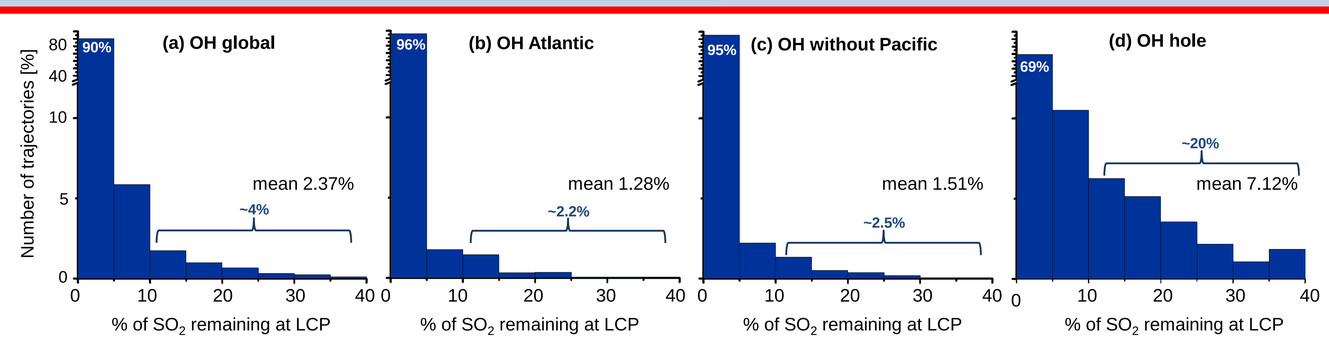

Figure 3: SO_2 remaining at LCP for 4 OH scenarios. High OH → ~2-4% of all trajectories deliver more than 10% of their initial SO_2 to the stratosphere. Low OH → ~20% of all trajectories deliver more than 10% of SO_2 to the stratosphere.

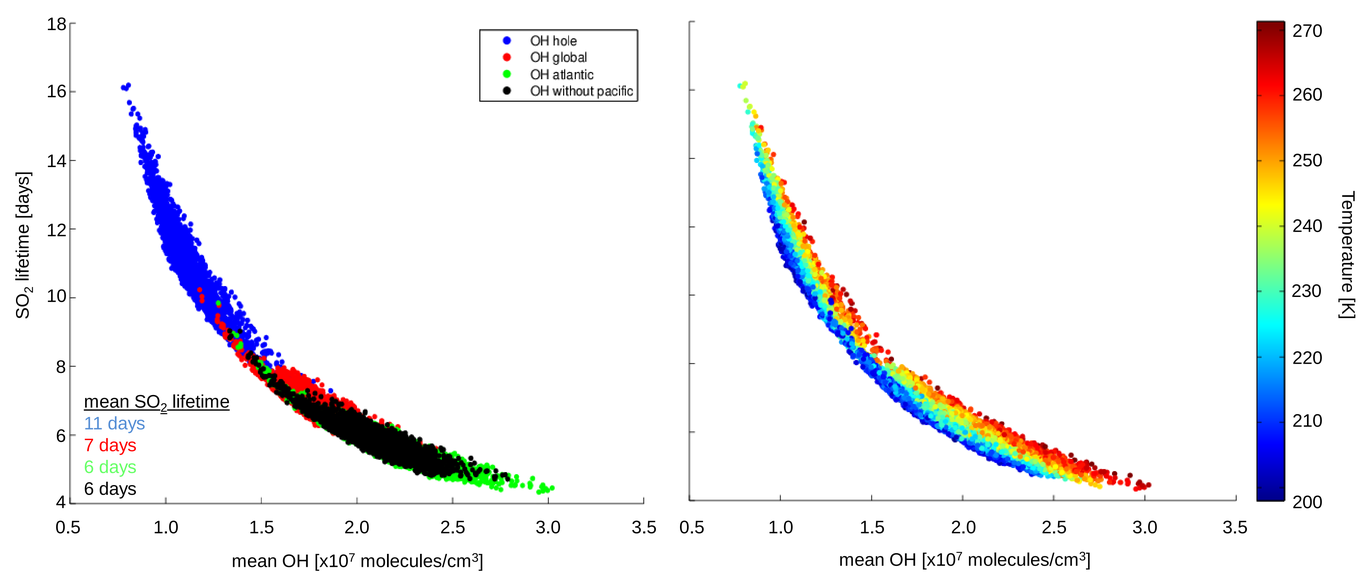

Figure 6: SO_2 lifetime vs. mean OH concentration along all trajectories for 4 OH scenarios. Low OH → lifetime can be twice that for high OH. Panel b: OH weighted mean temperature along each trajectory. Spread in the SO_2 lifetime related to temperature dependence of OH oxidation (R1).

Acknowledgements
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