The stratospheric sulfur burden: an assessment based on gas and particle phase measurements

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Outline for the paper including rough responsibilities:

Introduction (Markus R., Terry)
- Discuss the unique nature of sulfur in the stratosphere, sources, sinks.
- Define the base of the stratosphere which will be used – dynamical tropopause.

Gas phase measurements (Michael, Marc)

SO2 – MIPAS 2002-2012 (Michael, Norbert, Anika)
- Describe the measurements. Are there any in situ measurements for comparison?
- Figure 1 to show SO2 profiles with error/uncertainty bars on the profiles, very similar to figures already provided.
- Create data file for temporal history of sulfur column from SO2 in 30 degree latitude bands. These can be provided, as the figures now as column mass, but ultimately we will want a total burden in each latitude band as a function of time.

OCS

MIPAS 2002-2012 (Michael, Annika)
- Describe the measurements.
- Create data for Figure 2 with error/uncertainty bars on the profiles, very similar to figures already provided.
- Create data file for temporal history of sulfur column from OCS in 30 degree latitude bands. These can be provided, as the figures now as column mass, but ultimately we will want a total burden in each latitude band as a function of time.

ACE-FTS 2004-2012 (Marc, Corrina)
- Describe the measurements.
- Create OCS mixing ratio profiles representative of latitude bands and seasons similar *to MIPAS* Figures for inclusion in Figure 2. Add error/uncertainty bars to the profiles.
- Create data file for temporal history of sulfur column from OCS in 30 degree latitude bands. These can be provided as the figures now as column mass, but ultimately we will want a total burden in each latitude band.

Figure 2 to display mixing ratio profiles of OCS from both ACE and MIPAS by season and in different latitude bands.

Compare ACE and MIPAS mixing ratio profiles for OCS for selected measurements. Figure 3. (Corrina and Annika?)

Ground based columns (Stefanie, Justus, Jim Hannigan)
- Describe measurements
- Figure 3 to compare column OCS data from:
  - Jungfraujoch (Justus)
  - Manua Loa (Jim)
  - Wollongong (Stefanie)
  - Lauder (Stefanie)
  - Arrival Heights (Stefanie)
- Data file for temporal history of sulfur column from OCS over the data record.

Particle phase measurements (Terry, Larry)

Extinction – (Larry, Adam)
- Review the measurements that will be used and differences between them. Rationale for limiting to the measurements included.
- SAGE solar occultation
- OSIRIS limb scatter
- Figure 4 to compare SAGE and OSIRIS extinction at 525 nm

Backscatter - (John, Thomas)
- Review the measurements that will be used and differences between them. Rationale for limiting to the measurements included.
- Figure 6 to show the following records, north to south
  - Garmisch 1976-2016
  - OHP – newly available
  - Hampton 1974-2002
  - Tskuba 1982 - 2016
  - Mauna Loa 1975 – 2016
  - San Jose dos Campos 1974-2009
  - Lauder – 1993-2016

$$N_{tot} = \int \left[\sum dn_j / d\ln(a) \right] d\ln(a) = \int \left[\sum \frac{N_j}{\sqrt{2\pi} \bullet \ln(\sigma_j)} \exp\left(\frac{-\ln^2[a/\mu_j]}{2 \bullet \ln^2(\sigma_j)_j} \right) \right] d\ln(a)$$

Size distribution (Terry)
- Brief review of the measurements and records available (Terry, Markus H., Chuck)
- Figure 7 to compare some combination of the measurements available
  - Wyoming OPCs
  - Denver University FCAS
  - CARIBIC

Conversion to particle mass (Terry, Larry, Adam, John)
- Estimate volume from:
  - Extinction - varies between solar occultation and limb scatter (Larry, Adam)
  - Backscatter – requires backscatter to mass or backscatter to volume ratios – Jager and Deshler or others? (Terry, John, Thomas)
  - Size distribution - straight forward integral (Terry)
  - Figure 8 to compare the different volume estimates, satellite, lidar, in situ (Terry &)
- Estimate particle mass and sulfur content from density of sulfate aerosol. (Terry)
  - Sulfate density appears to be relatively insensitive to h2o climatology. Even low h2o partial pressures will provide enough h2o collisions with a particle for the h2so4 and h2o to reach the equilibrium dictated by the acid molecules and the temperature.
  - Estimate acid weight percent from sulfate density. Acid wt% = f(T)
  - Derive sulfur content of the aerosol
    - CARRIBIC - Particulate sulfur mass concentration in air from 0 to 3 km above the tropopause stratified by volcanism. Direct check on other conversions
  - Figure 9 to show particle sulfur mixing ratio can be derived from any measurement producing a volume.

Create particle sulfur mixing ratio profiles from: (Larry, Adam, Terry)
- SAGE
- OSIRIS
- Wyoming OPC
- Compare with measurements from FCAS, CARRIBIC
- Figure 10 to compare sulfur mixing ratios from various primary measurements.

Particle phase sulfur burden (Larry, Adam, Terry, John)
- Integrals of sulfur mixing ratio profile from
  - SAGE
  - OSIRIS
  - Wyoming OPC
  - Integrated backscatter
- Create data file for temporal history of sulfur column from particle measurements in 30 degree latitude bands. These can be provided, as the figures now as column mass, but ultimately we will want a total burden in each latitude band as a function of time.

Global zonal average mixing ratio profile combining gas and particle phase sulfur (All)

Time period beginning of ACE, 2004, to end of SAGE II, July 2005
- Justification
  - Provides a good overlap of MIPAS and ACE for OCS measurements.
  - SAGE II provides global aerosol coverage with reasonable volume estimates
  - Volcanically quiescent and at the end of the major background period following Pinatubo

Figure 11 to show altitude latitude zonal averages of sulfur mixing ratio, dynamic tropopause to 35 km. Consider a three panel plot with one for gas phase, one particle phase, one total
- Purpose to illustrate the differences of sulfur mixing ratio with other trace species which have little dynamic character in the stratosphere. Sulfur is different due to its particle phase which has a sink through sedimentation

Sulfur burden estimates – combine gas and aerosol (All)

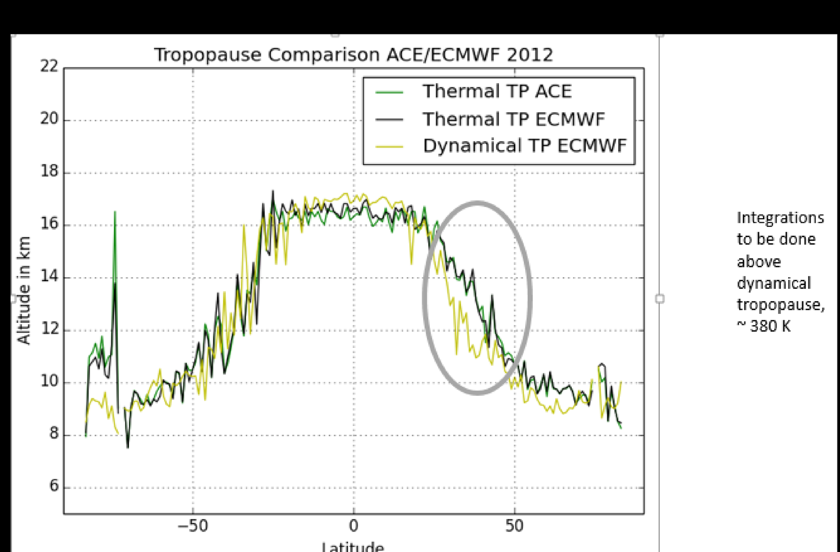
Specific locations with long term records
- Lidar
  - San jose dos Campos
  - Mauna Loa
  - Hampton
  - Garmisch
- In situ
  - Laramie
  - a/c limited to < 20 km

Global estimates – spatial and temporal distribution
- Thirty degree latitude bands using zonal averages and interpolated across the band to give column sulfur from: For each latitude band envision a figure showing the total as summed from the contributions from the two gas sources and the particles
  - OCS
  - SO2
  - Aerosol
    - SAGE
    - OSIRIS
- Compare
  - Non-volcanic, seasonal
  - Mildly volcanic
- Figure 12 to display results

Figure 13 to extend the record prior to the gas phase measurements beginning in 2002 with rough estimates of the COS and SO2 burden prior to measurements

Issues with estimates and uncertainties

Discussion
Conclusions

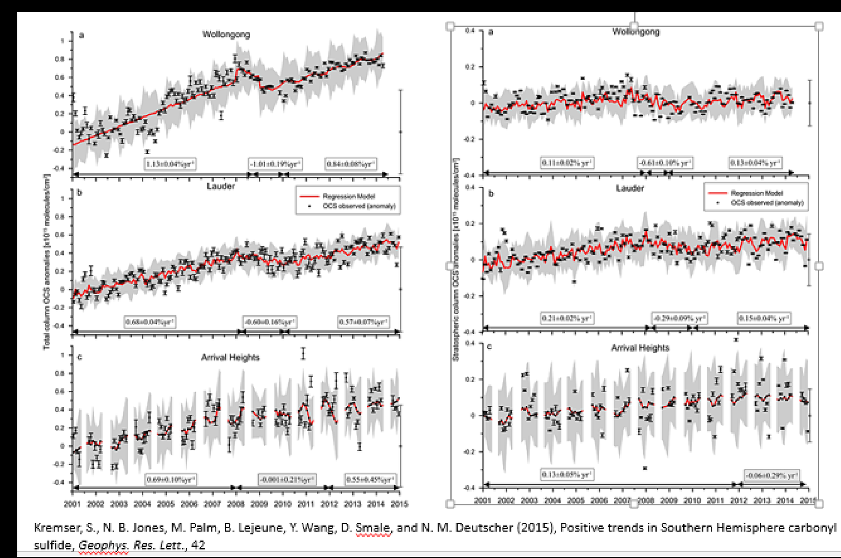


Integrations to be done above dynamical tropopause, ~ 380 K

Goals for the paper planned on: The measured stratospheric sulfur burden

- Provide a temporal history of the gas and particle phase sulfur burden in:
  - 30 degree latitude bands
  - Monthly to seasonal averages
  - Use measurements to create a global temporal / latitudinal burden
- Time period
  - 2002-2012 – Envisat period primarily
    - OCS (ACE, MIPAS, Ground Stations)
    - SO2 (MIPAS)
    - Particles – SAGE II, OSIRIS, Lidars, In situ
  - Extend the time period backwards to 1984 with particle measurements and some relevant gas phase components.

Gas Phase OCS, SO2



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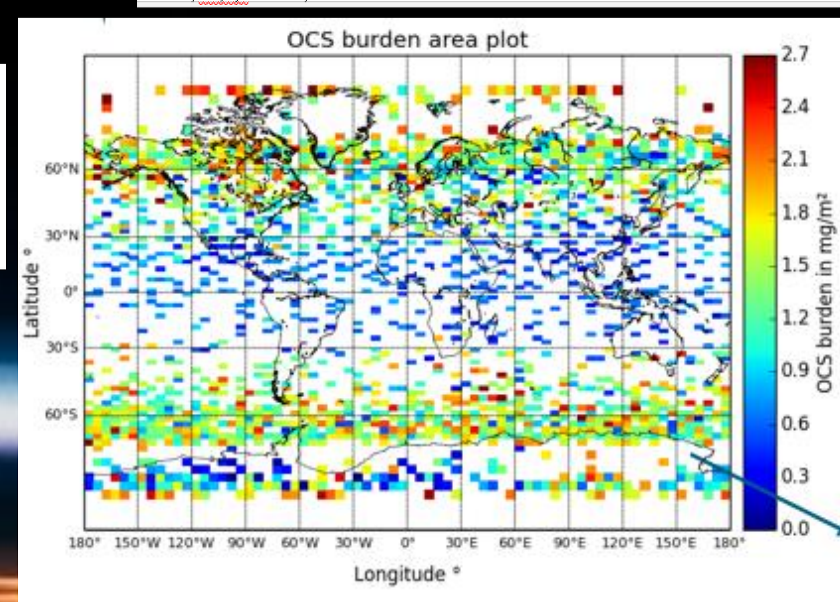
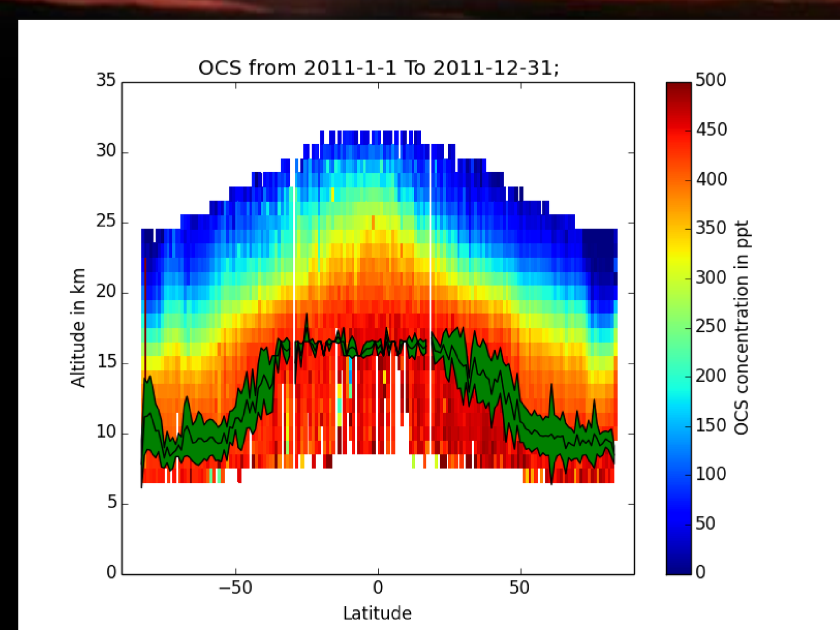
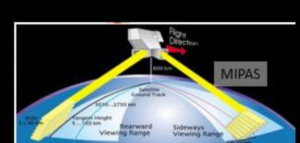
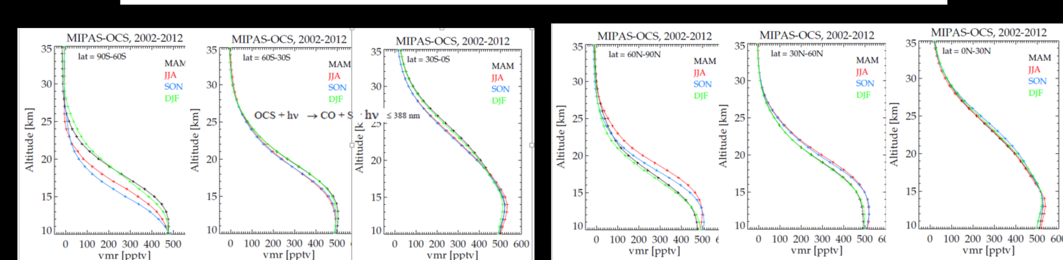
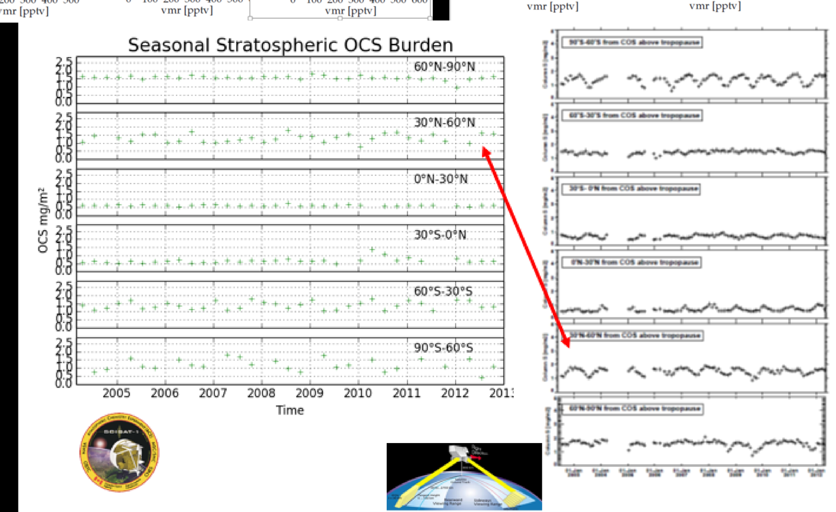


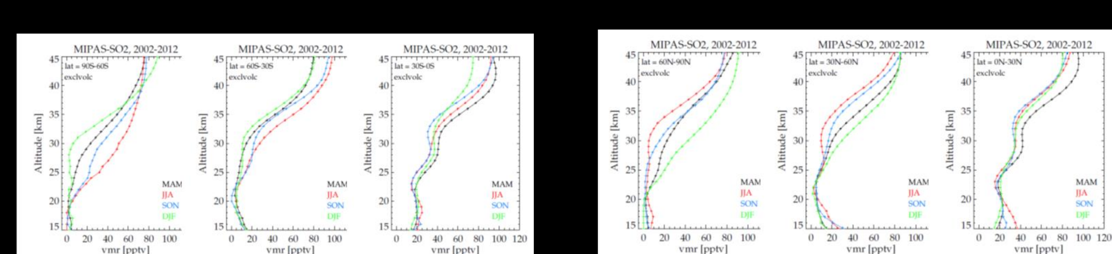




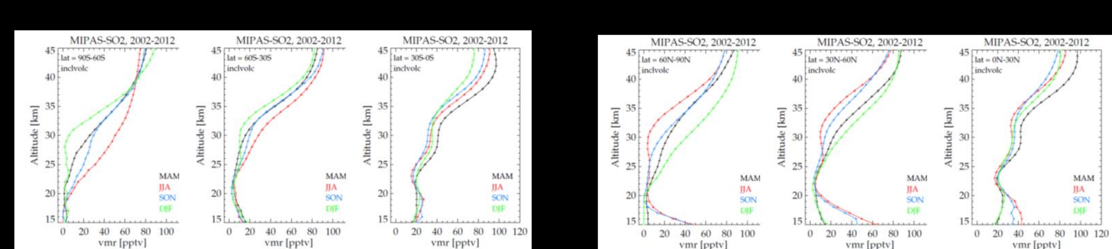


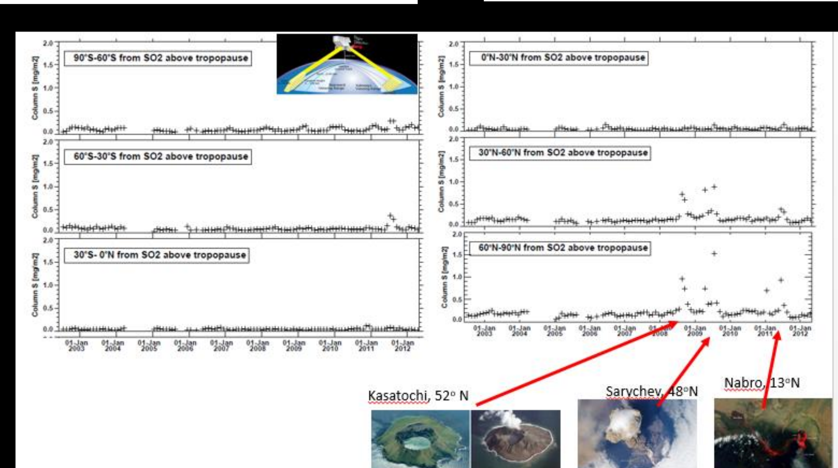


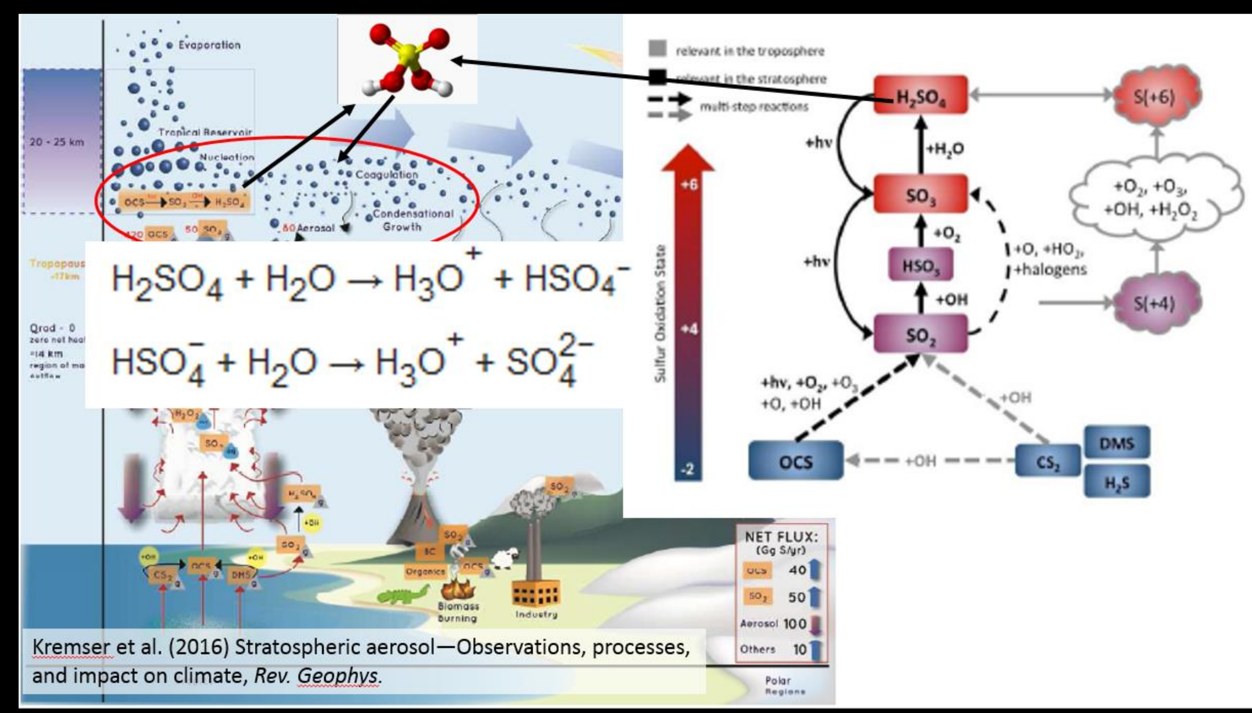
Excluding Volcanoes



Including Volcanoes







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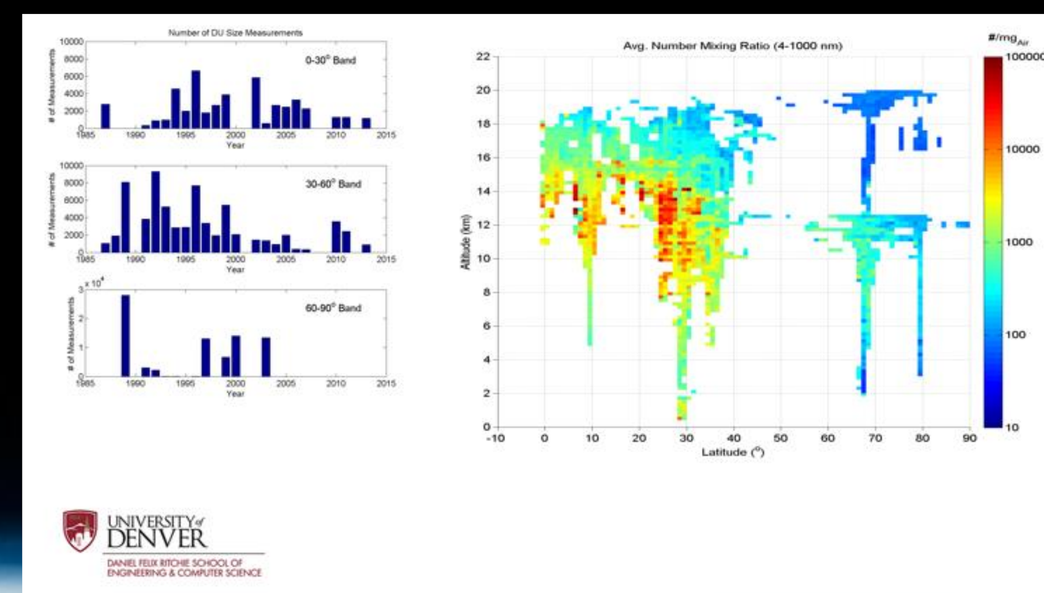
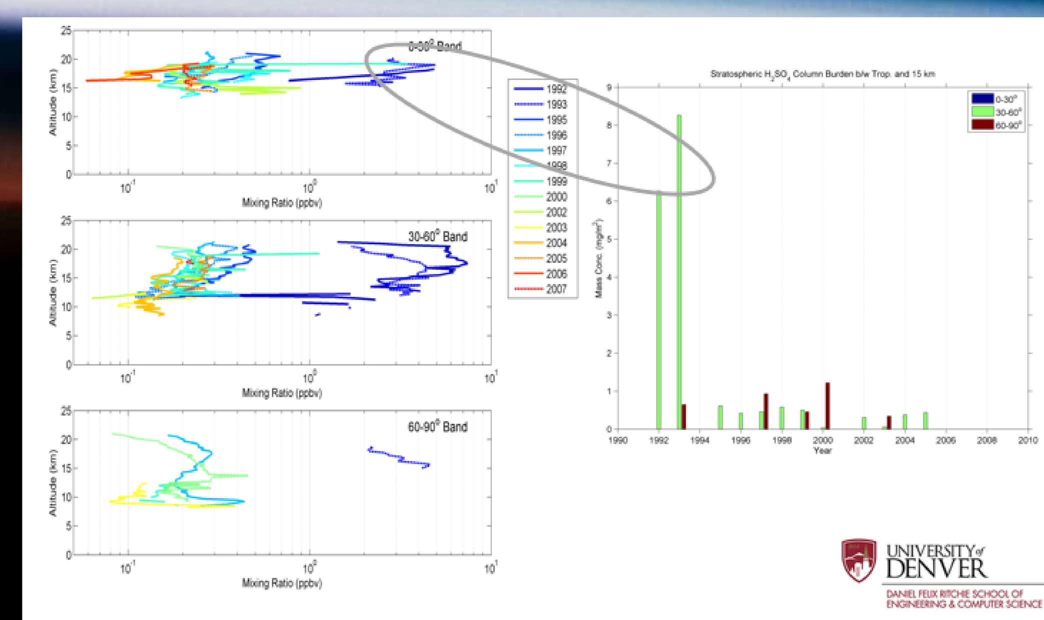
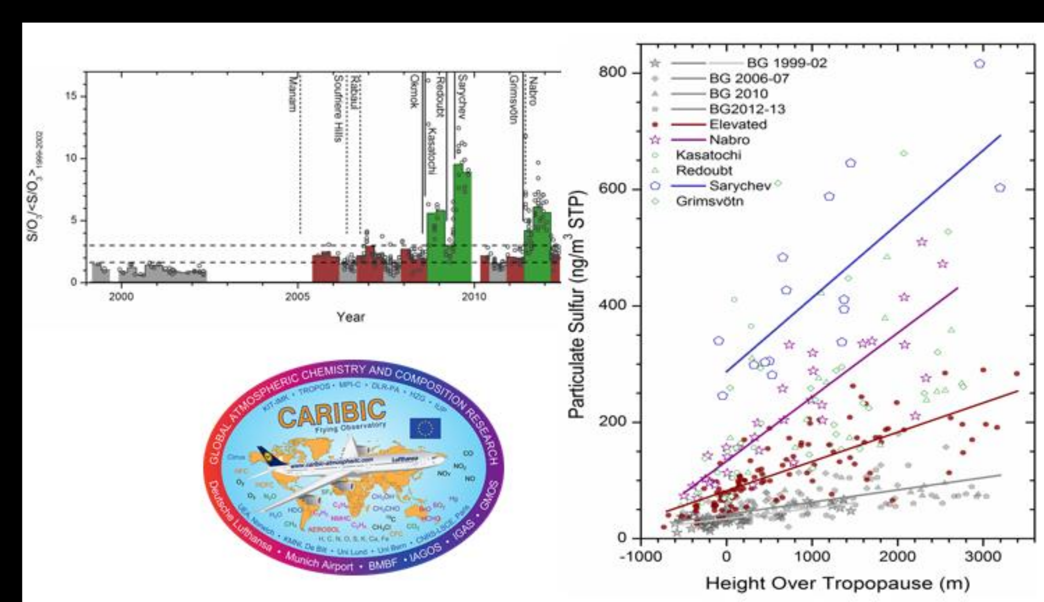
Particle phase

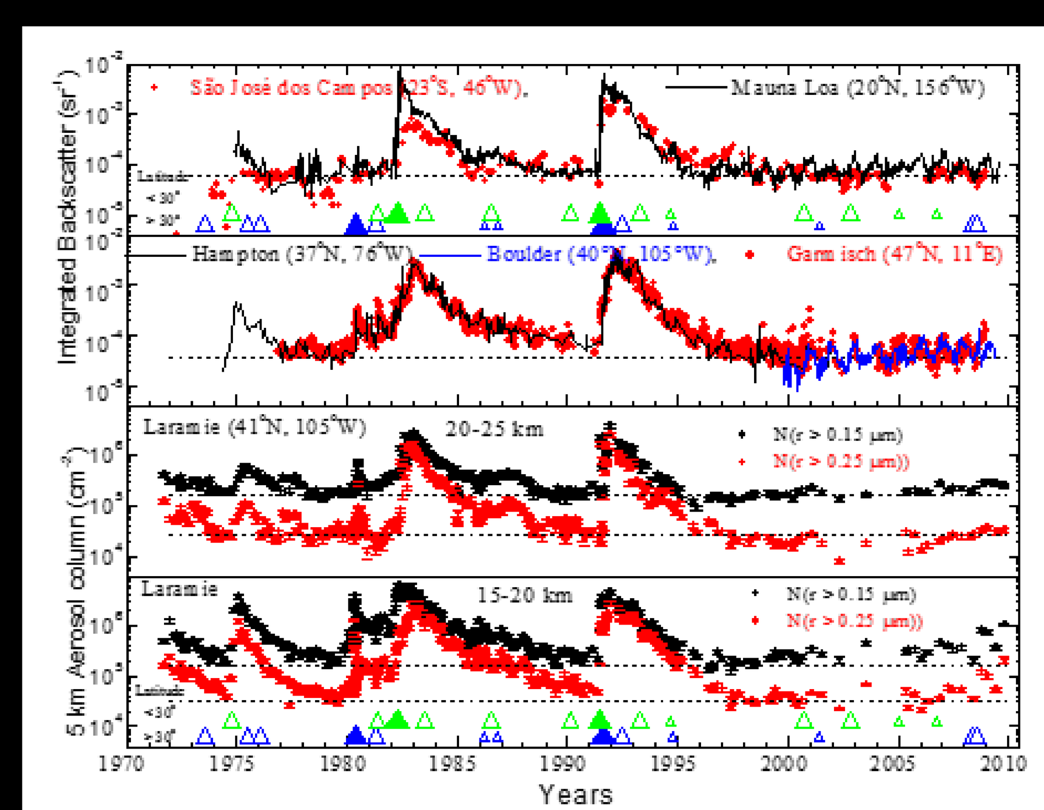
In situ aircraft

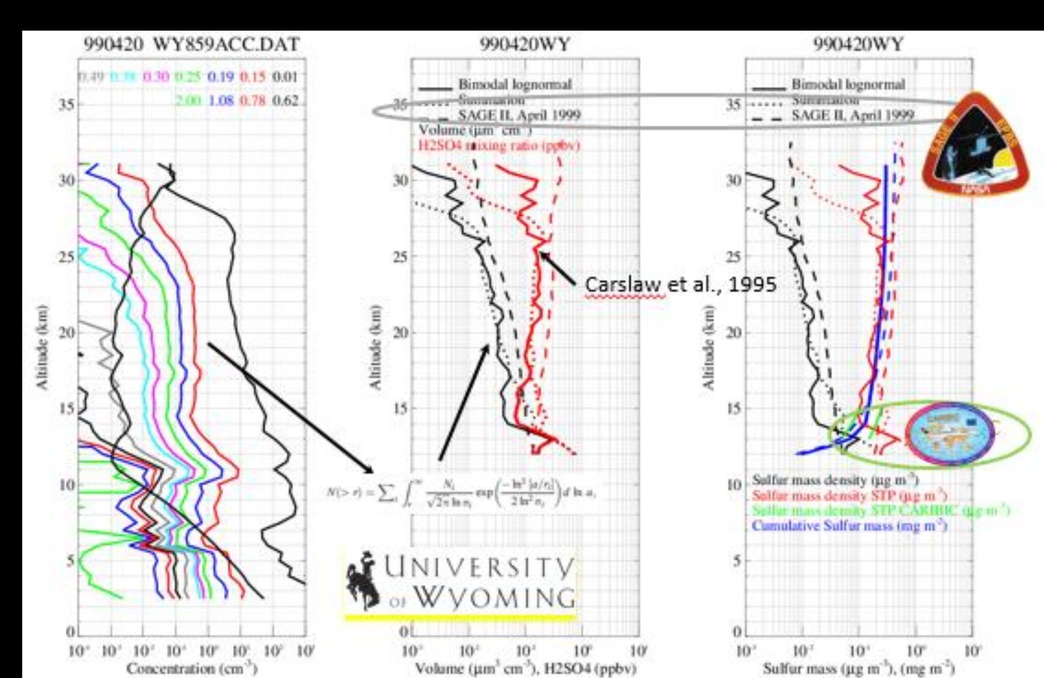


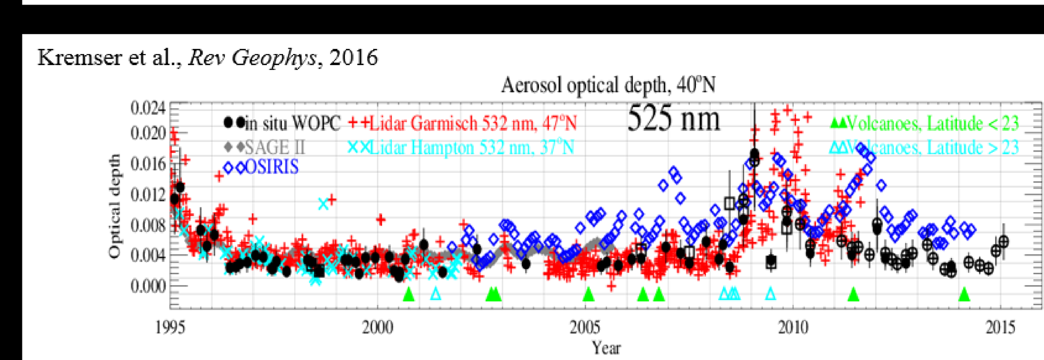




Ground based, balloonborne, satellite







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