Anthropogenic and volcanic contributions to aerosol composition and decadal variations in the upper troposphere and lower stratosphere

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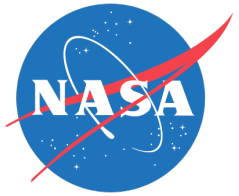


Introduction

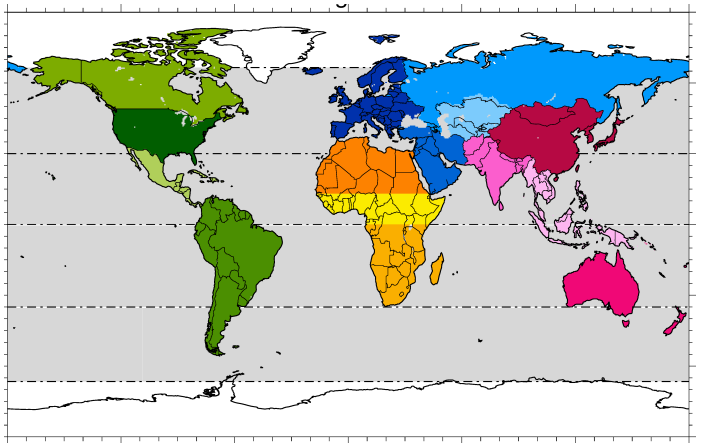
- The origin and variability of stratospheric aerosol have drawn considerable attention because the change of such aerosol could have long-term climate effects
- Recent observations seem to suggest that the stratospheric aerosol has been increasing in the past decade without major volcanic eruptions
  - Is the increase due to the Asian anthropogenic emission?
  - Or volcanoes?
- This work uses a global model to estimate the aerosol sources in the UTLS region and to elucidate the role of convective transport

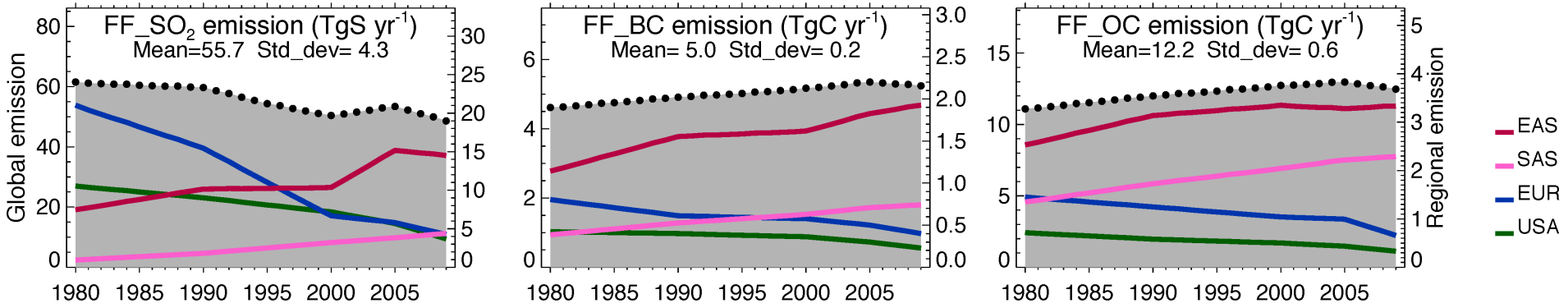
Model simulations

- Model simulations:
  - GOCART model simulations of atmospheric aerosols, driven by the MERRA meteorology, at 1.25°x1° horizontal resolution, 72 vertical layers
  - Anthropogenic and biomass burning emission: A2-ACCMIP (Granier et al., 2011)
  - Volcanic emission: A2-MAP (Diehl et al., 2012)
  - Sulfate from OCS oxidation taken from the GEOS-5/stratchem simulation (Valentina Aquila)
  - Simulations with all emissions (BASE) and with natural emission only (NAT), such that the source of aerosols at a location and time can be estimated
- Time period of this study: 2000-2009

Anthropogenic emissions

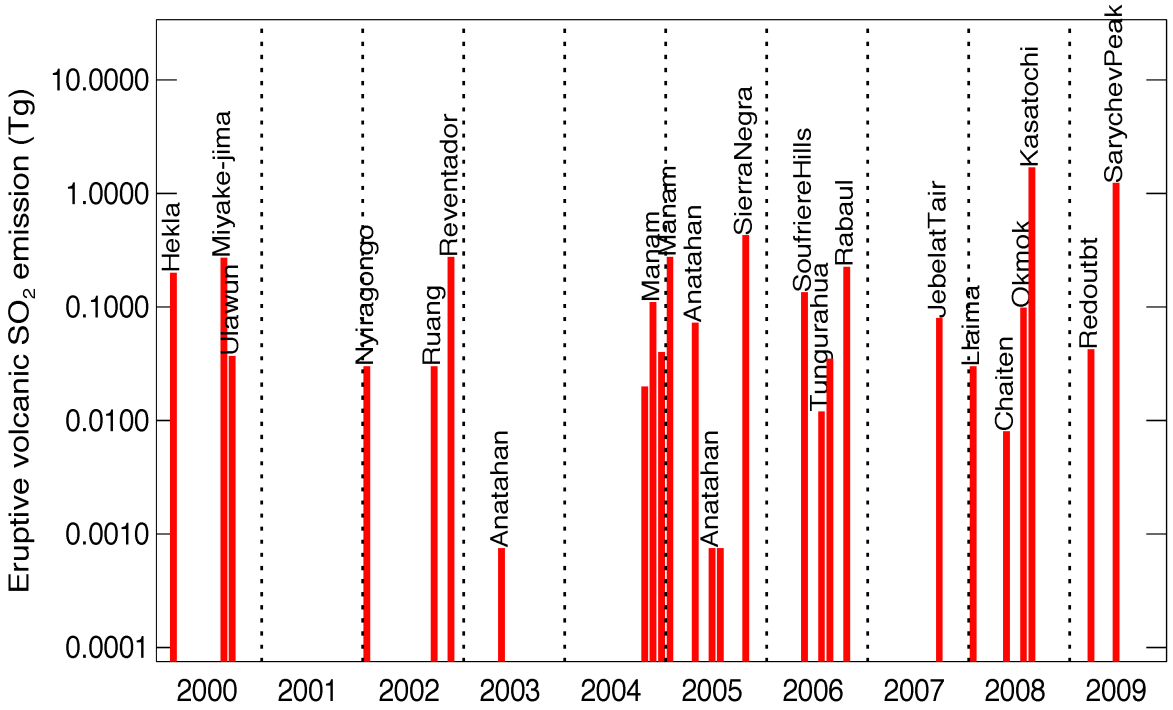
- Anthropogenic SO_2 (and other pollutants as well) emissions in East Asia and South Asia have increased significantly in the last decade
- EAS emission is much higher than SAS
- The question is: How efficient the transport is to lift Asia surface pollution to the stratosphere to control the stratospheric aerosol trend?





(Figures from Chin et al., 2014)

Volcanic emissions injecting to UTLS

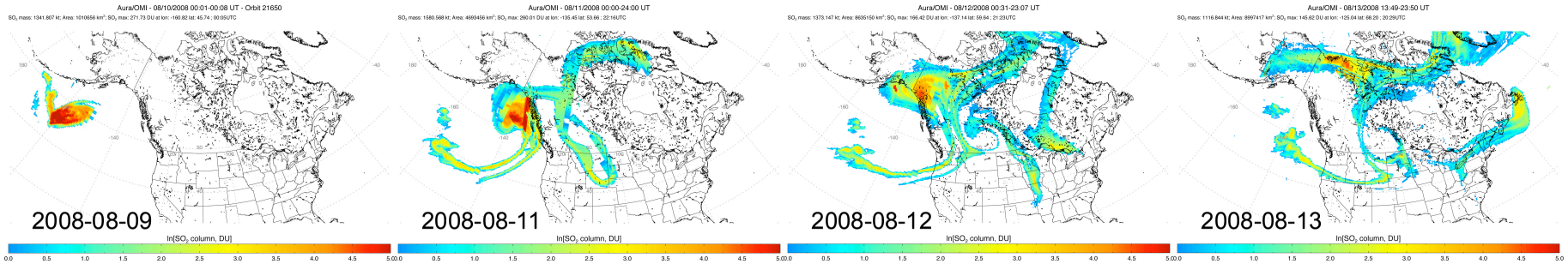


SO_2 emission from eruptive volcanoes from 2000 to 2009 with injection height above 10 km. Data source: OMI, GVP, and in-situ measurements reported in literature (Diehl et al., 2012)

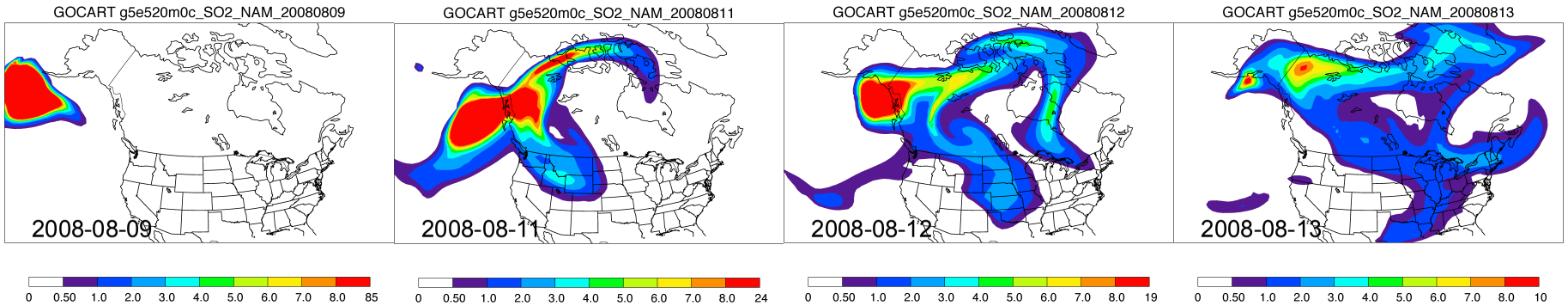
- Volcanic emissions that reach the UTLS seem to have a positive trend as well
- They release SO_2 at high altitudes to have a more direct influence than Asian anthropogenic sources

Example: Volcanic SO_2 – Kasatochi, Aug 2008

OMI column SO_2



GOCART column SO_2

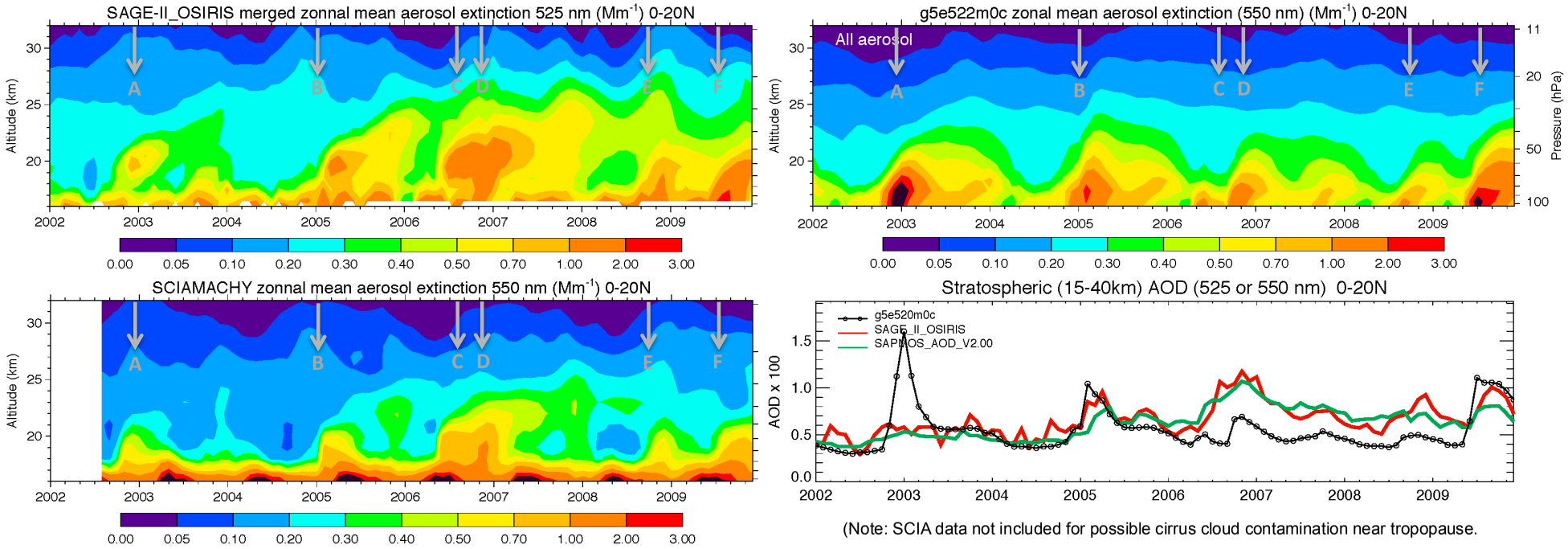


OMI: snapshot at ~1:40 pm local time GOCART: daily average
(note: unit and color scales are different between OMI and GOCART)

Comparison with satellite aerosol data

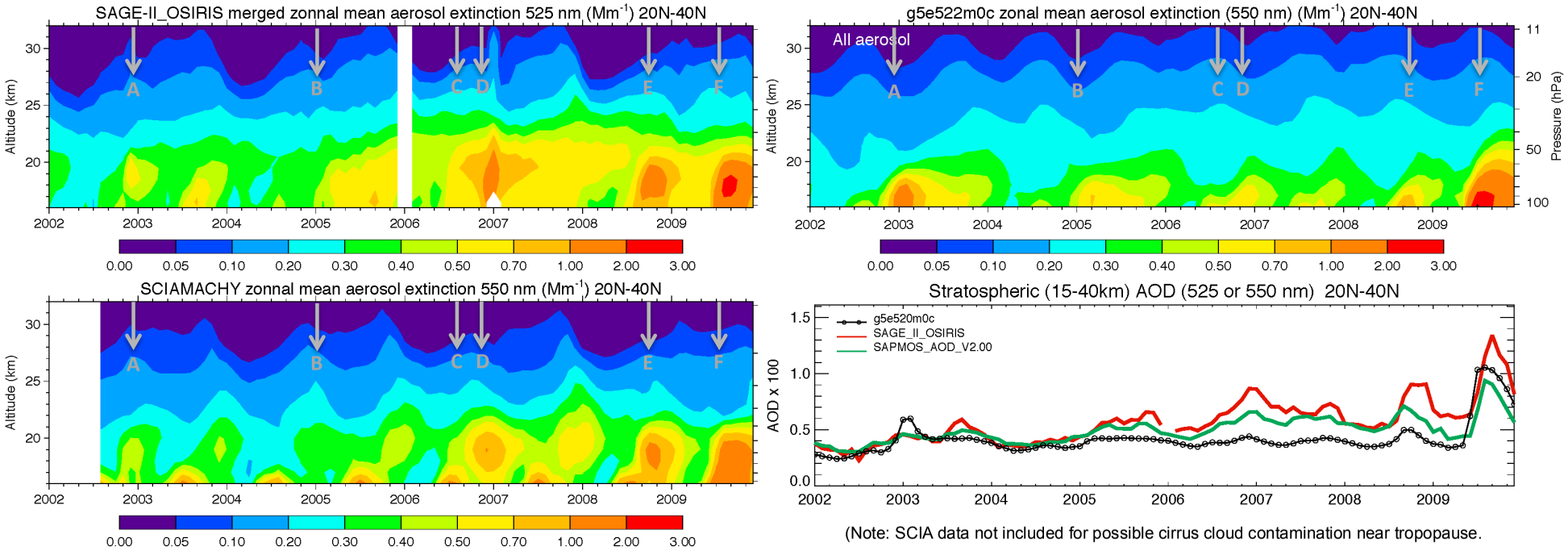
- OSIRIS:
  - V5-07 level 3 monthly zonal averages at 5° latitude resolution and 1-km vertical resolution from 0-40 km (provided by U. Saskatchewan group, POC: Landon Rieger)
  - Merged SAGE-II and OSIRIS: extinction at 525 nm
- SCIAMACHY:
  - V1.1. level 3 monthly averages at 5°x5° horizontal resolution and 1-km vertical resolution from 9-40 km (provided by U. Bremen group, POC: Alexei Pozanov)
  - 550 nm extinction was interpolated from 470 and 750 nm using the Angstrom Exponent
- CALIOP:
  - Stratospheric AOD V2.0, monthly zonal averages at 5° latitude resolution with extinction integrated from 15 to 40 km and converted to SAGE-II wavelength of 525 nm (provided by Jean-Paul Vernier, LaRC)
  - Time series before CALIPSO launch include SAGE-II (up to 2005), GOMOS (Sep. 2005 – May 2006), CALIOP (June 2006 –)

Zonal mean aerosol extinction at 550 nm (Mm^{-1}), 0-20N



A. Reventador (0.08°S, Nov 2002), B. Manam (4°S, Jan 2005), C. Soufriere Hills (16°N, May 2006), D. Tavurvur (4°S, Oct 2006), E. Kasatochi (52°N, Aug 2008), F. Sarychev Peak (48°N, July 2009)

Zonal mean aerosol extinction at 550 nm (Mm^{-1}), 20-40N



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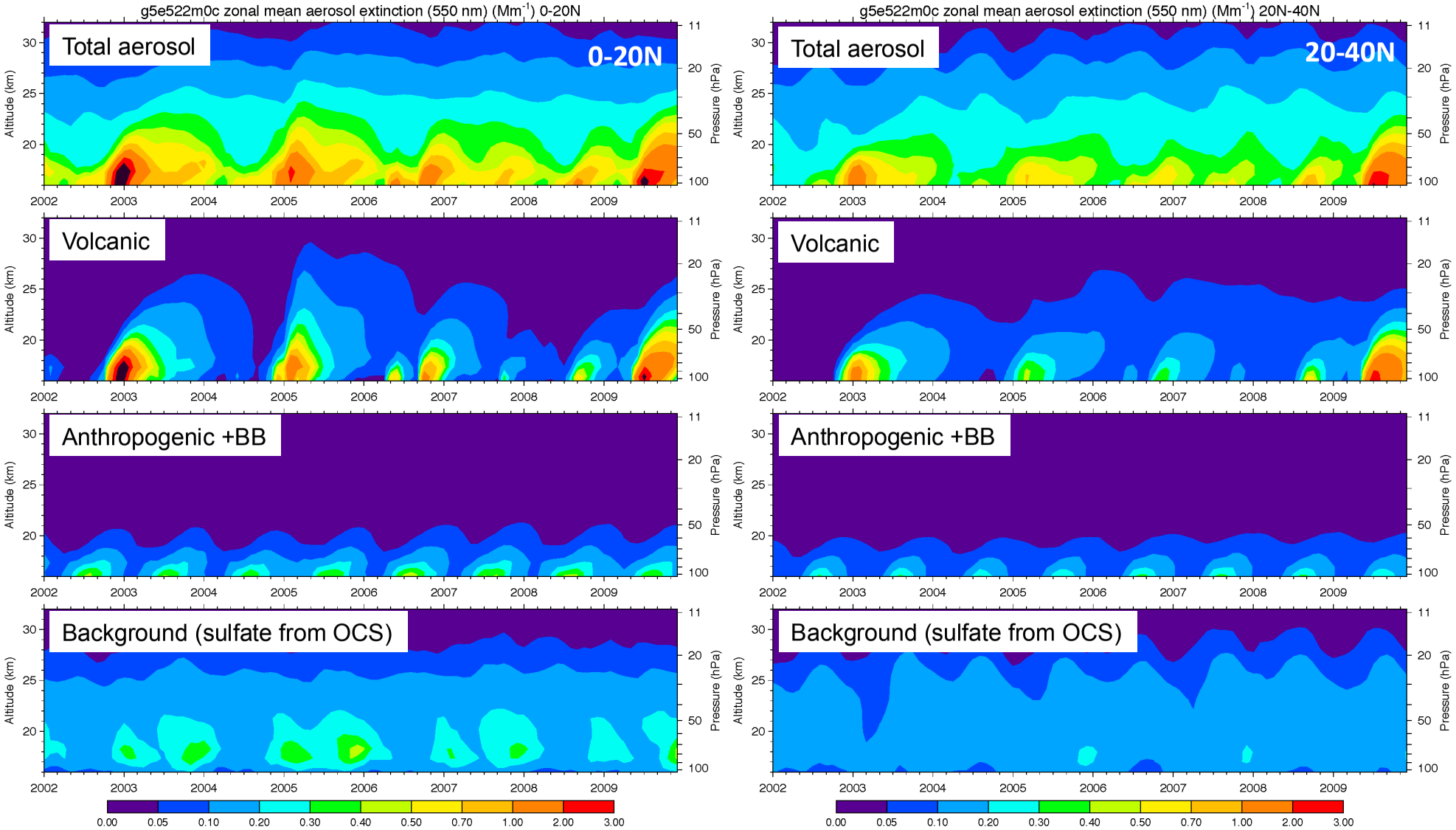
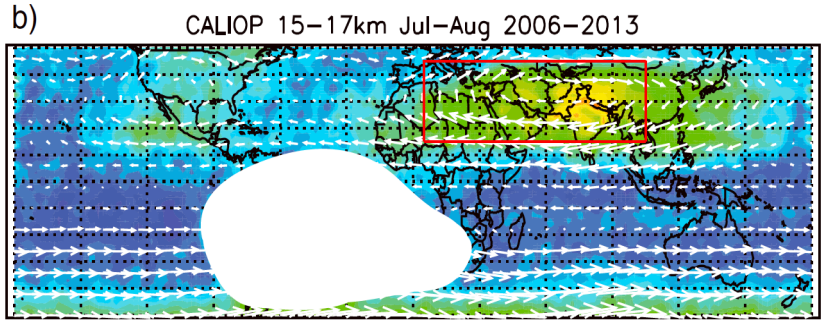
Source attribution – volcanic, anthropogenic, and background



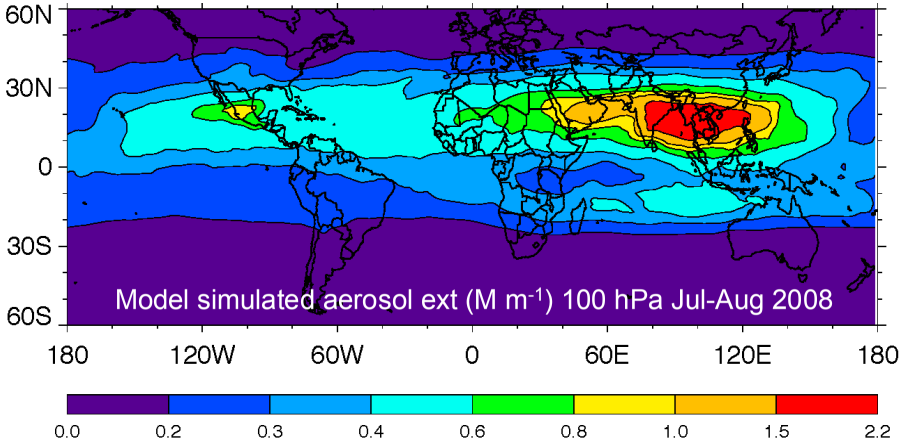
Source attribution – volcanic, anthropogenic, and background

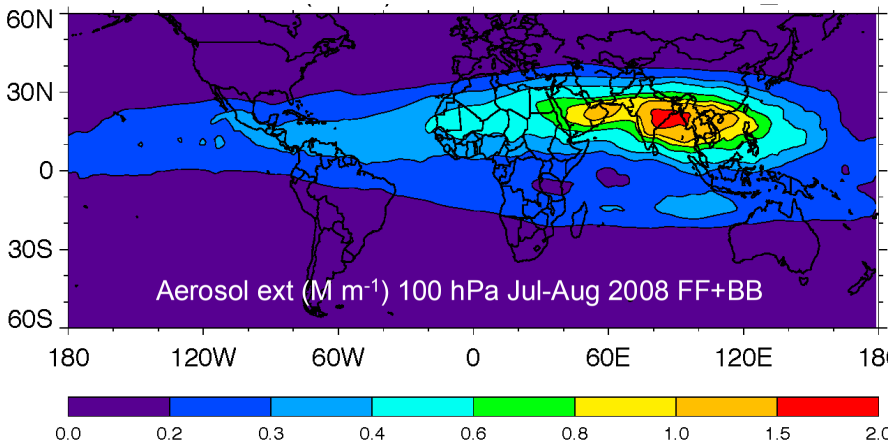
- Overall, the volcanic aerosol dominates the stratospheric aerosol loading even without Pinatubo-scale large eruption
- However, near the tropopause, anthropogenic aerosol shows a well organized seasonal cycle
- On the other hand, the "background" sulfate aerosol from OCS oxidation is more significant than anthropogenic aerosol transport

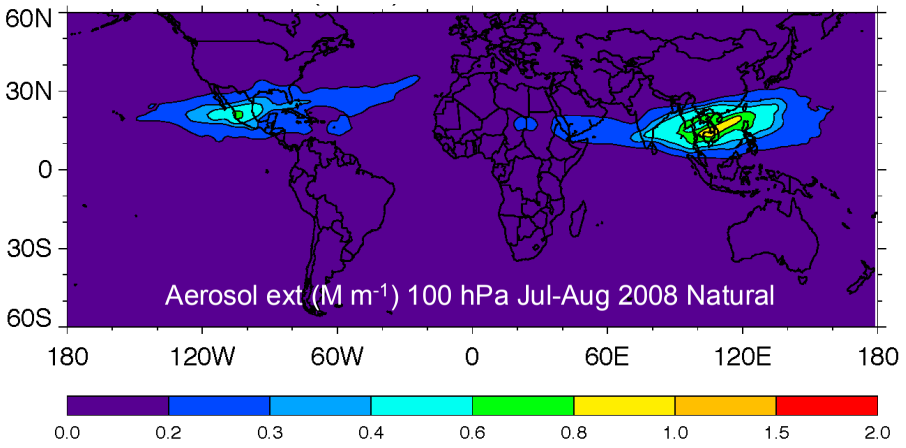
Maximum aerosol in ATAL



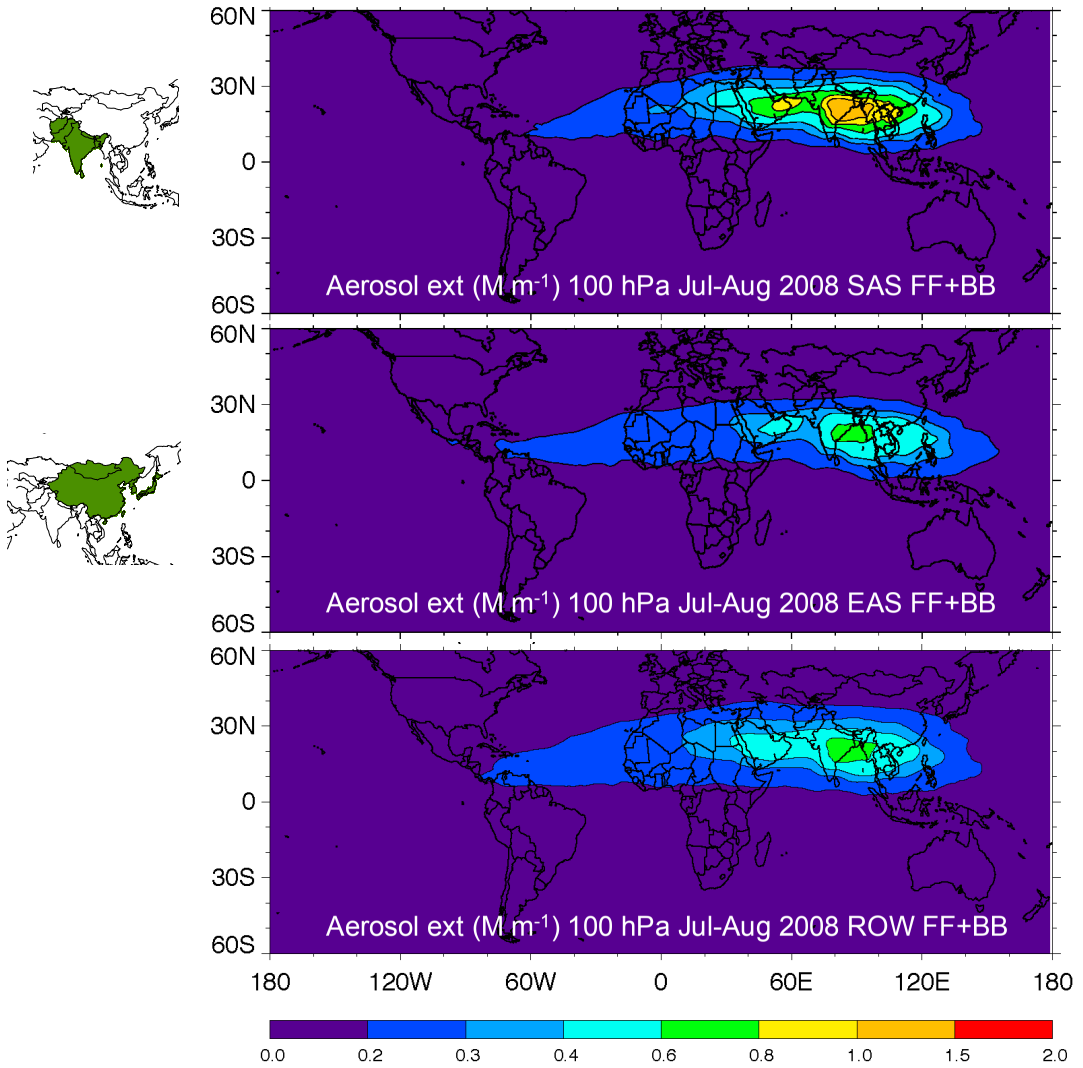
CALIOP aerosol SR 15-17 km Jul-Aug 2006-2013 (Vernier et al., 2015)

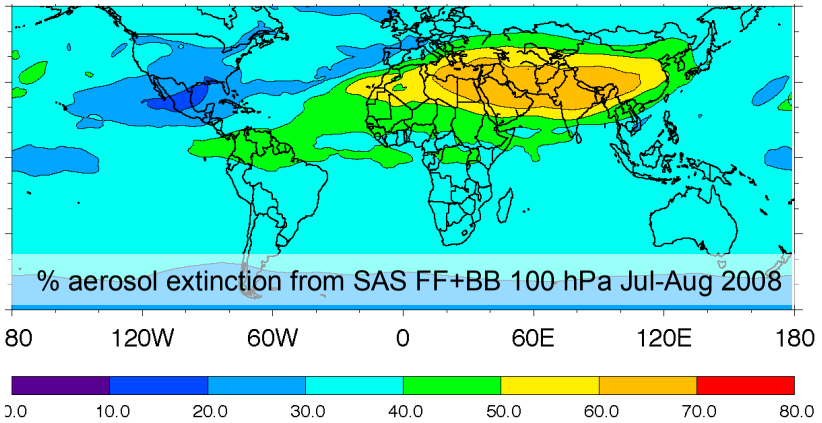






Asian monsoon convective transport – sending lower tropospheric material to UTLS





- SAS anthropogenic aerosol dominates the ATAL aerosol in summer (60-70%), even though EAS anthropogenic emission is much higher than SAS
- Not all aerosols in ATAL are from Asia – other regions contribute as well

Concluding remarks

- By model experiments separating anthropogenic and natural sources, we have found that
  - volcanic aerosol dominates the total stratospheric aerosol amount even without very large volcanic eruptions like Pinatubo
  - anthropogenic aerosol exhibits well organized seasonal cycle in the tropopause region
- Strong Asian monsoon convection and higher tropopause in the Asian summer monsoon region making transport of lower tropospheric material (from Asia and beyond) to UTLS effective in the summer
- Next: aerosol composition, transport pathways, precursor distributions

Proposed AeroCom phase III modeling and analysis relevant to SSiRC and ACAM

- Multiple model simulation for 2000-2012 (or 1998-2014, as required by SSiRC)
- Purpose:
  - To assess the origins (anthropogenic, biomass burning, and volcanic) and composition (sulfate, organics, BC, etc.) of aerosols in UTLS
  - To understand the chemical and transport processes that determine the aerosol characteristics in UTLS
- Coordinated with SSiRC MITAR model experiments and ACAM analysis
- Details are being worked out