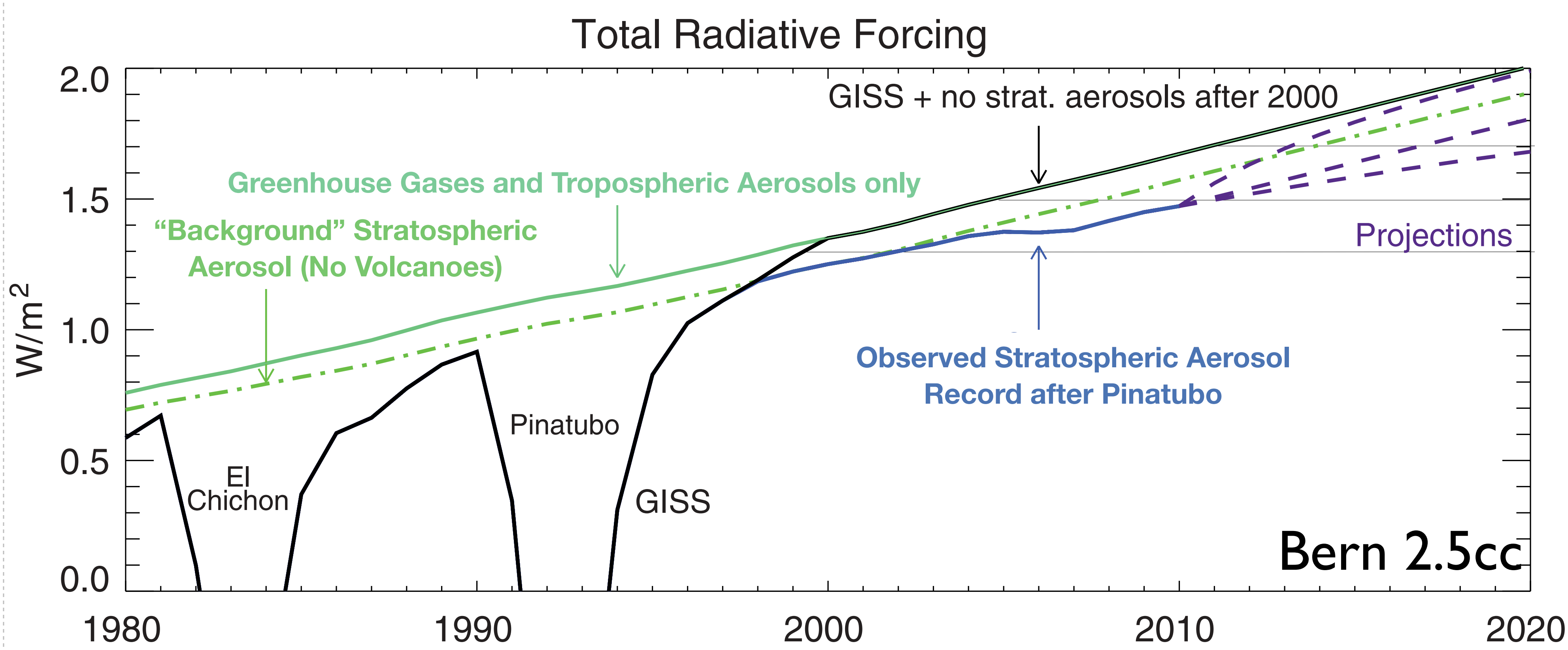
The Contribution of Anthropogenic SO_2 Emissions to the Asian Tropopause Aerosol Layer

Ryan R. Neely III (NCAR/ASP), , P. Yu, K. H. Rosenlof, O. Brian Toon, Susan Solomon, J. M. English, J. S. Daniel, H. L. Miller

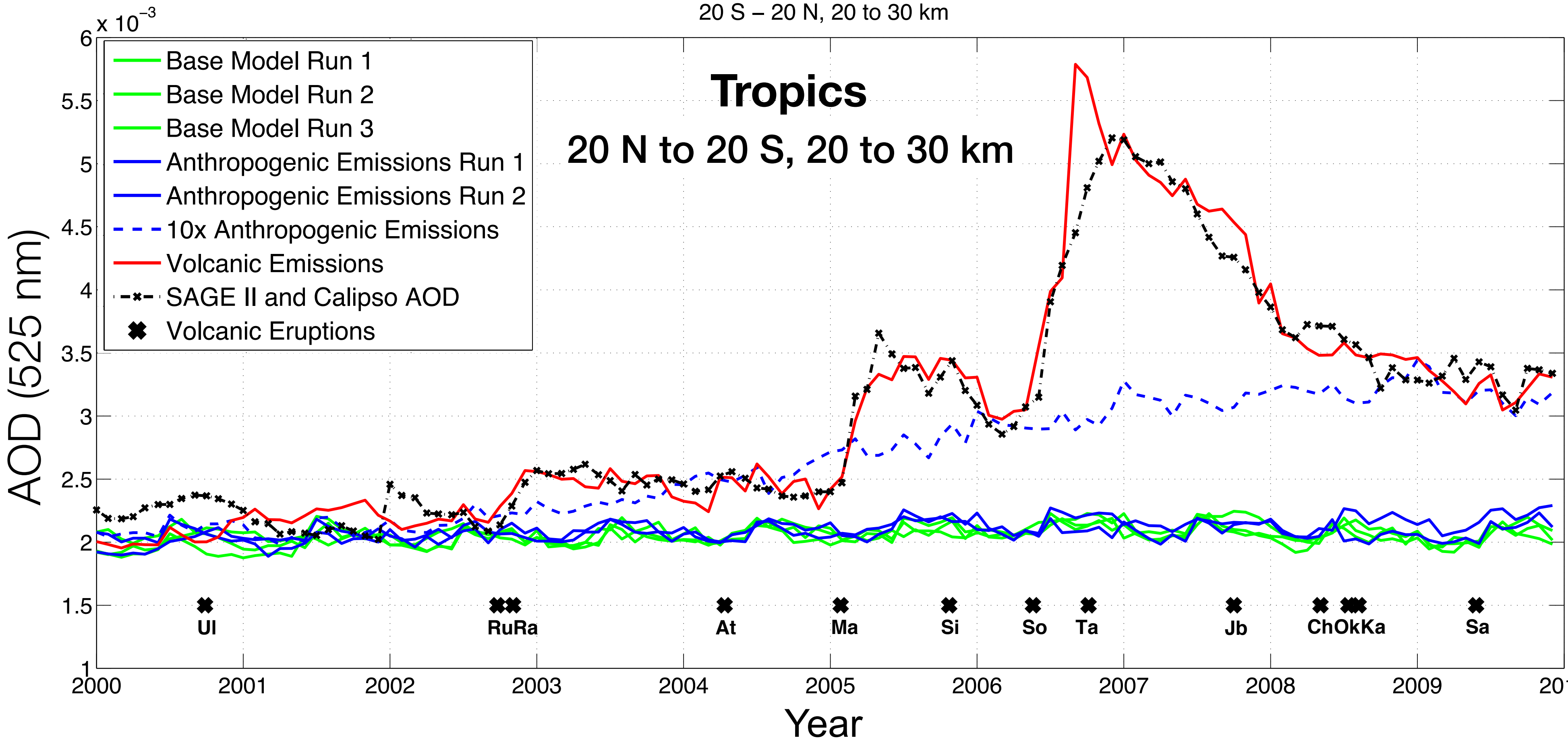
Adapted from: Neely et al., (2013), Regional Contributions of Anthropogenic SO2 Emissions to the Asian Tropopause Aerosol Layer, Geophys. Res. Lett, *in review*.

Variability in stratospheric aerosol impacts global radiative forcing



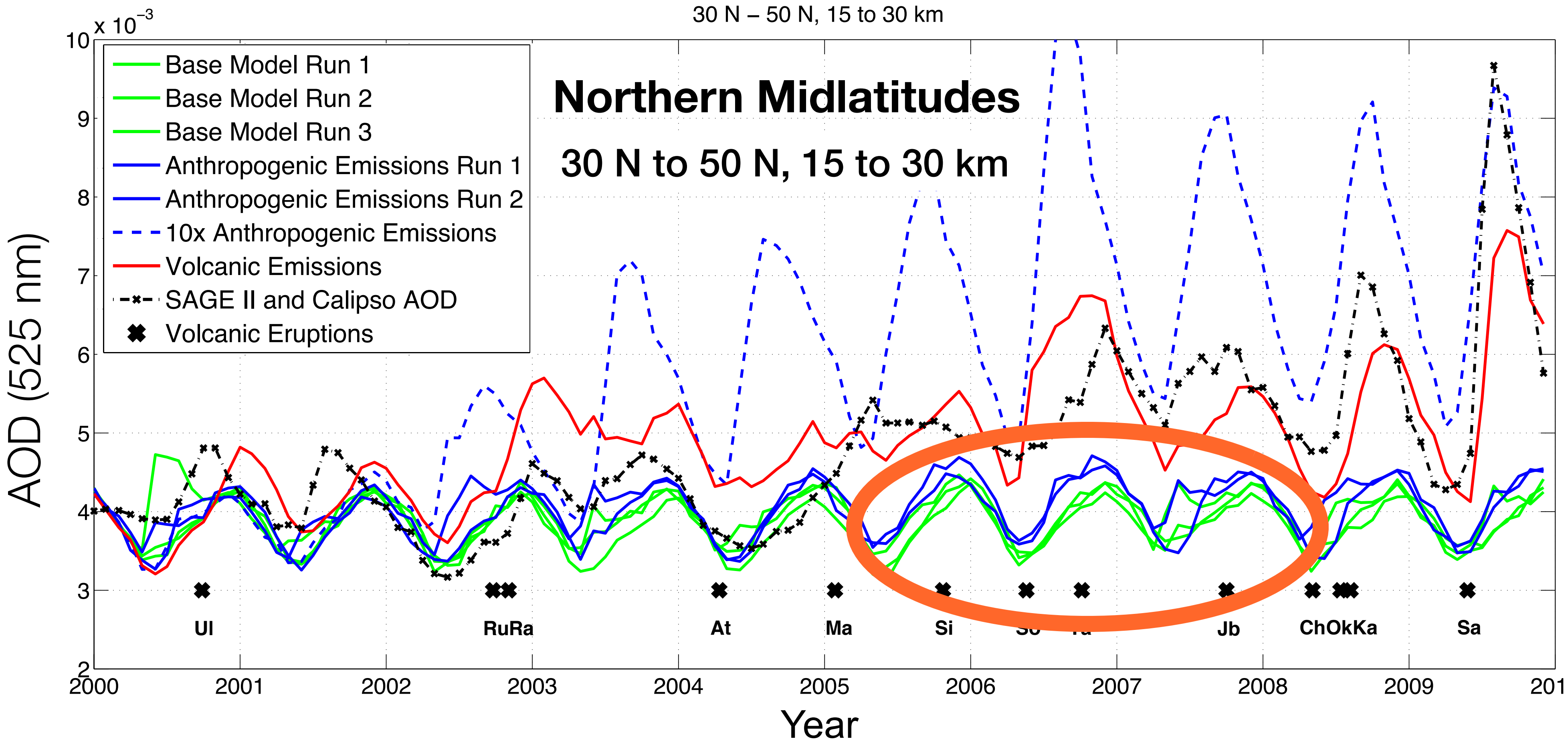
Adapted from Solomon et al. (2011), The Persistently Variable "Background" Stratospheric Aerosol Layer and Global Climate Change, *Science*.

Volcanoes drive stratospheric aerosol variability



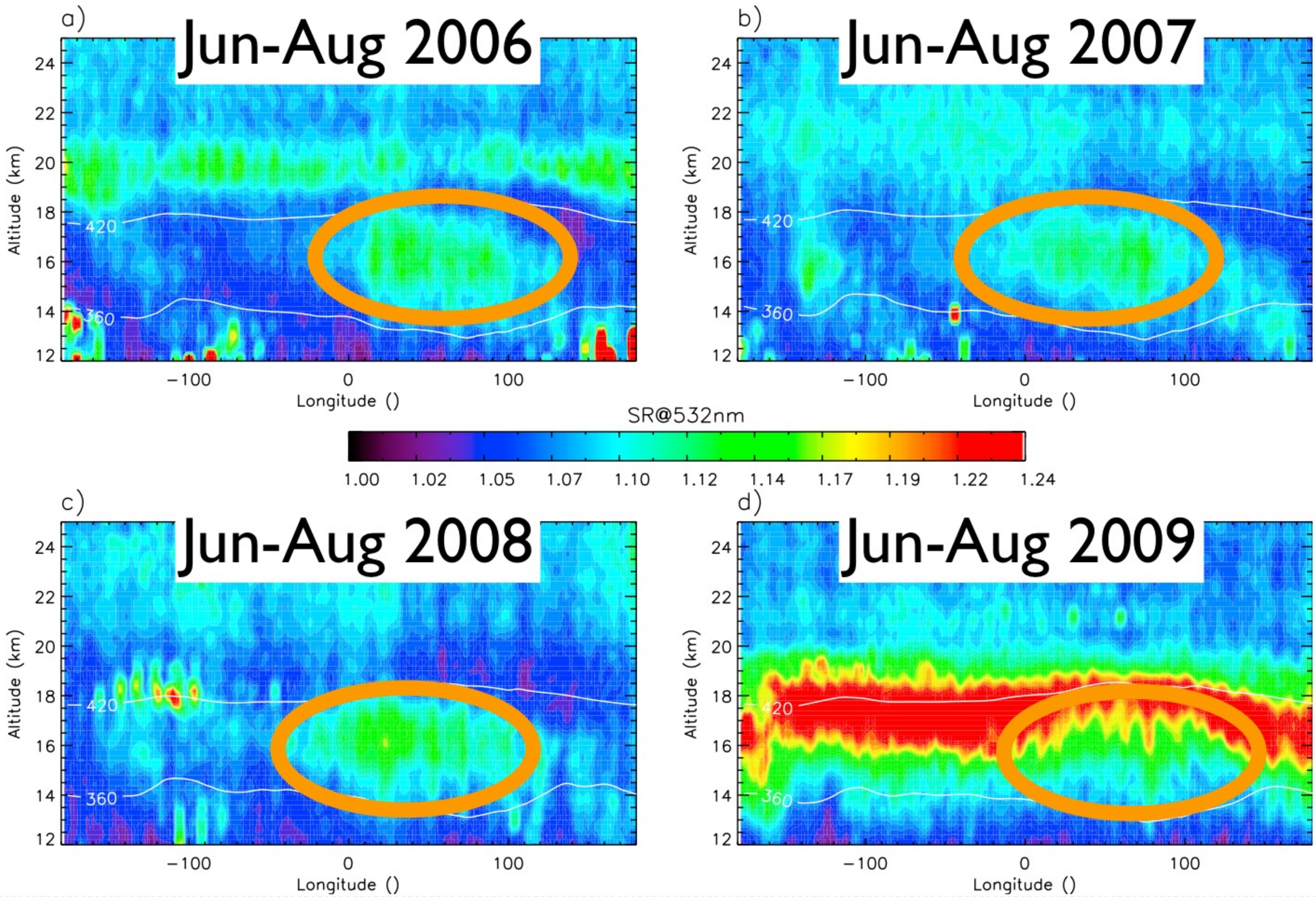
Adapted from Neely, R. R., III et al. (2013), Recent anthropogenic increases in SO2 from Asia have minimal impact on stratospheric aerosol, Geophys. Res. Lett, n/a–n/a, doi:10.1002/grl.50263.

Anthropogenic emissions may have some influence



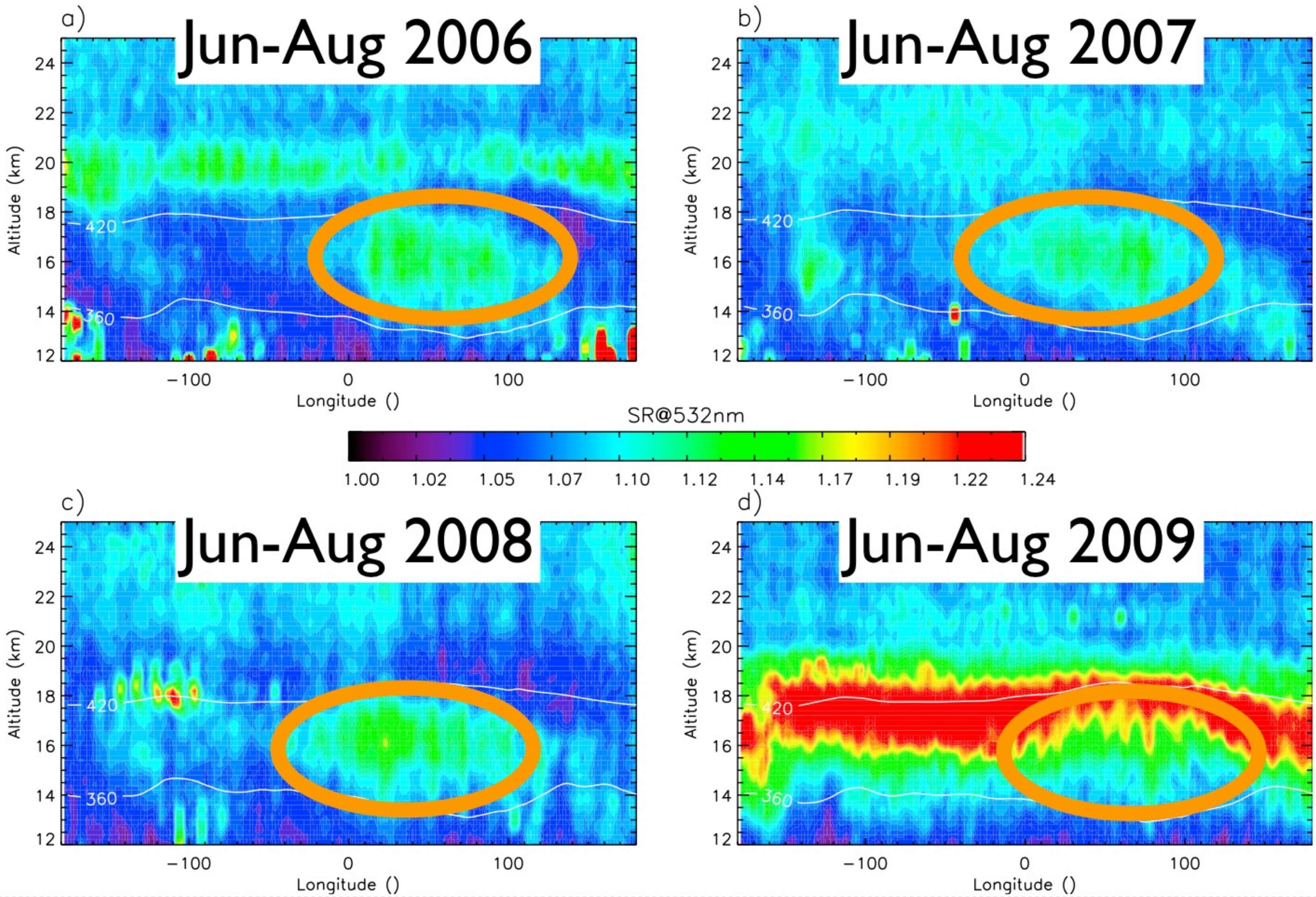
Recent variability in stratospheric aerosol is mostly due to volcanic eruptions but influences from anthropogenic emissions may have an impact on the lower troposphere (Neely et al. GRL 2013)

The Asian Tropopause Aerosol Layer (ATAL)
Mean Scattering Ratio (SR) from CALIPSO at 532 nm from 15N to 45N



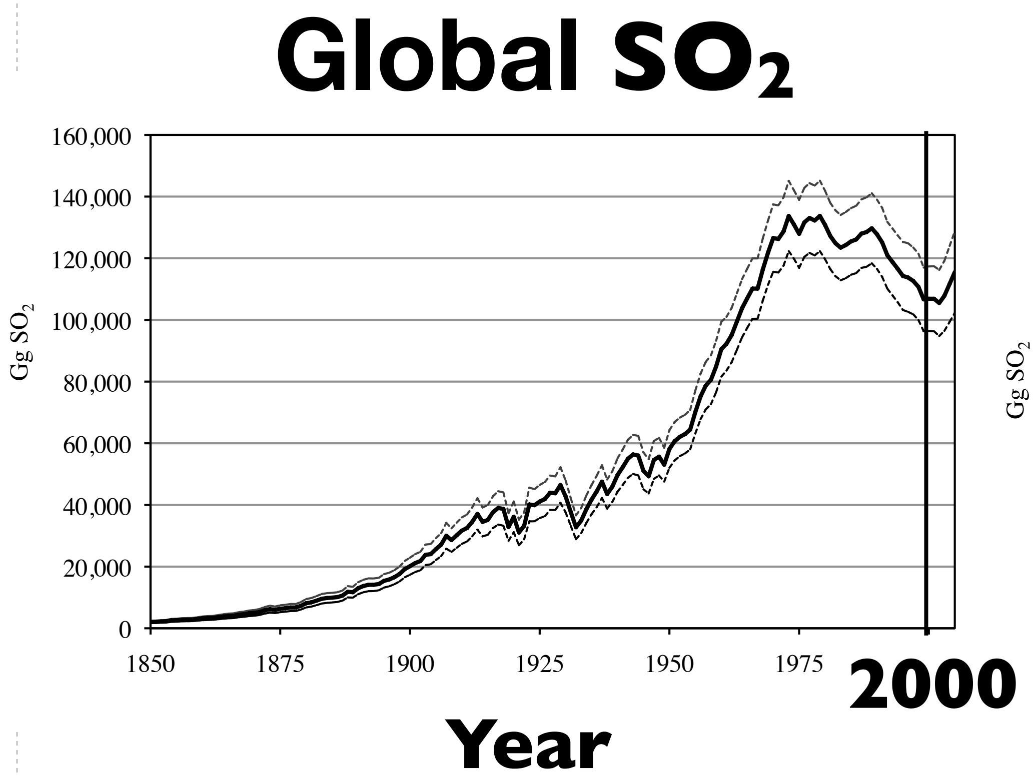
Adapted from: Vernier, J. P., L. W. Thomason, and J. Kar (2011), CALIPSO detection of an Asian tropopause aerosol layer, Geophys. Res. Lett, 38(7), doi:10.1029/2010GL046614.

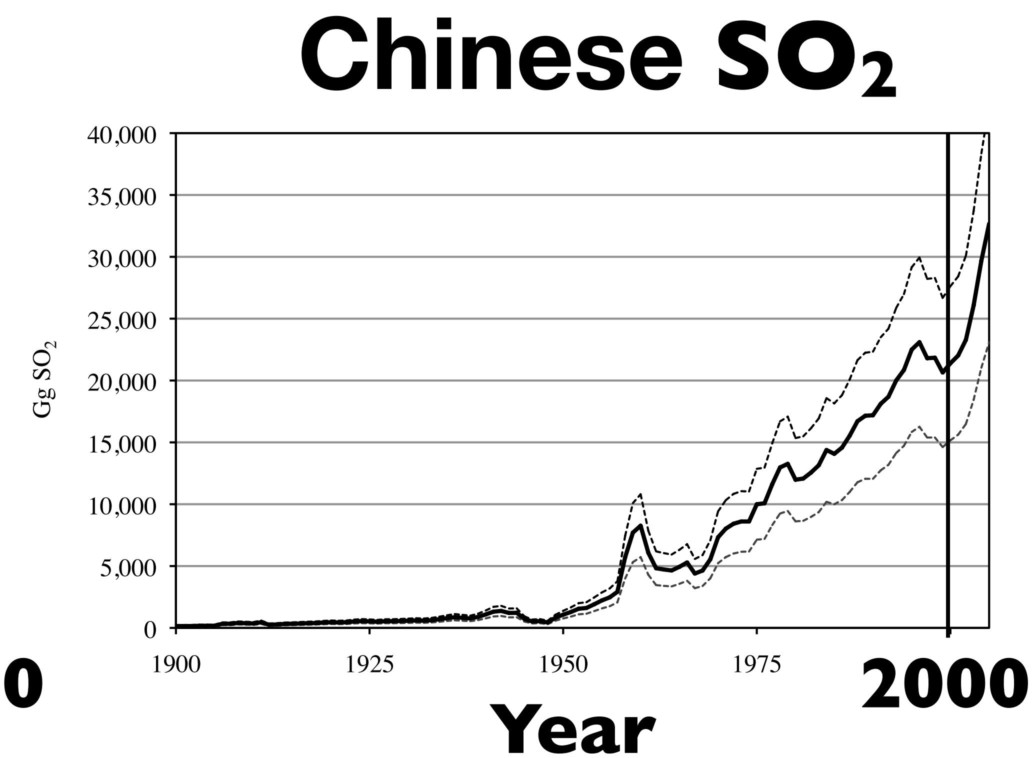
Current observations cannot attribute the observed variability to sources



Adapted from: Vernier, J. P., L. W. Thomason, and J. Kar (2011), CALIPSO detection of an Asian tropopause aerosol layer, Geophys. Res. Lett, 38(7), doi:10.1029/2010GL046614.

Hypothesis: Asian Emissions Lead to ATAL



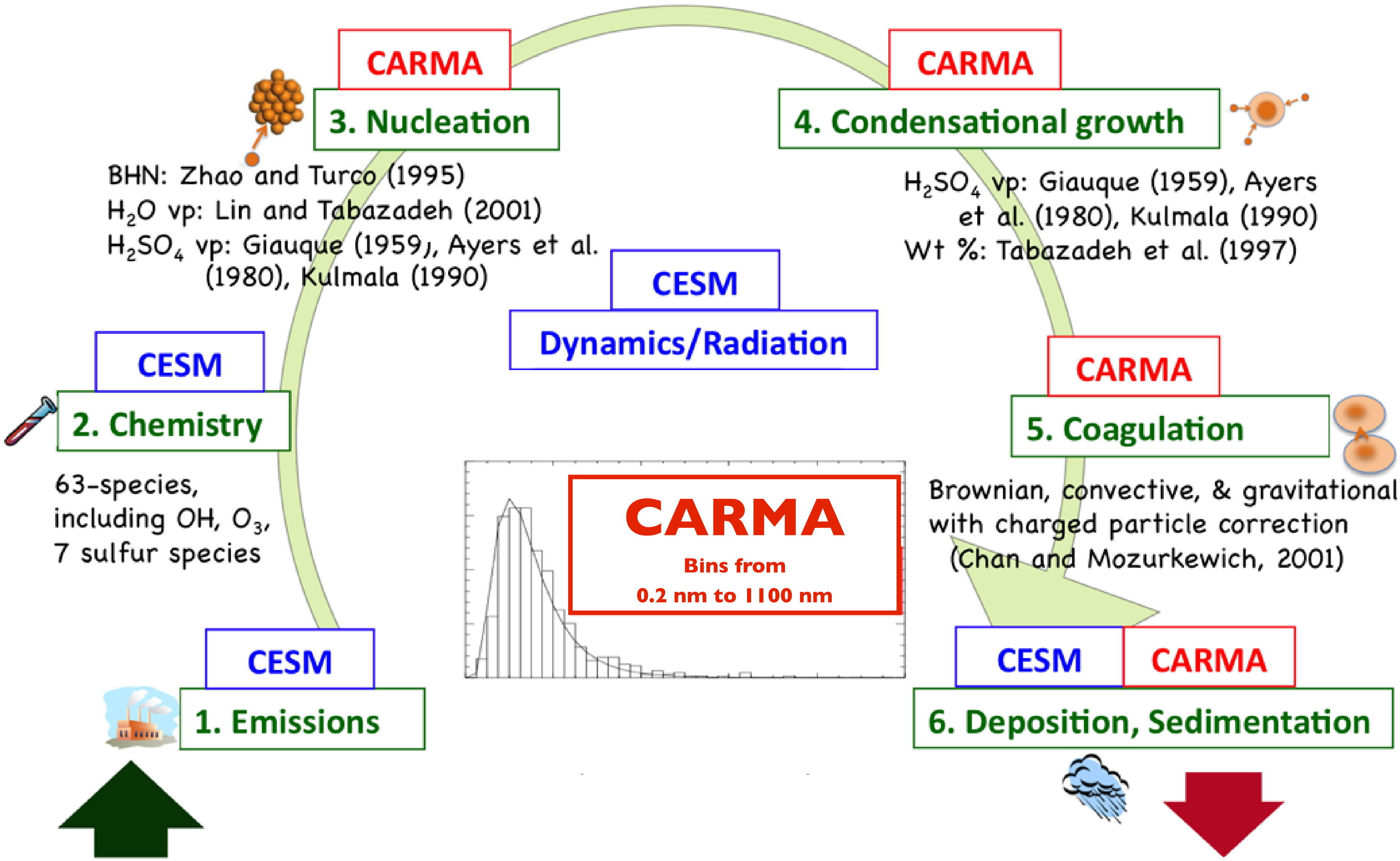


Increases in other constituents of carbonaceous aerosol (POA, SOA) are also thought to contribute

Adapted from Smith et al. ACP 2011

The Model:

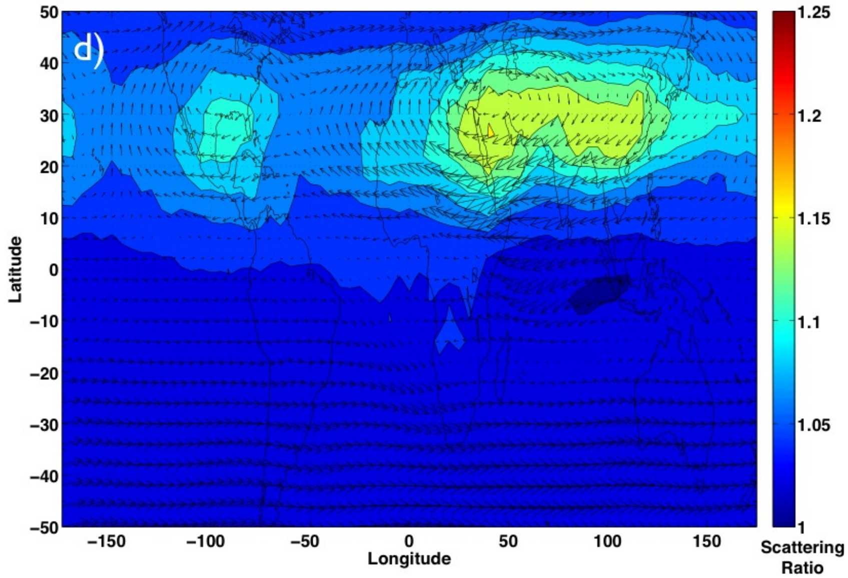
Non-Sulfates: Primary Organics (POA), Secondary Organics (SOA), Black Carbon (BC), Wind Blown Dust, Sea Salt
Sulfates: Meteoritic Smoke, Pure Sulfates, Mixed Sulfates

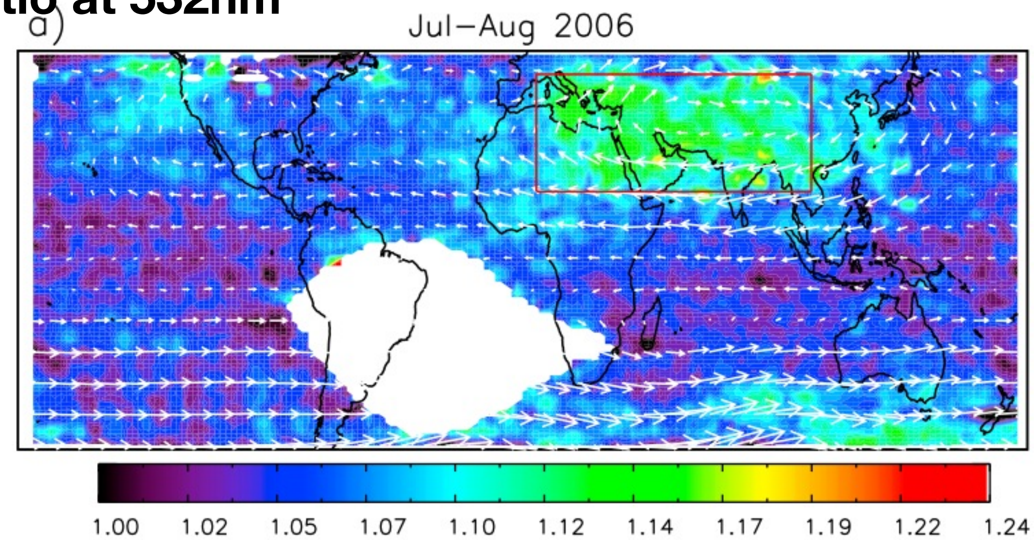


Neely, R. R., III, J. M. English, O. B. Toon, S. Solomon, M. Mills, and J. P. Thayer (2011), Implications of extinction due to meteoritic smoke in the upper stratosphere, Geophys. Res. Lett, 38(24), doi:10.1029/2011GL049865.

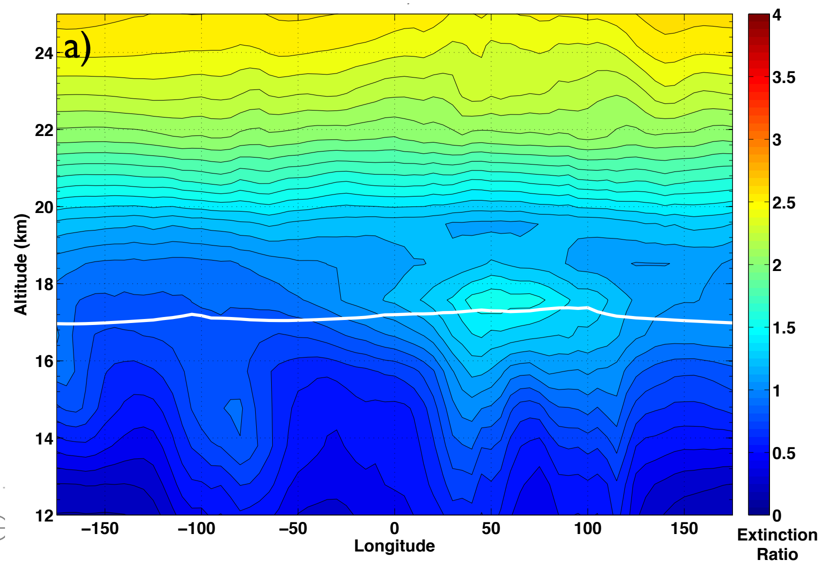
Model Captures Spatial Pattern and Magnitude of Observations

Mean 100 hPa JJA Lidar Backscatter Ratio at 532nm

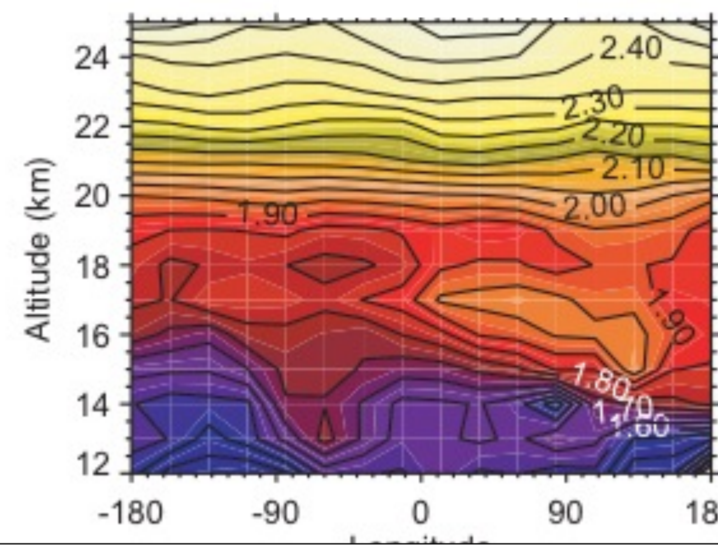




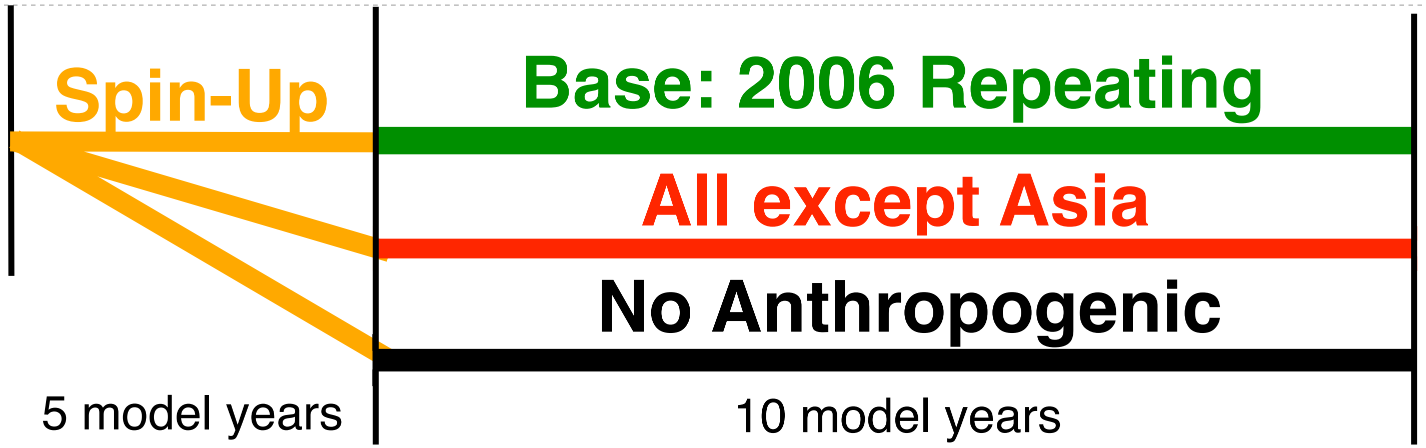
Mean 1020 nm Extinction Ratio from 14N to 46N, June-August



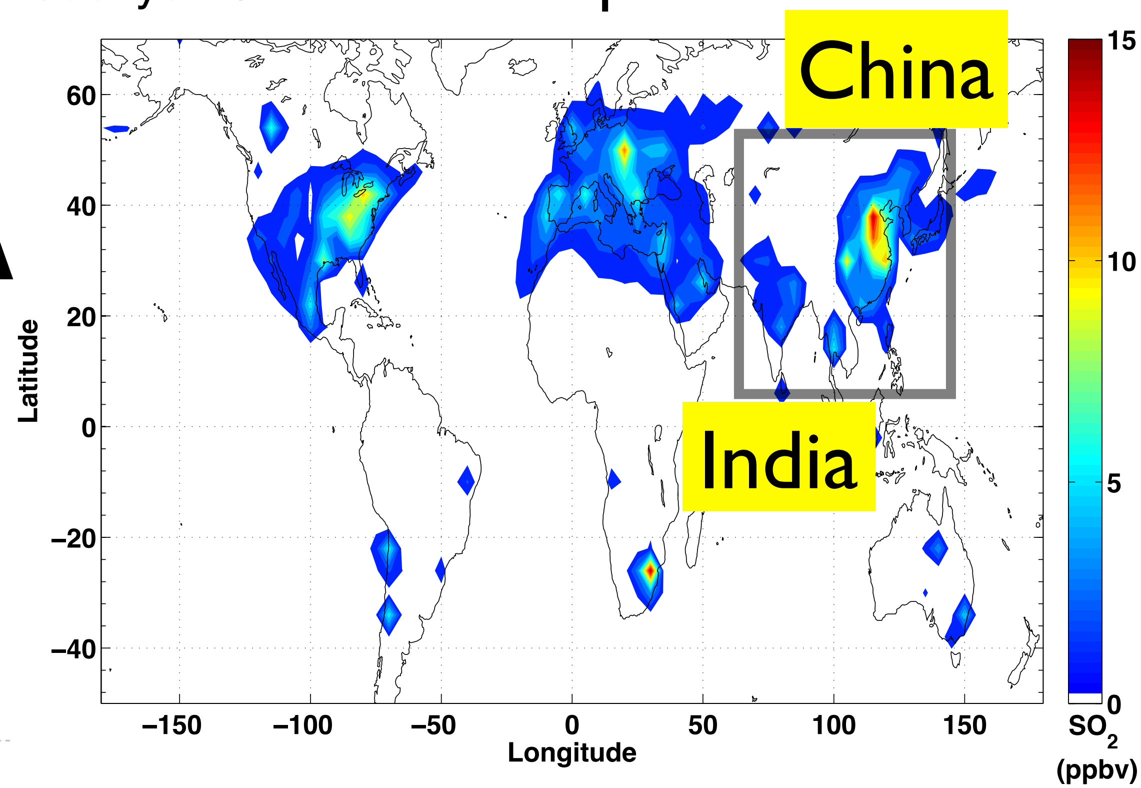
SAGE II



Model Experiment: SO_2 Schemes

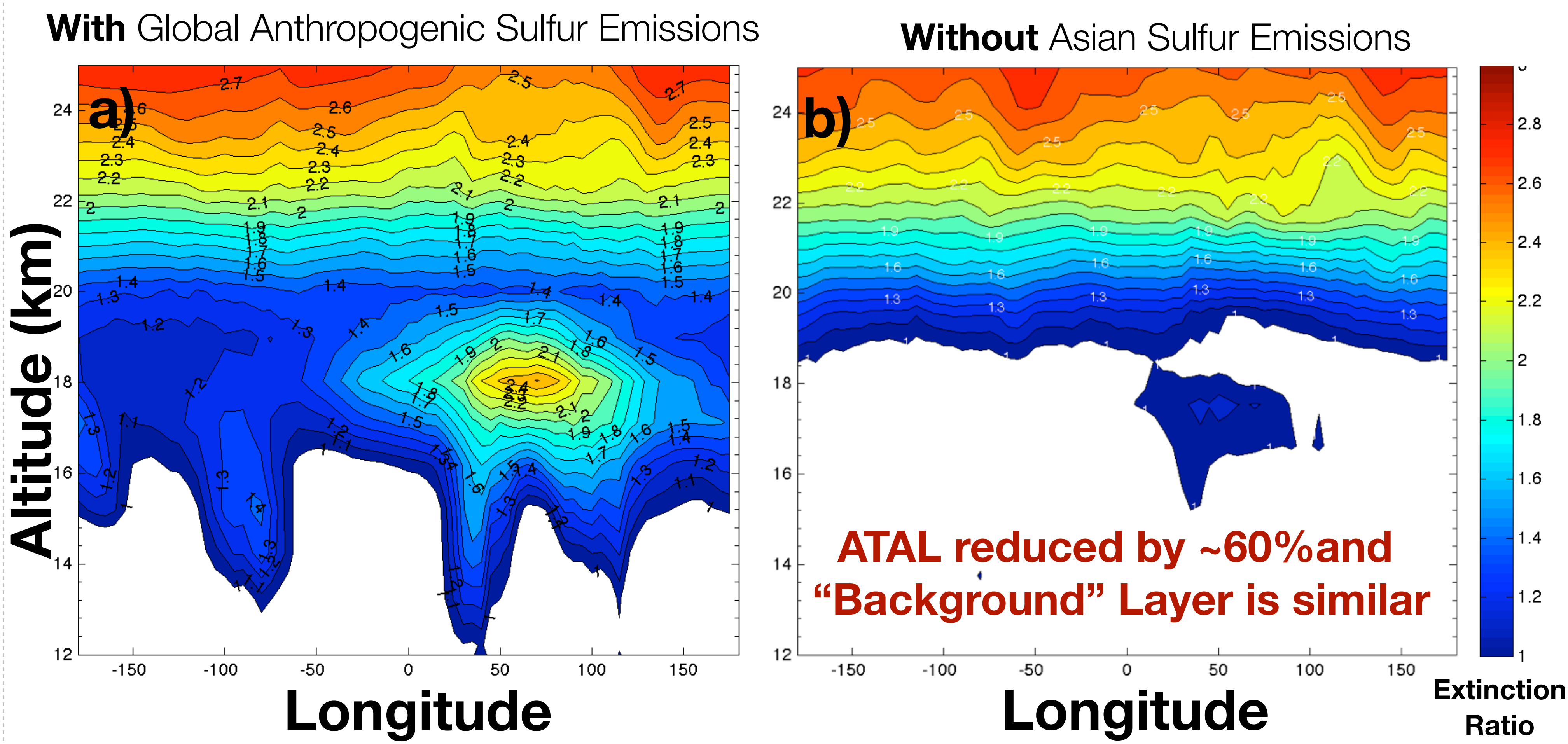


Baseline Surface JJA SO_2 Concentrations



Asian Anthropogenic Influence on the ATAL

Modeled Mean 1020 nm Extinction Ratio from 14N to 46N, June-August

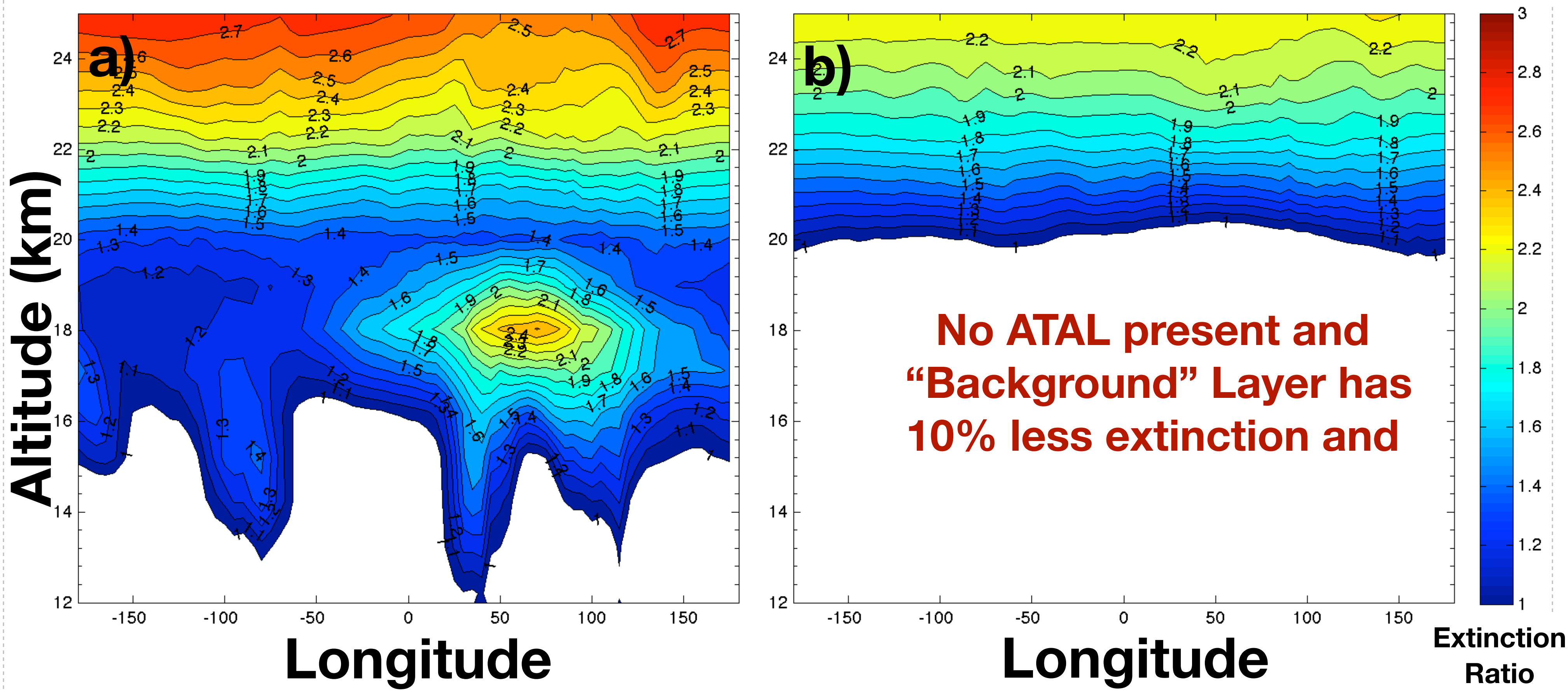


Total Anthropogenic Influence

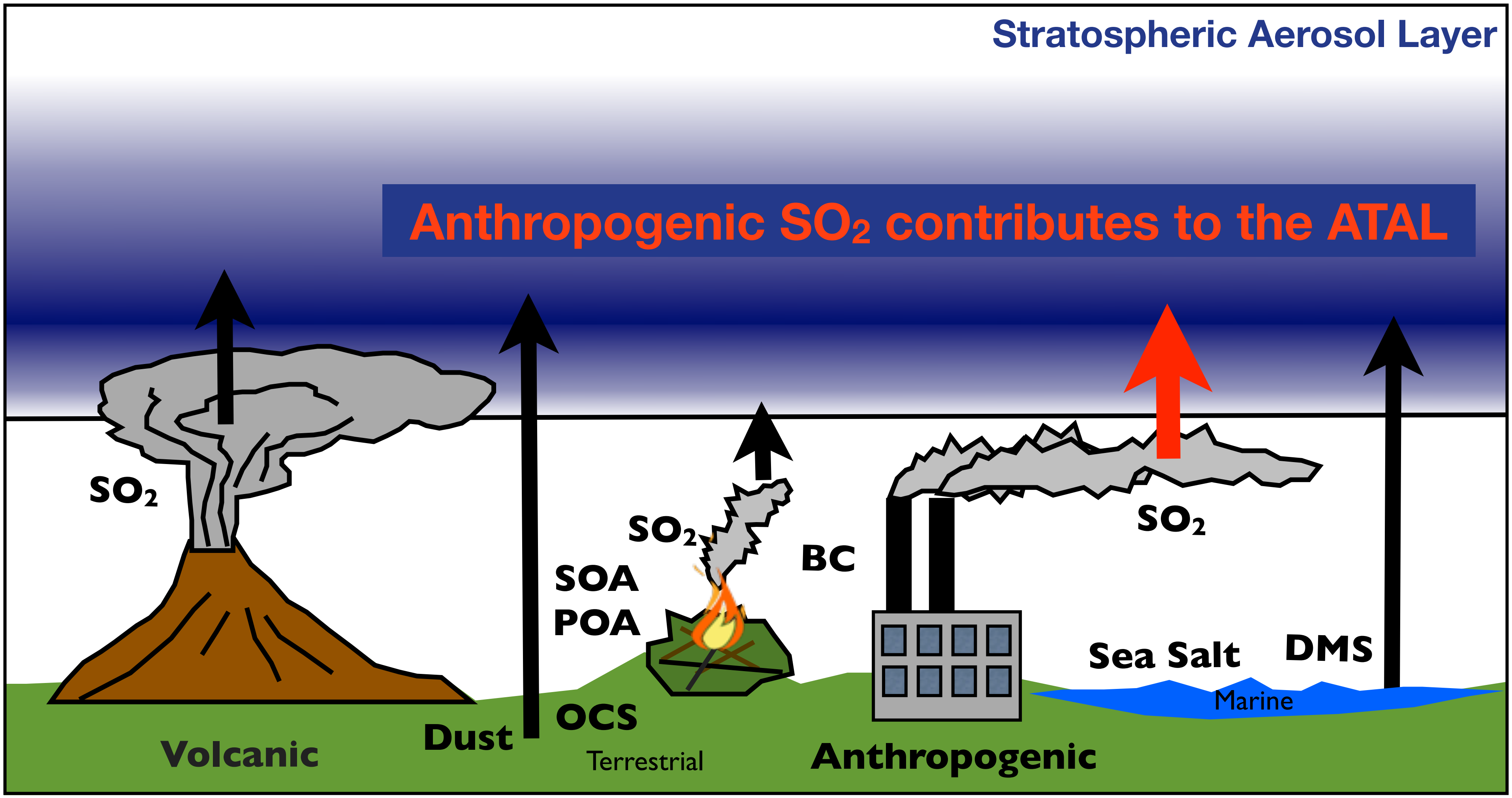
Modeled Mean 1020 nm Extinction Ratio from 14N to 46N, June thru August

With Global Anthropogenic Sulfur Emissions

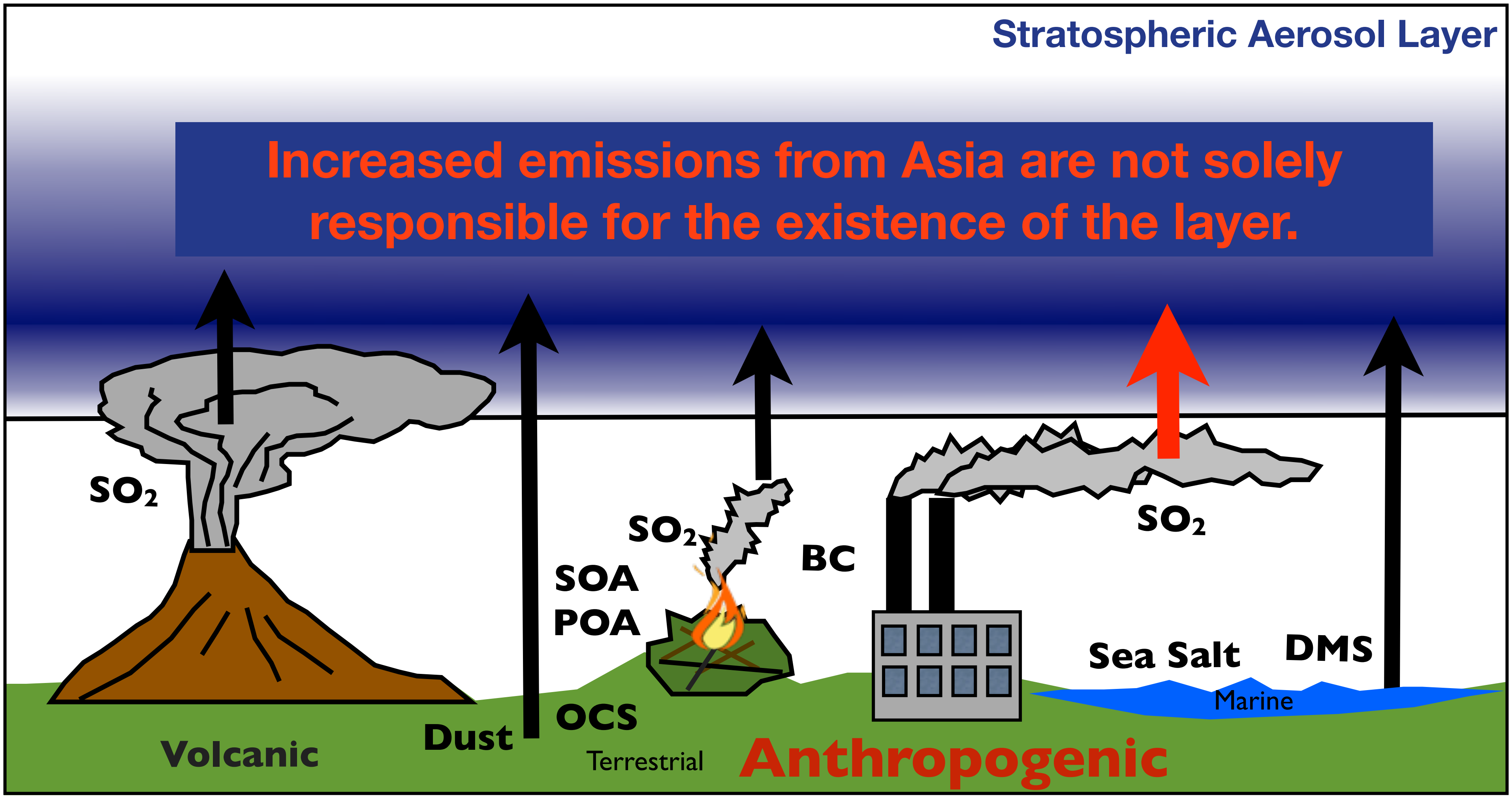
Without Global Anthropogenic Sulfur Emissions



Conclusions



Conclusions



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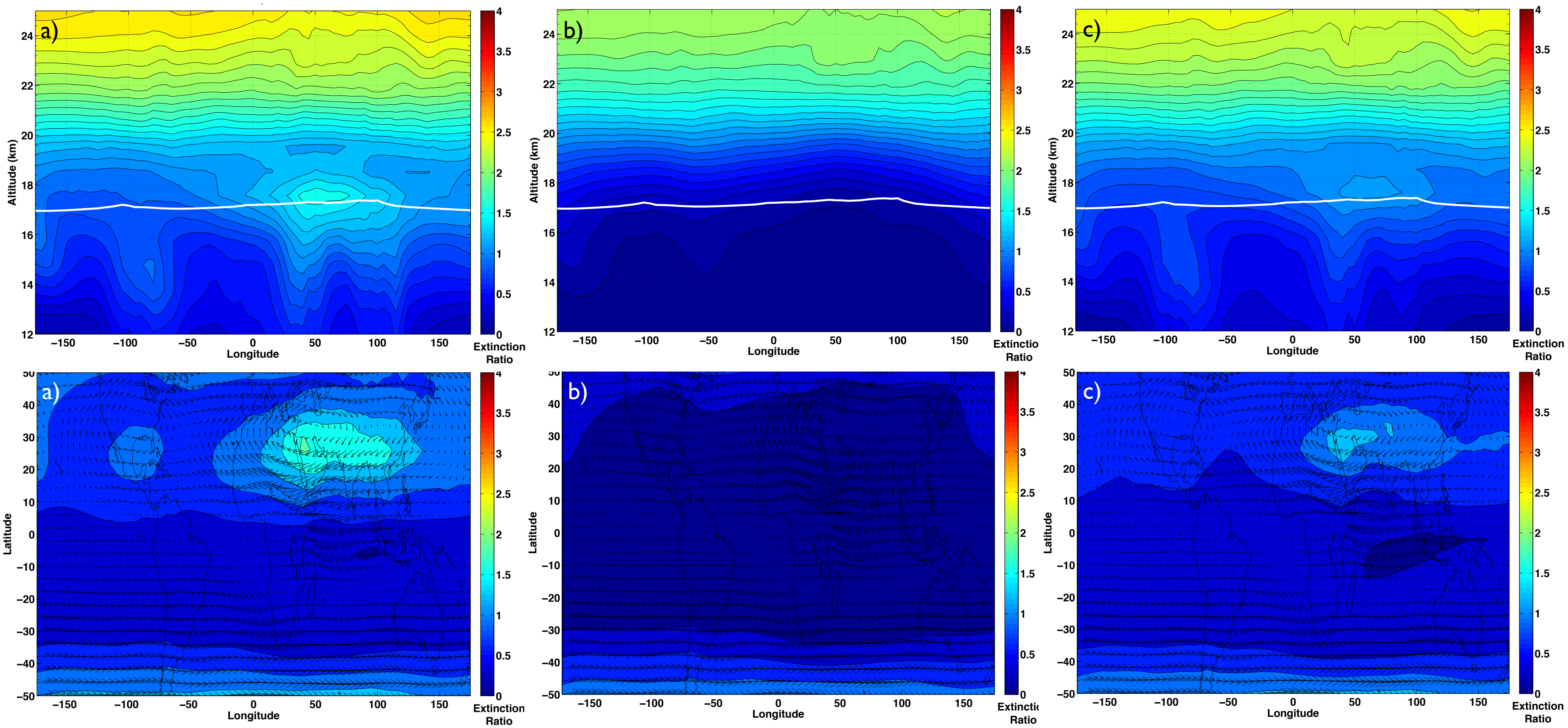
Anthropogenic Influence Summary

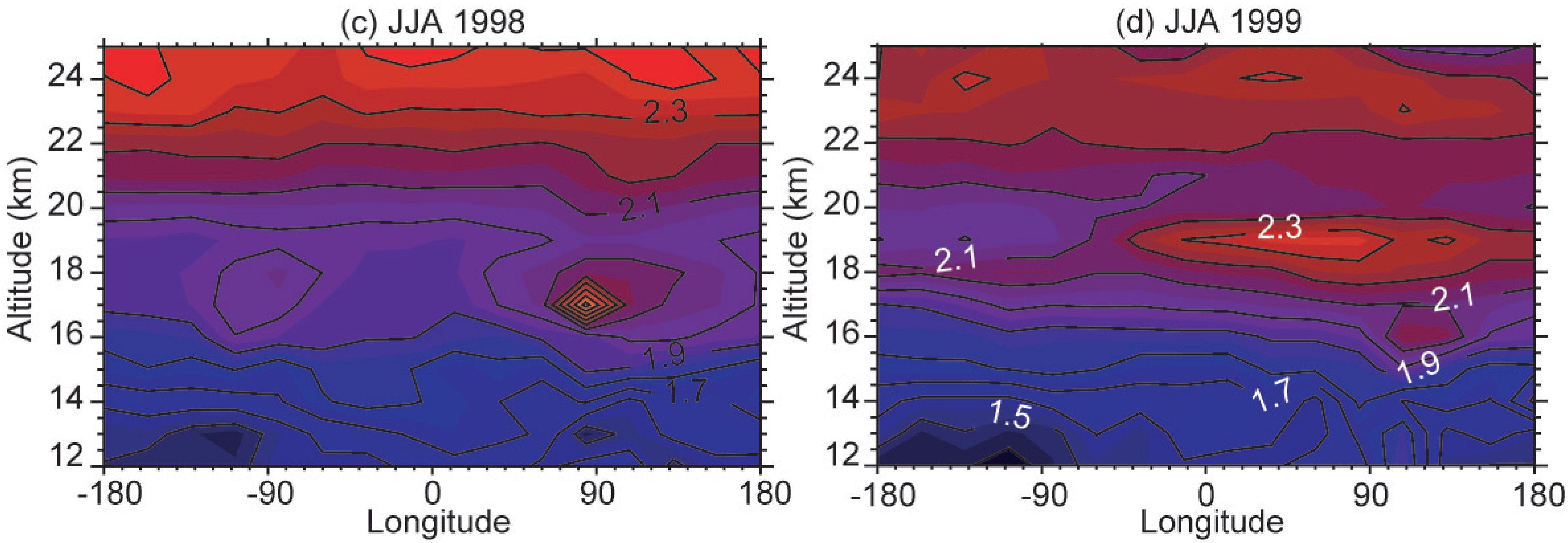
2000	No Anthropogenic SO_2	No China and India



	No ATAL present	ATAL reduced by ~60%

First Observed in 1998

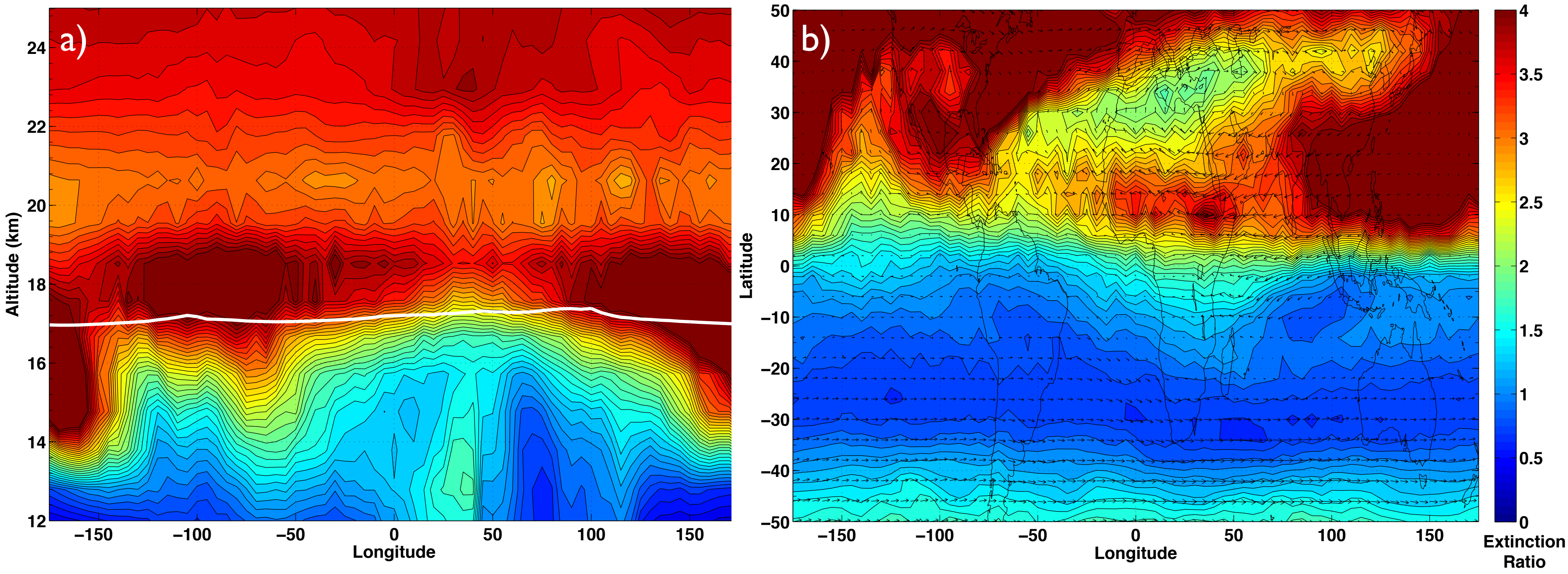
Median 1020 nm Extinction Ratio Observed by SAGE II from 15N to 45N, June thru August



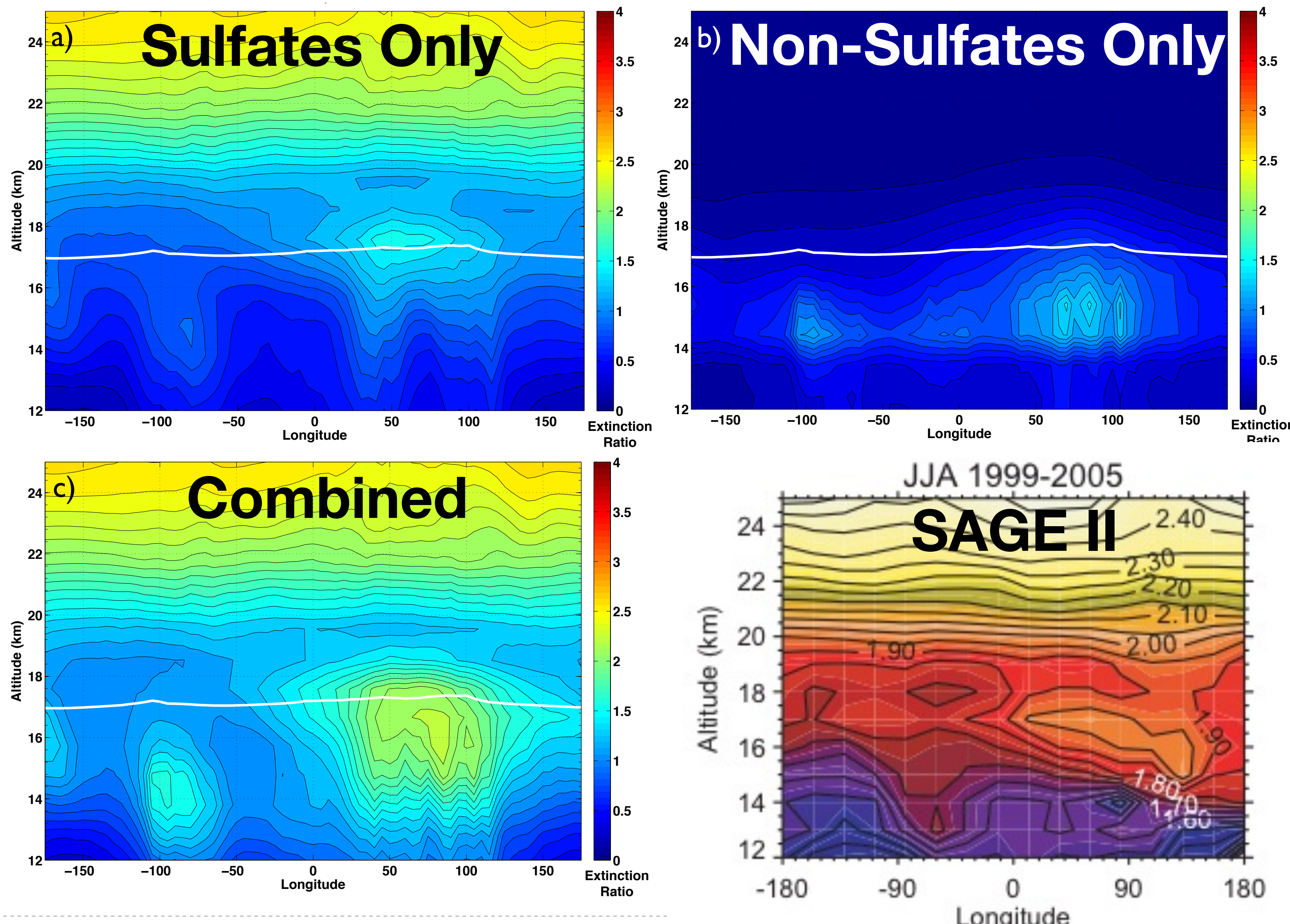
Hypothesis: Asian Emissions Lead to ATAL

Plot adapted from Thomason, L. W. and Vernier, J.-P.: Improved SAGE II cloud/aerosol categorization and observations of the asian tropopause aerosol layer: 1989–2005, Atmos. Chem. Phys. Discuss., 12, 27521-27554, doi:10.5194/acpd-12-27521-2012, 2012.

Why was the ATAL Unobserved Until 1998?

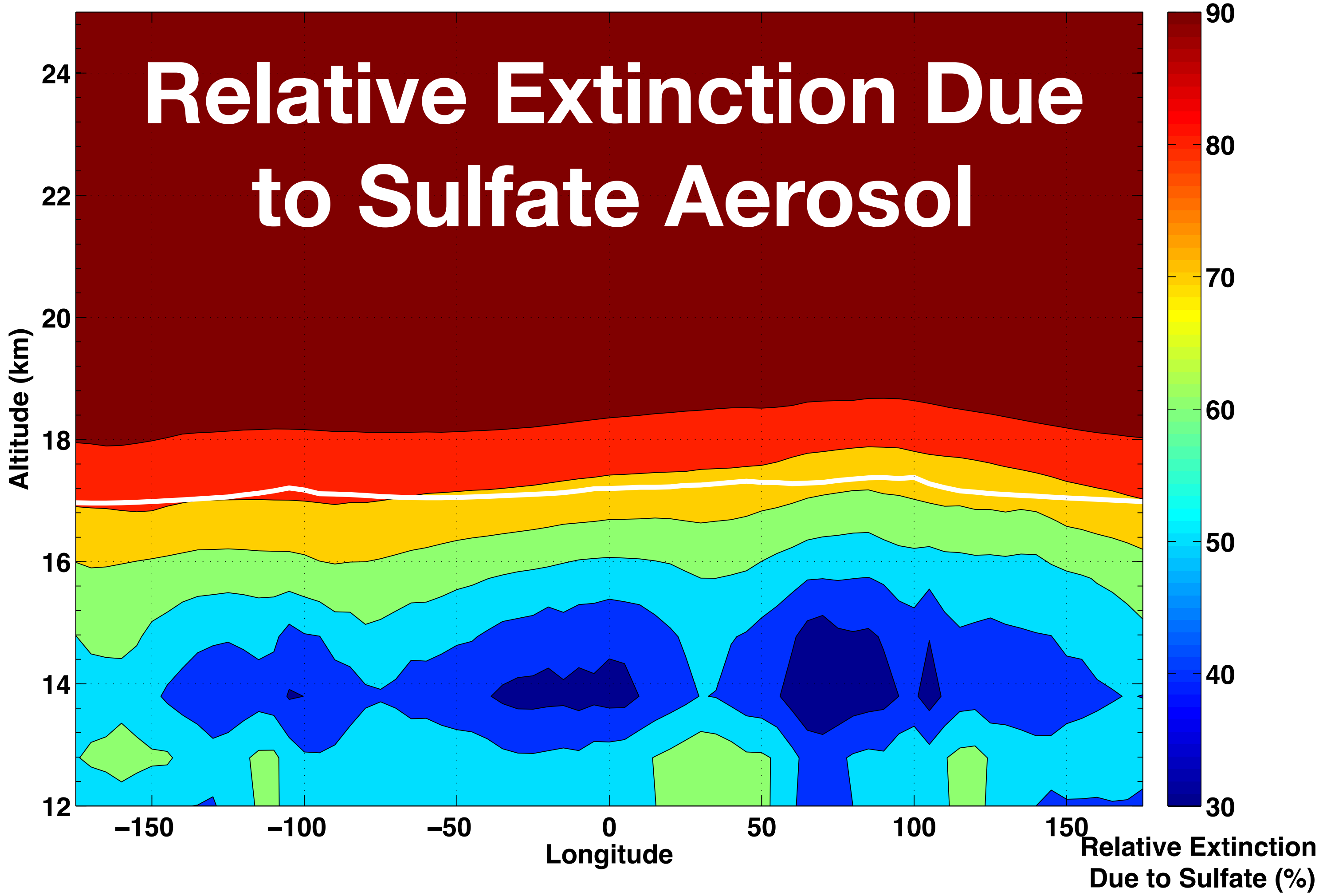


Vertical Comparison to SAGE II (14N to 46N)



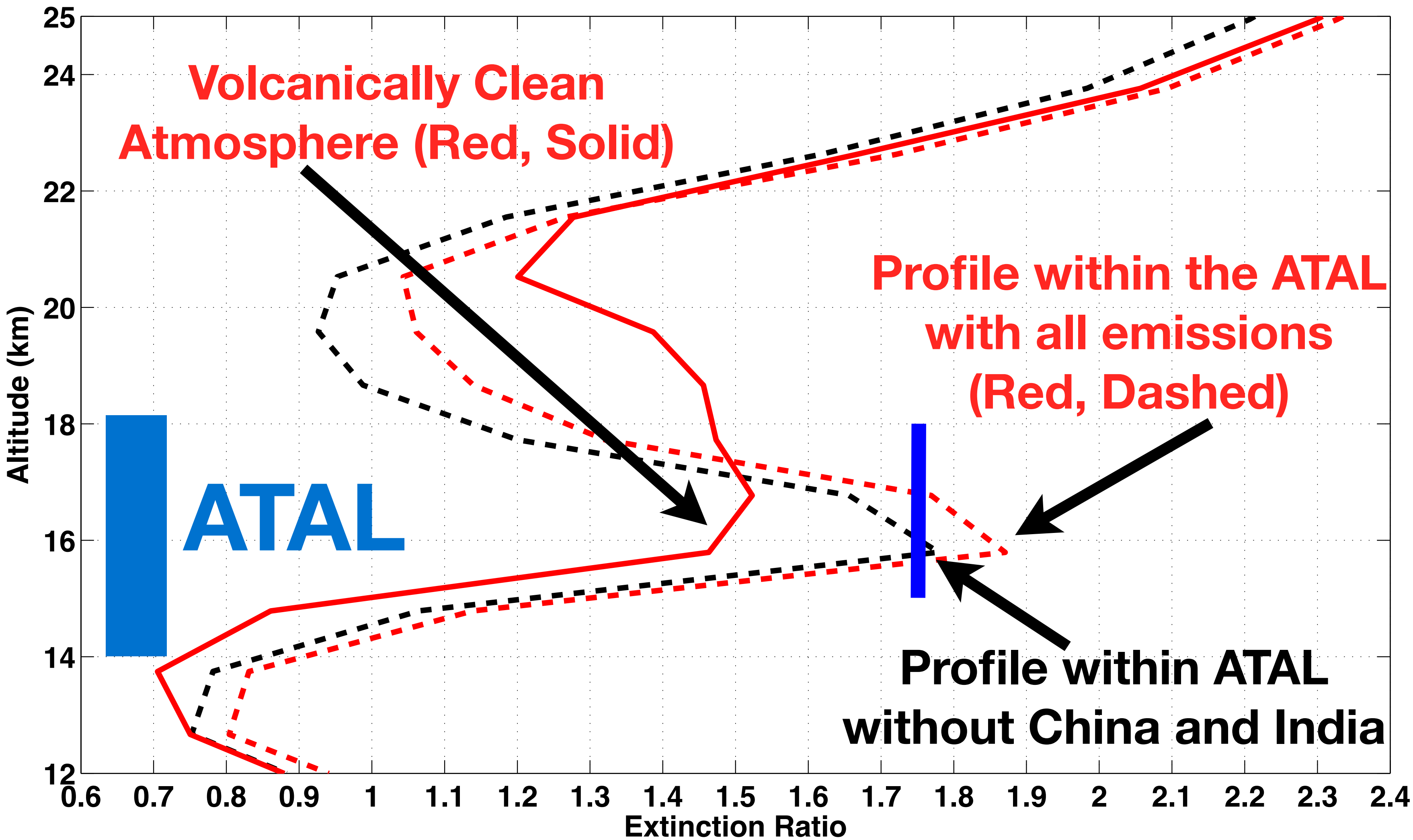
All plots show mean JJA extinction ratio at 1020 nm

ATAL Composition: Sulfate Splits With Non-Sulfate at Tropopause



Agrees with near tropopause *in situ* observations of CARIBIC and PALMS (Papaspiropoulos et al., 2002 and Murphy et al. 1998; 2007)

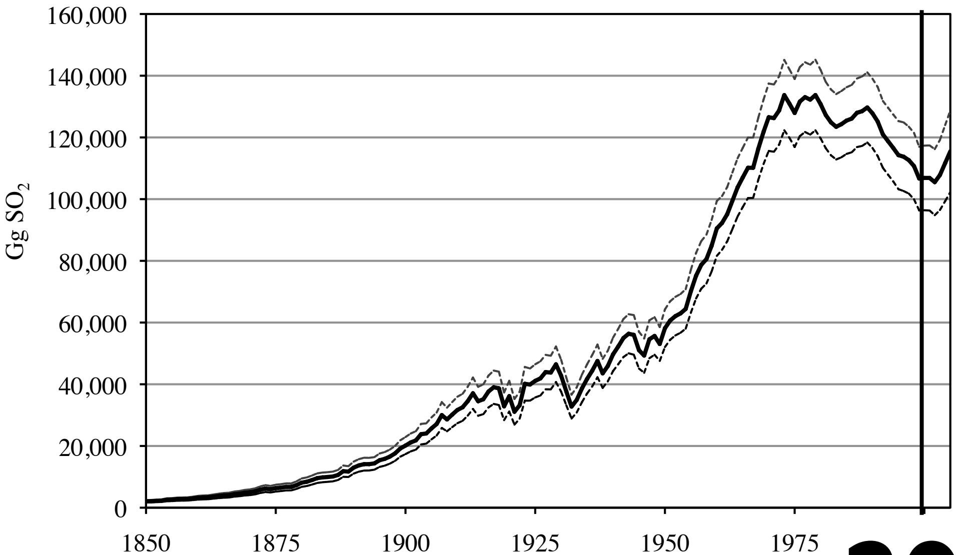
Why was the ATAL Unobserved Until 1998?



Lower Bound of Observed 1989 Extinction Ratio from 16 to 18 km
(Thomason et al, 1997)

Hypothesis: Asian Emissions Lead to ATAL

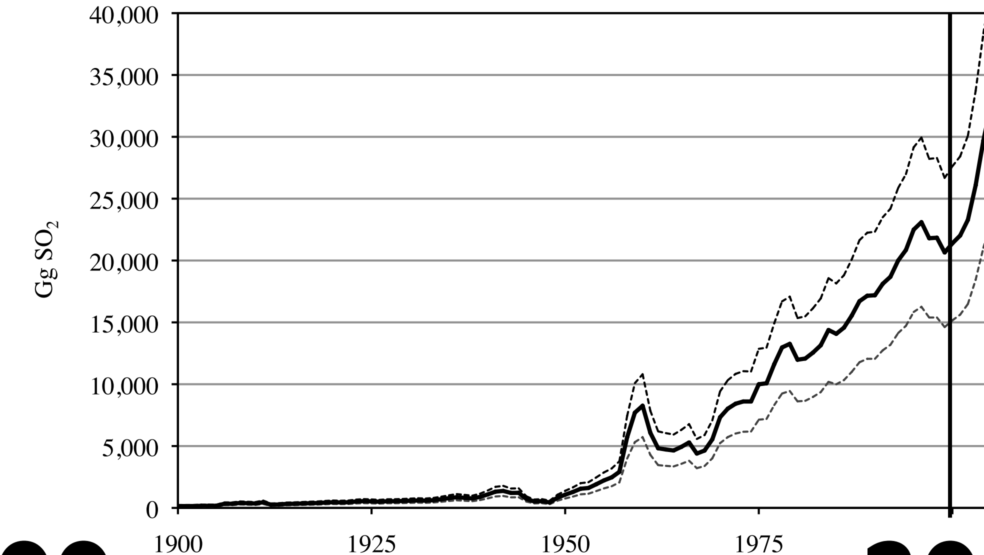
Global SO_2



2000

Year

Chinese SO_2



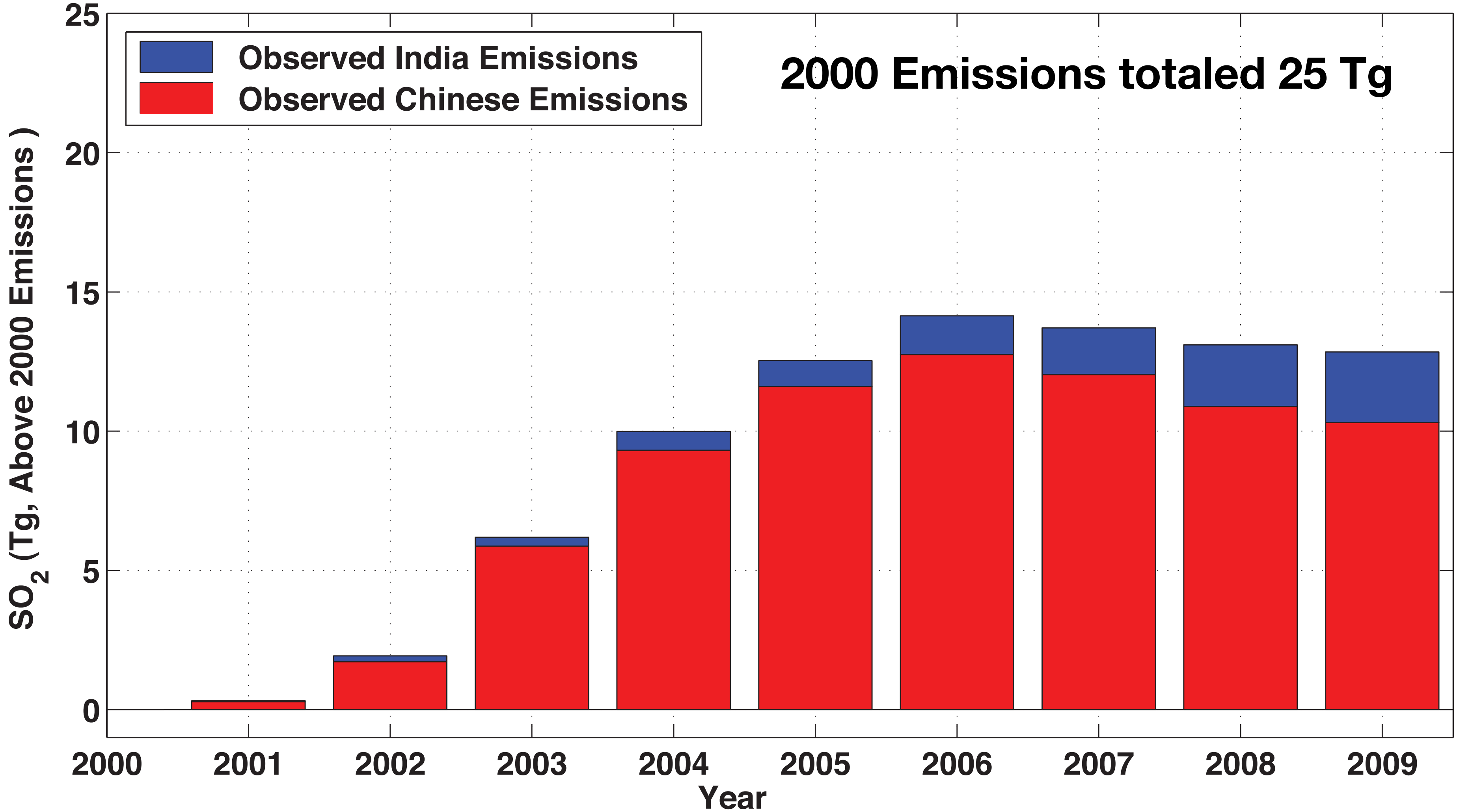
2000

Year

Increases in other constituents of carbonaceous aerosol (POA,

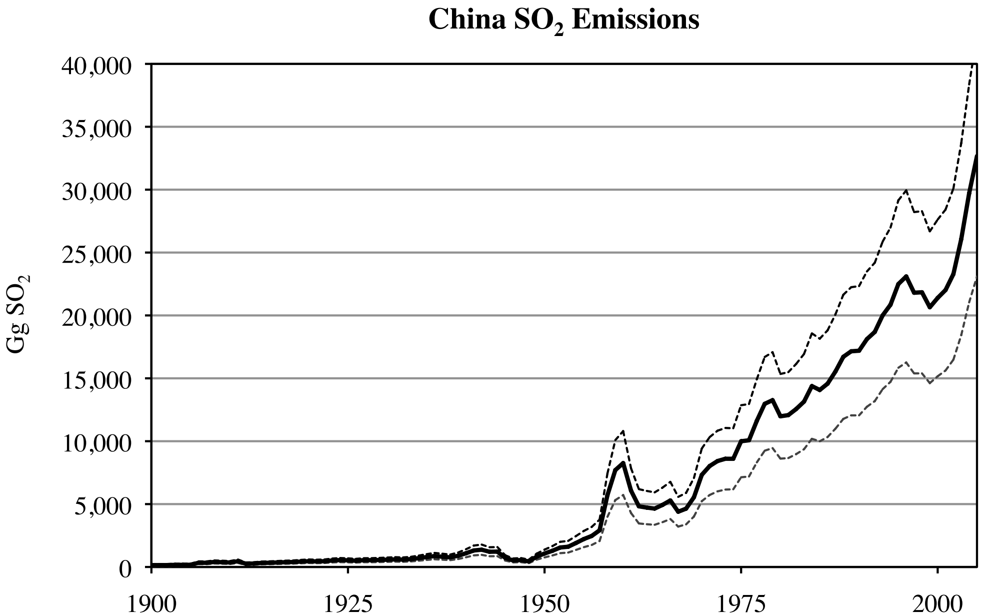
Adapted from Smith et al. ACP 2011

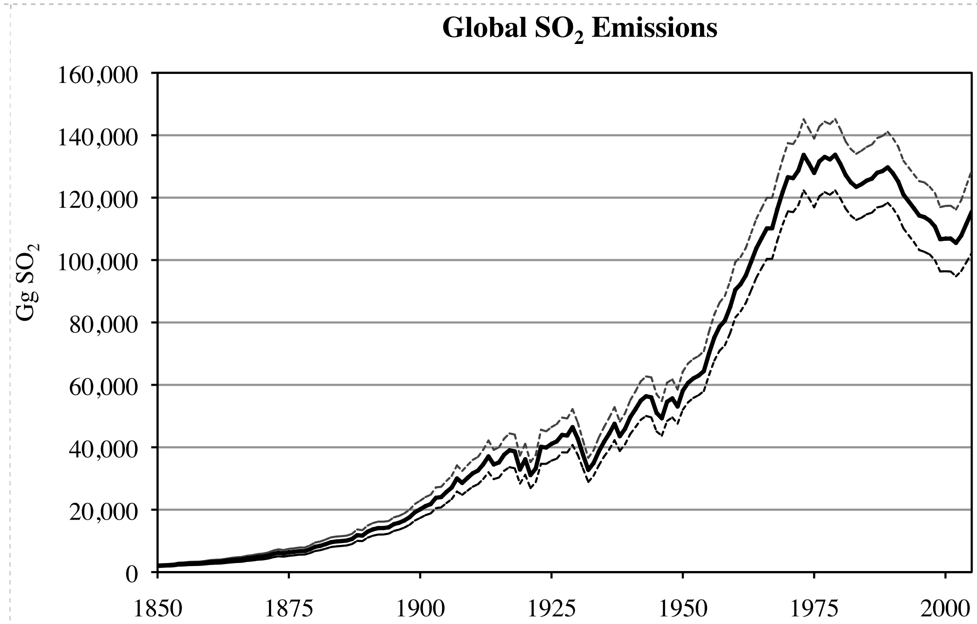
Increases in Asian Anthropogenic Emissions since 2000



Data source: Lu, Z., Zhang, Q. & Streets, D. G. Sulfur dioxide and primary carbonaceous aerosol emissions in China and India. 1996–2010, Atmospheric Chemistry and Physics 11, 9839–9864 (2011).

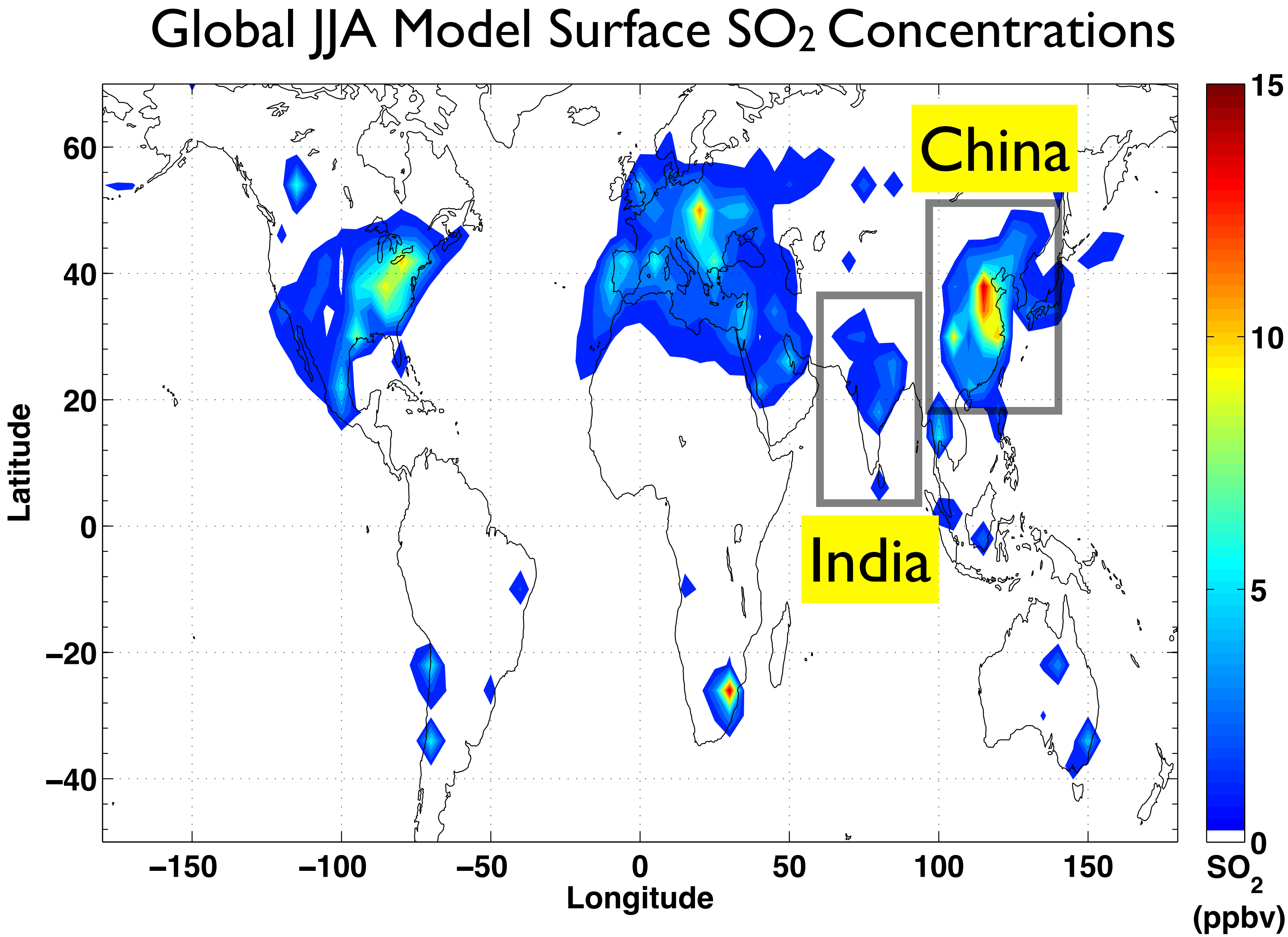
Asian Emissions



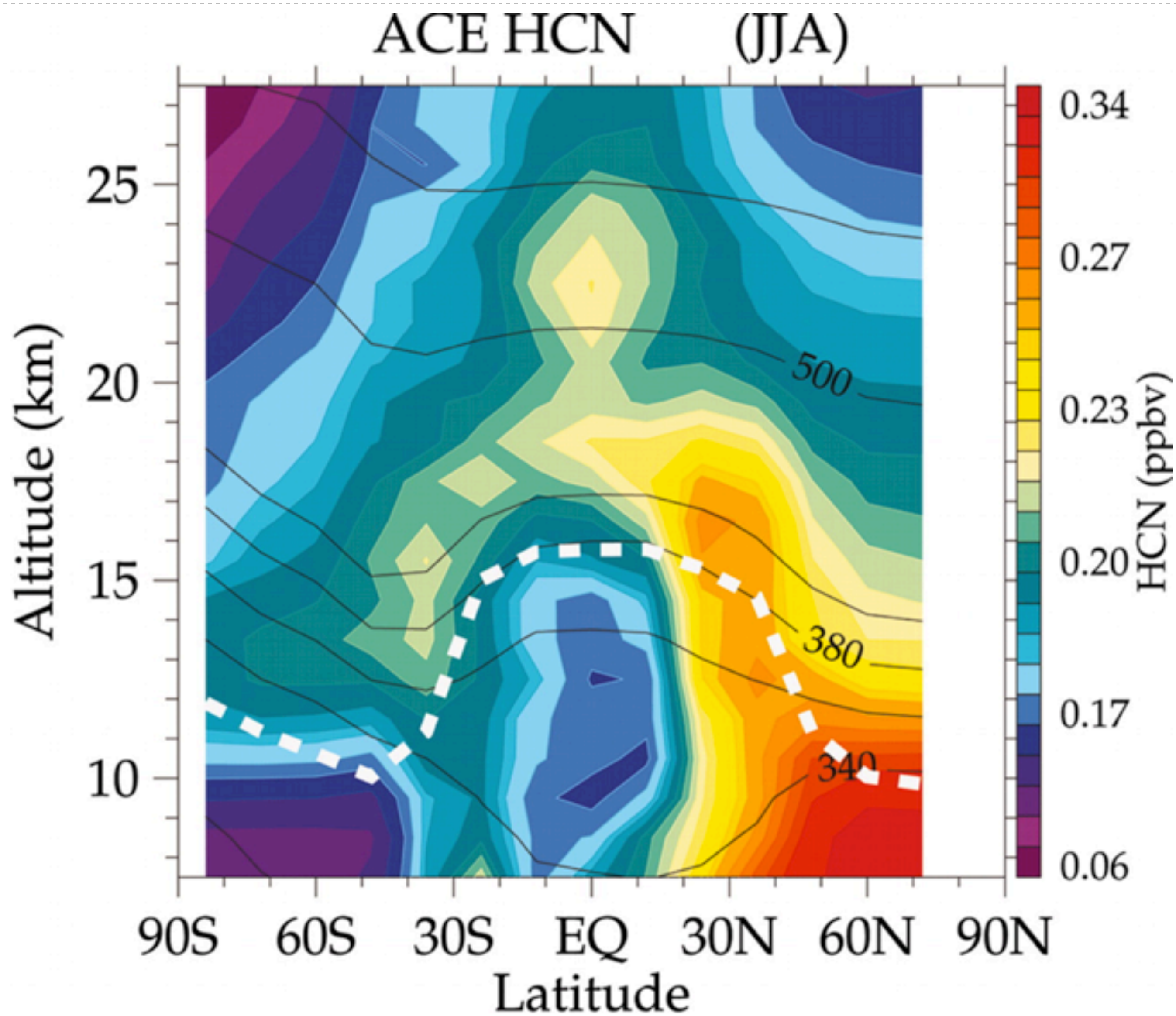


Adapted from Smith, S. J., J. van Aardenne, Z. Klimont, R. J. Andres, A. Volke, and S. Delgado Arias (2011), Anthropogenic sulfur dioxide emissions: 1850–2005, Atmos. Chem. Phys, 11(3), 1101–1116, doi:10.5194/acp-11-1101-2011.

Experiment: Modulate Global Anthropogenic Emissions

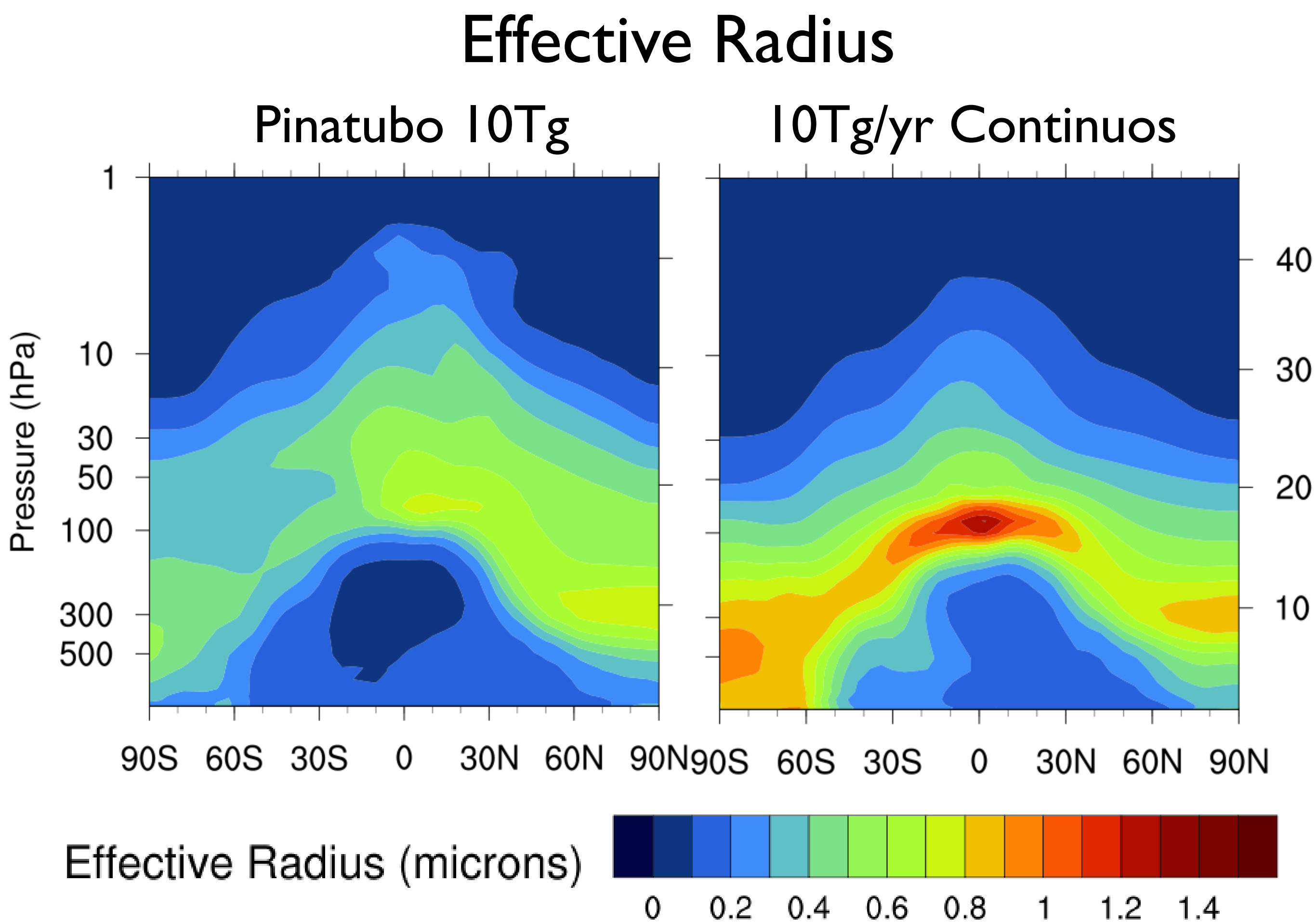


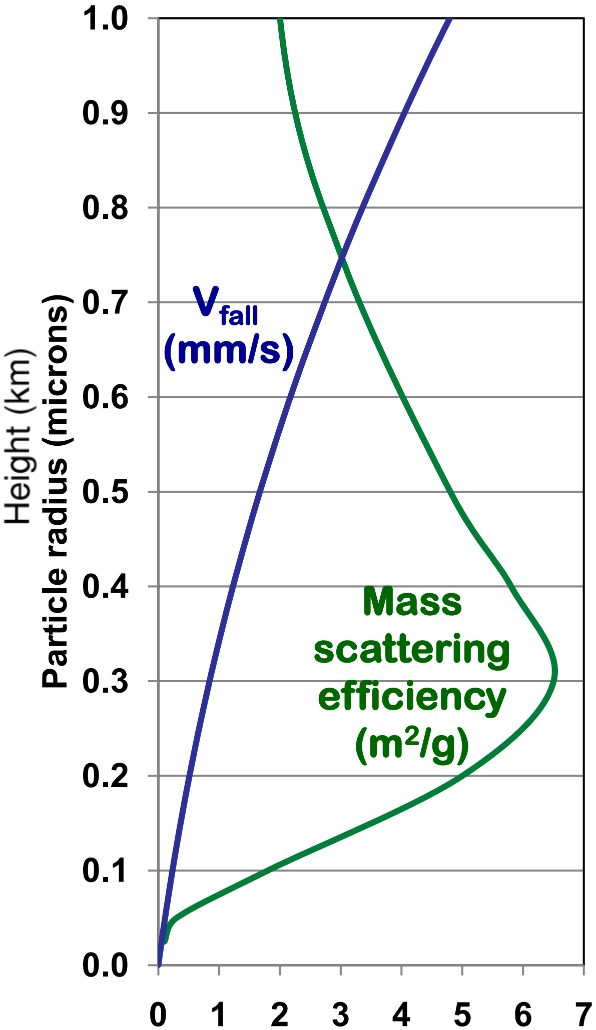
Anthropogenic emissions transported to the stratosphere via the Asian Monsoon



Plot from Randel et al. (2010), Asian Monsoon Transport of Pollution to the Stratosphere, *Science*.

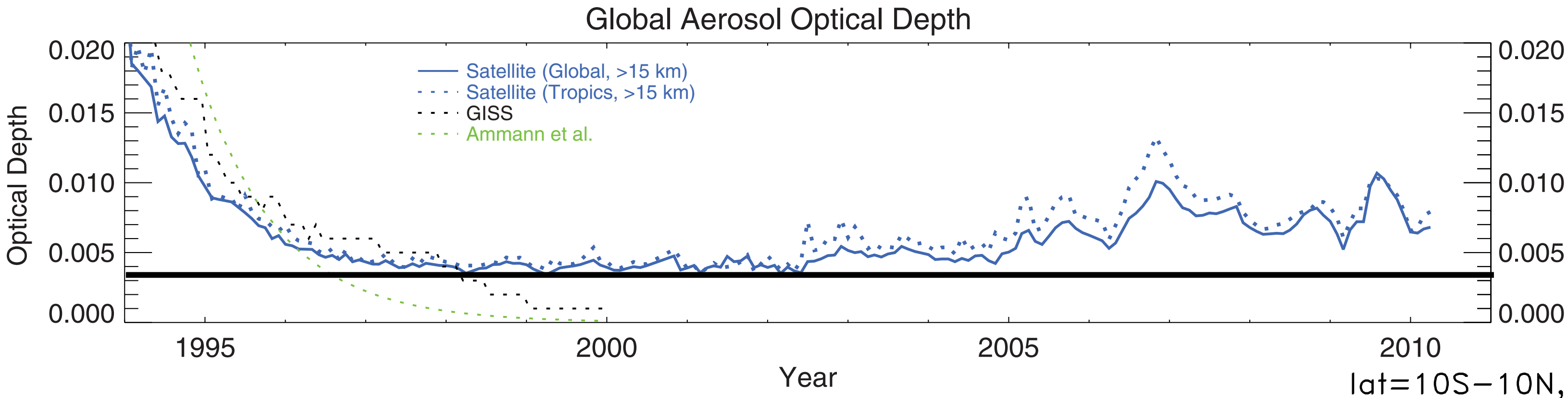
Why Are Volcanoes More Efficient at Making Aerosol?





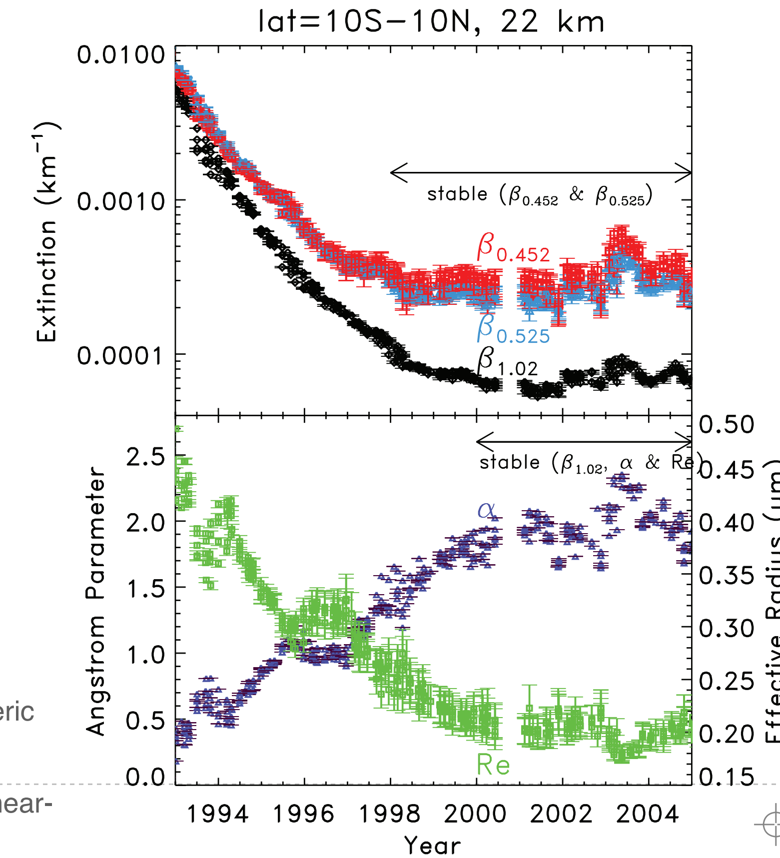
Plots adapted from English, J. M., O. B. Toon, and M. J. Mills (2011), Microphysical simulations of stratospheric sulfur geoengineering, *J. Geophys. Res.*, under review.

2000 to 2010 is an unprecedented "background" period



Layer became stable only in 2000.

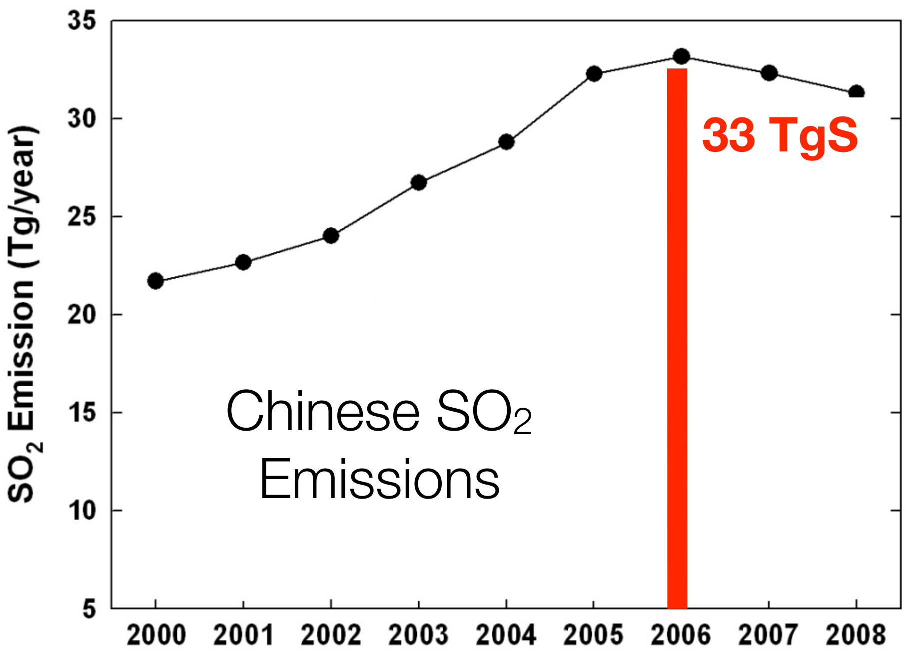
Previous observations will be influenced by 1991 Pinatubo eruption.



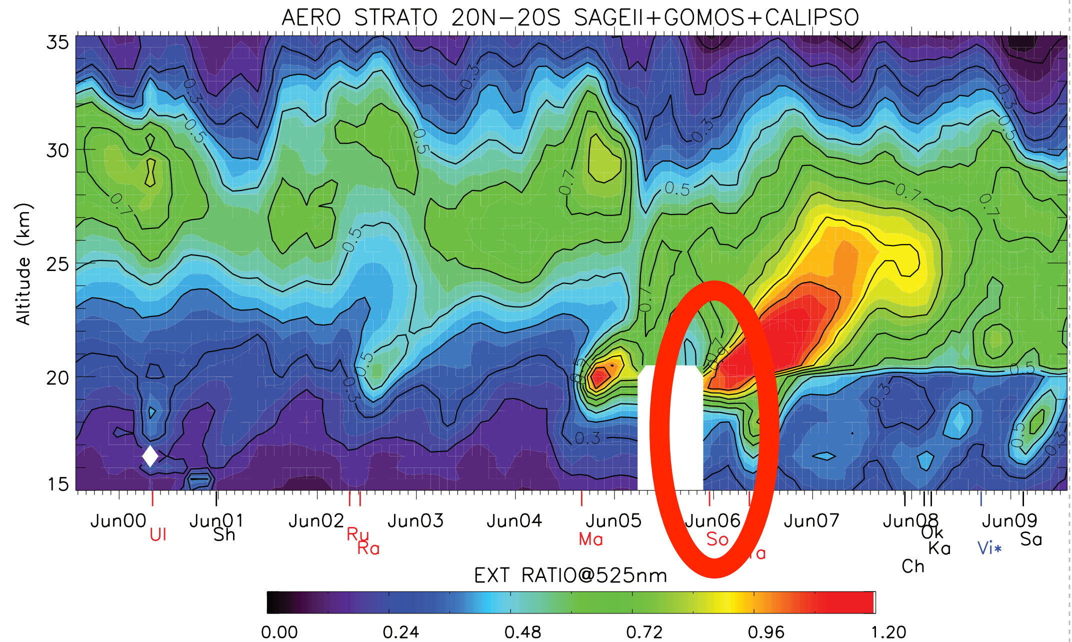
Adapted from Solomon et al. (2011), The Persistently Variable "Background" Stratospheric Aerosol Layer and Global Climate Change, *Science*

Niwano et al. (2009), Seasonal cycles of Stratospheric Aerosol and Gas Experiment II near-background aerosol in the lower stratosphere, J. Geophys. Res, 114(D14), D14306.

Anthropogenic vs Volcanic emissions



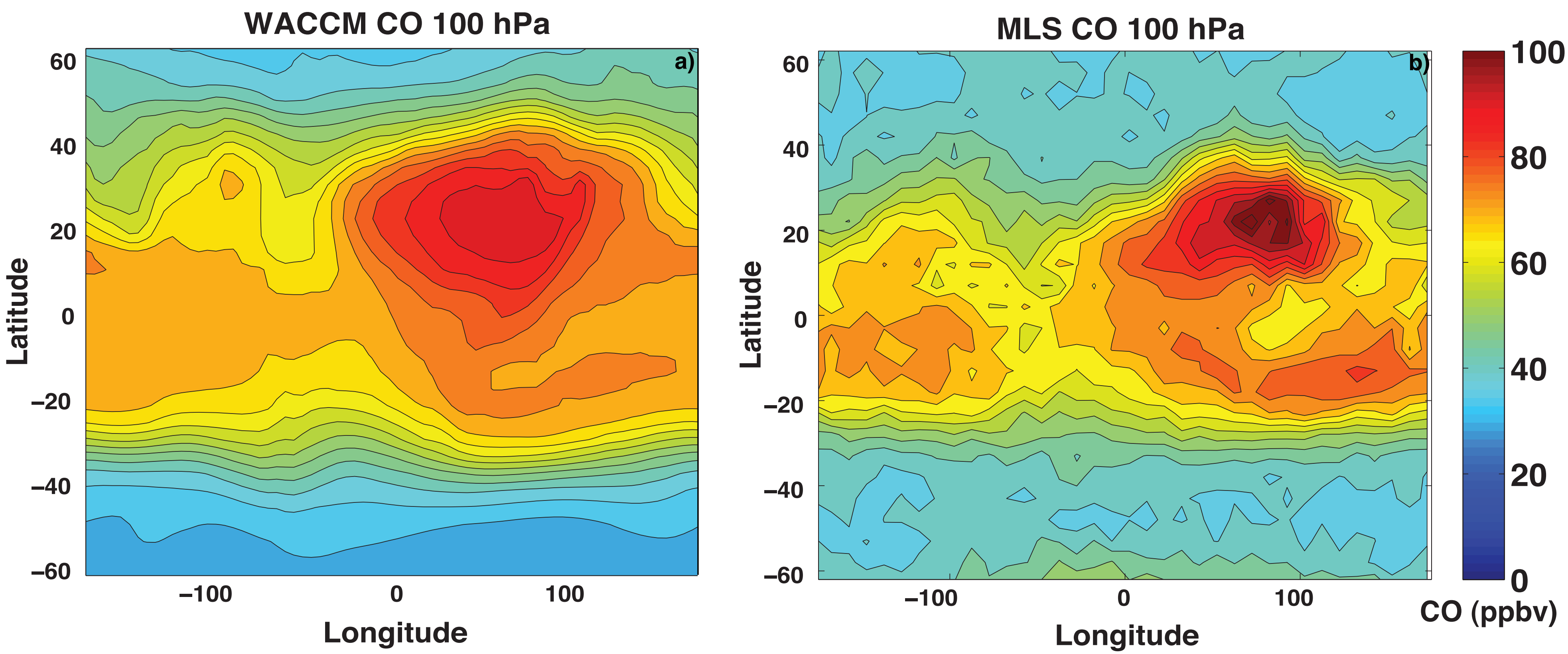
Lu et al. (2010), Sulfur dioxide emissions in China and sulfur trends in East Asia since 2000, *Atmos. Chem. Phys*, *10*(13)



Estimated Emission to Stratosphere
(0.6% of Global Emissions must make it to stratosphere to maintain sulfur burden (Hofmann et al. 2009))

Year	China	Volcano
2006	0.2 TgS	Soufrière Hills 0.17 TgS

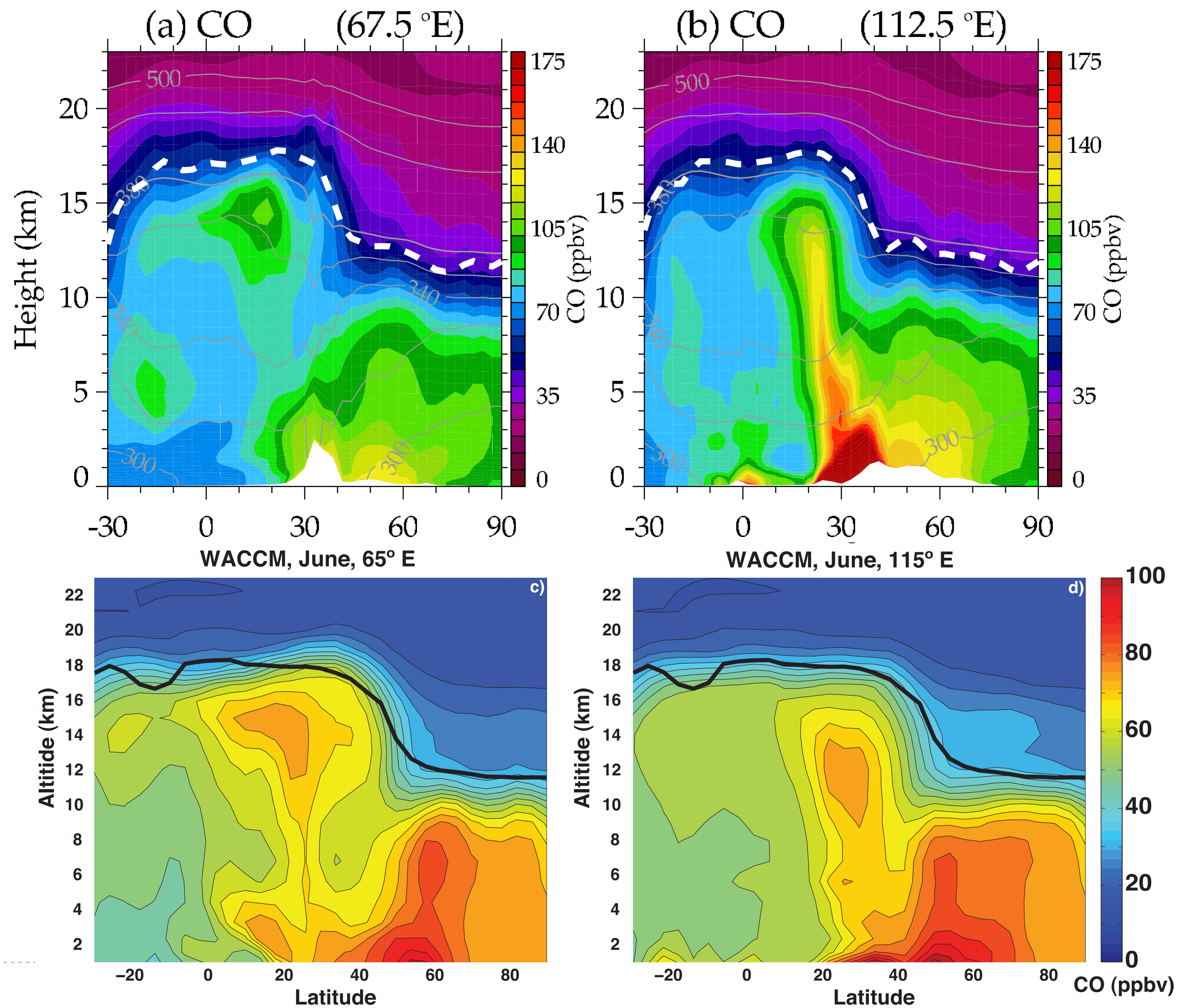
Transport in WACCM



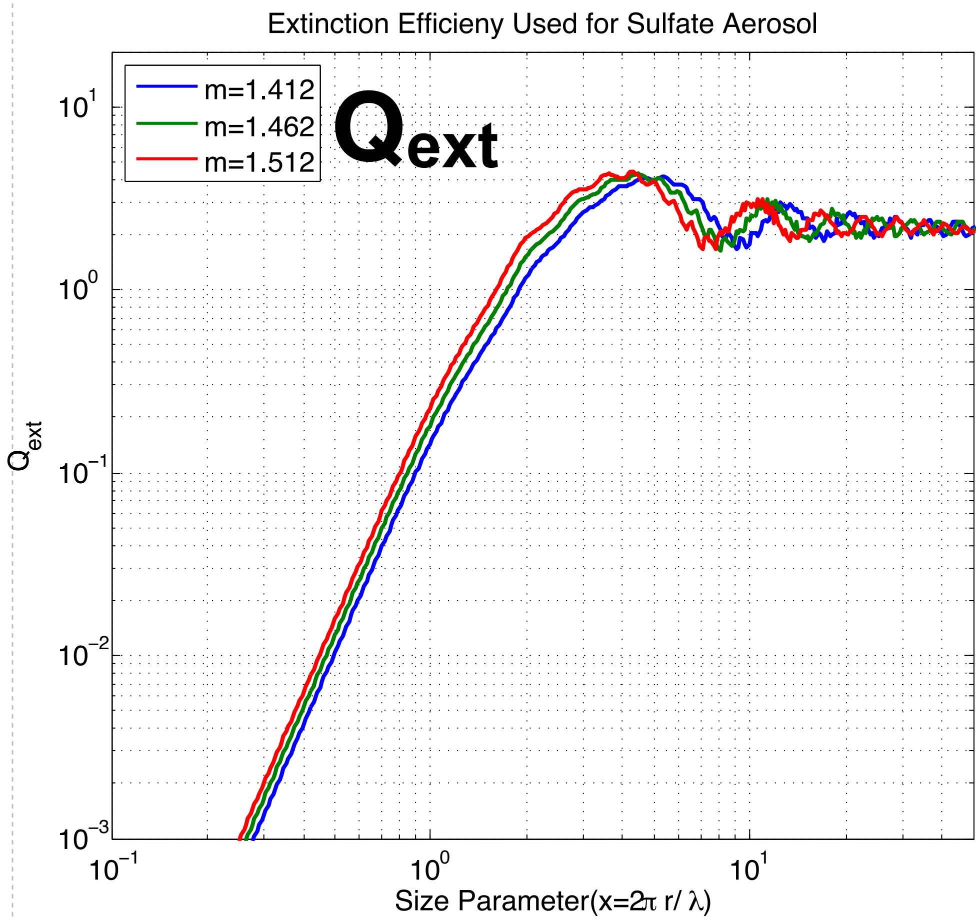
Transport pathways of carbon monoxide in the Asian summer monsoon diagnosed from Model of Ozone and Related Tracers (MOZART)

Mijeong Park,[1] William J. Randel,[1] Louisa K. Emmons,[1] and Nathaniel J. Livesey[2]

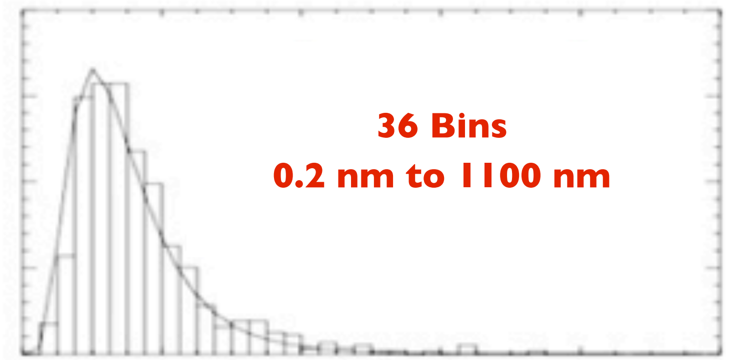
Figure 7. Latitude-altitude cross-sections of monthly mean MOZART-4 CO at the (a) western (67.5°E) and (b) eastern (112.5°E) sides of the monsoon maximum in June 2005. Thermal tropopause derived from the model temperature profile is denoted as thick dashed lines. Thin solid lines are isentropes (320, 340, 360, 380, 450, and 500 K).



Scattering Calculations



$n(r, z) =$



$$\beta_a(z,t) = \pi \int_0^\infty r^2 Q_\pi(\tilde{m}, x) n(r, z) dr$$

Bohren and Huffman (1983)