

Aerosols, Clouds and CLimAte Interactions Mission

An airborne science mission to study the processes and properties of aerosol in the upper troposphere and lower stratosphere

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The diagram illustrates the sulfur cycle in the atmosphere, showing the flow of sulfur species from the surface to the stratosphere and back. The cycle is divided into three main regions: Tropics, Tropopause, and Polar Regions.

Key Processes and Fluxes:

- Evaporation:** Sulfur species (OCS, SO₂, H₂SO₄) are evaporated from the surface into the atmosphere.
- Nucleation:** Sulfur species form new aerosol particles in the stratosphere.
- Coagulation:** Aerosol particles grow by colliding and sticking together.
- Condensation:** Sulfur species condense onto existing aerosol particles.
- Sedimentation:** Aerosol particles fall out of the atmosphere and return to the surface.
- Cloud Scavenging:** Aerosol particles are removed from the atmosphere by clouds.
- Meteoritic Dust:** Dust particles from space enter the atmosphere.
- Wintertime Nucleation:** Sulfur species form new aerosol particles in the stratosphere during winter.

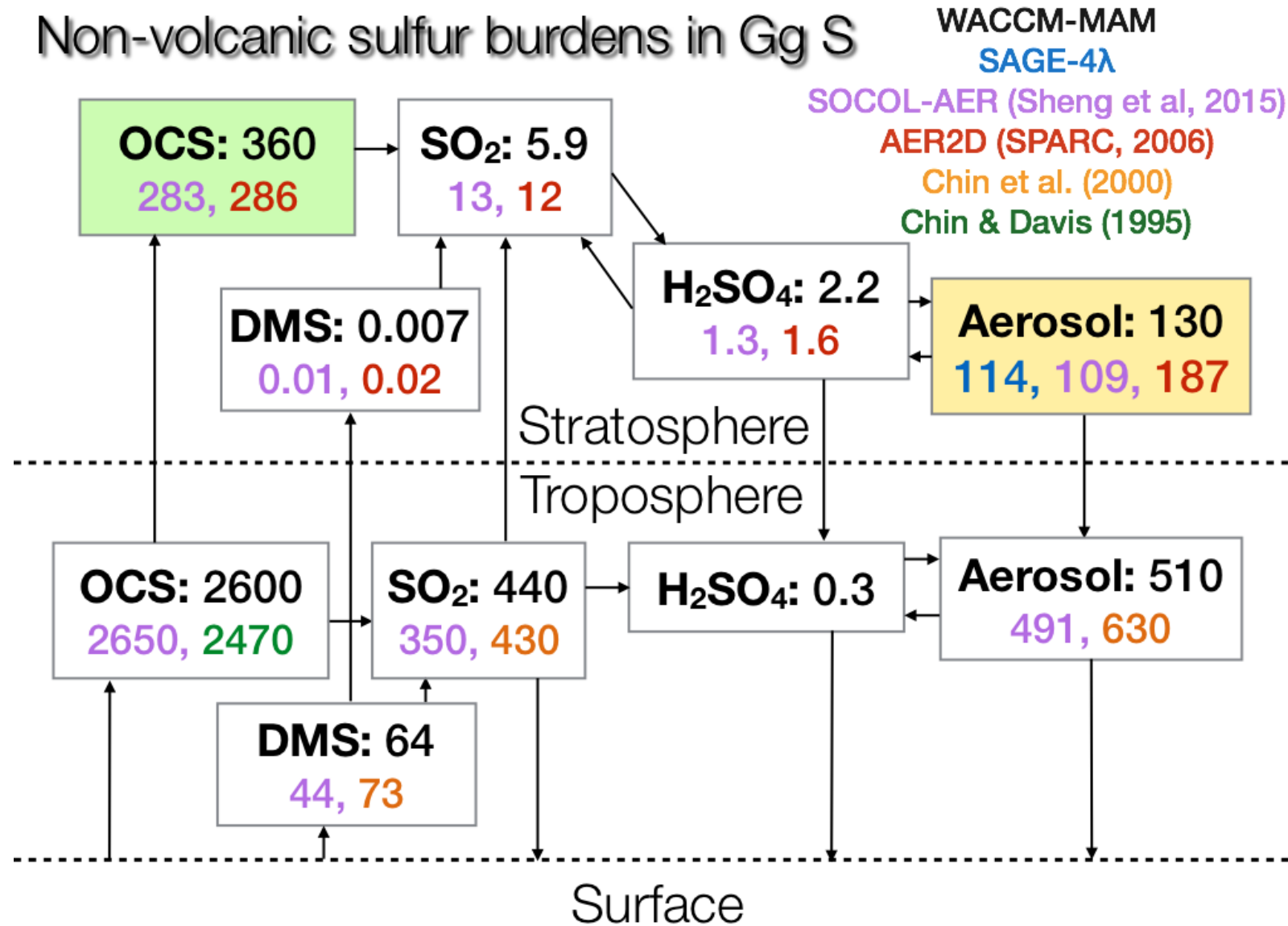
NET FLUX: (Gg S/yr)

OCS	40	↑
SO ₂	50	↑
Aerosol	100	↓
Others	10	↑

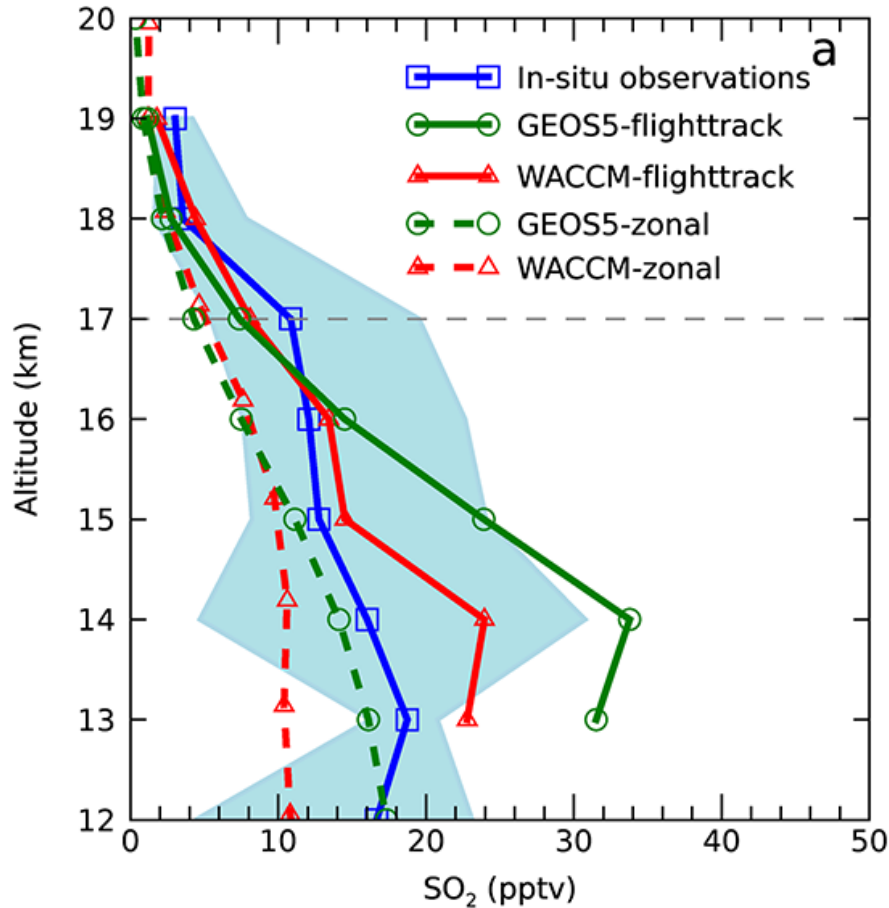
- Uncertainties in UTLS aerosol/sulfur budgets
- Uncertainties in UTLS aerosol physical processes and properties
- Uncertainties in UTLS sources (Asian monsoon, anthropogenic contribution, organics, volcanoes)
- Potential that someone will attempt/ implement SA geoengineering

Kremser et al., Rev. Geophys., 2016, Figure 1

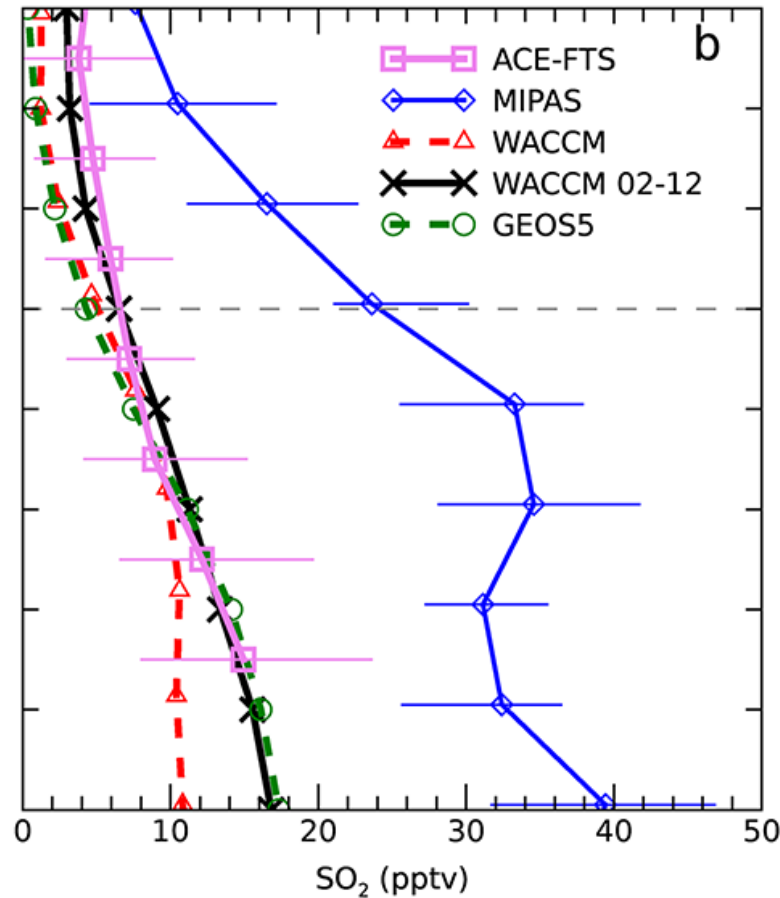
Global Stratospheric Sulfur Budget



SO₂ in the Tropical UT/LS



Comparison of in situ measured SO₂ profile (10°-25° N, 85°-110° W, October 2015) with global models sampled along the flight track and zonal averages.

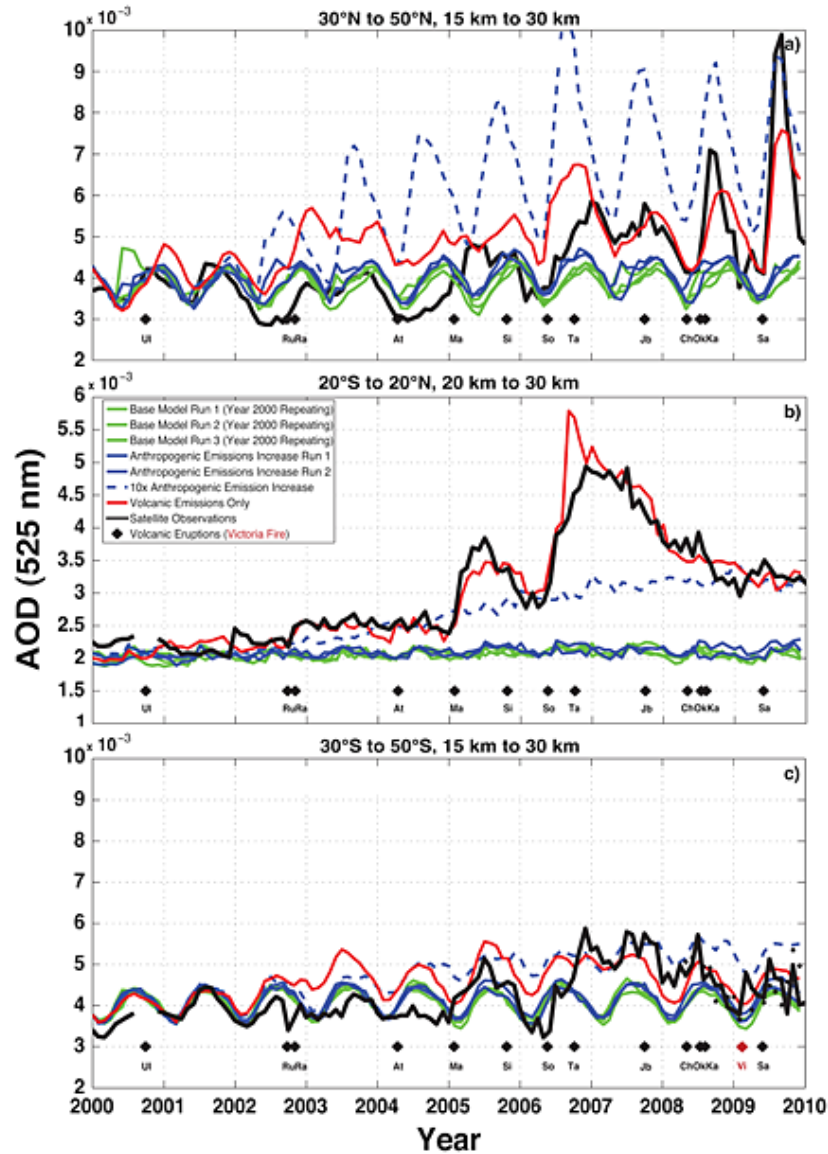


Comparison of model zonally averaged SO₂ profiles with those retrieved from the ACE-FTS and MIPAS satellite measurements (omitting periods of significant volcanic influence)

Rollins et al., *Geophys. Res. Lett.*, 2017, Figure 3

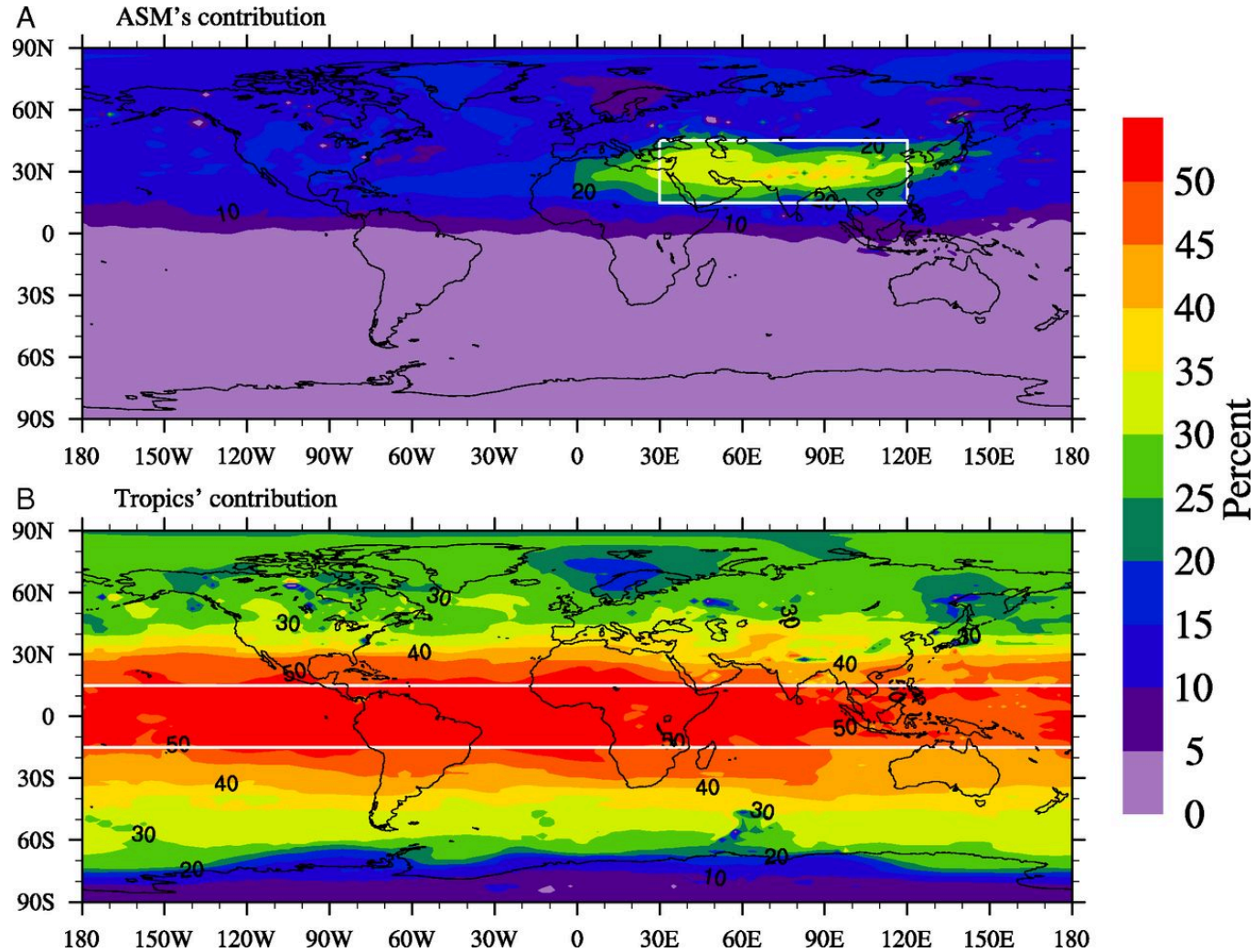
Drivers of Trends in Stratospheric Aerosol

Neely et al. (2013) modeled stratospheric AOD and concluded that an increasing frequency of small-moderate volcanic eruptions explains the observed increase over the period 2000 – 2010. Increasing anthropogenic emissions in Asia were not found to contribute significantly.



Neely et al., *Geophys. Res. Lett.*, 2013, Figure 1

Asian Summer Monsoon as a Source of Stratospheric Aerosol

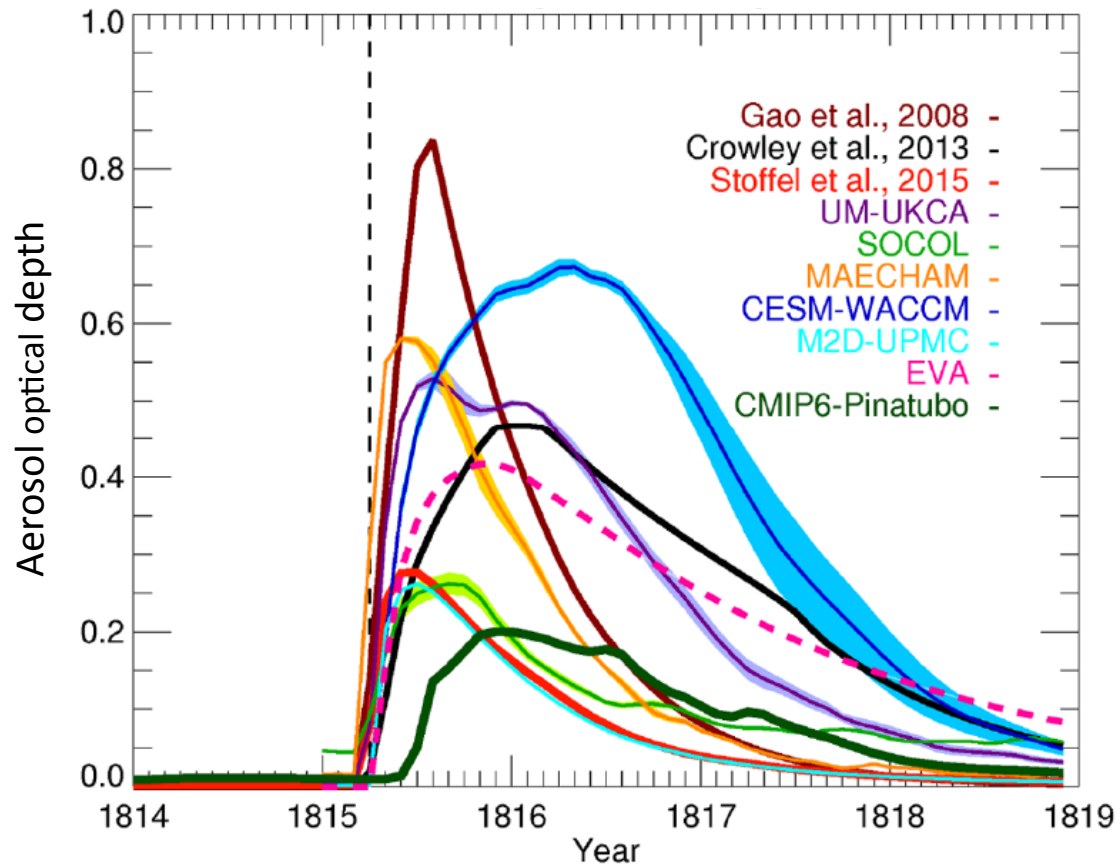


(A) Contribution (percent) to the annual mean particle surface area in the stratosphere from aerosol that is transported through the ASM 3D box (15°–45°N, 30°–120°E, June–September).

The white box shows the spatial extent of the region included in the 3D box where we scrub the aerosol and aerosol precursors.

(B) Contribution to the annual mean particle surface area in the stratosphere from aerosol that is transported through the tropics (white lines, 15°S – 15°N, 0° – 360°E, entire year).

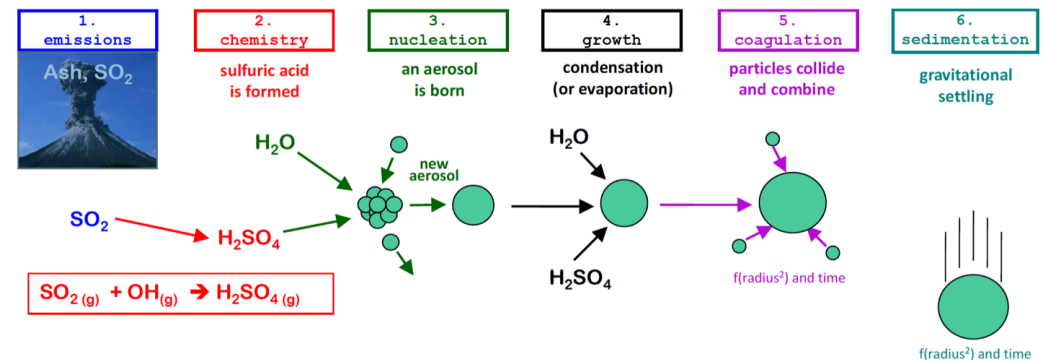
Modeling of Stratospheric Aerosol Perturbations



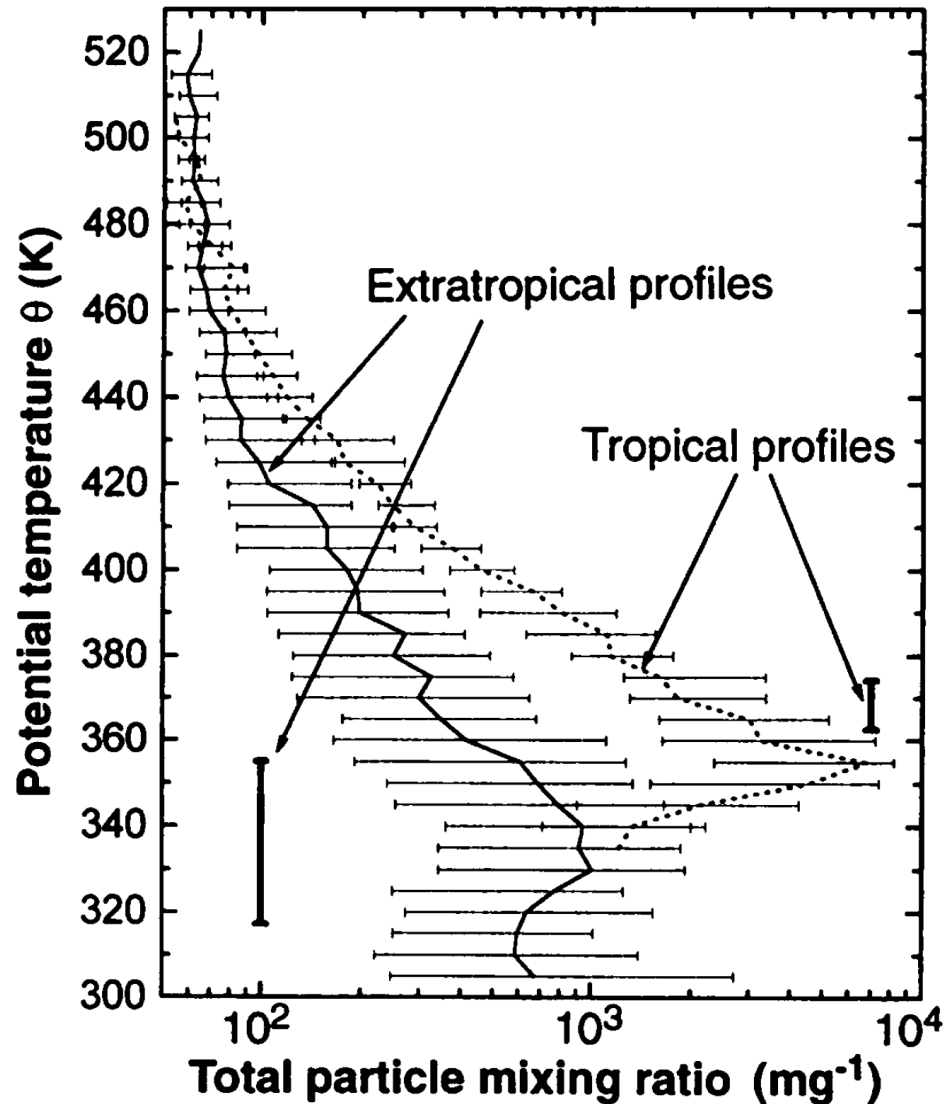
Zanchettin *et al.*, *Geosci. Model Dev.*, 2016, Figure 3a

- Simulation of optical depth caused by the 1815 Tambora eruption by an ensemble of global aerosol models.
- Global aerosol models show large differences in predicting aerosol radiative effects, indicating significant uncertainties in understanding and parameterization of the sulfur-to-aerosol conversion process.

Need to get this part right

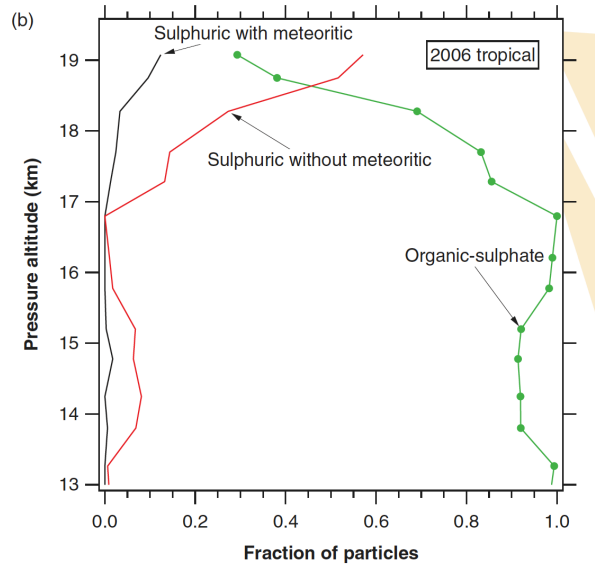


Particle Nucleation and Growth in the UTLS



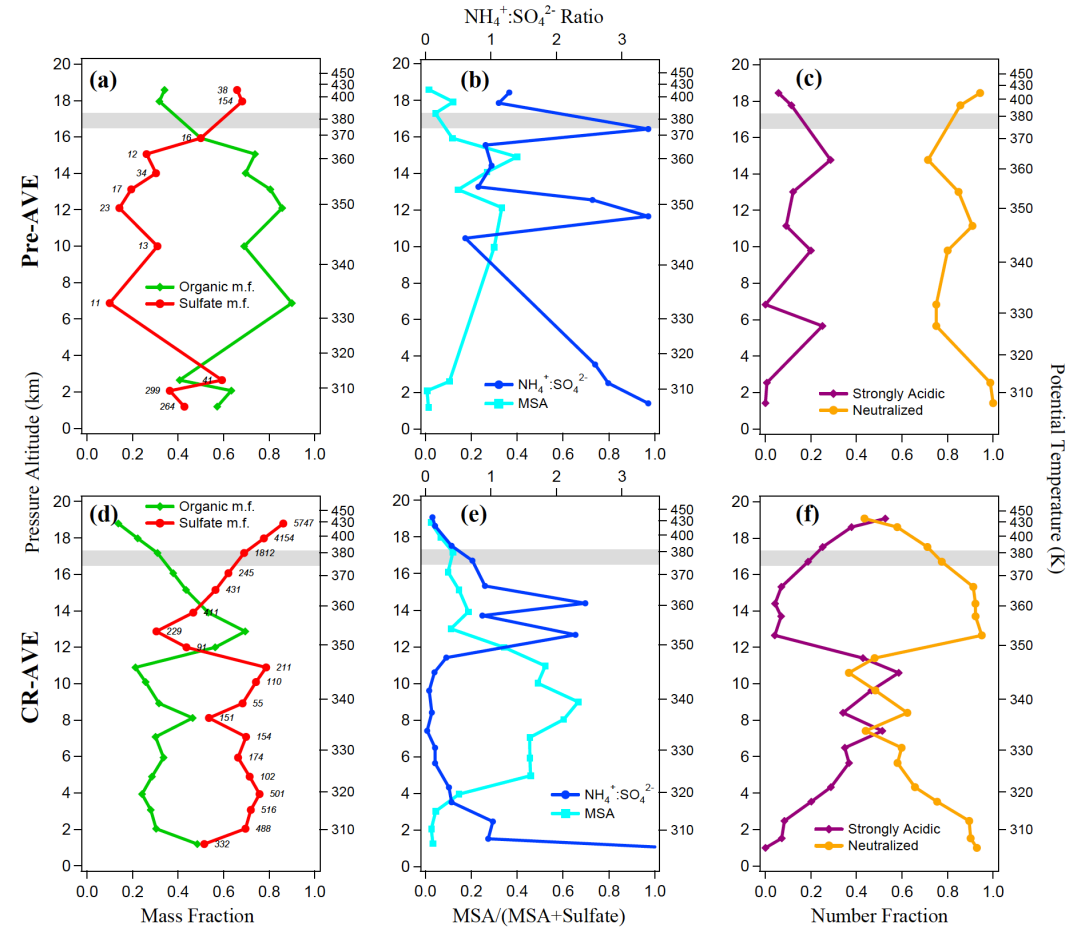
- Brock et al (1995) presented measurements in the tropical UTLS that showed particle nucleation occurring below the tropopause
- Modeling (and volatility measurements) were used to infer a H_2SO_4 - H_2O binary nucleation mechanism

Composition of UT/LS Aerosol - Observations



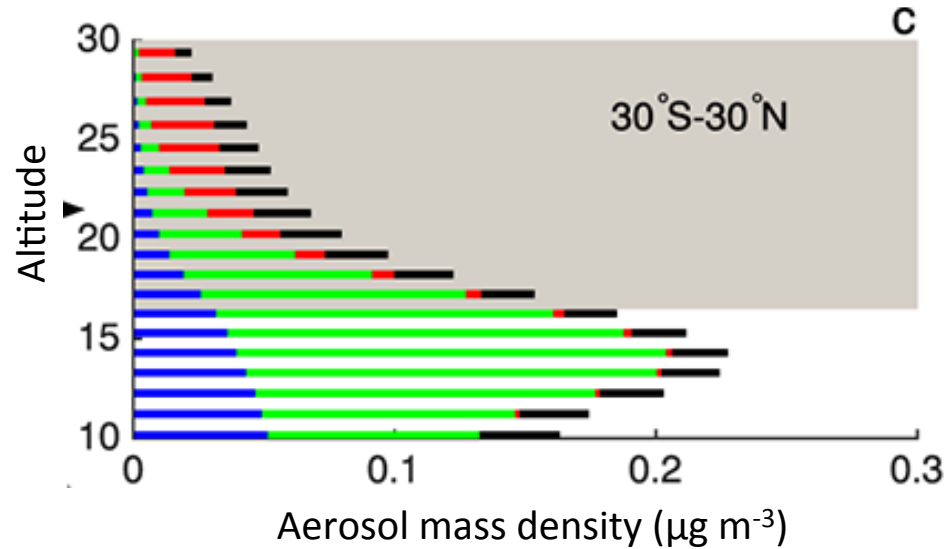
Murphy et al., *QJRMS*, 2013, Figure 3

- Composition measurements of individual particles in the tropical UT/LS by PALMS indicate a significant contribution from organics to aerosol mass
- Sulfate-organic aerosol near the tropopause was mostly neutralized

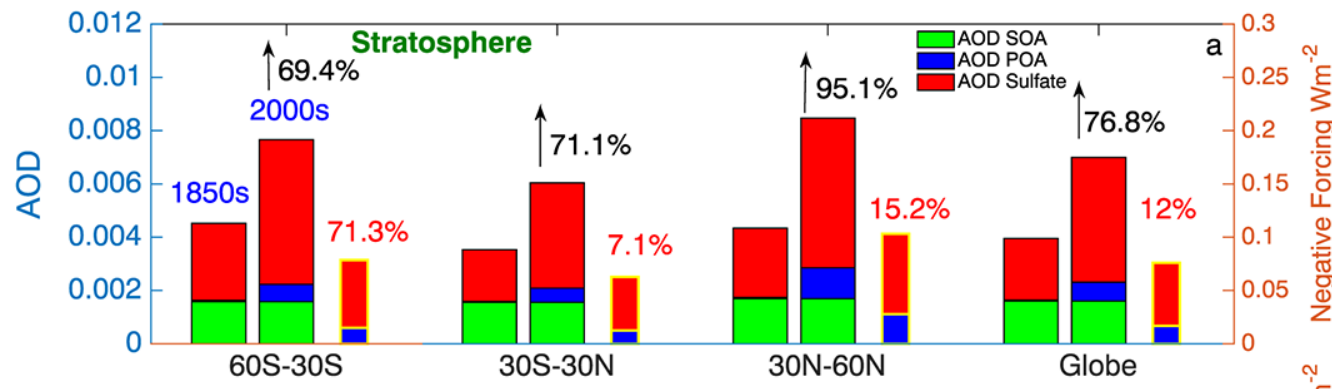


Froyd et al., *Atmos. Chem. Phys.*, 2009, Figure 5

Composition of UT/LS Aerosol - Modeling



- CESM-WACCM modeled aerosol mass in the tropical UT/LS is dominated by organics
- Mass fraction from organics falls off with increasing altitude above the tropopause, in agreement with PALMS observations



- Model runs comparing pre-industrial to modern emissions suggest that anthropogenic emissions have increased stratospheric sulfate aerosol AOD

Yu et al., *GRL*, 2016, Figure 2c (top) and Figure 3a (bottom)

ACCLAIM:

A NASA EV-S 3 Proposal to Study the Processes and Climate Impacts of UT/LS

PI: Eric Jensen (NASA Ames)

Deputy PI: Karen Rosenlof (NOAA ESRL)

Aerosol
Science team includes members with extensive experience and expertise in UTLS in situ measurements, aerosol process analysis, global modeling, and satellite retrieval and data analysis

Overarching NASA Earth Science Goal: *Advance the understanding of changes in the Earth's radiation balance, air quality, and the ozone layer that result from changes in atmospheric composition*

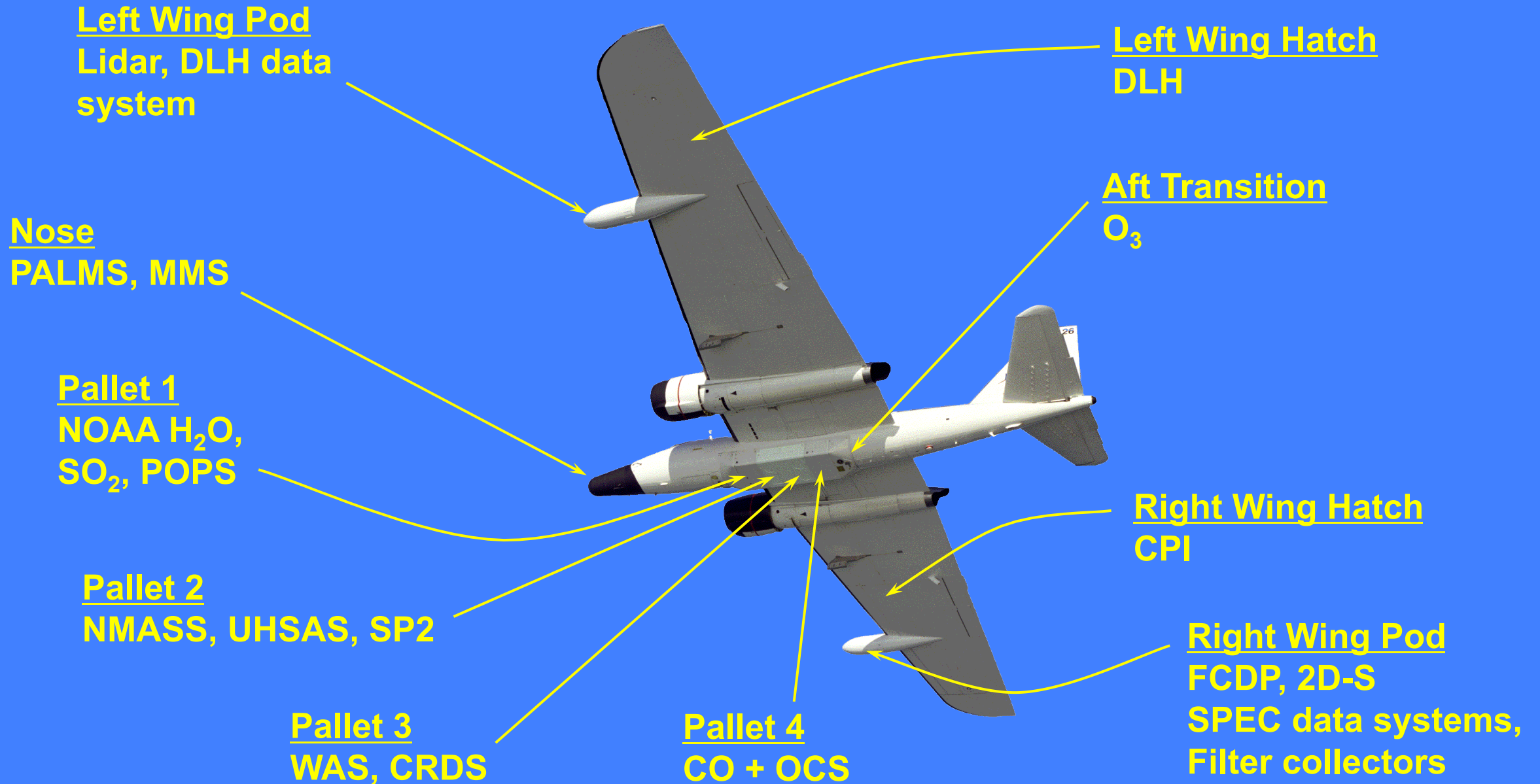
Motivation

- Changes in source gas distributions affecting stratospheric aerosols
- Increasing awareness of organic contributions to UT/LS aerosols
- Potentially large climate impacts from volcanoes and intentional solar radiation management

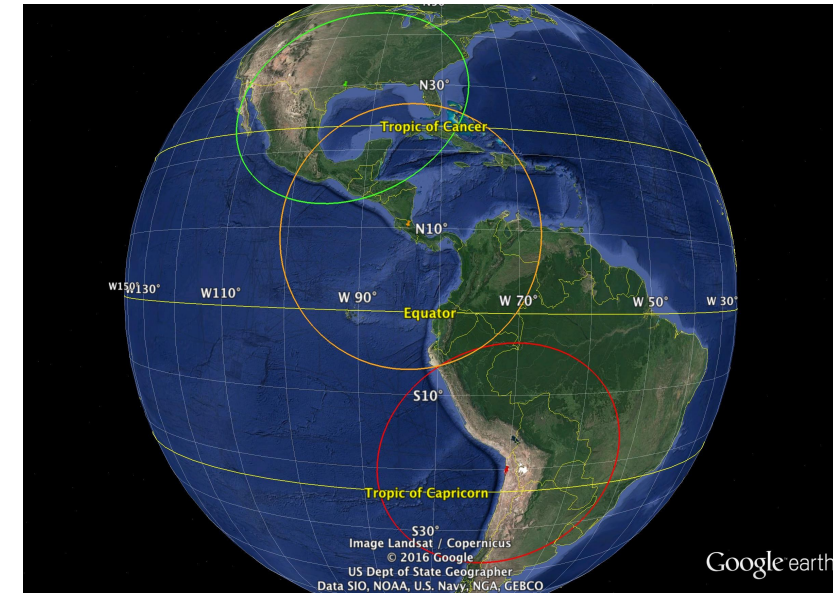
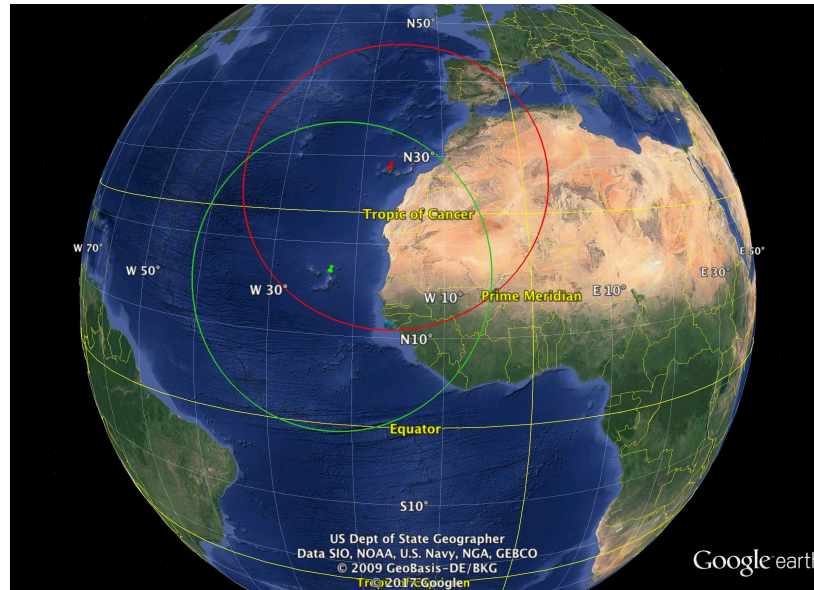
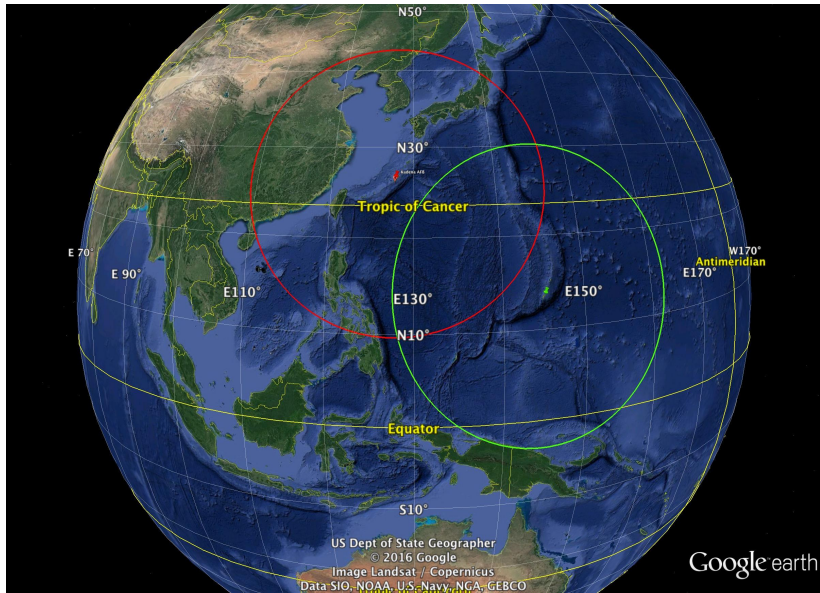
Objectives

1. In situ measurements with the WB-57
 - ~4 deployments (WestPac, E Atlantic, Central America, latitudinal coverage)
 - on the order of 100 flight hours/deployment
 - Typically profiling between ~14 km and ceiling (~19 km)
2. Use the in situ data to
 - Evaluate/improve NASA satellite (e.g. SAGE III, OMPS) retrievals of stratospheric aerosol properties
 - Evaluate/improve global model representations of UTLS aerosol sources, formation and growth
 - Investigate effects of aerosols on cirrus cloud distribution and optical properties
3. Use the improved global models to assess the radiative forcing and climate impacts of UTLS aerosol

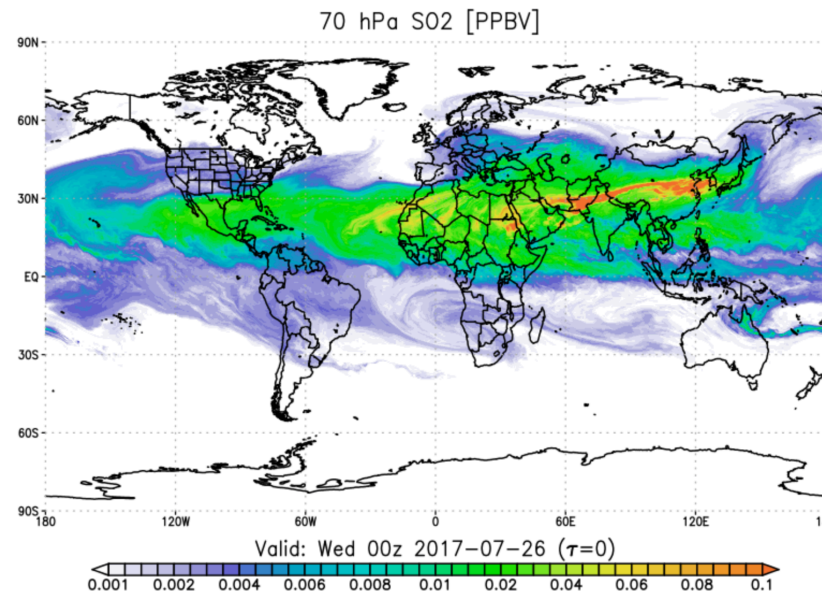
ACCLAIM Payload



ACCLAIM Deployment Concepts



- ❑ Western Pacific and Asian Summer Monsoon outflow
 - Kadena AFB, Okinawa, July – September
 - High latitude sampling on transit?
- ❑ Westerly shedding of the Asian Summer Monsoon
 - Cape Verde, July – September
 - African convective contribution?



- ❑ EFD, Costa Rica
 - Latitudinal coverage: tropical focus, but NH-SH differences? Continental vs maritime sources?
 - seasonal coverage (e.g. convection)?

GEOS-5 Model forecast, July 2017

ACCLAIM Science Questions (and how we would address them)

UTLS Aerosol budget

1. How do the magnitude and occurrence frequency of new particle formation events depend on existing aerosol SAD, meteorological conditions, and gas-phase precursor concentrations?

In situ measurements of aerosol size distribution, met fields, and sulfur species; parcel-models run along trajectories.

2. How important are organics for the TTL aerosol budget (particle size, SAD, mass)?

Single-particle composition measurements; gas-phase organics measurements; constrained global-model simulations.

3. How much do aerosols from the Asian monsoon affect the stratospheric aerosol surface-area budget?

In situ measurements of aerosol properties and sulfur species in air masses recently shed from the ASM anticyclone; parcel models and global models constrained by in situ and satellite measurements of aerosol composition and size distribution.

4. What are the relative contributions of different sulfur species and organics to stratospheric aerosol SAD and mass? What are the relative contributions of natural and anthropogenic emissions to the stratospheric aerosol loading?

In situ measurements of aerosol composition, sulfur species, organics, and air mass tracers; parcel models and global models constrained by in situ and satellite measurements of aerosol composition and size distribution.

ACCLAIM Science Questions (and how we would address them)

TTL aerosol-cloud interactions

1. How does the comparison between ice crystal residual (ice nuclei) composition and ambient aerosol composition vary with location in the tropics, season, and proximity to potential ice nuclei sources?

In situ measurements of ambient aerosol and ice residual composition in different tropical locations and seasons; tracer measurements of air mass origin

2. How do the statistics of ice supersaturation and cirrus microphysical properties depend on aerosol composition and size distribution?

In situ measurements of ice supersaturation, cloud properties, aerosol composition, and aerosol size distribution; detailed transport and cloud microphysics simulations

3. Is the aerosol size distribution altered by TTL cirrus? Do TTL cirrus effectively scavenge trace gases before entry to the stratosphere?

Measurements of interstitial aerosol size distribution and cirrus properties; tracer measurements

ACCLAIM Science Questions (and how we would address them)

Stratospheric aerosol physical properties and climate impacts

1. How does the stratospheric aerosol extinction across the solar spectrum depend on aerosol physical properties (size distribution and composition)?

In situ measurements of aerosol composition, size distribution, and extinction

2. How do stratospheric aerosol radiative forcing and climate impact depend on changes in source strengths and aerosol physical properties? How important are the associated climate feedbacks?

Radiative transfer calculations and global-model simulations constrained by in situ and satellite measurements

ACCLAIM Science Questions (and how we would address them)

Satellite aerosol retrieval evaluation/improvement

1. How well do satellite retrievals represent UTLS aerosol properties (SAD, mass, extinction, etc.)?

Comparison between in situ and satellite measurements

2. How sensitive are satellite retrievals of aerosol extinction to [assumed] aerosol size distribution?

Use of in situ size distribution measurements to assess and improve aerosol model used in satellite retrievals

ACCLAIM Science Questions (and how we would address them)

Global model evaluation/improvement

1. How well do operational (modal aerosols) and specialized (bin microphysics) global models represent the UTLS aerosol size distribution and composition?

Comparisons between global-model aerosol properties and measurements (both in situ and aircraft); deployments to regions/seasons with greatest model discrepancy/variation

2. How well are aerosol processes (new particle formation, coagulation, condensation, and transport) represented in global models?

Comparison of ultrafine particle concentrations with in situ measurements; comparison between aerosol evolution in global models and parcel models; comparison of global model regional, vertical, and temporal distributions with satellite and in situ measurements