Chemistry, Aerosols, and WACCM

Presented by Simone Tilmes
NESL: ACD/CGD

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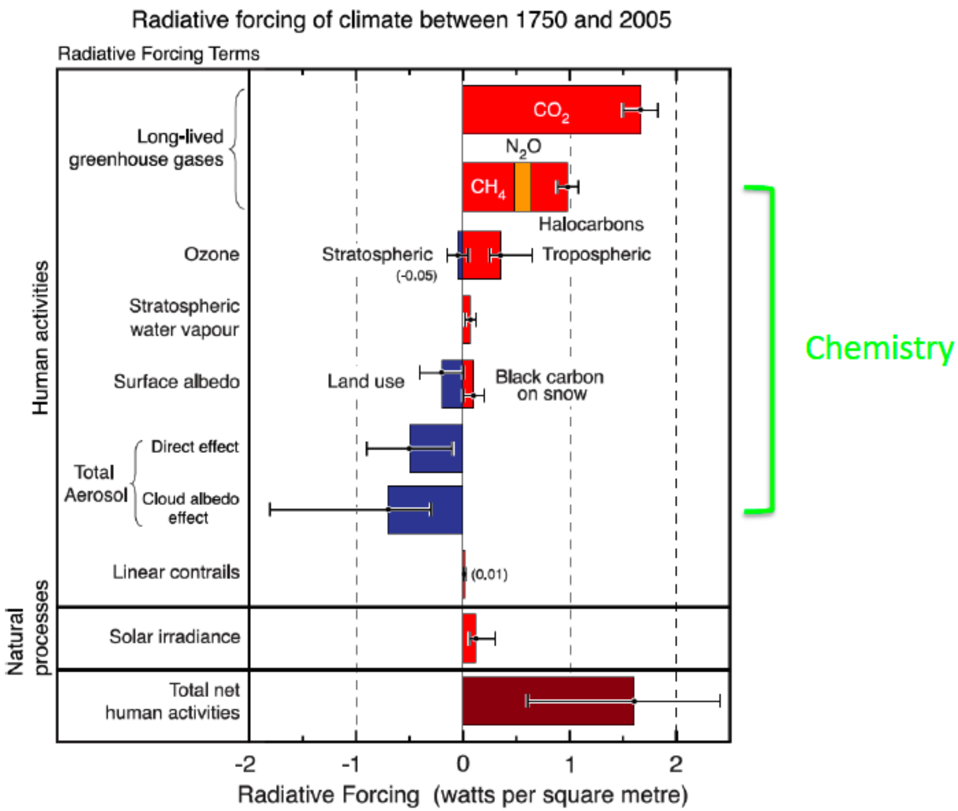
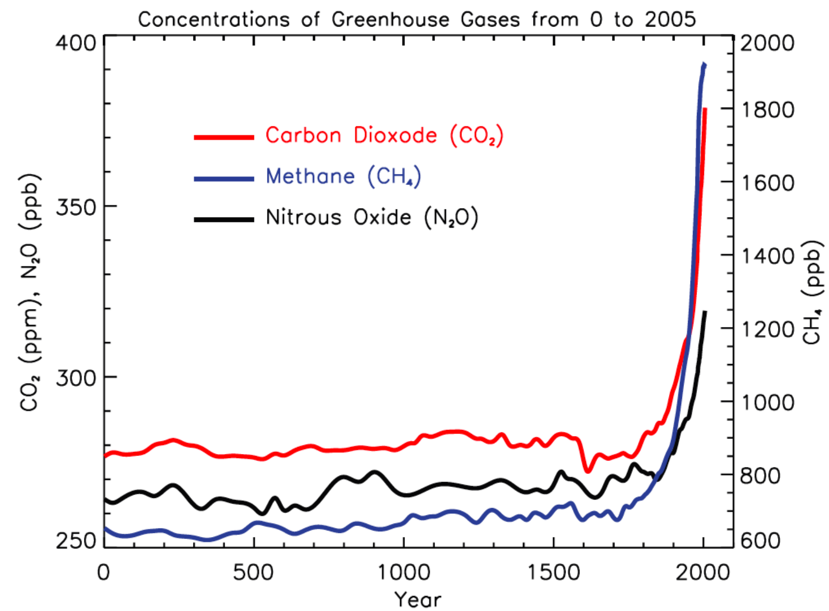
CAMChem Liaison: Simone Tilmes

Aerosol Liaison: Po-Lun Ma

Motivation: Modeling with Chemistry



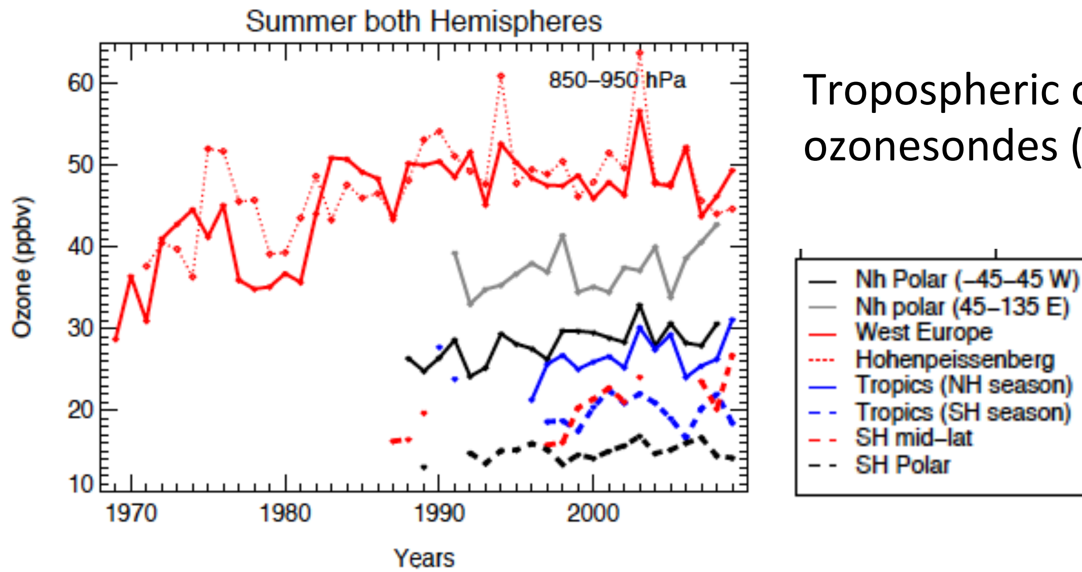
Forster and Ramaswamy. AR4 Chapter 2



ice core measurements

Importance to represent climate gases for radiative forcing: (CO_2), CH_4, O_3, H_2O

Motivation: Modeling with Chemistry

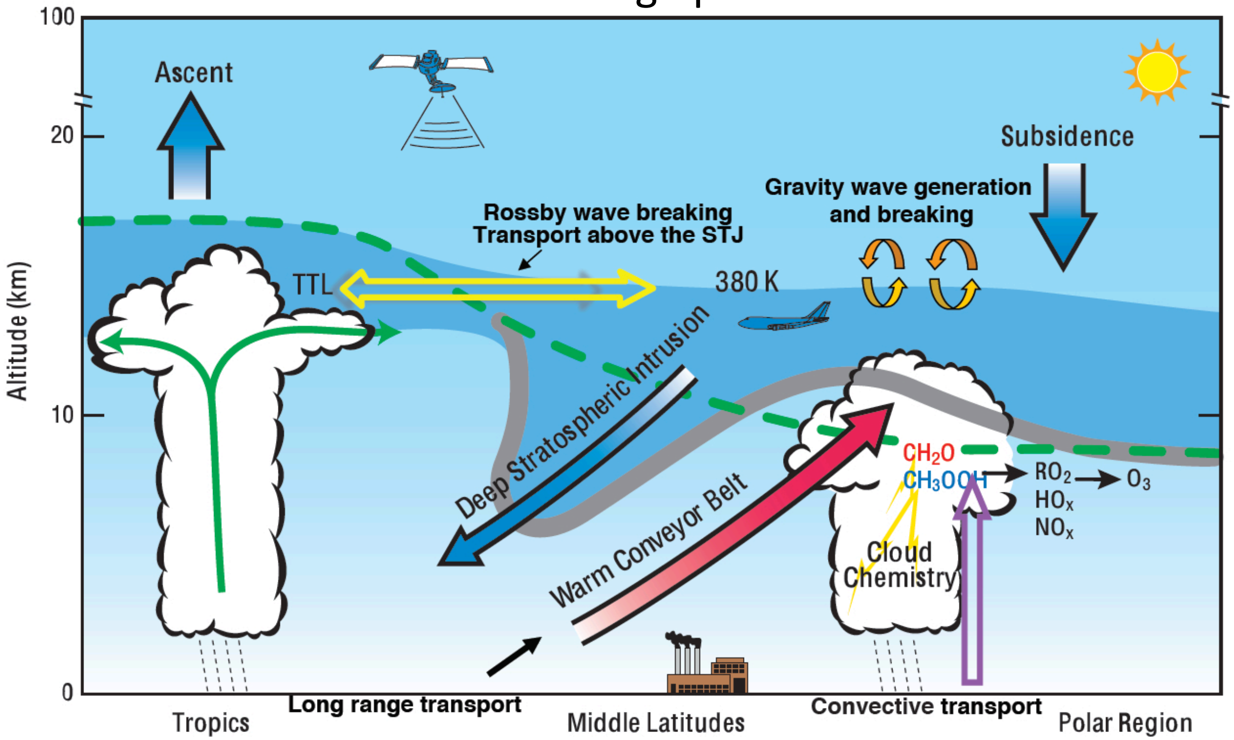


Tropospheric ozone evolution based on ozonesondes (1968-2011)

- Tropospheric ozone is controlled by changes in precursors: NO_x ($NO + NO_2$), CO, volatile organic compounds (VOCs) and therefore emissions
- Implications for air quality and ecosystem: EPA defines standards for air quality (currently 75 ppb)
- OH determines oxidation capacity, CH_4 lifetimes
- Importance of primary and secondary organic aerosols, produced from volatile organic compounds (VOC's)

Motivation: Modeling with Chemistry

UTLS exchange processes



Stratosphere-Troposphere Analyses of Regional Transport (2008)

- Exchange of chemistry and aerosol due to stratospheric/ tropospheric transport
- Impact of halogen loading on stratospheric ozone (ozone hole) and impact on climate (importance of very short-lived species)

-> local changes of short time scales are important (not captured by climatologies)

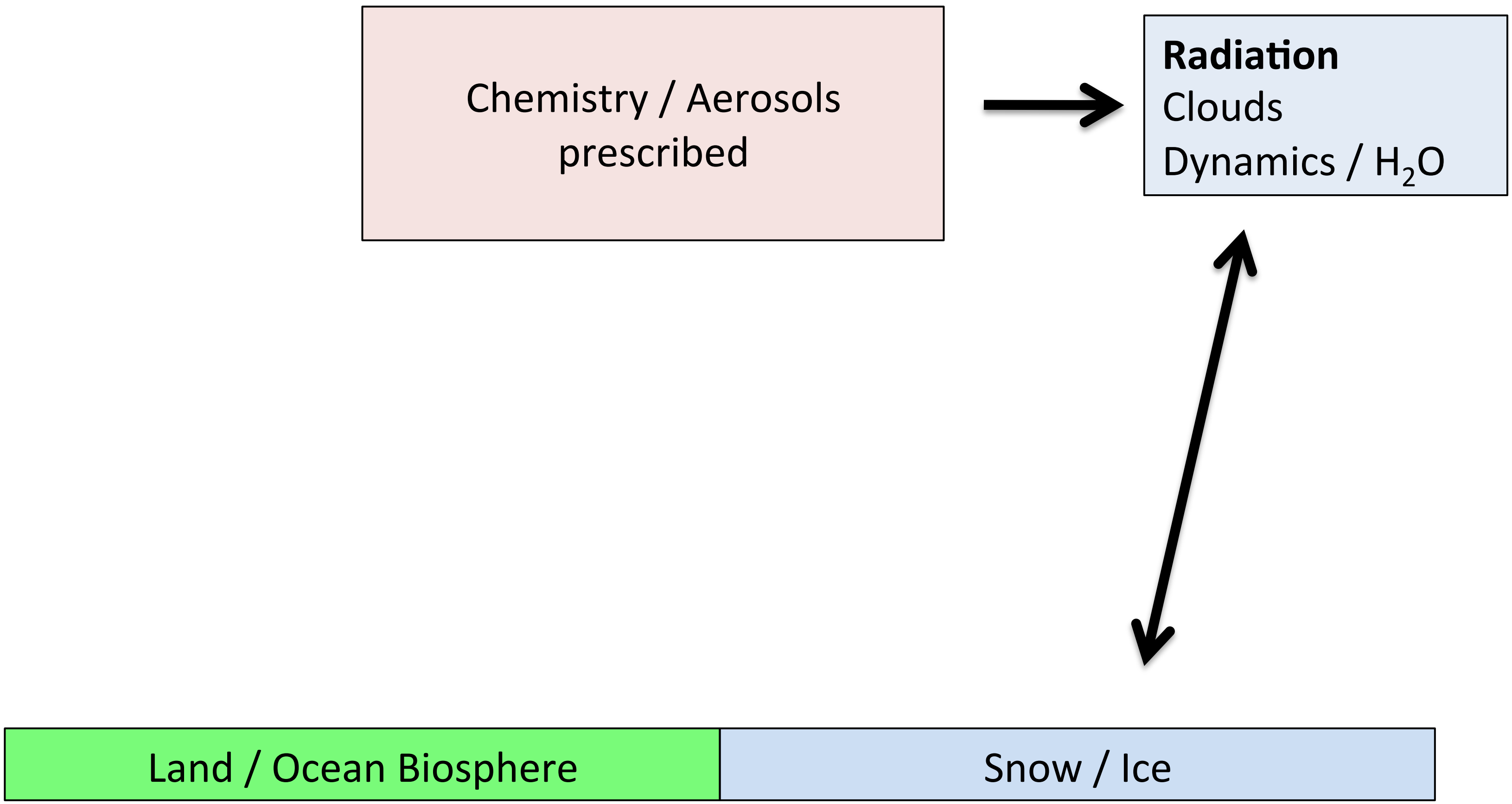
Modeling without Chemistry-Climate interactions in CESM

- Chemistry and aerosols are prescribed in CAM4: (prescribed monthly fields of CO_2, CH_4, O_3, N_2O, CFCs)

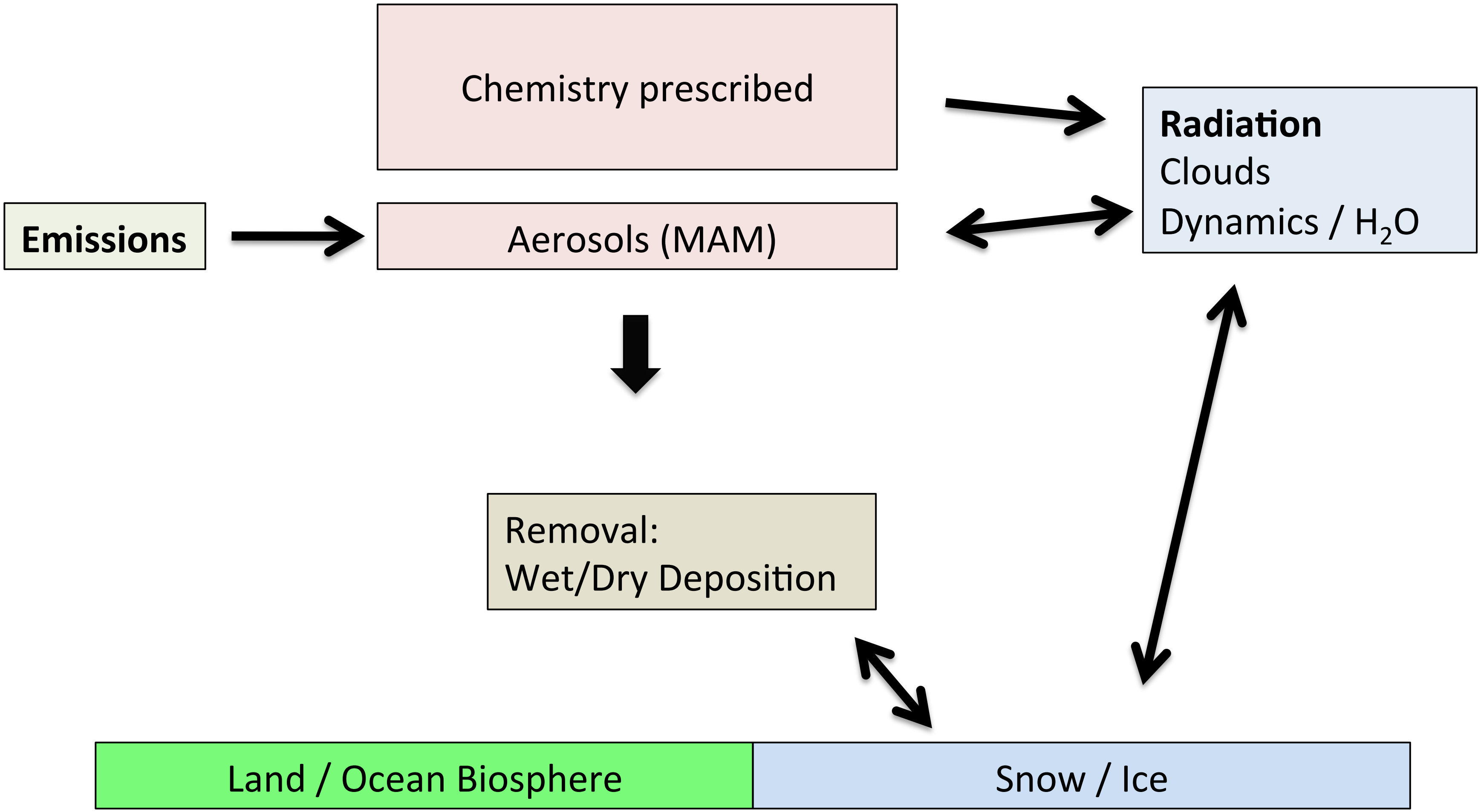
- Aerosols are calculated in CAM5 (Modal Aerosols Model MAM), but not coupled with chemistry, simple chemistry is added ("fixed" oxidants)

No interaction between Chemistry and Climate

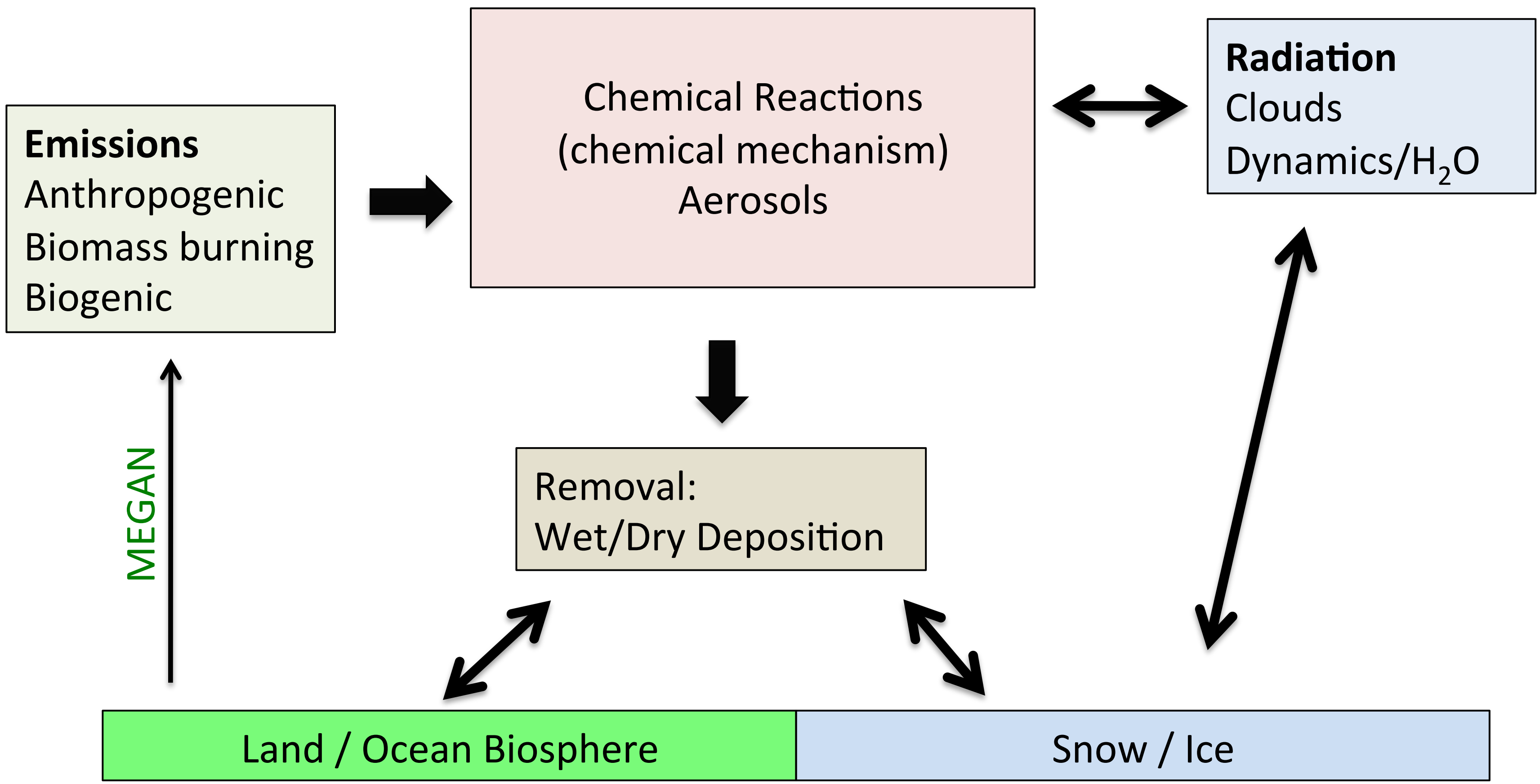
Chemistry in CESM CAM4



Chemistry in CESM CAM5



Chemistry in CESM CAM4/5-Chem



Modeling with Chemistry

Chemistry: more and less complex mechanisms are available

Emissions: surface emissions fields, fixed boundary conditions, calculated using vegetation type (biogenic, VOC) , external forcings (aircraft emissions)

Dry Deposition: uptake of chemical constituents by plants and soil (CLM), depending on land type, roughness of surface, based on resistance approach

Wet Deposition: uptake of chemical constituents in rain or ice (linked to precipitation, both large-scale and convective).

Modeling with Chemistry

Available chemical mechanisms in CAM-Chem

Superfast Chemistry (CAM4):

12 species, simple chemistry mechanism, CH_4 prescribed
LINOZ + Cariolle in stratosphere, fully coupled

Bulk Aerosol Model (BAM) (CAM4):

Includes Black Carbon, Organic Carbon, Sea Salt, Dust
(prescribed monthly fields of CO_2, CH_4, O_3, OH, HO_2, NO_2, N_2O, SO_2/SO_4)

Tropospheric chemistry (trop_mozart) (CAM4/5):

Tropospheric mechanism, ~103 species (MOZART: *Emmons et al., 2010*)
Stratospheric chemistry is prescribed about 50 hPa: (O_3, HNO_3, CH_4, CO)
Emissions, Dry/Wet Deposition
Secondary Organic aerosols

Plus stratospheric chemistry (trop-strat mozart) (CAM4/5):

Tropospheric and Stratospheric mechanism (~122 species) including stratospheric heterogeneous reactions, about 300 reactions (similar to WACCM)

Modeling with Chemistry

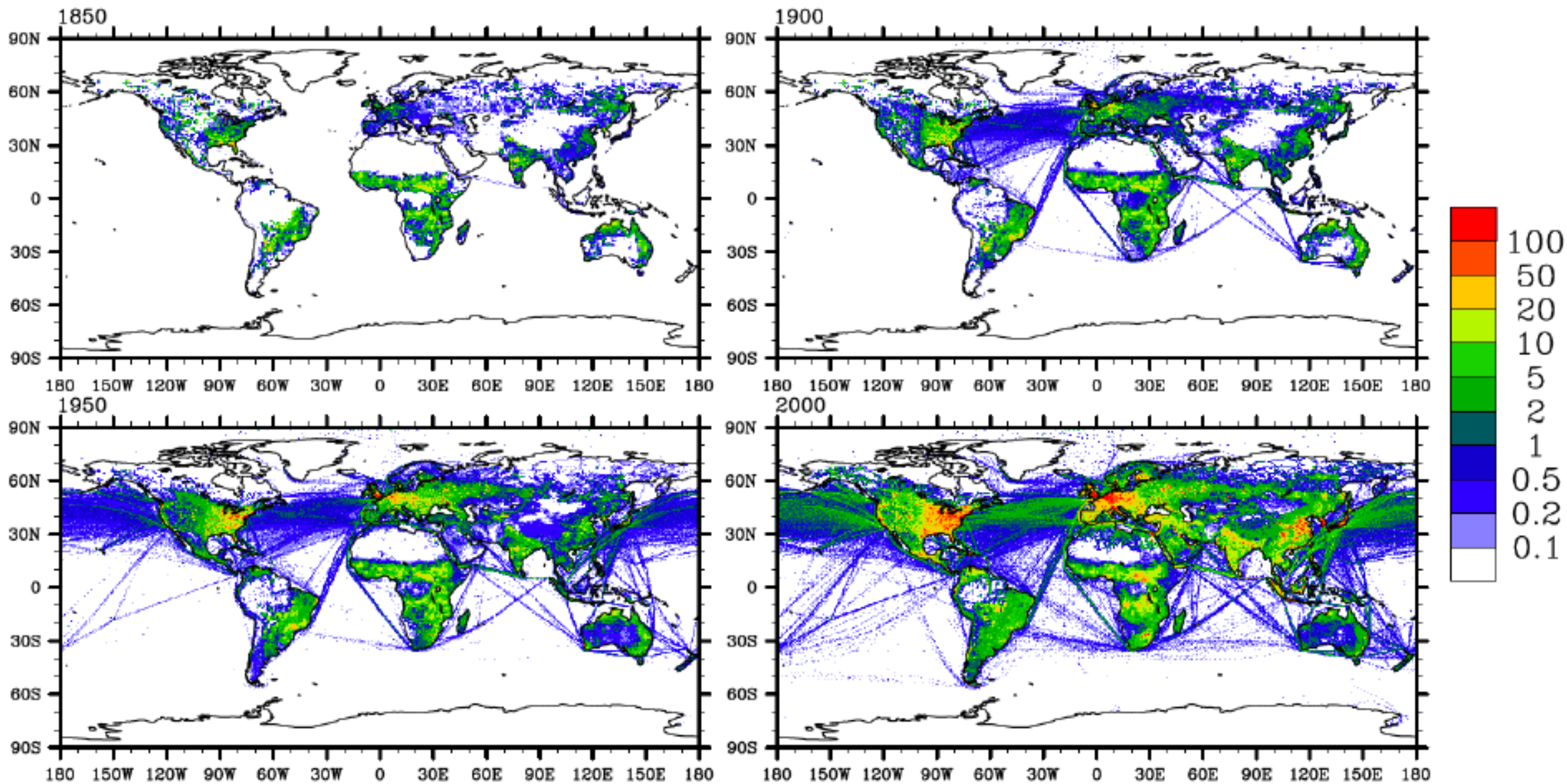
Chemistry: more and less complex mechanisms available

Emissions: surface emissions fields, fixed boundary conditions, calculated using vegetation type (biogenic, VOC) , external forcings (aircraft emissions)

Dry Deposition: uptake of chemical constituents by plants and soil (CLM), depending on land type, roughness of surface, based on resistance approach

Wet Deposition: uptake of chemical constituents in rain or ice (linked to precipitation, both large-scale and convective).

Example: NO_x emissions



Lamarque et al., 2010

Anthropogenic + biomass burning + ships: kg(N)/year

Modeling with Chemistry

Chemistry: more and less complex mechanisms available

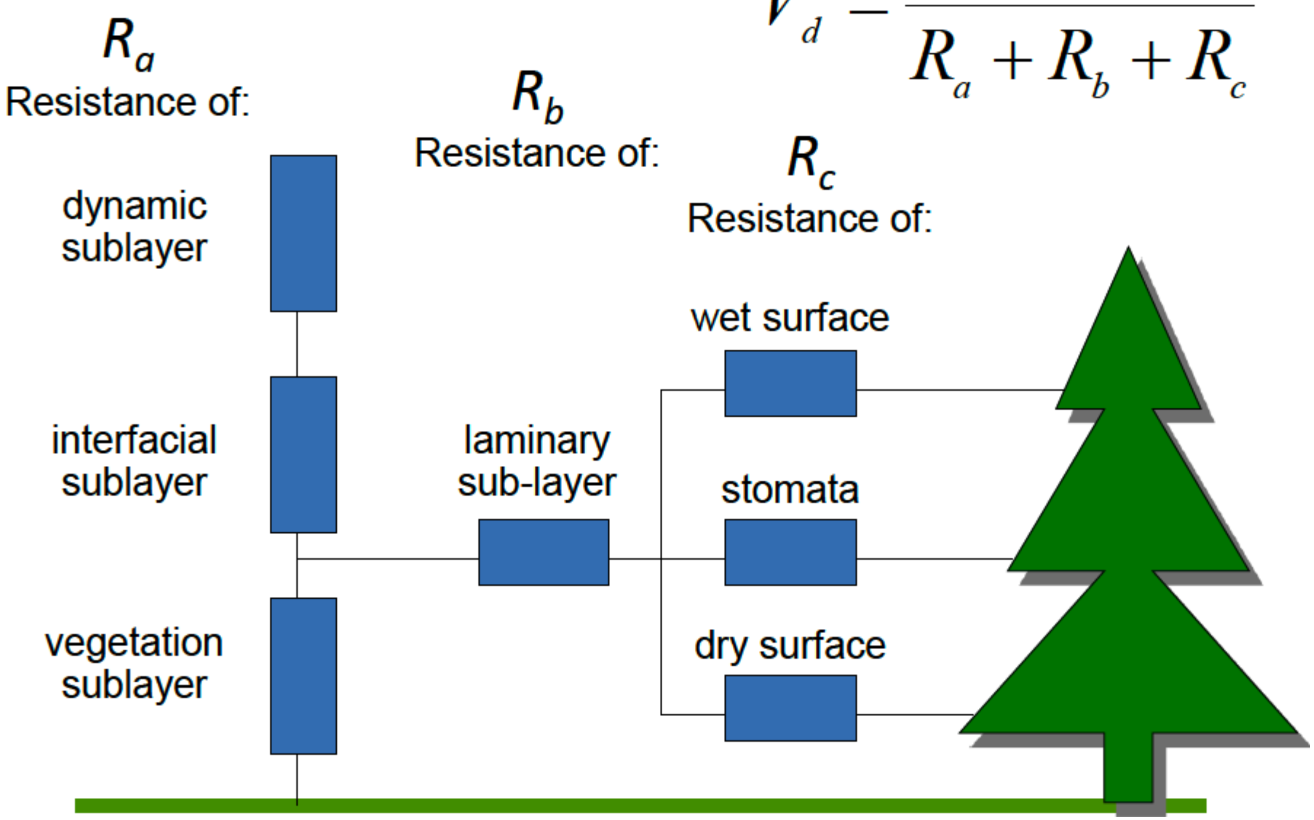
Emissions: surface emissions fields, fixed boundary conditions, calculated using vegetation type (biogenic, VOC), external forcings (aircraft emissions)

Dry Deposition: uptake of chemical constituents by plants and soil (CLM), depending on land type, roughness of surface, based on resistance approach

Wet Deposition: uptake of chemical constituents in rain or ice (linked to precipitation, both large-scale and convective).

Dry Deposition Velocity

$$V_d = \frac{1}{R_a + R_b + R_c}$$



Deposition flux: $$F = -v_d C$$

C: concentration of species am 10m

Modeling with Chemistry

Chemistry: more and less complex mechanisms available

Emissions: surface emissions fields, fixed boundary conditions, calculated using vegetation type (biogenic, VOC) , external forcings (aircraft emissions)

Dry Deposition: uptake of chemical constituents by plants and soil (CLM), depending on land type, roughness of surface, based on resistance approach

Wet Deposition: uptake of chemical constituents in rain or ice (linked to precipitation, both large-scale and convective).

- Removal is modeled as a simple first-order loss process

$$X_{\mathbf{iscav}} = X_i \times F \times (1 - \exp(-\lambda \ \Delta t))$$

- X_{iscav} is the species mass (in kg) of Xi scavenged in time
- F is the fraction of the grid box from which tracer is being removed, and λ is the loss rate.

Aerosols

Direct Effects:

- Radiation (scattering/absorbing)

Indirect Effects:

- Changes in cloud properties (consistency, reflectivity), precipitation

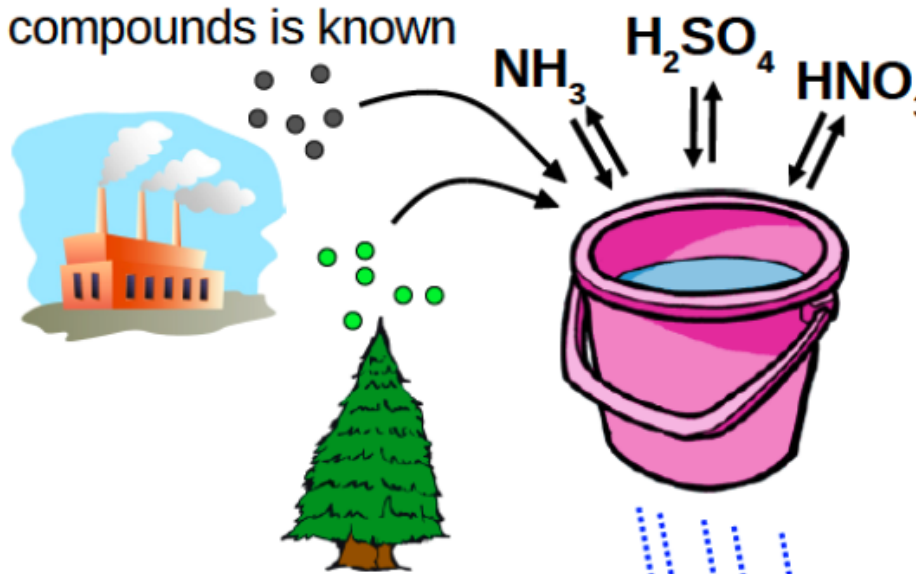
Controlled by: Emissions, nucleation processes, deposition, chemistry

Aerosols in CESM:

- Bulk Aerosols Model (BAM)
- Modal aerosol Model (MAM)
- Secondary Organic Aerosols (require chemistry)

Bulk aerosol scheme

- Only total mass of aerosol compounds is known
- No information on
  - Particle number
  - Aerosol size distribution



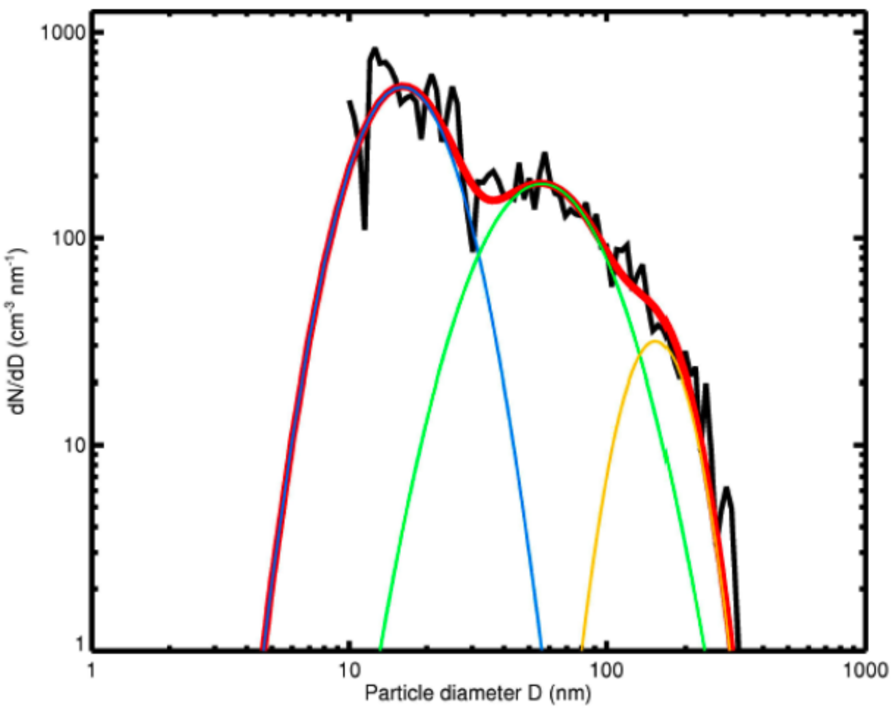
- Aerosol size distribution needs to be assumed for ...
  - radiative transfer
  - response of cloud properties to aerosol number
- Can't do aerosol nucleation
- **Numerically efficient**
- **Useful when focus is on complex gas phase / aerosol chemistry**

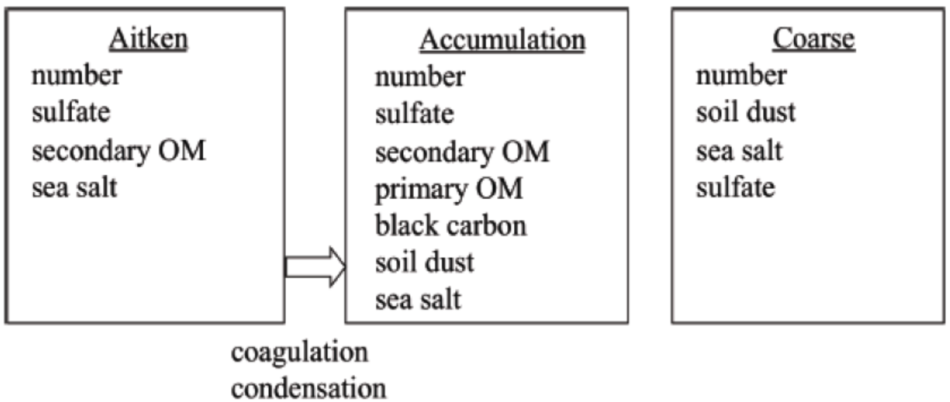
From J. Kazil, CIRES

Modal Aerosol Model (MAM3)

CESM CAM5
Aerosol size distribution using 3 modes
New (PNNL) version exists with 4 and 7 modes.

Twin Otter data (black)

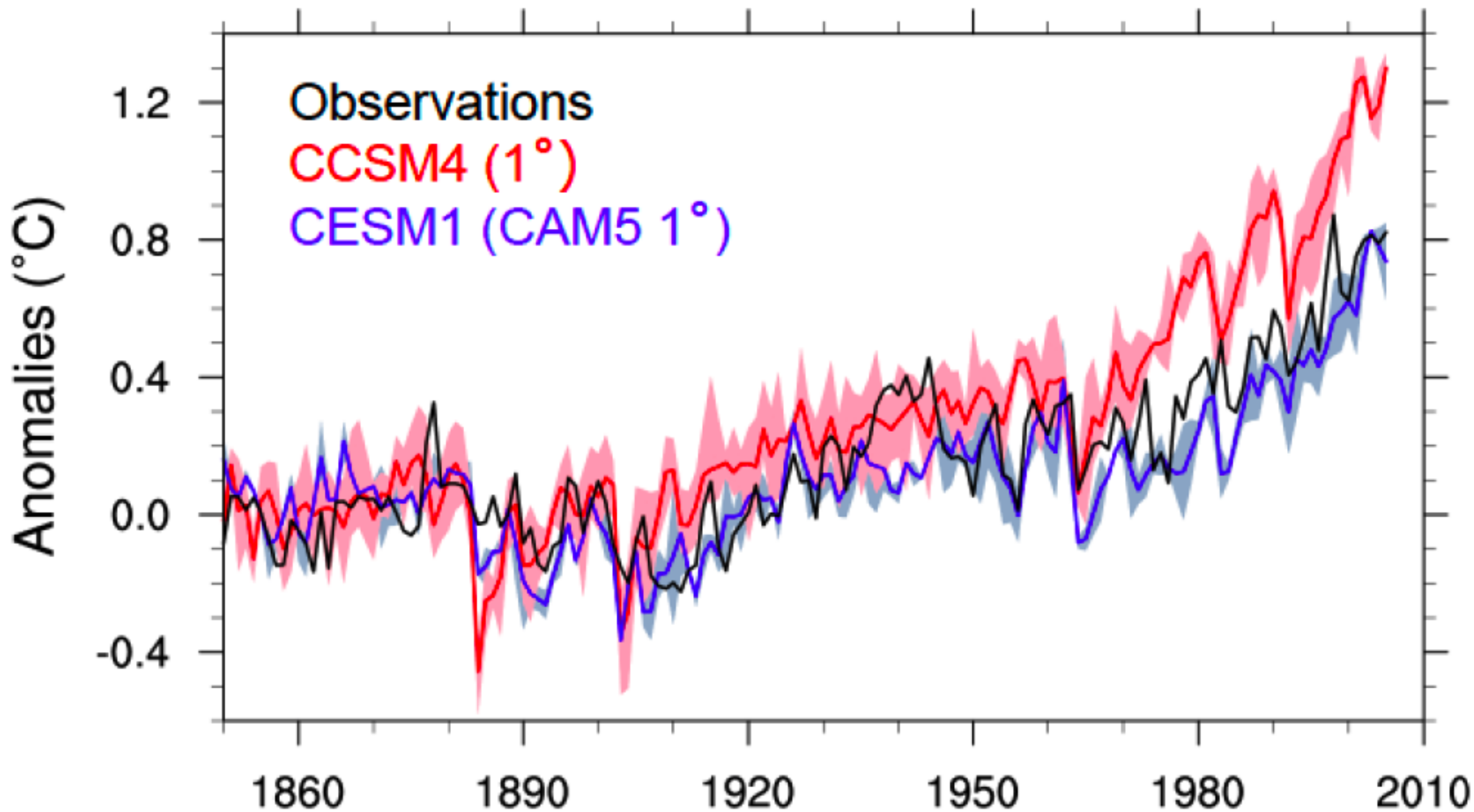




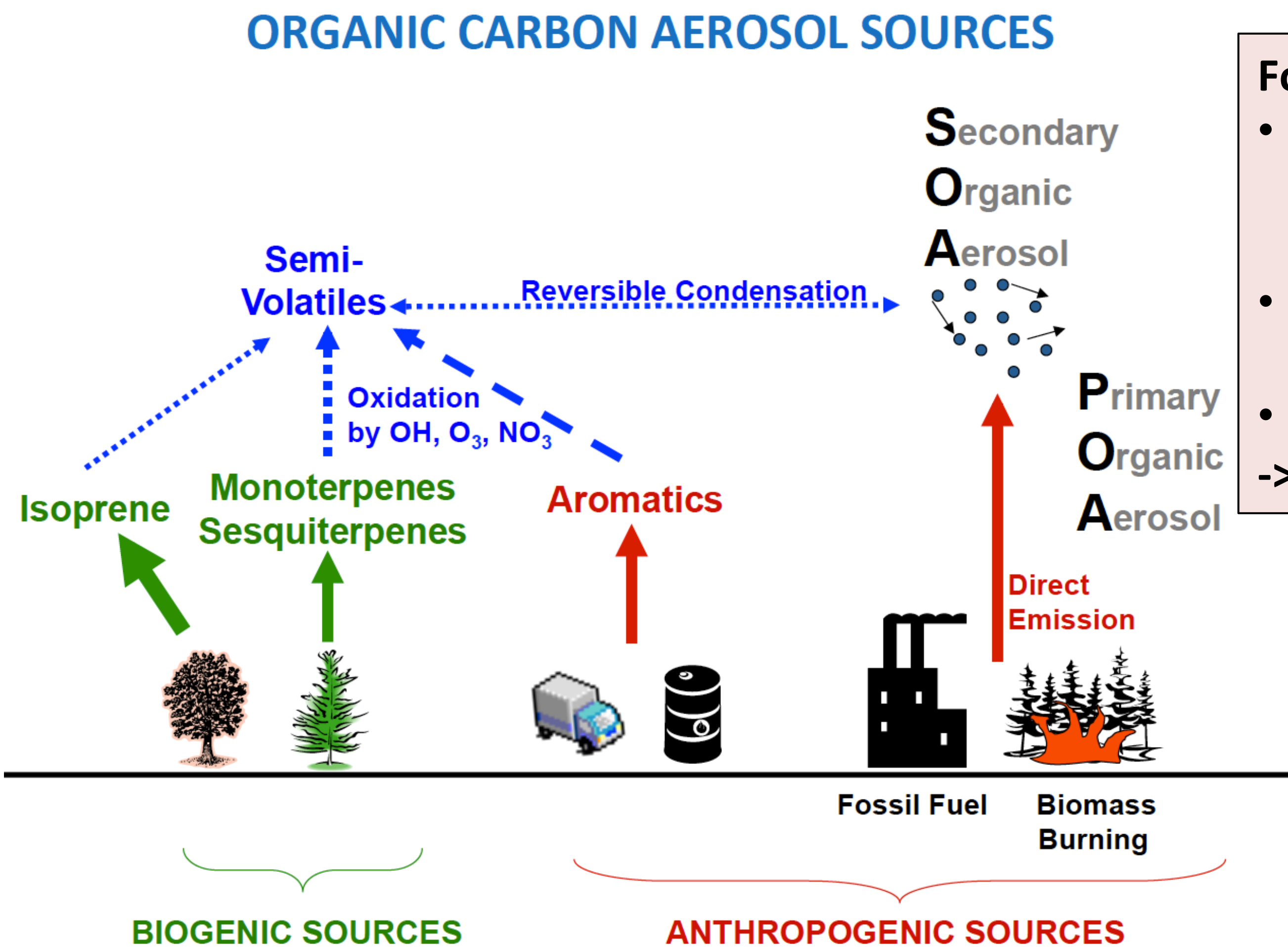
Liu et al., 2011

From J. Kazil, CIRES

Modal Aerosol Model (in CESM)



Organic Aerosols (simulated in CAM4-Chem)



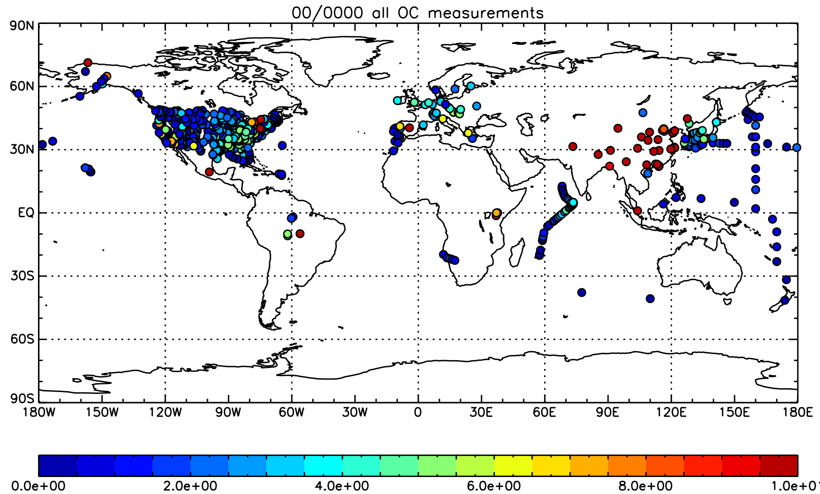
Formation of SOA

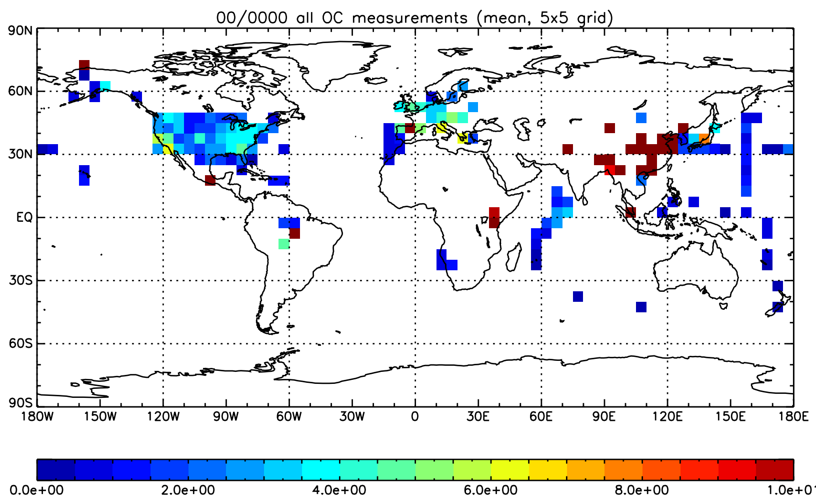
- Emissions of volatile organic carbons
- Formation of Semi-Volatiles
- Emissions of POM

-> SOA

From C. Heald, MIT Cambridge

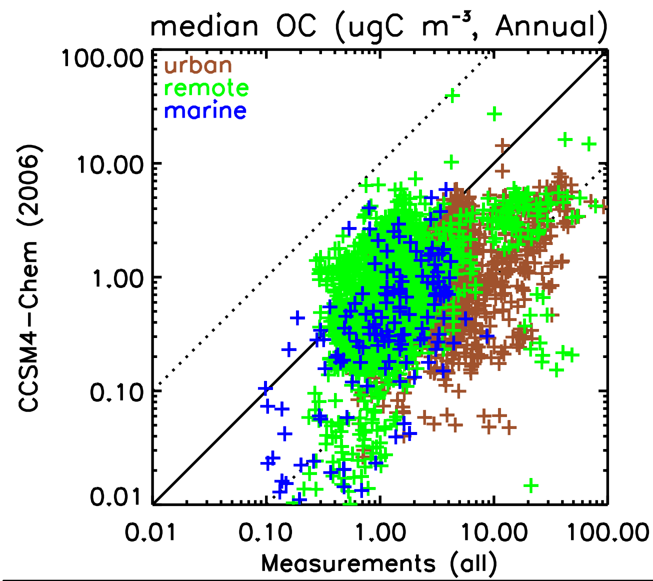
Evaluation of Organic Aerosols





Tsigaridis et al., to be submitted: An AeroCom intercomparison exercise on organic aerosol global modeling (to ACP, 70+ authors, 40+ affiliations)

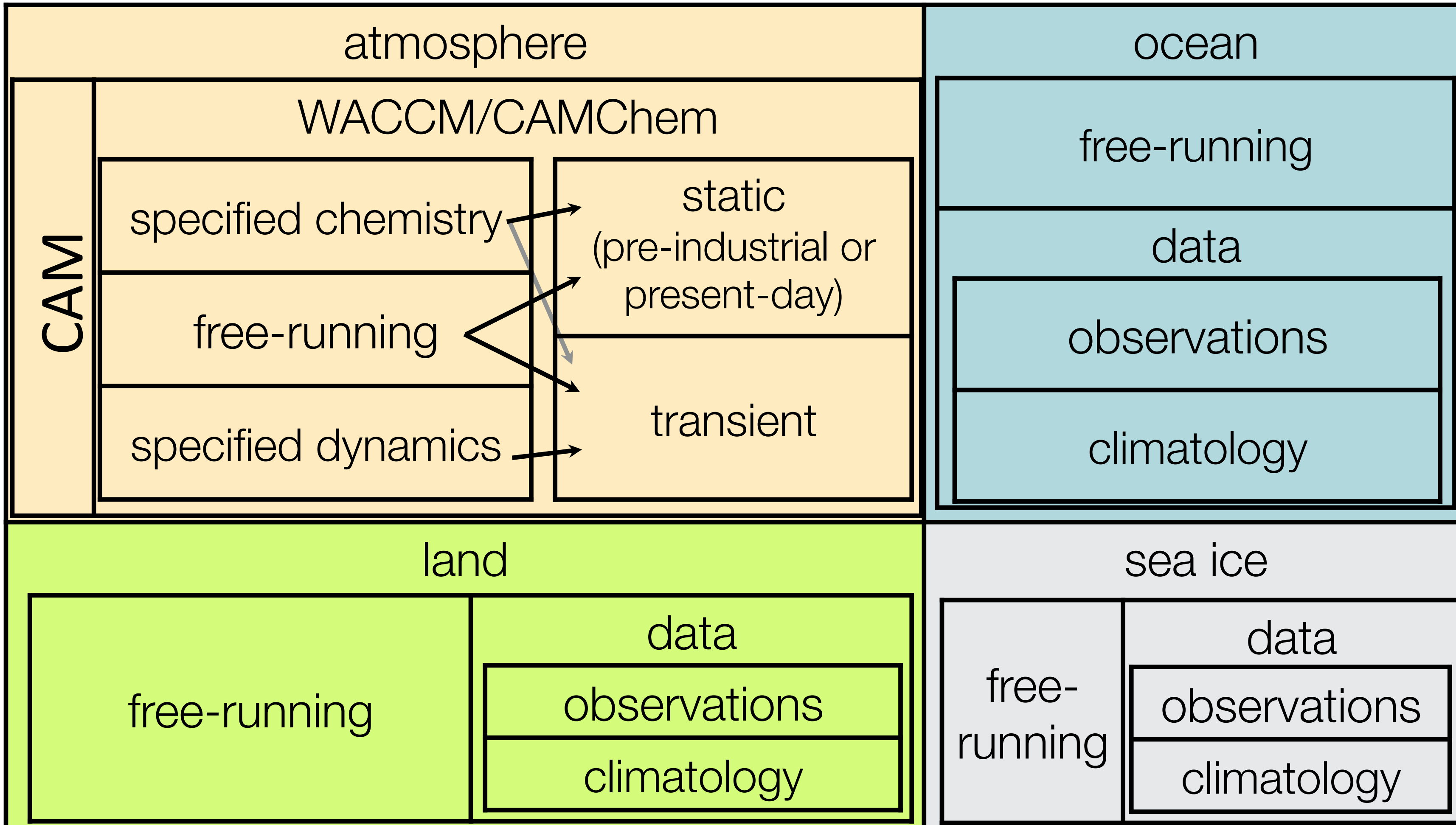
- Uses ground based, filters, period anywhere between 1980 and 2007
- CAM4Chem, SOA advanced chemistry



Work is in progress to further improve SOA in CAMChem

WACCM /CAMChem Configurations and Applications

WACCM /CAMChem component configurations



CAM-Chem with Online/Offline Meteorology

Fully coupled model:

Chemistry interacts with radiation, atmosphere, land, ocean

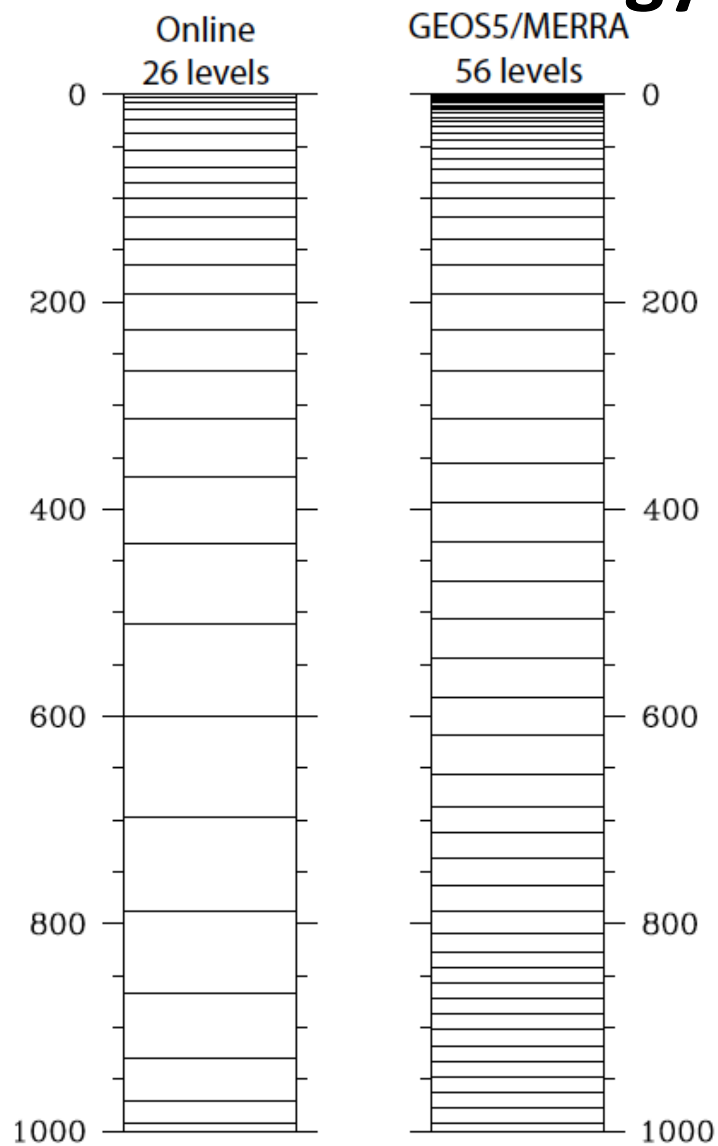
Specified sea/ice distribution

Specified Dynamics (offline) model:

Meteorological data are prescribed or nudge (%) for defined altitudes

Nudging: the amount and altitudes of nudging can be defined

Can be run in WACCM with 88 levels instead



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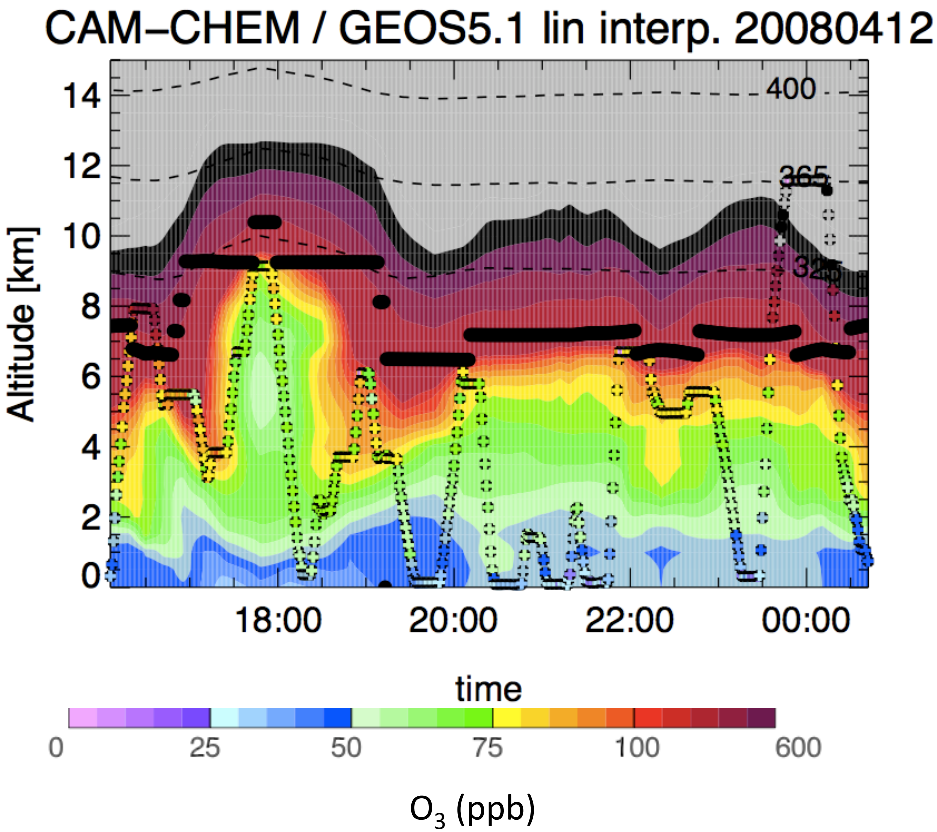
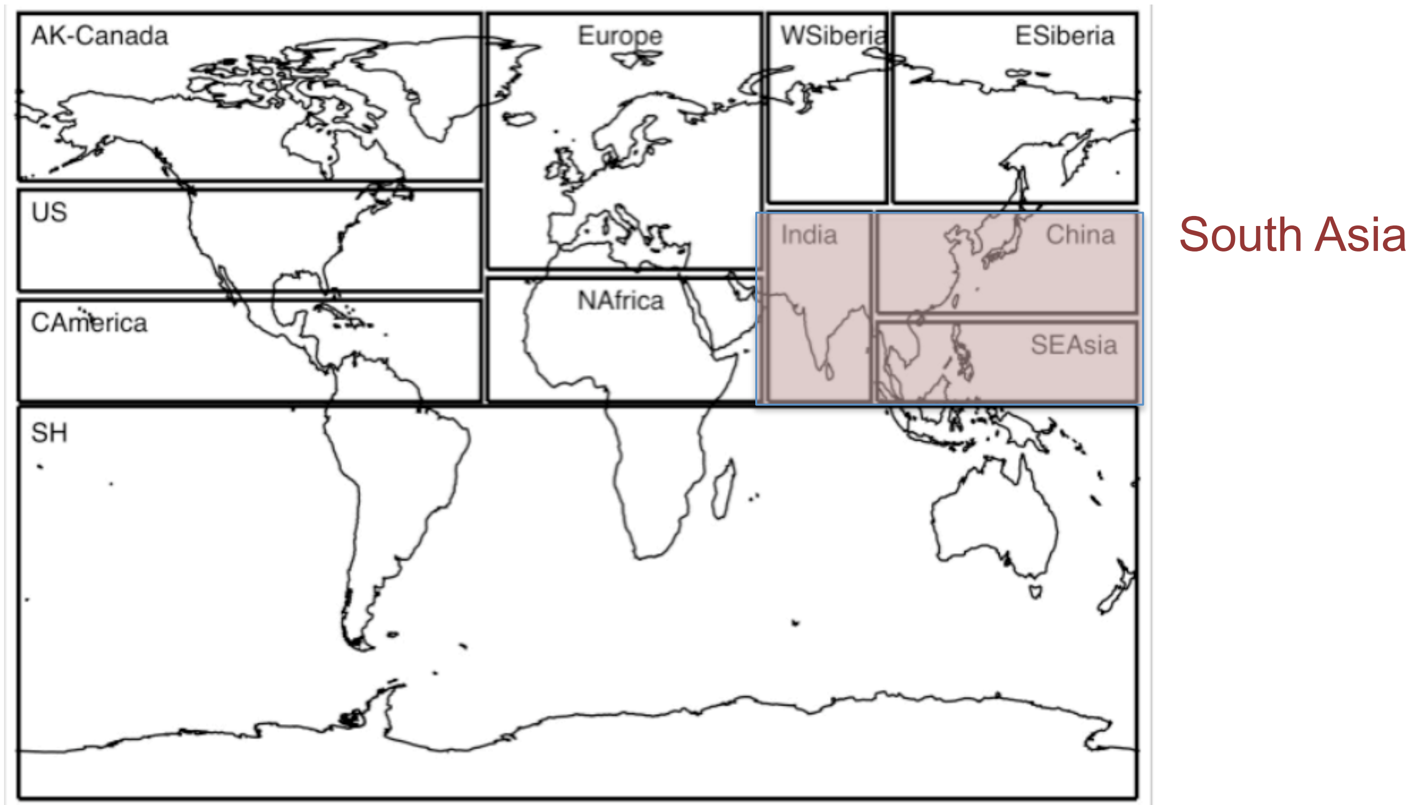
CAM-Chem with Offline Meteorology



Support of aircraft campaigns:
Model output on the
flight path for direct comparison

O_3, CO, BC tags with Offline Meteorology

Emmons et al., 2012, GMD



The Model for Ozone and Related chemical Tracers (MOZART4)

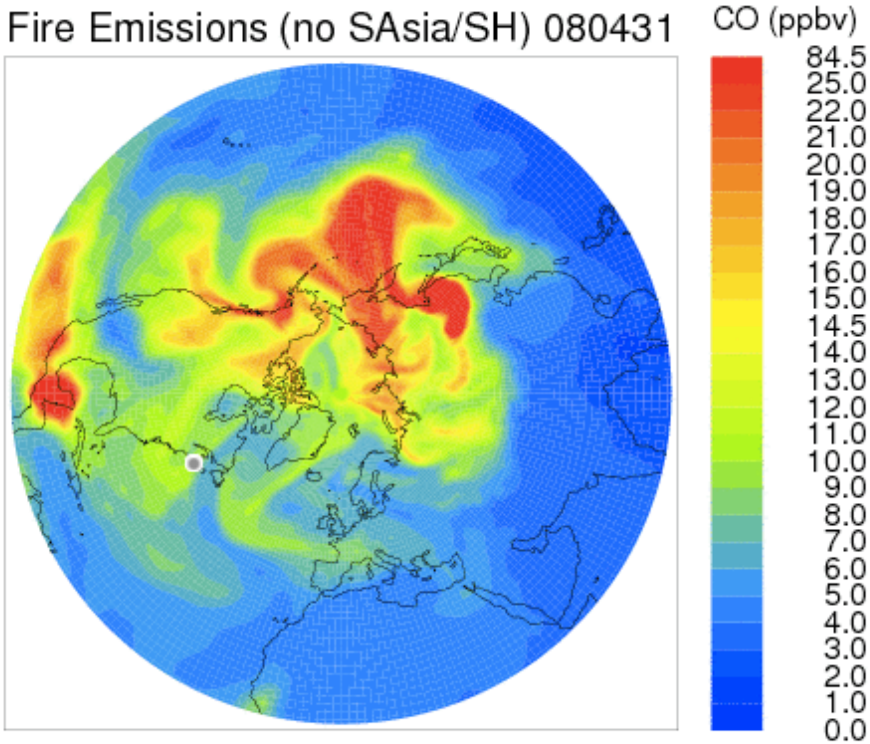
Emissions: Streets ARCTAS emissions + daily fires (C. Wiedinmyer)

Vertical Injection of Fire Emission between 0-6 km

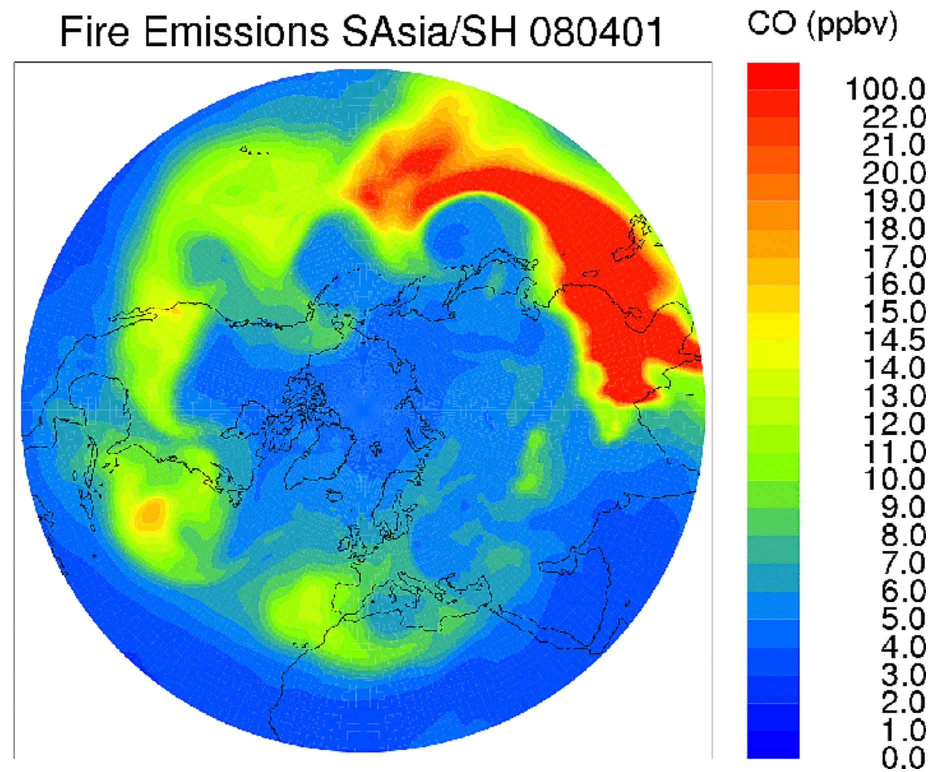
Importance of Fire Emissions

April

without South Asia and SH



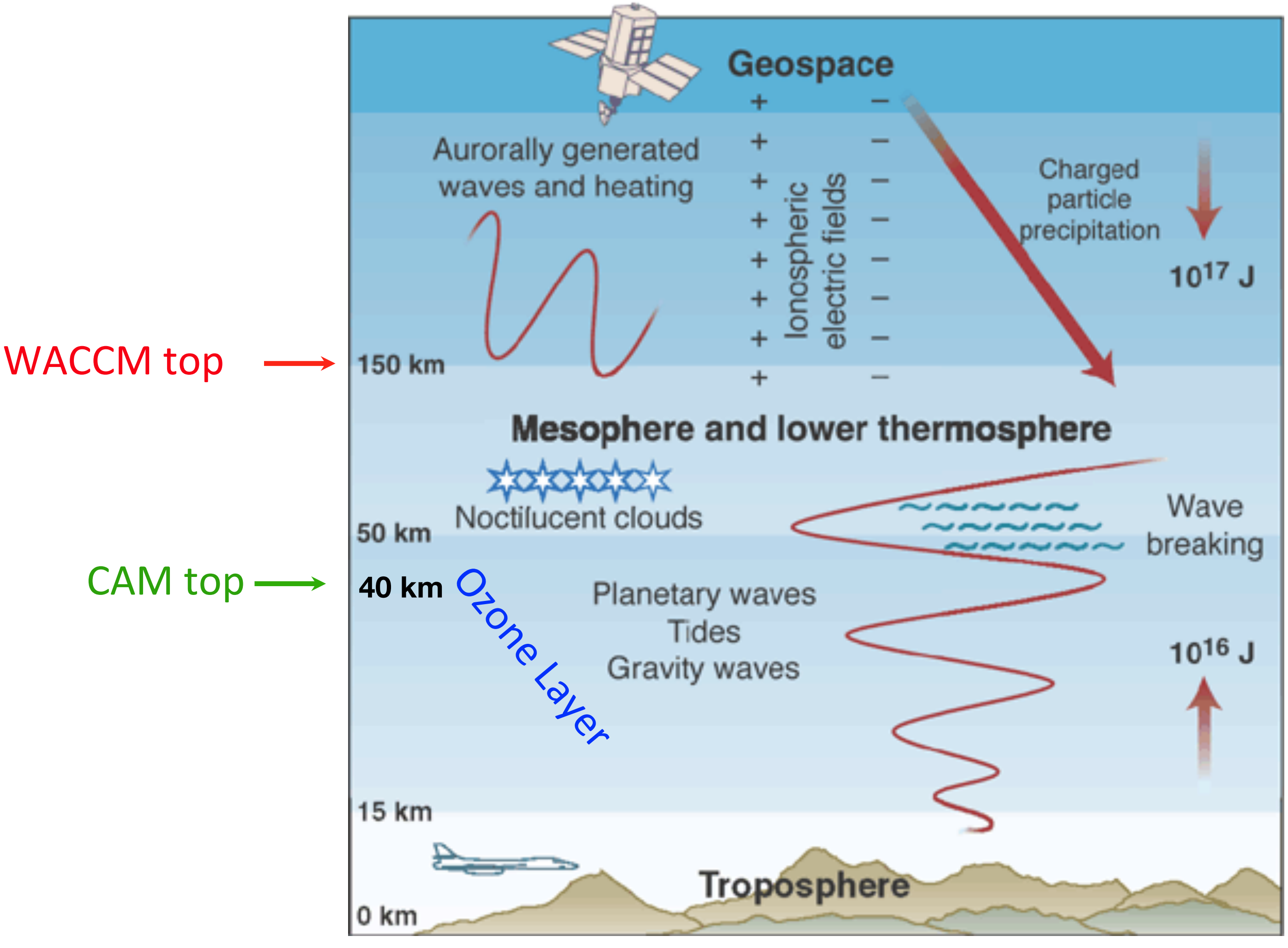
South Asia and SH only



CO averaged column between surf. and 200 hPa

Tilmes et al., 2011

WACCM: The High-Top Model

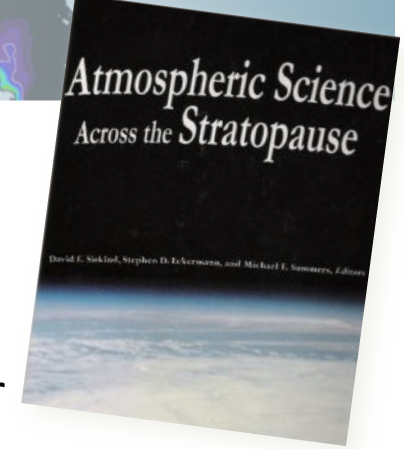


WACCM: additions to CAMChem

- Extends from surface to 5.1×10^{-6} hPa (~150 km), with 66 vertical levels
- Detailed neutral chemistry model for the middle atmosphere,
  - heterogeneous chemistry on PSCs and sulfate aerosol
  - heating due to chemical reactions
  - can be run with extended tropospheric chemistry
- Model of ion chemistry in the mesosphere/lower thermosphere (MLT), ion drag, auroral processes, and solar proton events
- EUV and non-LTE longwave radiation parameterizations
- QBO imposed, based on cyclic, fixed-phase, or observed winds
- Volcanic aerosol heating calculated explicitly
- Gravity wave drag deposition from vertically propagating GWs generated by orography, fronts, and convection
- Molecular diffusion and constituent separation
- Thermosphere extension (WACCM-X) to ~500 km

WACCM: Motivation

Roble, Geophysical Monograph, **v. 123**, p. 53, 2000

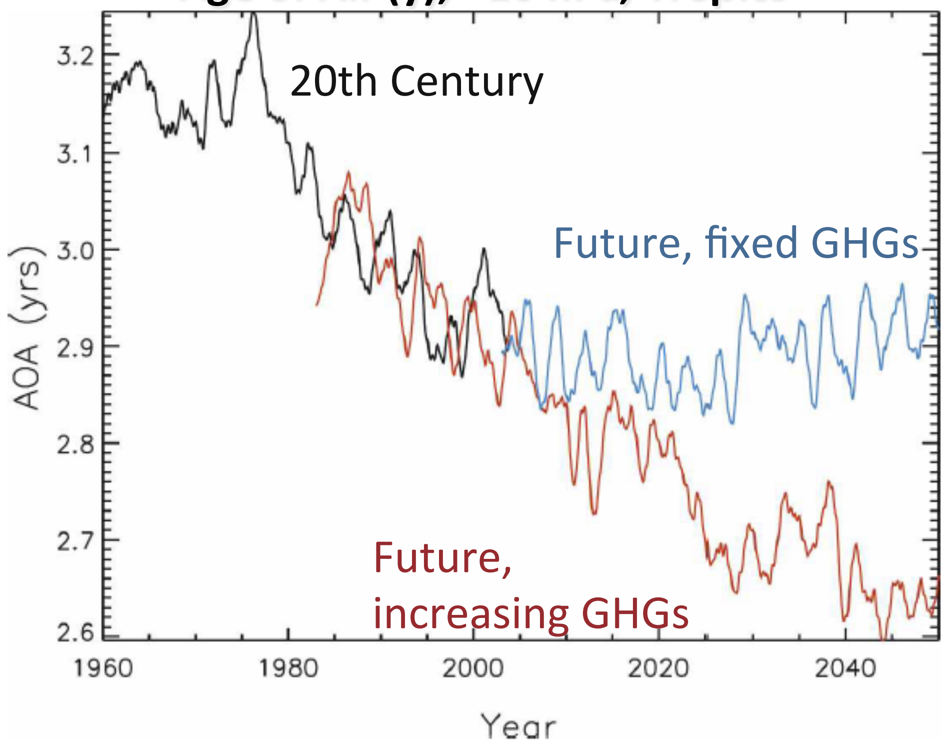


- **Coupling between atmospheric layers:**
  - Waves transport energy and momentum from the lower atmosphere to drive the QBO, SAO, sudden warmings, mean meridional circulation
  - Solar inputs, e.g. auroral production of NO in the mesosphere and downward transport to the stratosphere
  - Stratosphere-troposphere exchange
- **Climate Variability and Climate Change:**
  - What is the impact of the stratosphere on tropospheric variability?
  - How important is coupling among radiation, chemistry, and circulation? (e.g., in the response to O_3 depletion or CO_2 increase)
  - Response to solar variability: impacts mediated by chemistry?
- **Interpretation of Satellite Observations**

Stratospheric Dynamics

Acceleration of the Brewer-Dobson Circulation due to increases in Greenhouse Gases, Garcia and Randel, JAS, 2008

Age of Air (y), ~10 hPa, Tropics



Increasing GHGs:

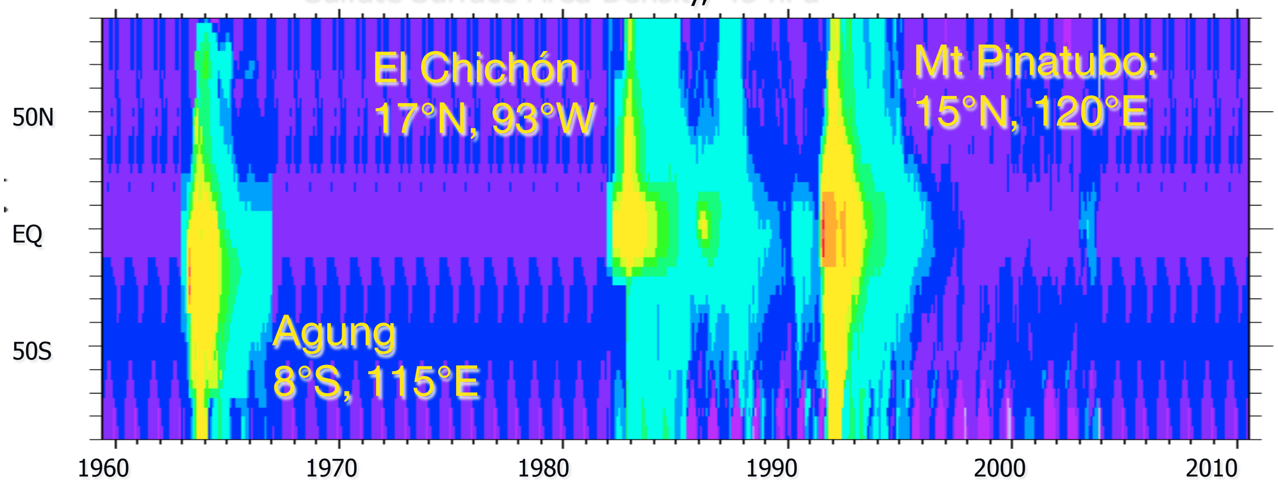
- Enhanced propagation of wave activity into the lower stratosphere and its dissipation in the subtropics
- Changes in meridional temperature gradient affect zonal winds, which change the regions where waves dissipate, increasing momentum deposition

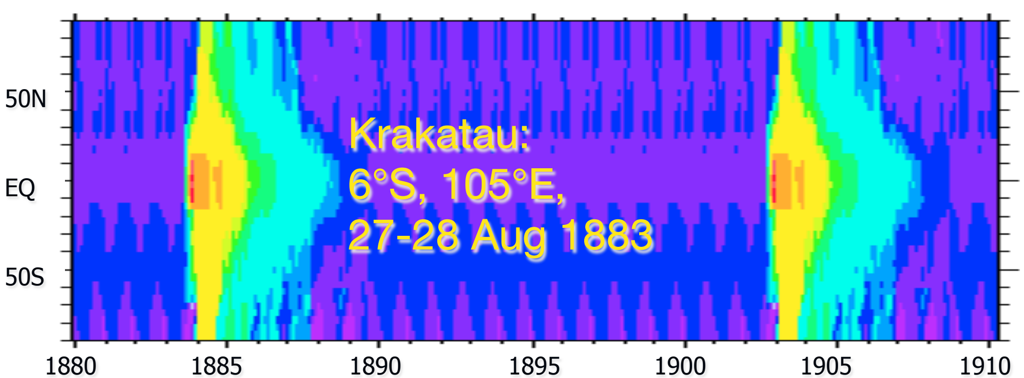
-> impact on stratospheric/ tropospheric exchange

Stratospheric Aerosols (past and present day)

- Described by a stratospheric sulfate surface area density climatology

Sulfate Surface Area Density, 43 hPa



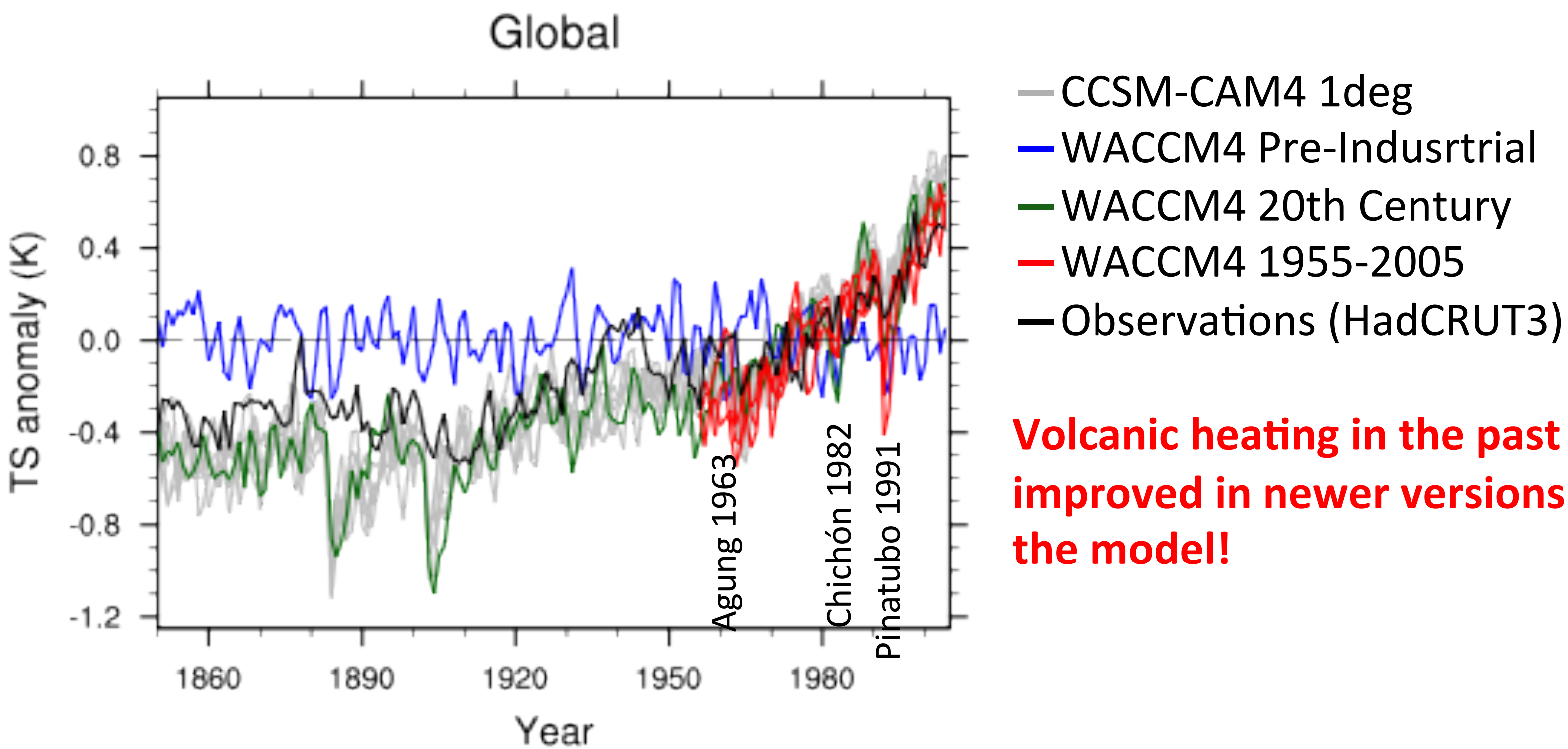


Santa Maria: 14°N, 91°W, 24 Oct. 1902

- Observations used: SAGE I, SAGE II, SAM II, and SME instruments
- Updated version available including increasing background aerosols after 2000, based on CALIPSO
- Used Pinatubo aerosol for Krakatau and Santa Maria.

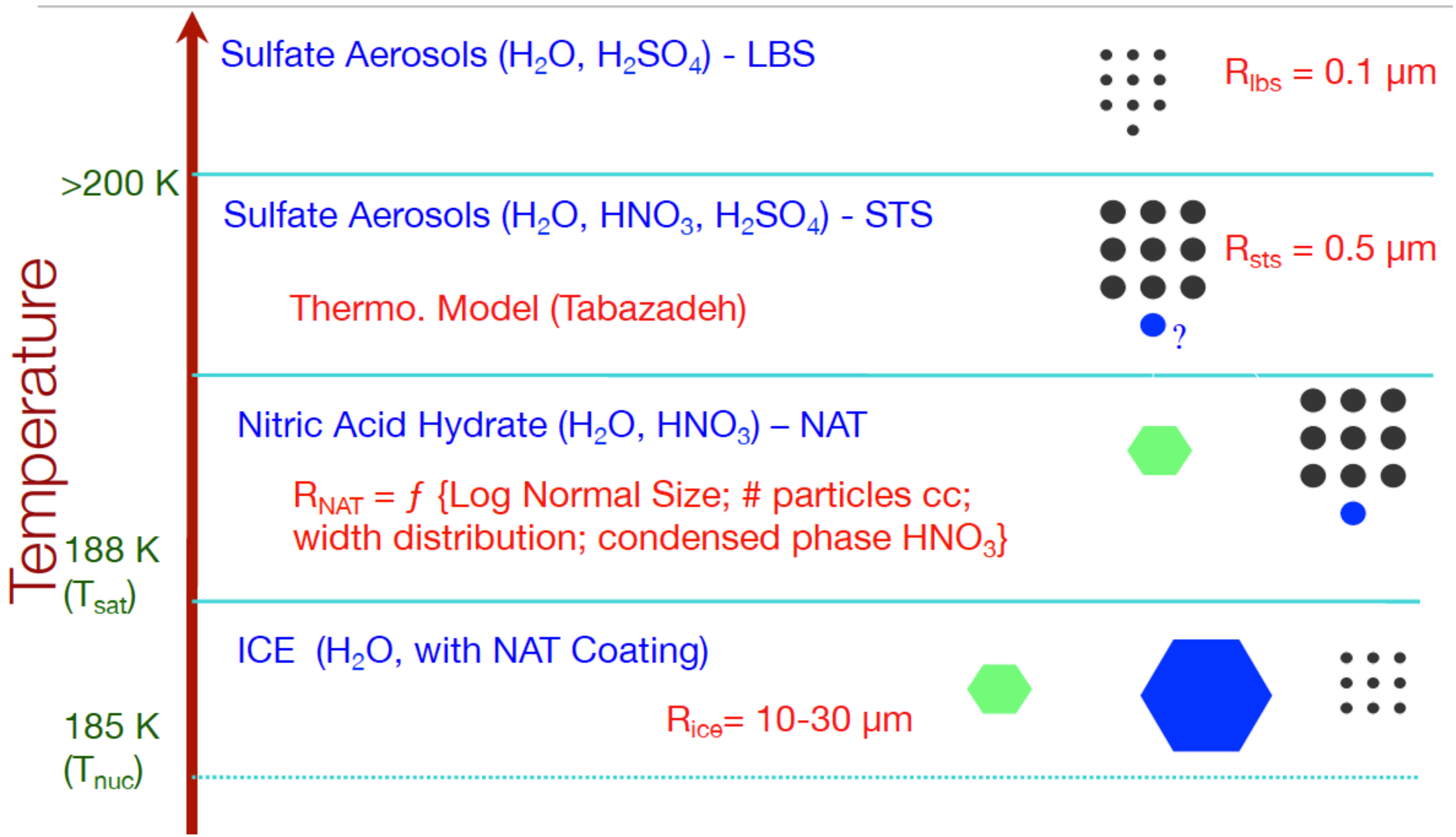
Stratospheric Aerosol Heating

Surface Temperature Anomalies normalized to 1961-90



Volcanic heating in the past will improved in newer versions of the model!

WACCM Heterogeneous Chemistry Module



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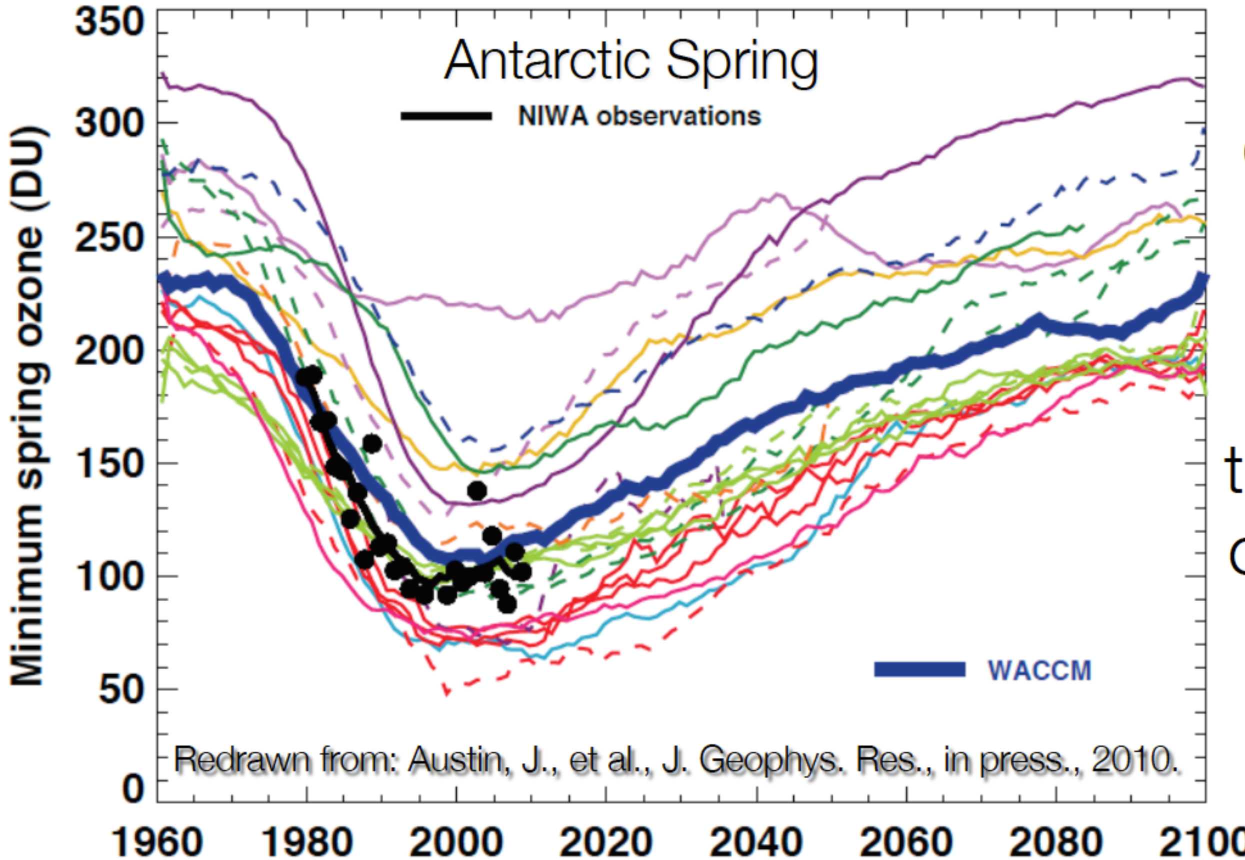
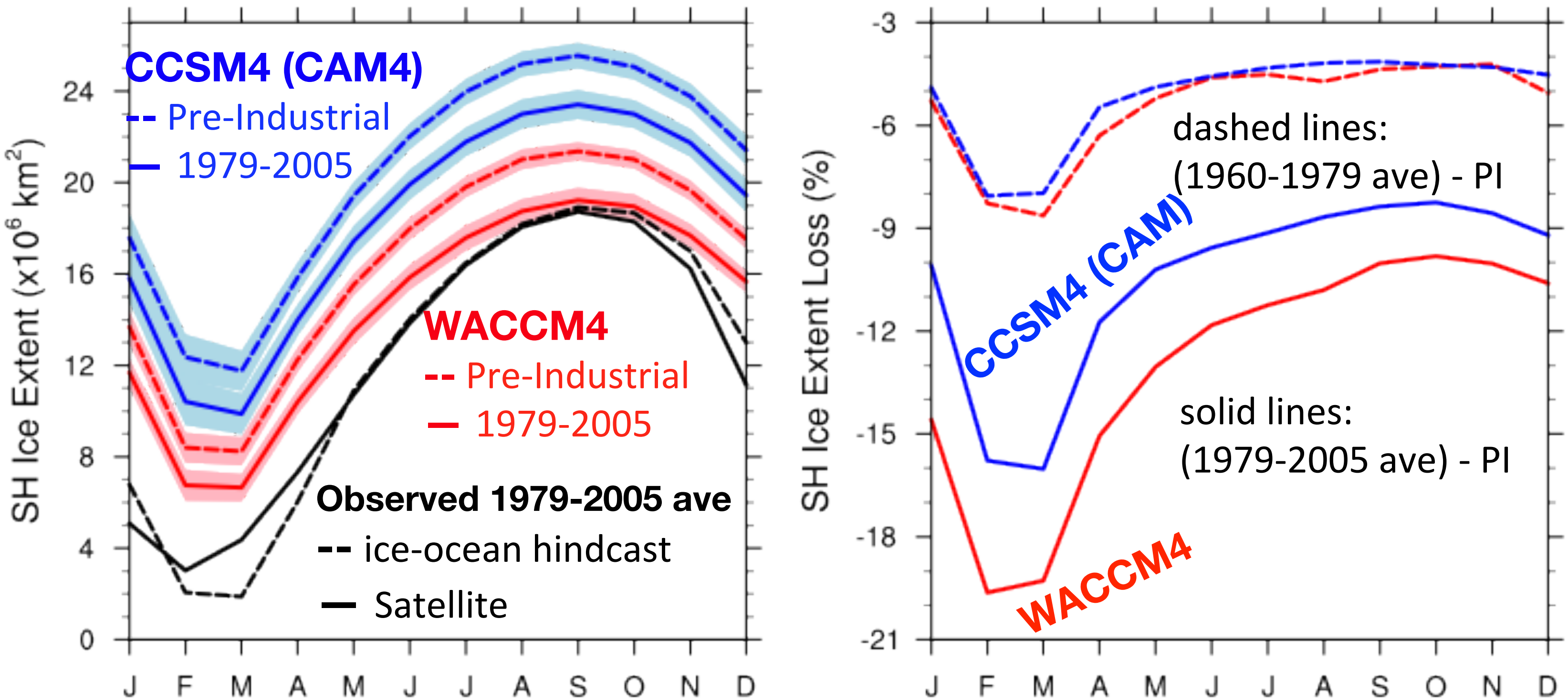
WACCM Polar Ozone Trend



WACCM does better than most models at calculating the evolution of the ozone hole.

Austin et al., 2010

Response to Arctic Sea-Ice Extent



More realistic ozone loss in WACCM drives changes in winds that enhance sea-ice loss, producing sea-ice extent closer to modern observations.

Marsh et al., 2012

WACCM and CAM-Chem Customer Support

CGD Forum: http://bb.cgd.ucar.edu/

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