Description of Chemistry, Aerosols in CESM and WACCM

Presented by Simone Tilmes, Mike Mills
NESL: ACD/CGD

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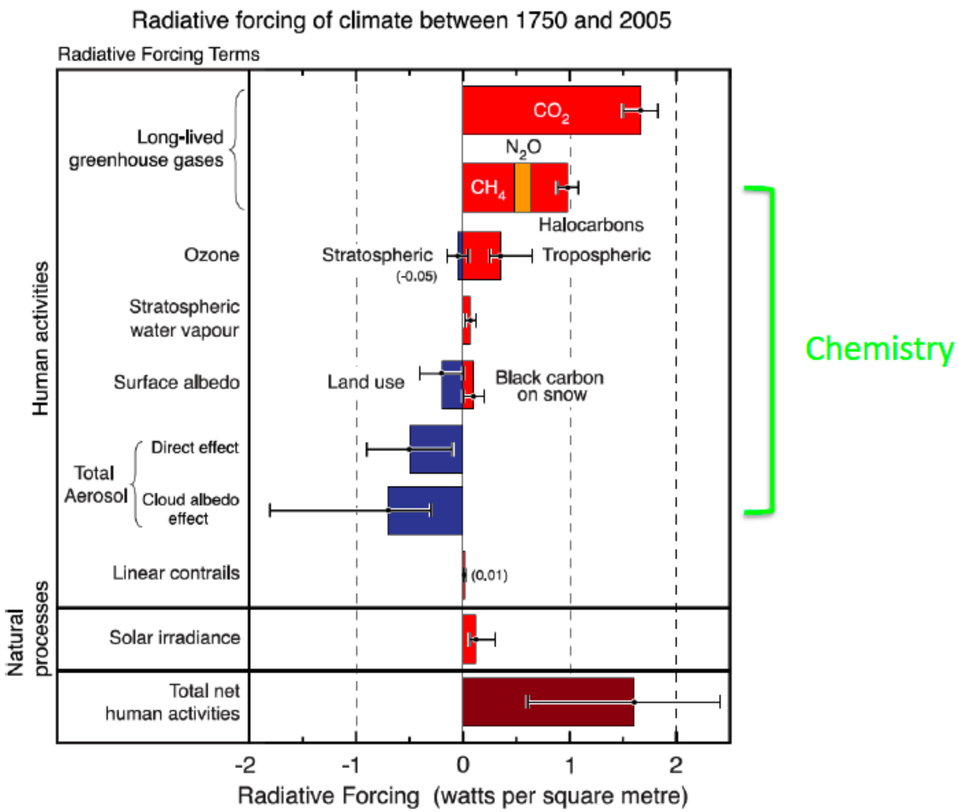
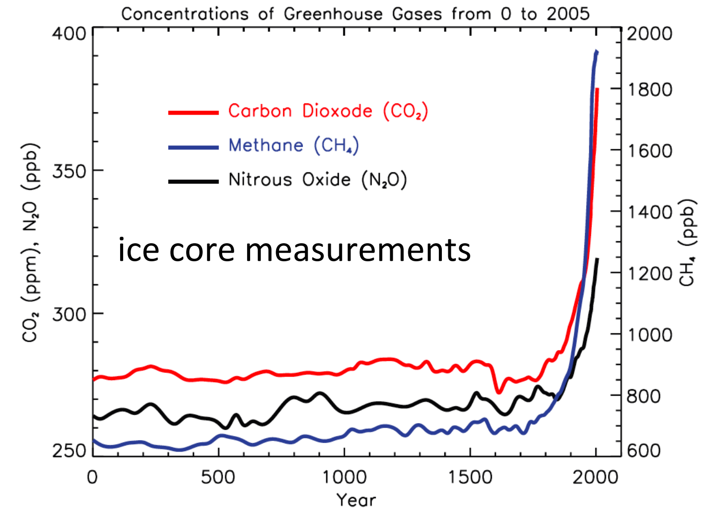
Software Engineers: Francis Vitt , Sean Santos
CAMChem Liaison: Simone Tilmes
Aerosol Liaison: Po-Lun Ma
WACCM Liaison: Mike Mills

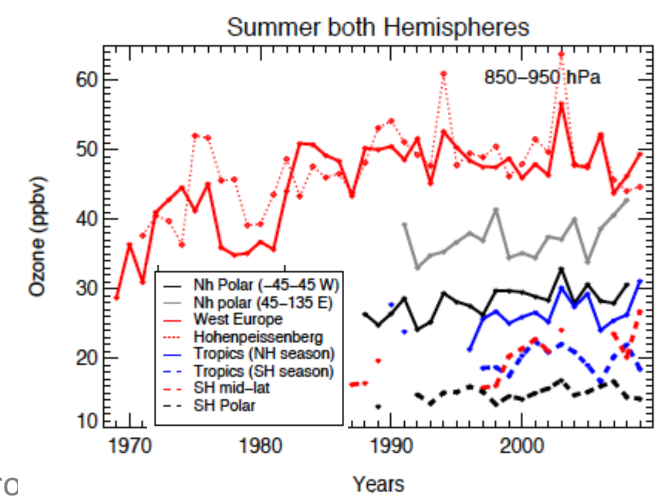
Motivation: Modeling with Chemistry



Forster and Ramaswamy. AR4 Chapter 2



Tropospheric ozone evolution based on ozonesondes (1968-2011)



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

Office of Science

Tropospheric Chemistry

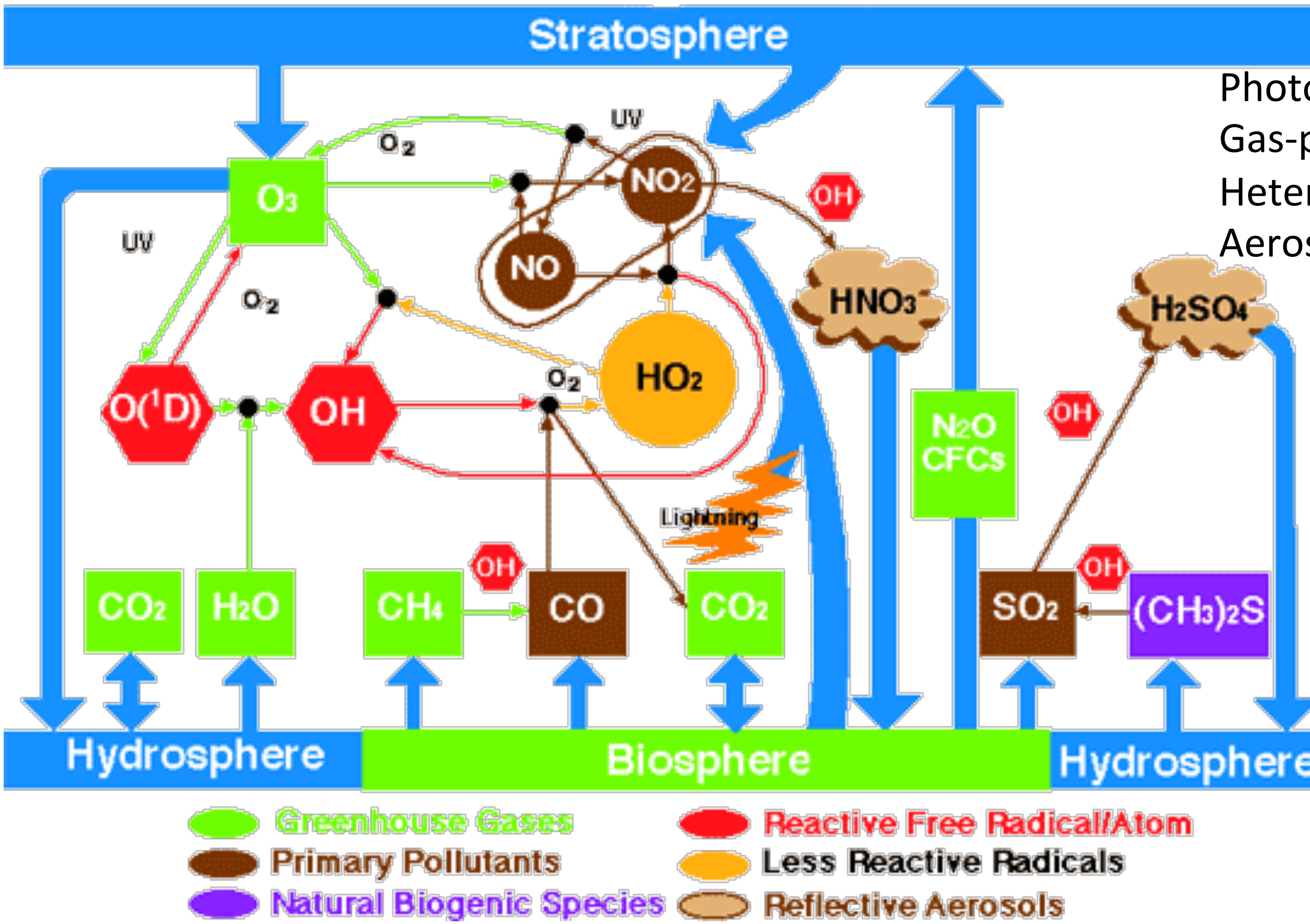


Photo-chemistry
Gas-phase chemistry
Heterogeneous chemistry
Aerosol formation

OH determines the lifetime of Methane

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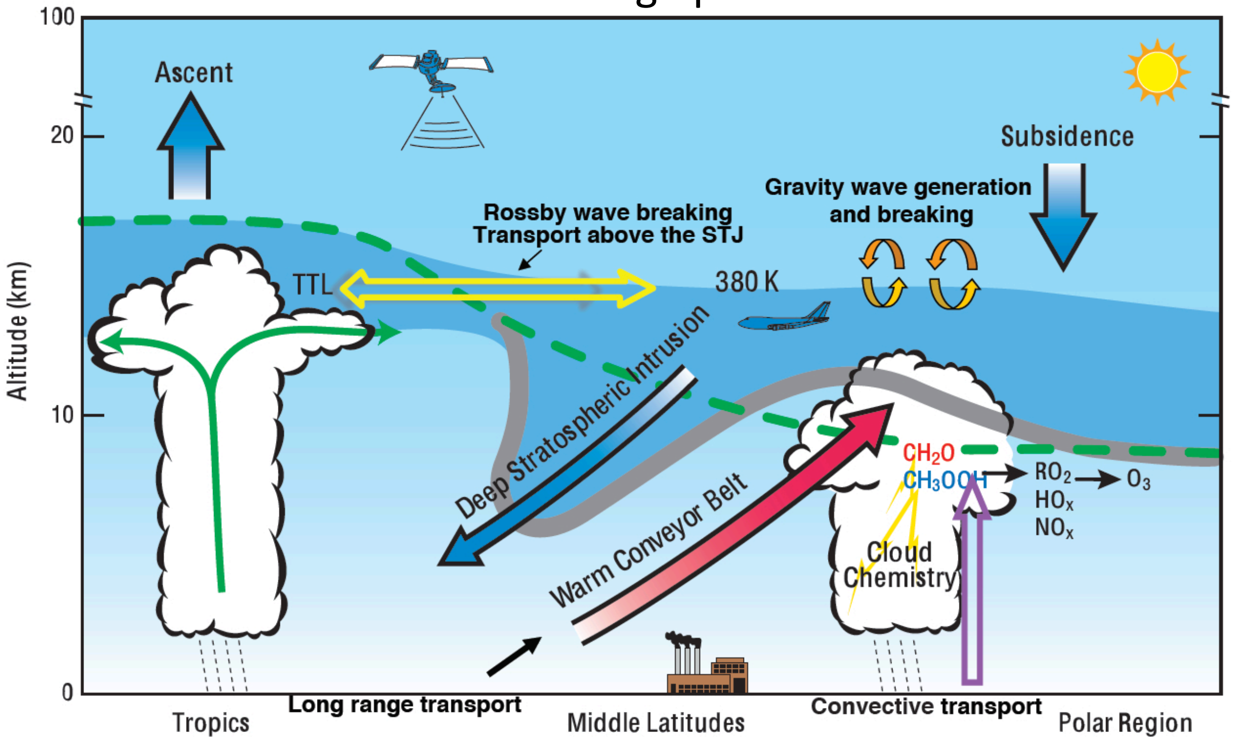
Importance of the Stratosphere, and Exchange Processes

UTLS exchange processes

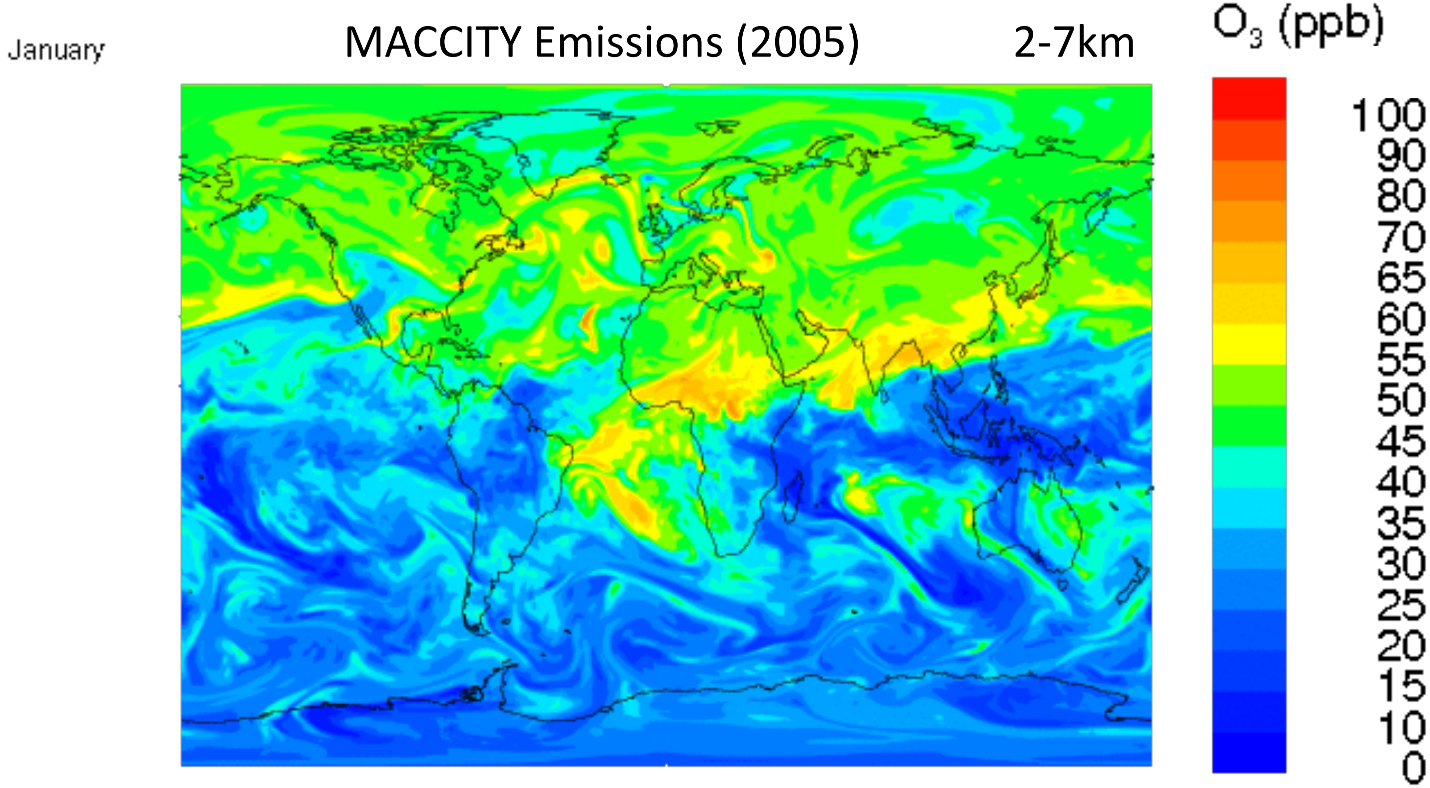


Stratosphere-Troposphere Analyses of Regional Transport (2008)

- 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

Interactive Modeling with Chemistry



Tropospheric ozone distribution is dependent on:

- changes in precursors: NO_x ($NO + NO_2$), CO, volatile organic compounds (VOCs) and other emissions
- Implications for air quality and ecosystem: EPA defines standards for air quality (75 ppb)
- Meteorology and Removal

-> Interaction with Radiation (climate gas)

Modeling without Chemistry-Climate Interactions in CESM

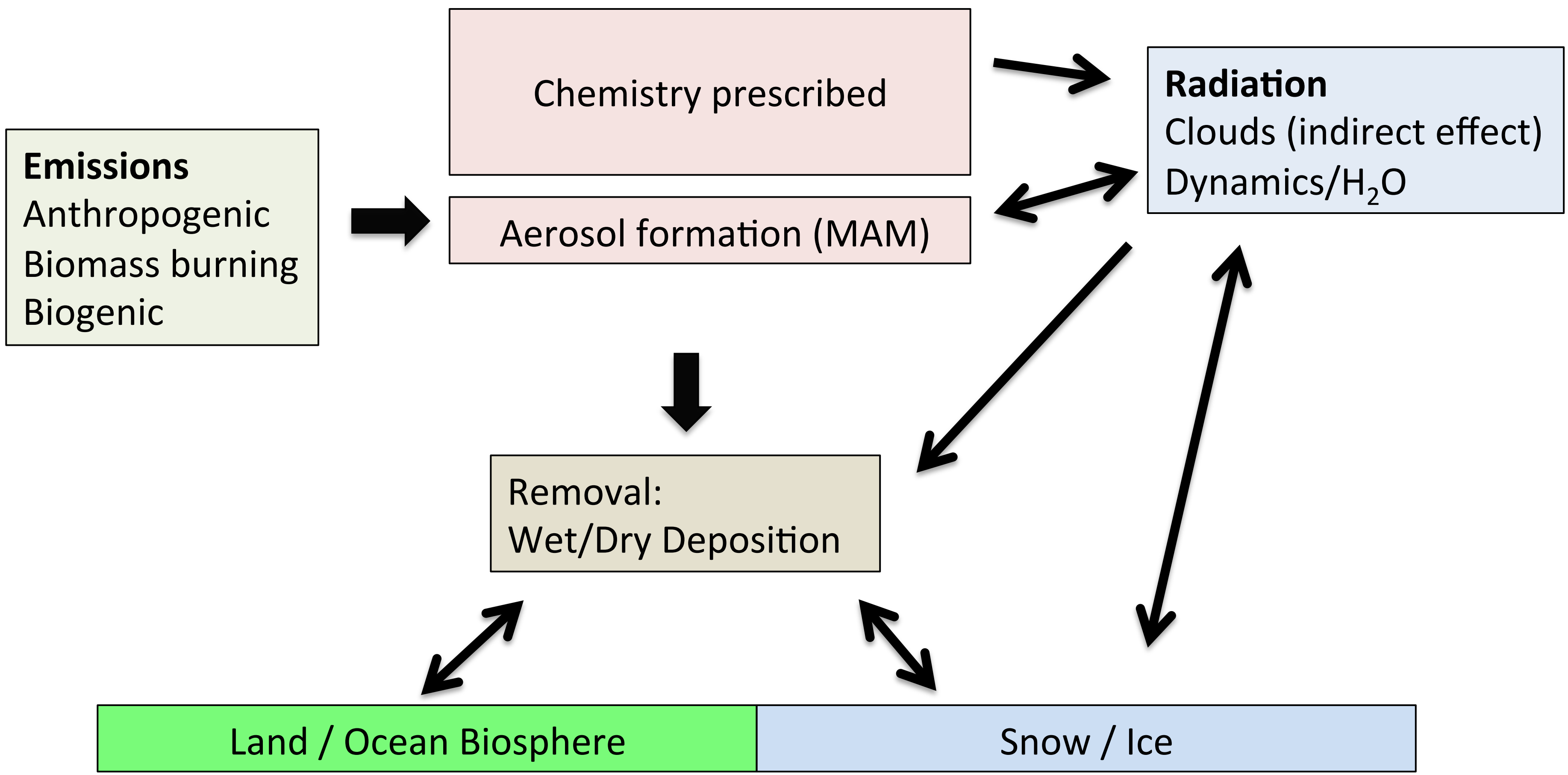
- Chemistry and aerosols are prescribed in **CAM4**: (prescribed monthly fields of CO_2, CH_4, O_3, N_2O, CFCs)

- Aerosols are calculated in **CAM5** (Modal Aerosols Model MAM), but not coupled with chemistry, simple chemistry is added ("fixed" oxidants) (prescribe: N_2, O_2, H_2O, O_3, OH, NO_3, HO_2; chemically active: H_2O_2, H_2SO_4, SO_2, DMS, SOAG)

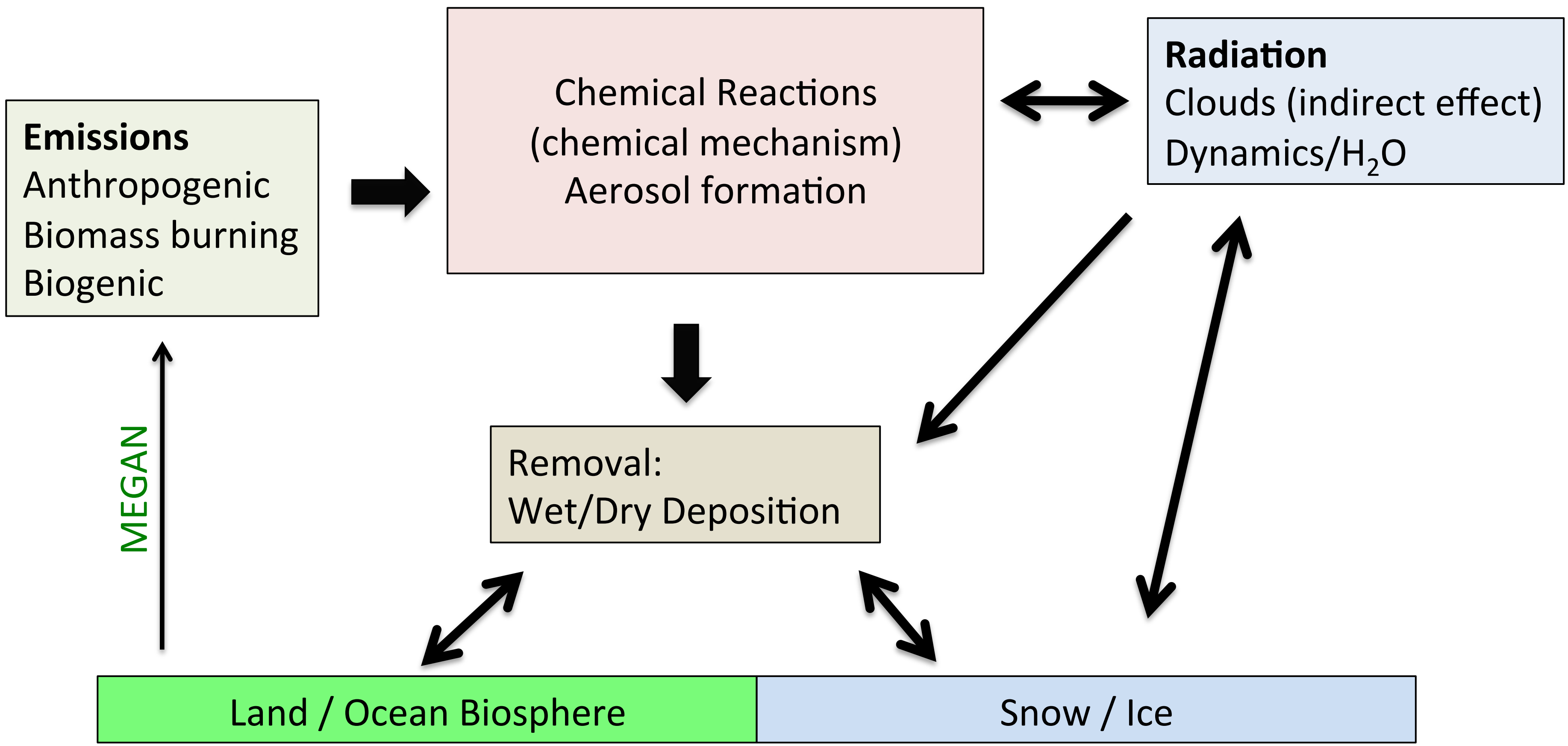
No interaction between Chemistry and Climate

-> prescribed fields have to be derived using chemistry-climate simulations

Chemistry in CESM CAM5



Chemistry in CESM CAM5-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

Chemical Mechanism: includes set of equations

rates (temperature dependence etc.)

Photolysis
End Photolysis
Reactions
End Reactions
Heterogeneous
End Heterogeneous
Ext Forcing
End Ext Forcing

```
* ---------------------------------------------------------------
* Odd-Oxygen Reactions (O1D only)
* ---------------------------------------------------------------
 [O1D_N2]            O1D + N2 -> O + N2                         ; 2.15e-11,   110.
 [O1D_O2b]           O1D + O2 -> O + O2                         ; 3.30e-11,    55.
 [ox_l1]             O1D + H2O -> 2*OH                          ; 1.63e-10,    60.
 [O1D_N2Oa]          O1D + N2O -> 2*NO                          ; 7.25e-11,    20.
 [O1D_N2Ob]          O1D + N2O -> N2 + O2                       ; 4.63e-11,    20.
 [O1D_O3]            O1D + O3 -> O2 + O2                        ; 1.20e-10
 [O1D_CFC11]         O1D + CFC11 -> 3*CL                        ; 2.02e-10
 [O1D_CFC12]         O1D + CFC12 -> 2*CL                        ; 1.204e-10
 [O1D_CFC113]        O1D + CFC113 -> 3*CL                       ; 1.50e-10
 [O1D_CFC114]        O1D + CFC114 -> 2*CL                       ; 9.75e-11
 [O1D_CFC115]        O1D + CFC115 -> CL                         ; 1.50e-11
 [O1D_HCFC22]        O1D + HCFC22 -> CL                         ; 7.20e-11
 [O1D_HCFC141B]      O1D + HCFC141B -> 2*CL                     ; 1.794e-10
 [O1D_HCFC142B]      O1D + HCFC142B -> CL                       ; 1.628e-10
 [O1D_CCL4]          O1D + CCL4 -> 4*CL                         ; 2.84e-10
 [O1D_CH3BR]         O1D + CH3BR -> BR                          ; 1.674e-10
 [O1D_CF2CLBR]       O1D + CF2CLBR -> CL + BR                   ; 9.60e-11
 [O1D_CF3BR]         O1D + CF3BR -> BR                          ; 4.10e-11
```

The chemistry preprocessor: tool that generates CAM Fortran source code; numerically solve a set of differential equations which represent the chemical reactions -> temporal evolution of the chemical tracers

Modeling with Chemistry

Available chemical mechanisms in CAM-Chem

Superfast Chemistry (CAM4/5):

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

Bulk Aerosol Model (BAM) (CAM4/5):

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, over 100 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

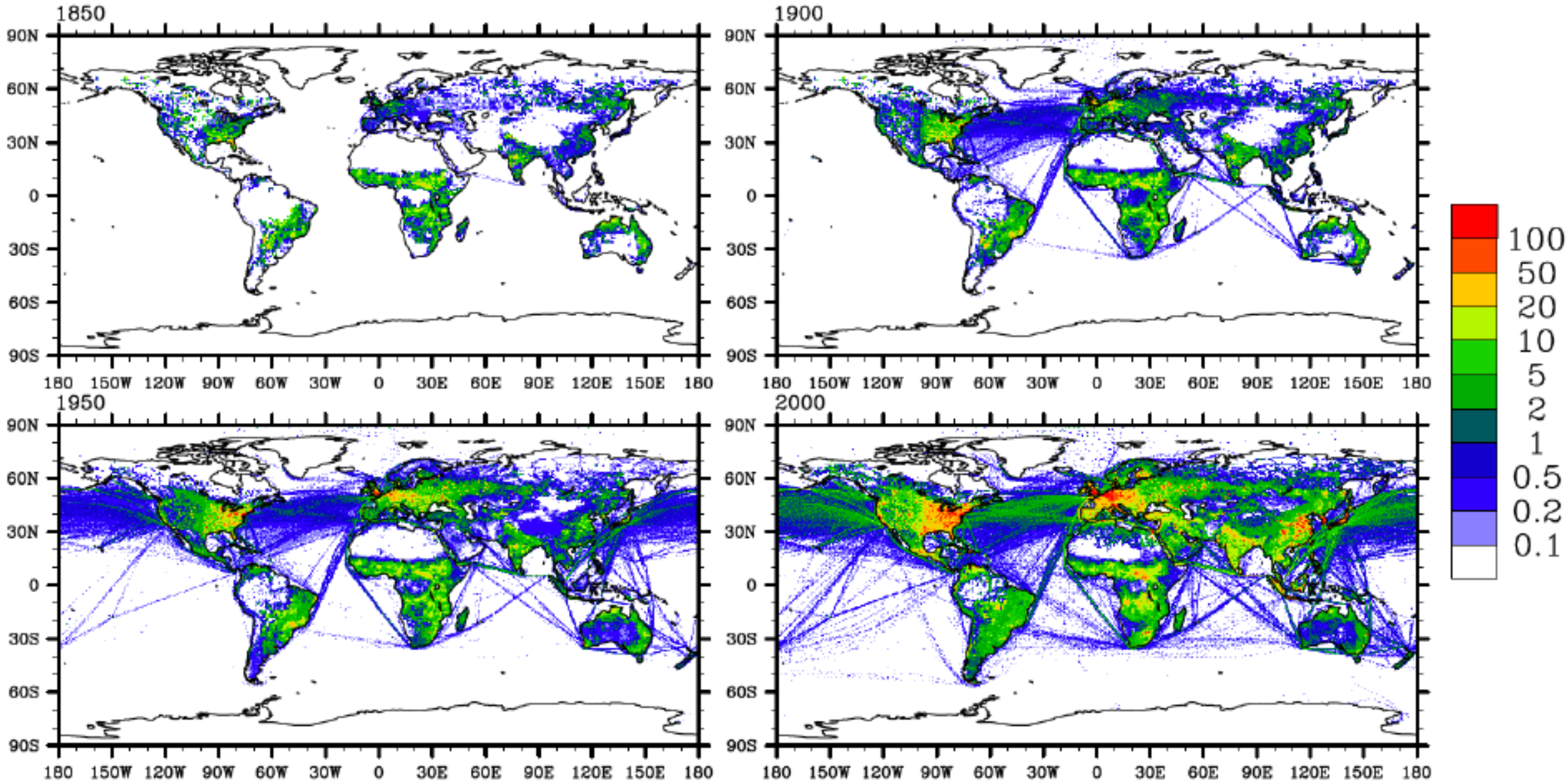
Chemistry: more and less complex mechanisms available

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

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Modeling with Chemistry

Chemistry: more and less complex mechanisms available

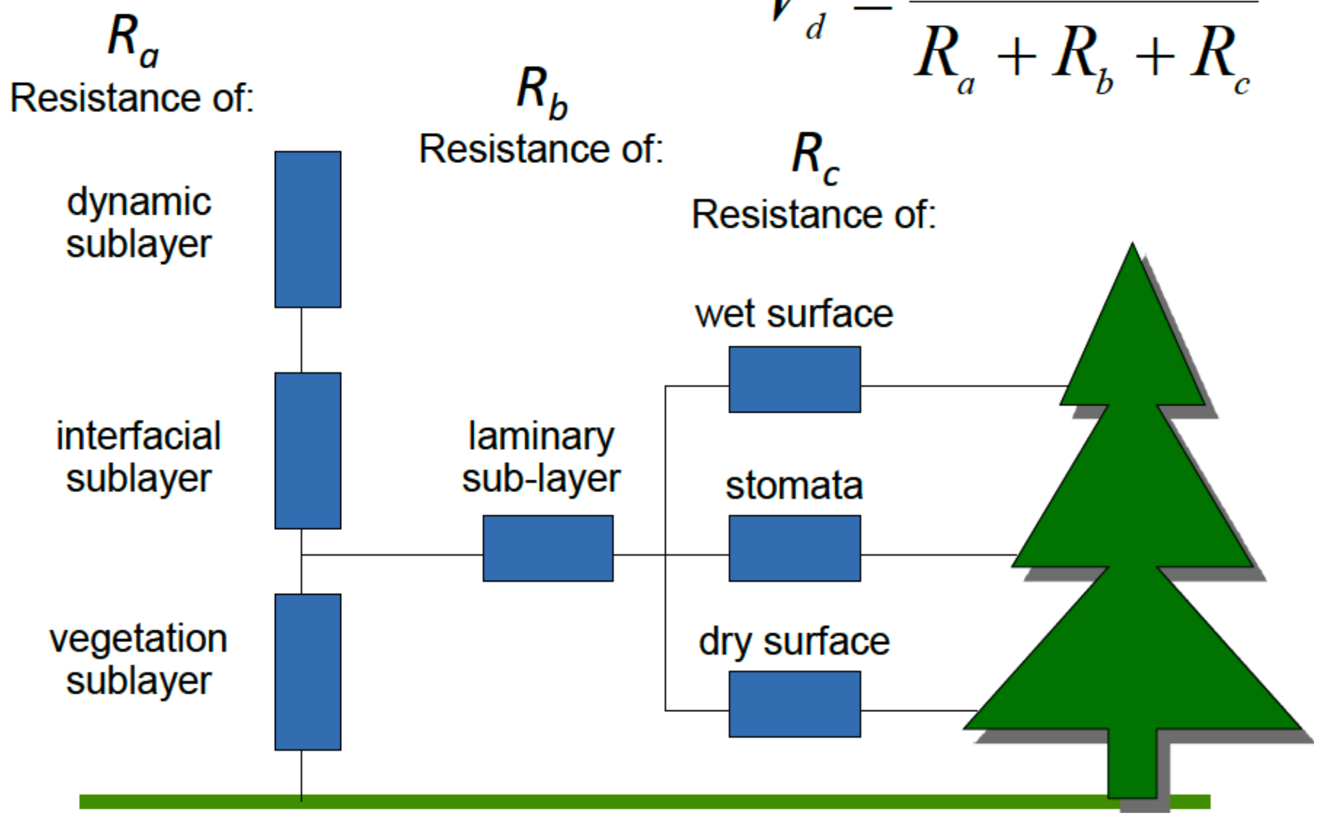
Emissions: surface emissions fields, fixed boundary conditions, calculated using vegetation type (biogenic, VOC), external forcings (aircraft emissions)

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

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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

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

- 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

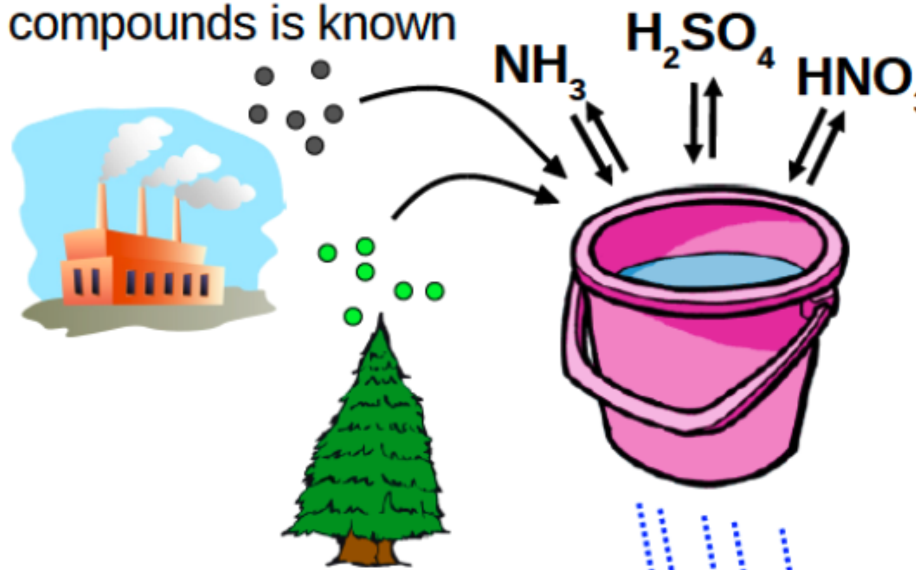
Controlled by: Emissions, nucleation processes, deposition, chemistry

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



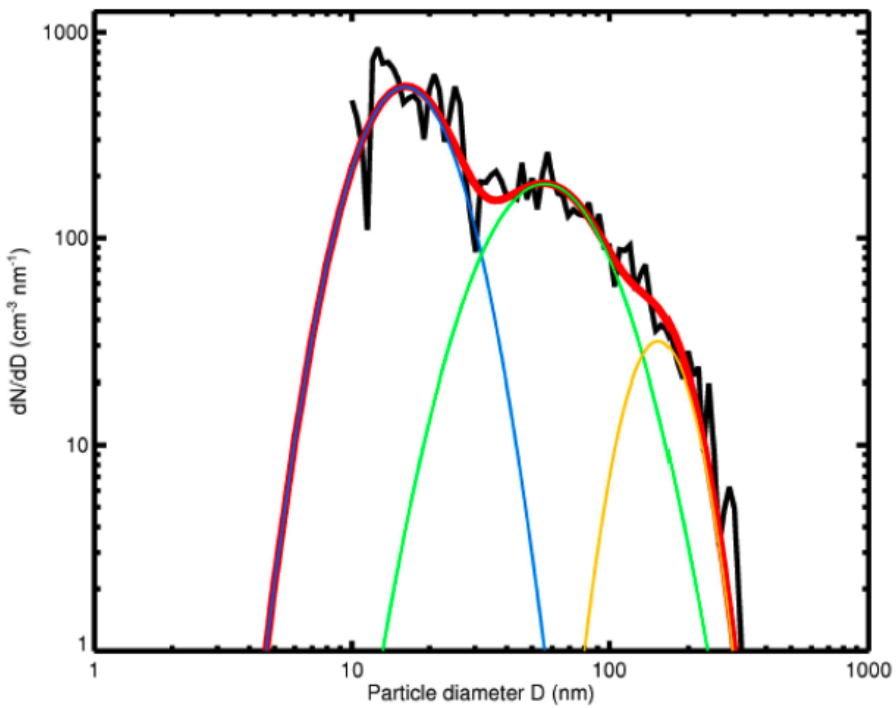
- Aerosol size distribution needs to be assumed for ...
  - radiative transfer
  - response of cloud properties to aerosol number
- Can't do aerosol nucleation
- **Numerically efficient**
- **Useful when focus is on complex gas phase / aerosol chemistry**

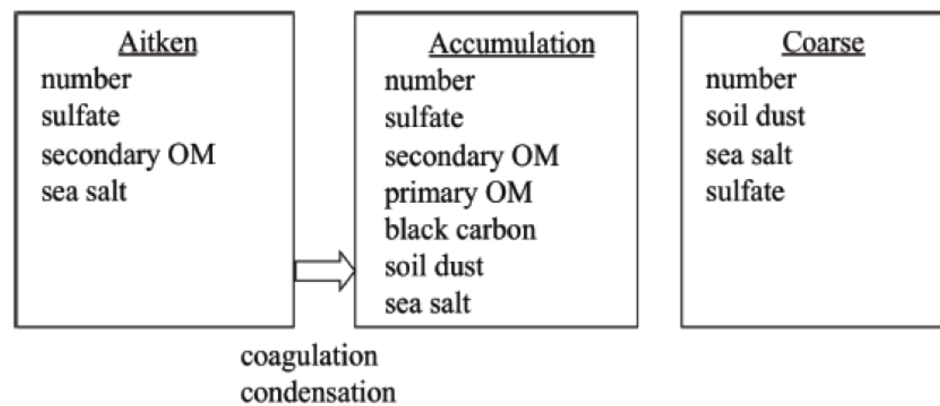
From J. Kazil, CIRES

Modal Aerosol Model (MAM3)

CESM CAM5
Aerosol size distribution using 3 modes
Other versions exist with 4 and 7 modes.

Twin Otter data (black)

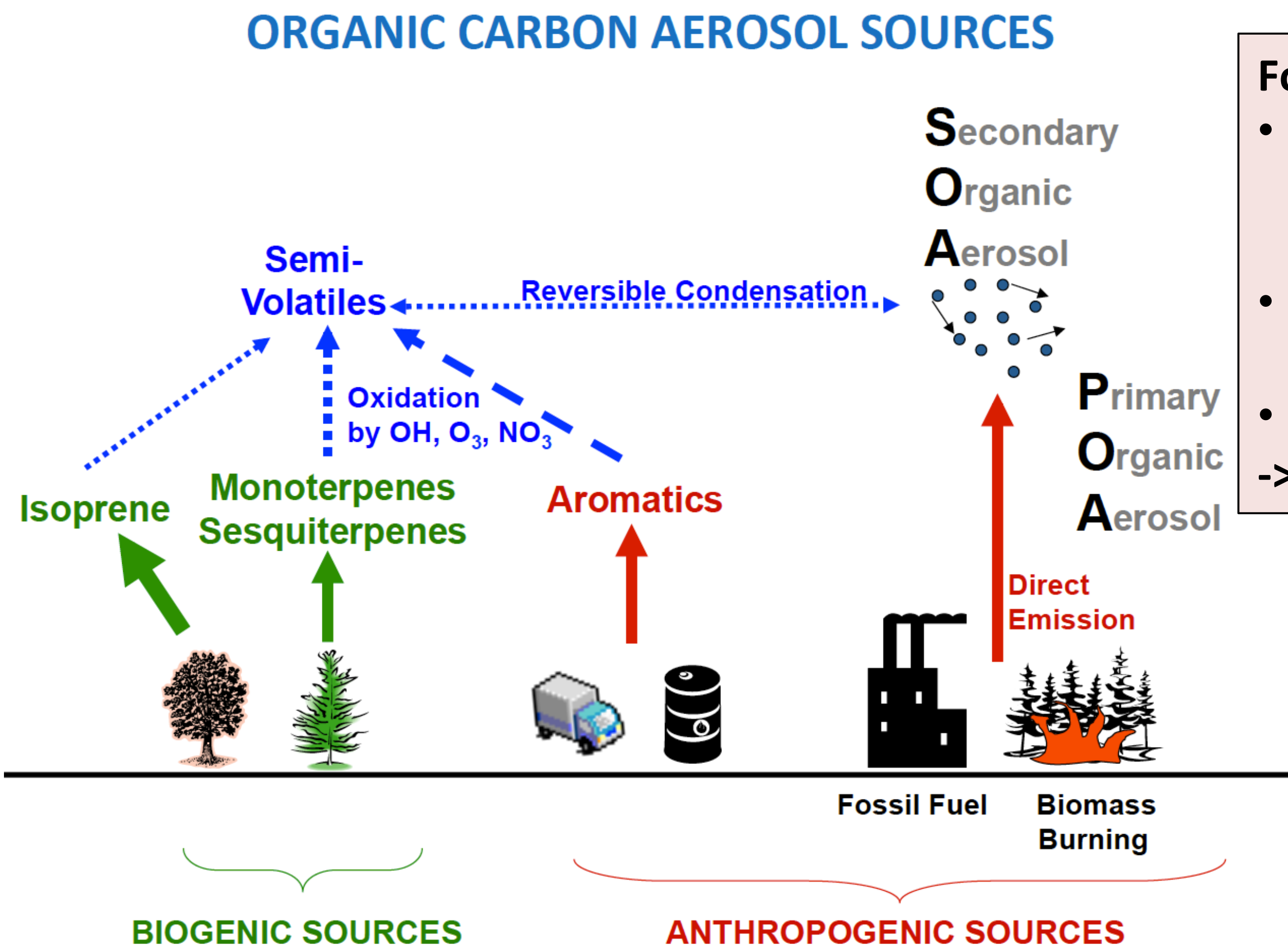




Liu et al., 2011

From J. Kazil, CIRES

Organic Aerosols (simulated in CAM4-Chem)



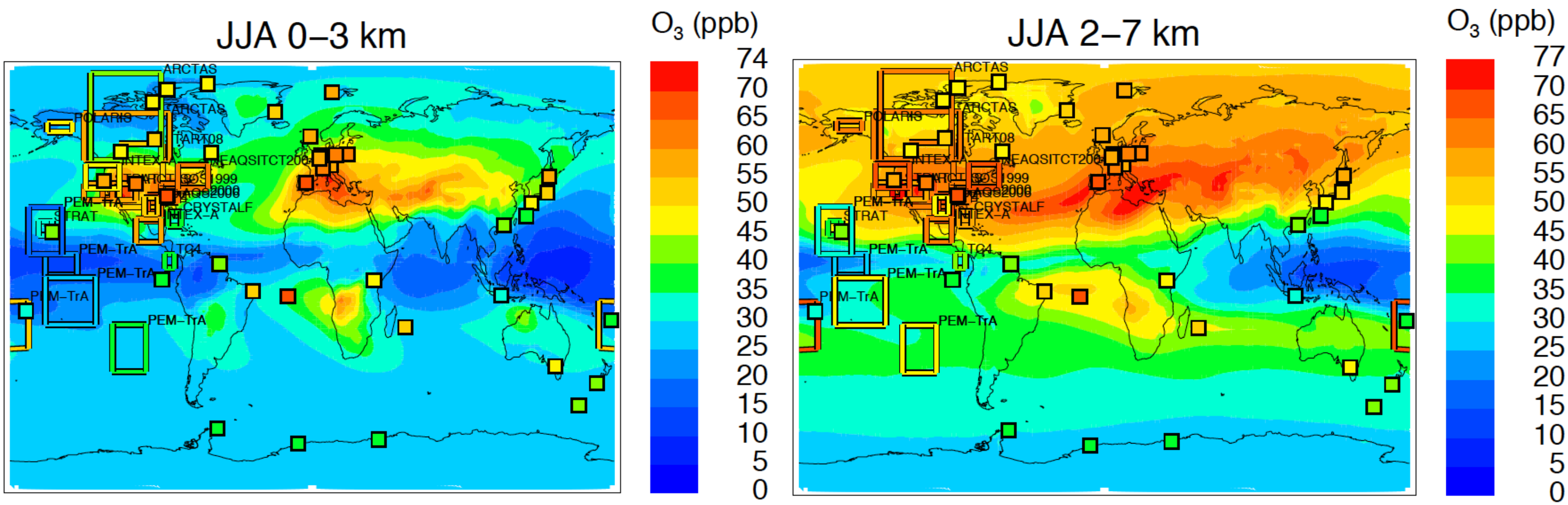
Formation of SOA

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

-> SOA

From C. Heald, MIT Cambridge

Performance of the Models: Ozone in Comparison to Ozonesondes and Aircraft



Simulated Ozone (background),
Ozonesonde observations (squares), Aircraft observations (color of framed regions)

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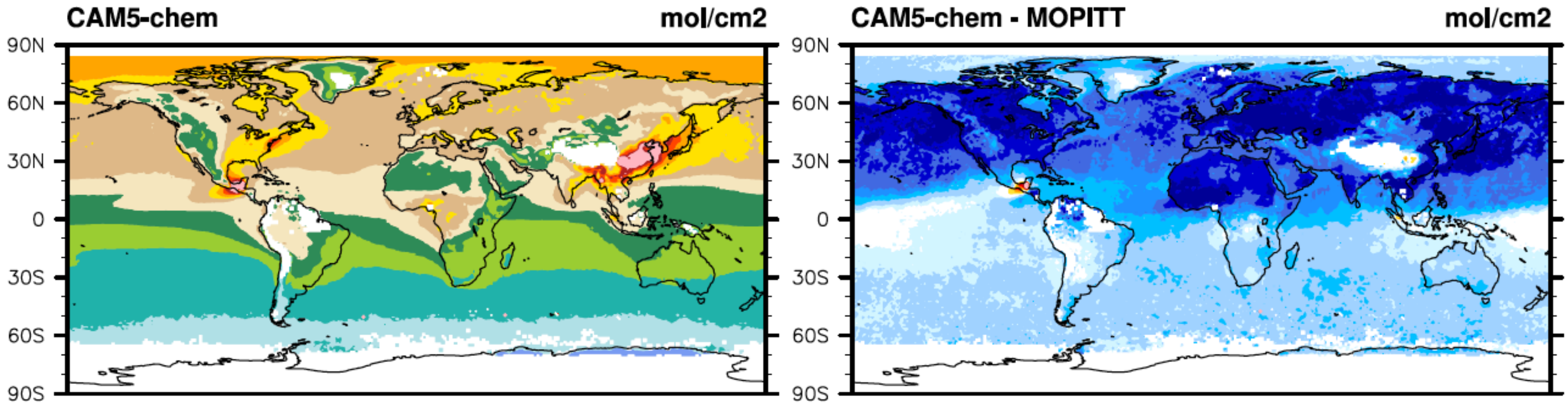
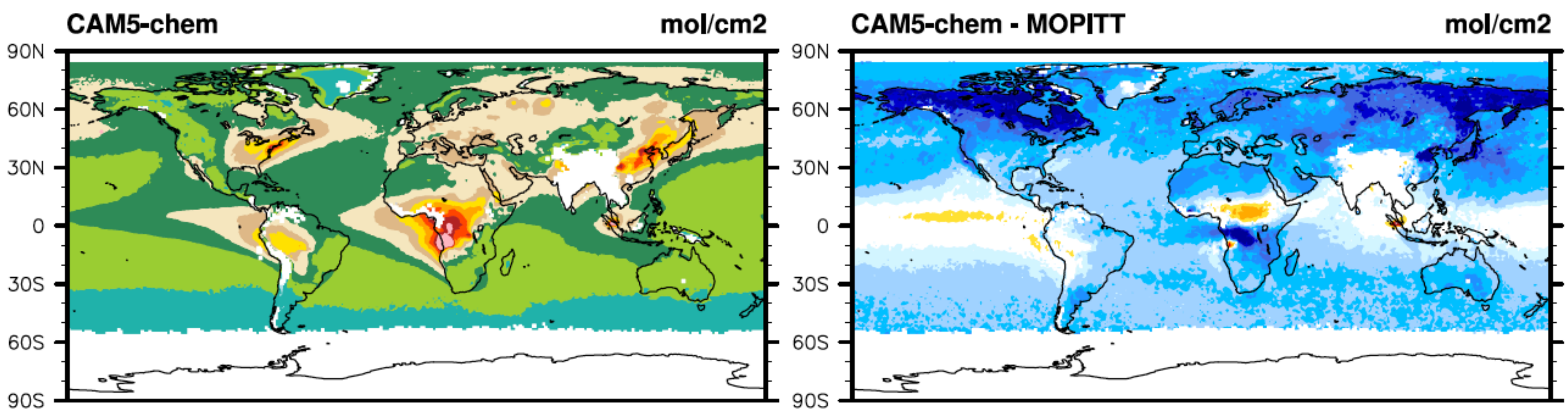
Performance of the Models: Carbon Monoxide in Comparison to MOPITT

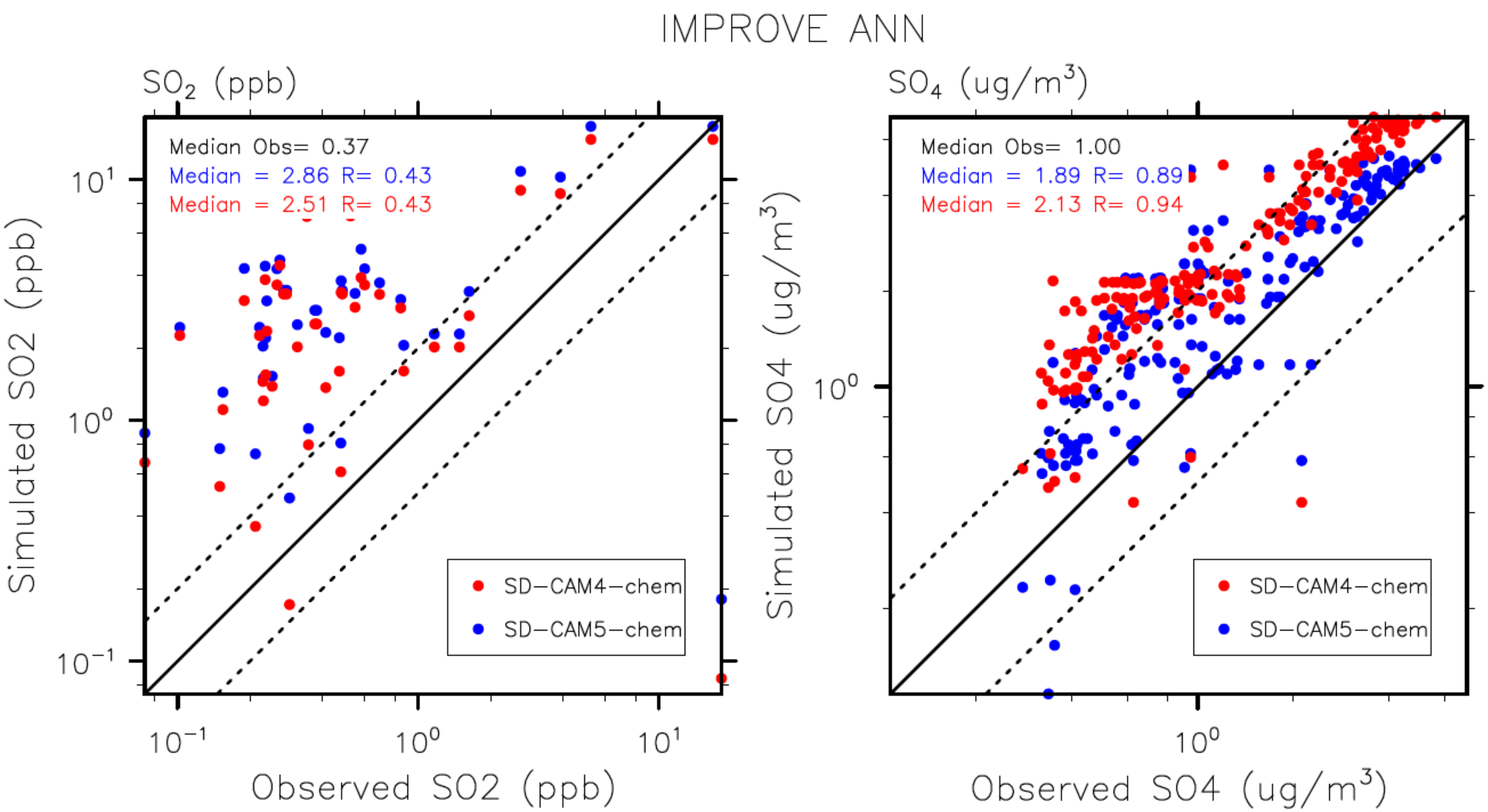
April



July



Performance of the Models: SO_2 and SO_4 in Comparison to IMPROVE



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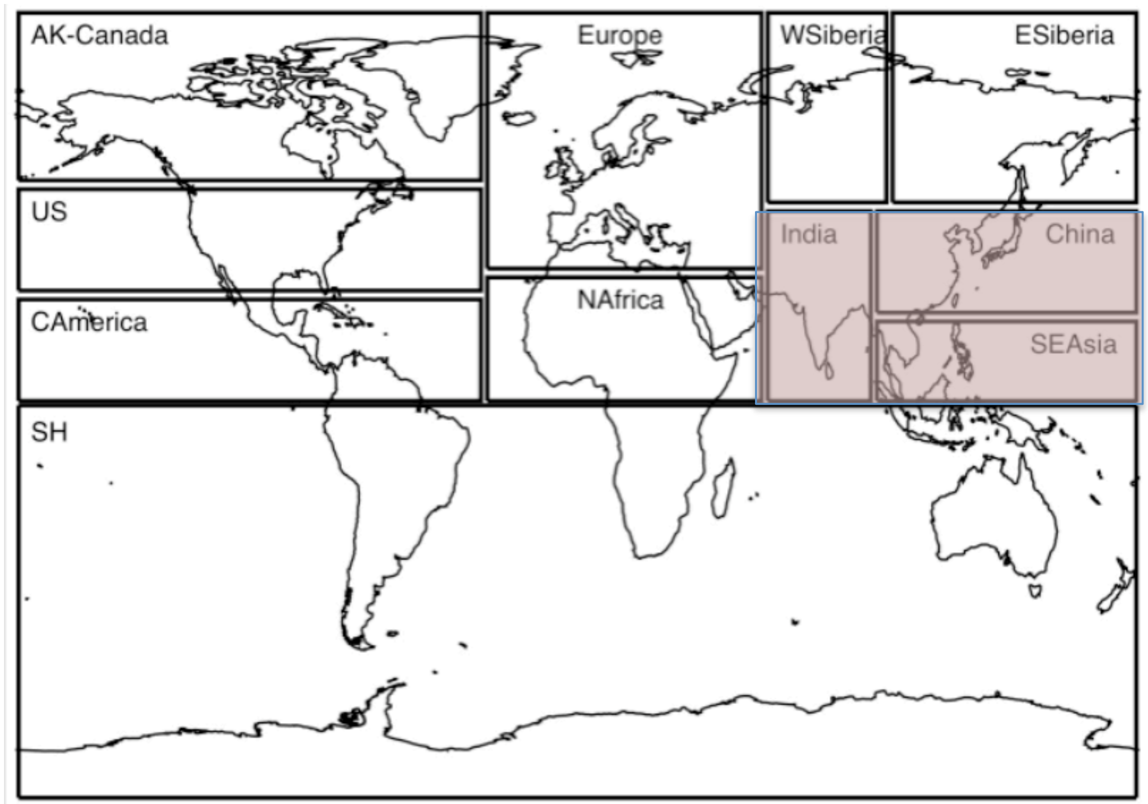
O_3, CO, BC tags with Offline Meteorology

Emmons et al., 2012, GMD



South Asia

The Model for Ozone and Related chemical Tracers (MOZART4)

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

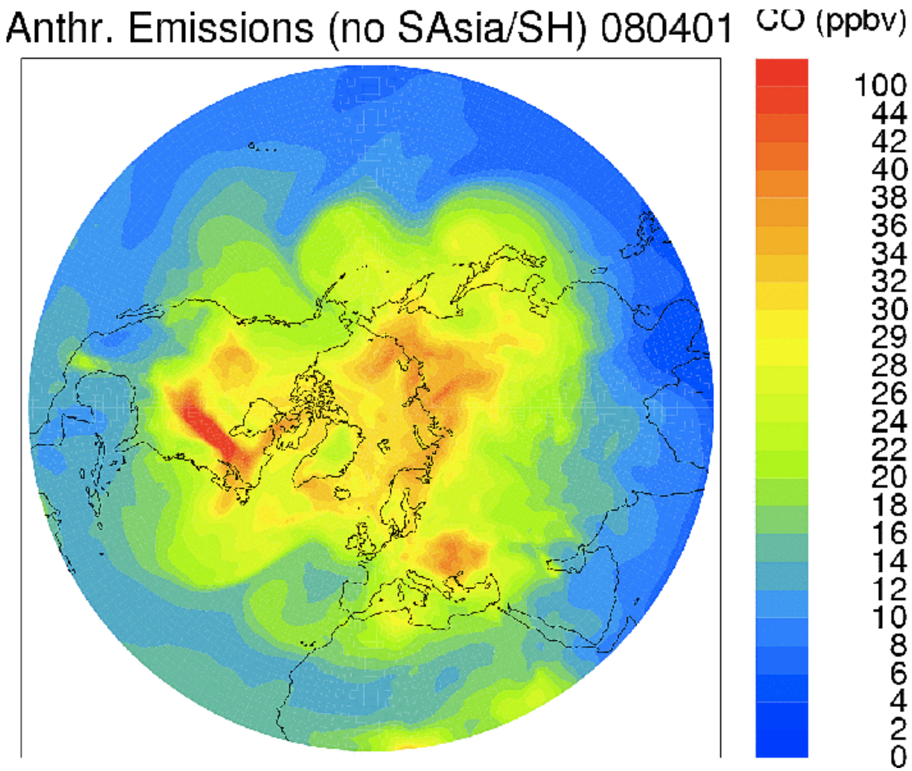
Vertical Injection of Fire Emission between 0-6 km

Importance of Anthropogenic CO Emissions

April

without South Asia and SH

South Asia and SH only



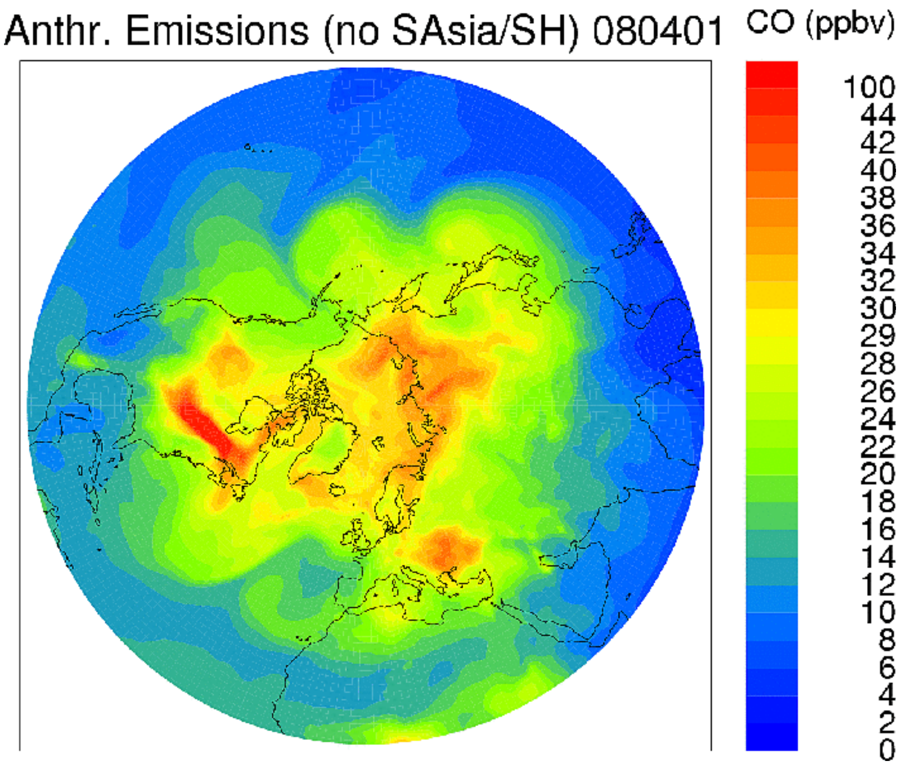
CO averaged column between surf. and 200 hPa

Importance of Anthropogenic CO Emissions

April

without South Asia and SH

South Asia and SH only

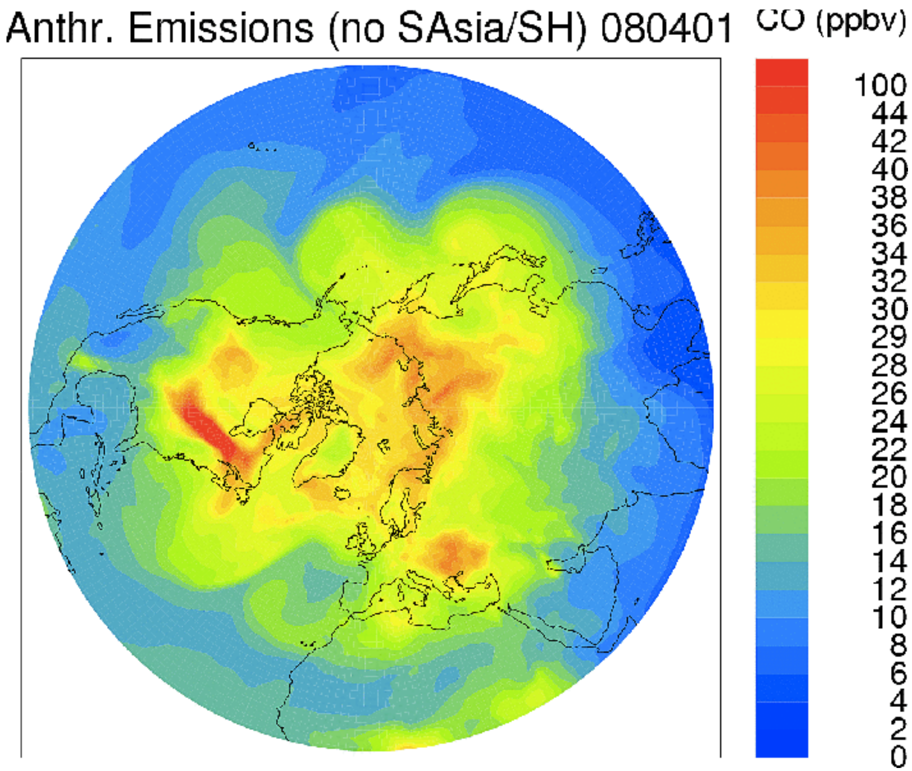


CO averaged column between surf. and 200 hPa

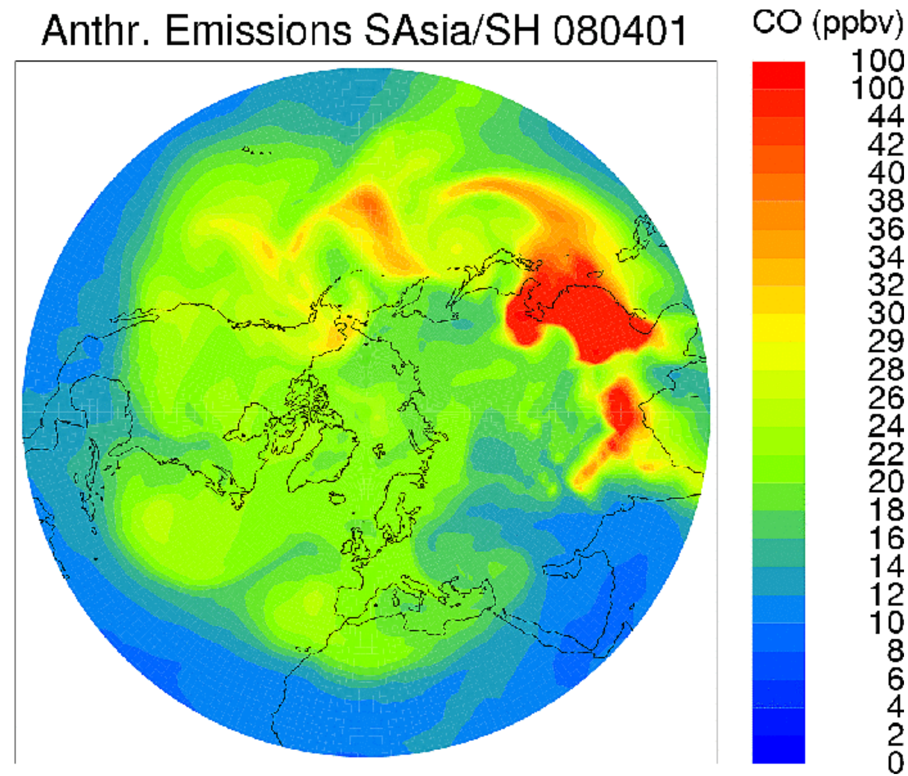
Importance of Anthropogenic CO Emissions

April

without South Asia and SH



South Asia and SH only



CO averaged column between surf. and 200 hPa

WACCM and CAM-Chem Customer Support

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

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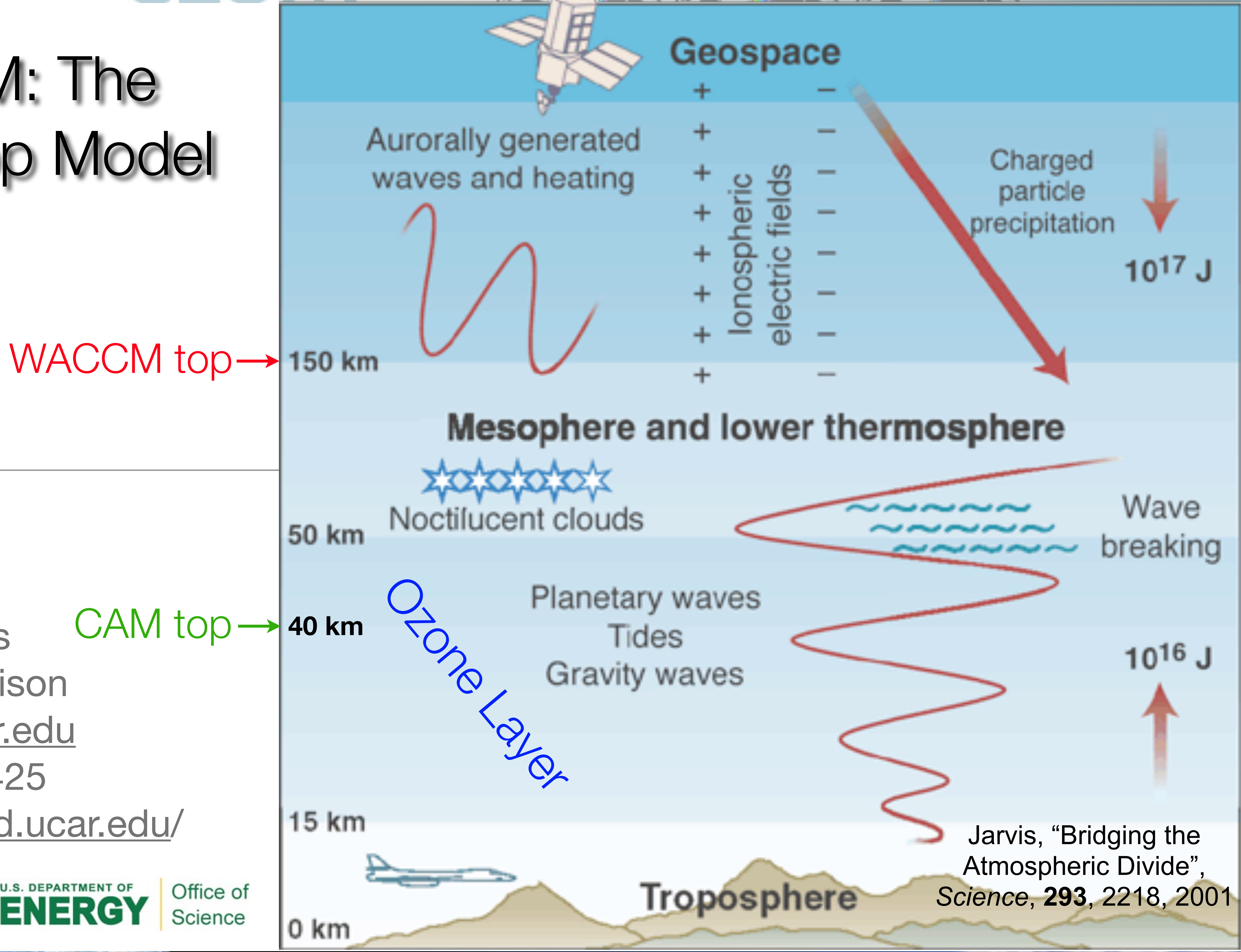
WACCM: The High-Top Model



Jarvis, "Bridging the Atmospheric Divide", *Science*, **293**, 2218, 2001

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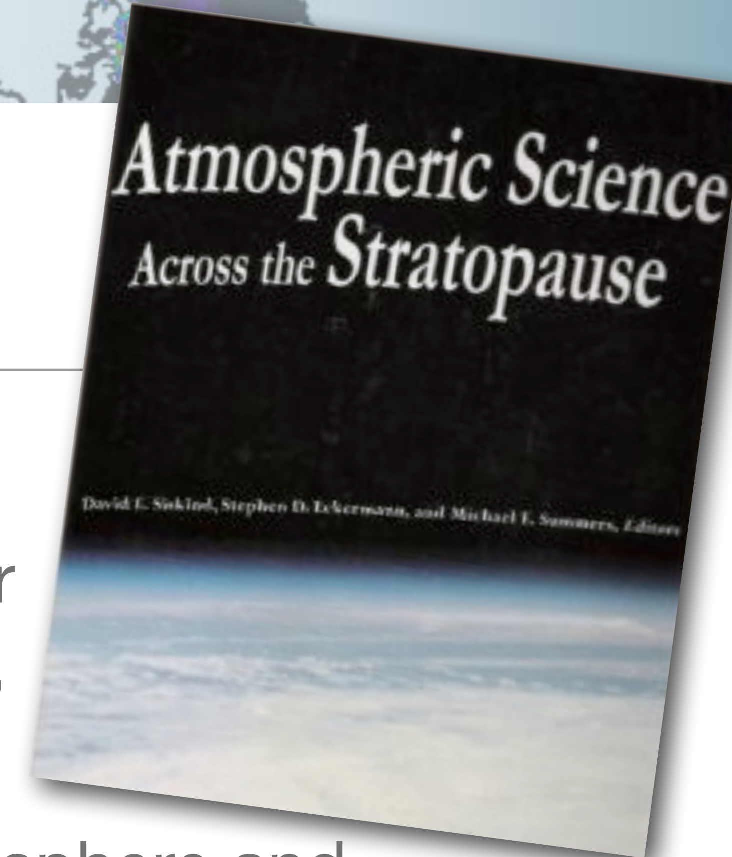
http://bb.cgd.ucar.edu/

WACCM Additions to CAM

- Extends from surface to 5.1×10^{-6} hPa (~150 km), with 66 vertical levels
- Detailed neutral chemistry model for the middle atmosphere,
  - catalytic cycles affecting ozone
  - heterogeneous chemistry on PSCs and sulfate aerosol
  - heating due to chemical reactions
- 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
- Imposed QBO, 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

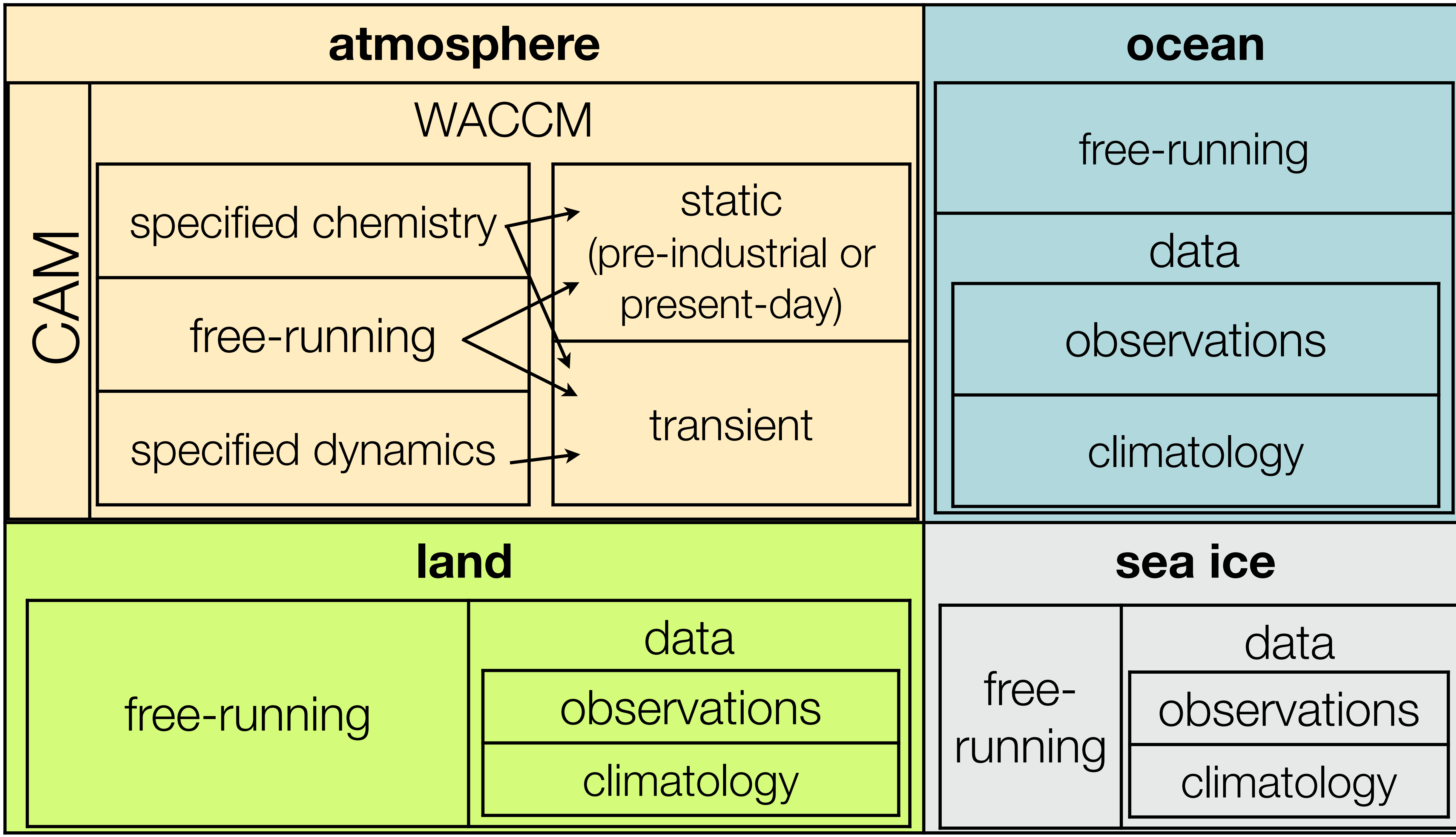
WACCM Motivation

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

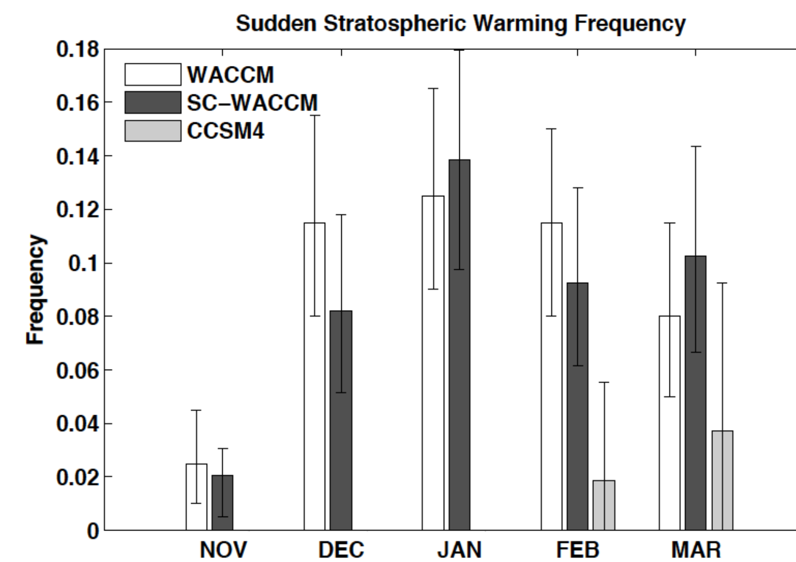
CESM-WACCM component configurations



WACCM Specified Chemistry (WACCM-SC)

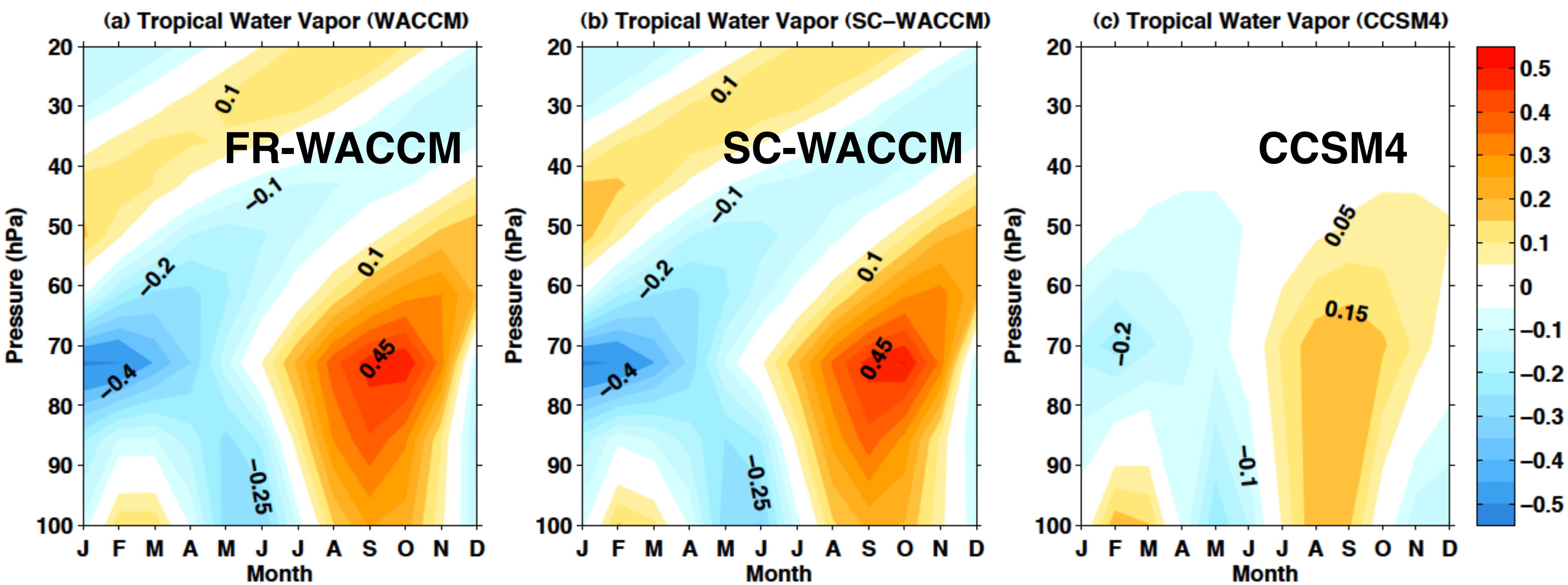
- Specifies Ozone (among other species)
- 2x as fast as WACCM: for stratospheric dynamics studies, with nearly identical results

Model	# cores	simulated years/day	core-hrs/simulated year
WACCM	352	7.5	1130
SC-WACCM	352	14.8	573
CCSM4 1°	352	19.6	432
CCSM4 2°	416	42.0	237



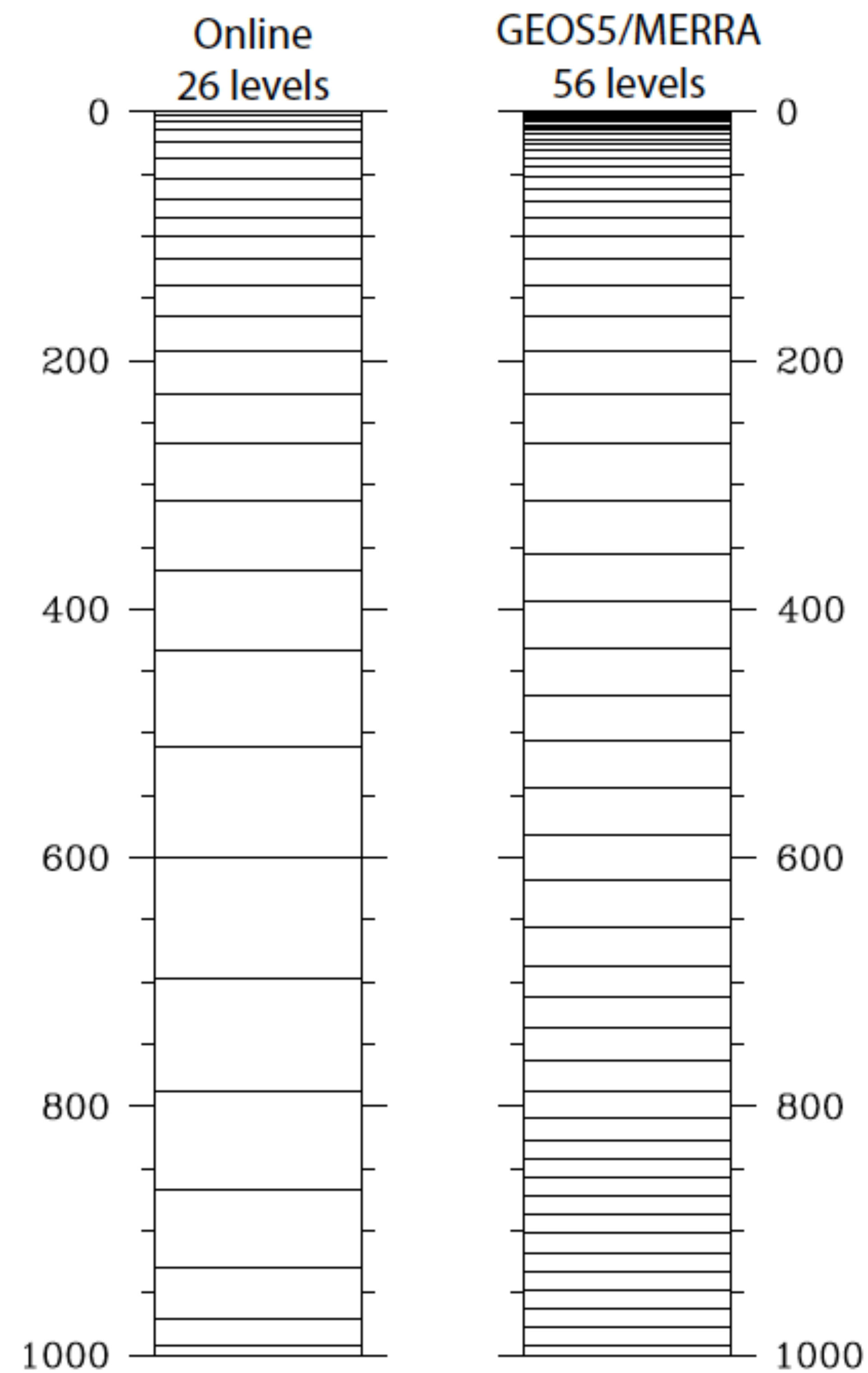
Above: WACCM4-SC also gets sudden stratospheric warming frequency right

Below: Tropical H_2O Tape Recorder looks like WACCM (good), not CCSM4 (bad)

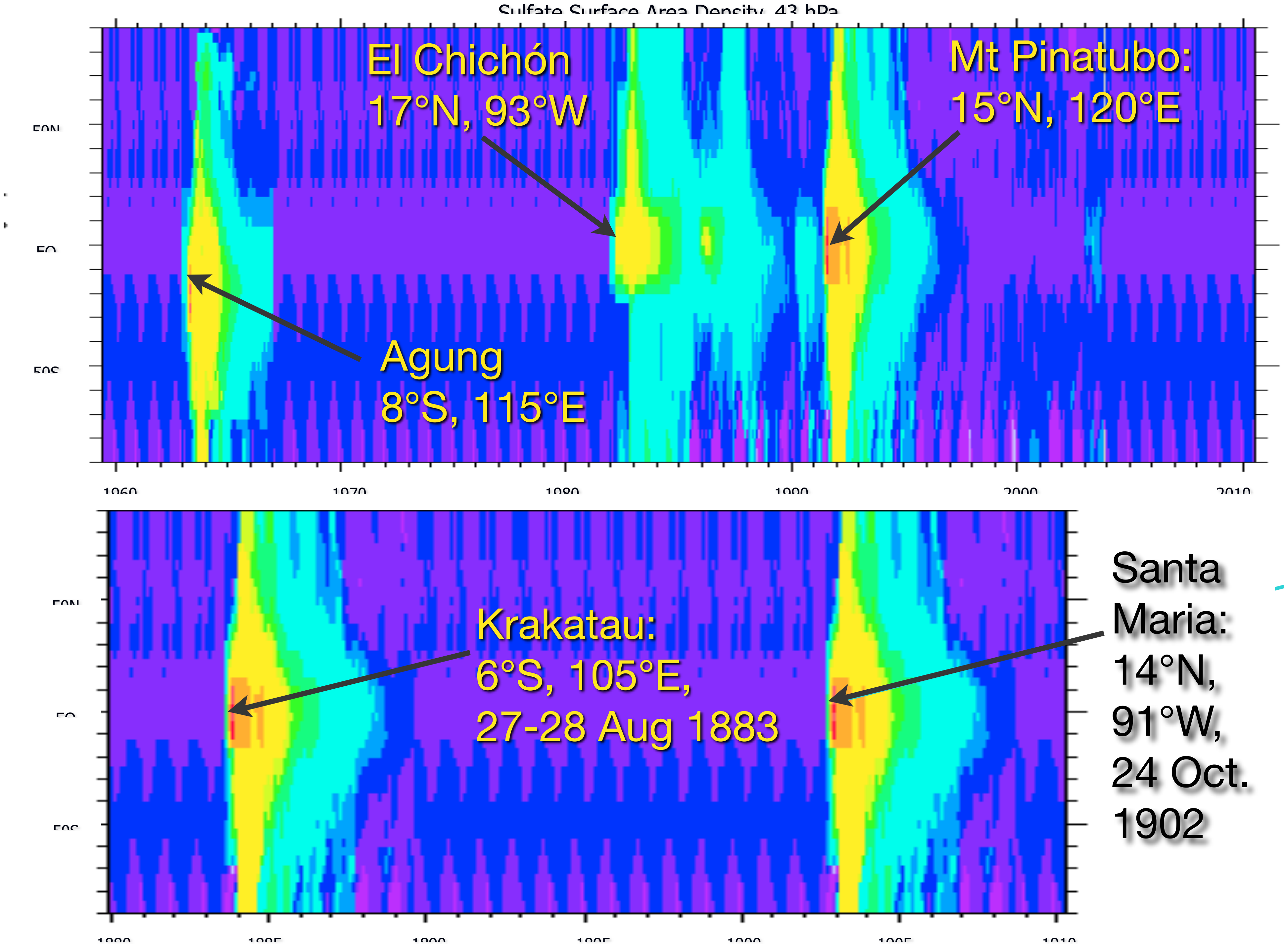


Specified Dynamics: SD-WACCM and SD-CAM-Chem

- Reproduce winds and temperatures from specific periods in analyses from GEOS5 (2004-present) or MERRA (1979-present).
- **FSDW** compset starts on 1 Jan 2005, uses GEOS5, out of the box.
- Increased vertical resolution
  - CAM-Chem: 26 levels → SD-CAM-Chem: 56 levels
  - WACCM: 66 levels → SD-WACCM: 88 levels
- Nudge T, U, V, PS towards analyses at every dynamics timestep. Nudging strength (i.e. 1%, 10% each timestep) and top altitude (50 km default for WACCM) can be adjusted.
- Chemistry interacts with radiation, atmosphere, land, ocean
- Data ocean and sea ice components



Stratospheric Sulfate Surface Area Density Climatology

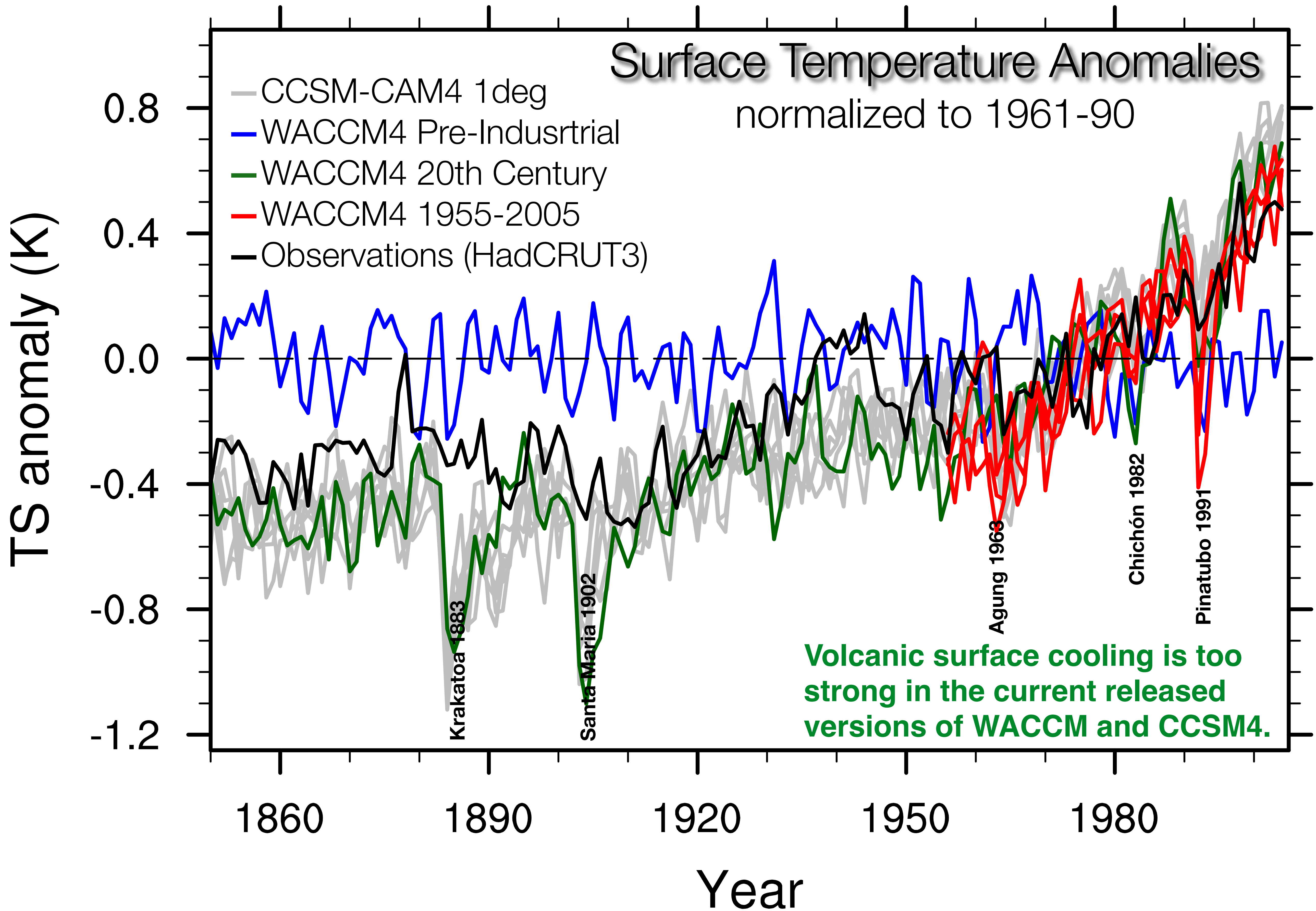


- Observations used: SAGE I, SAGE II, SAM II, and SME instruments.
- Non-volcanic periods filled with monthly mean of 1998-2002 values.
- Used Pinatubo aerosol for Krakatau and Santa Maria.

NCAR

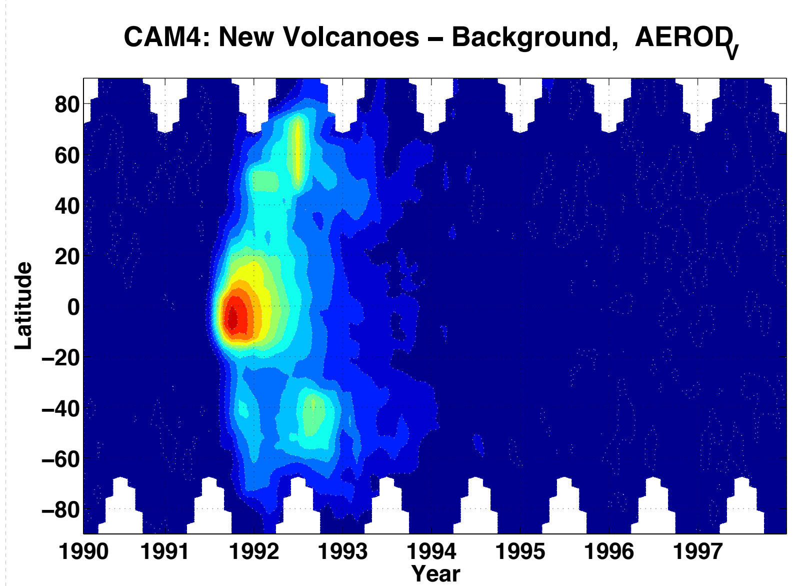
WACCM

Whole Atmosphere Community Climate Model

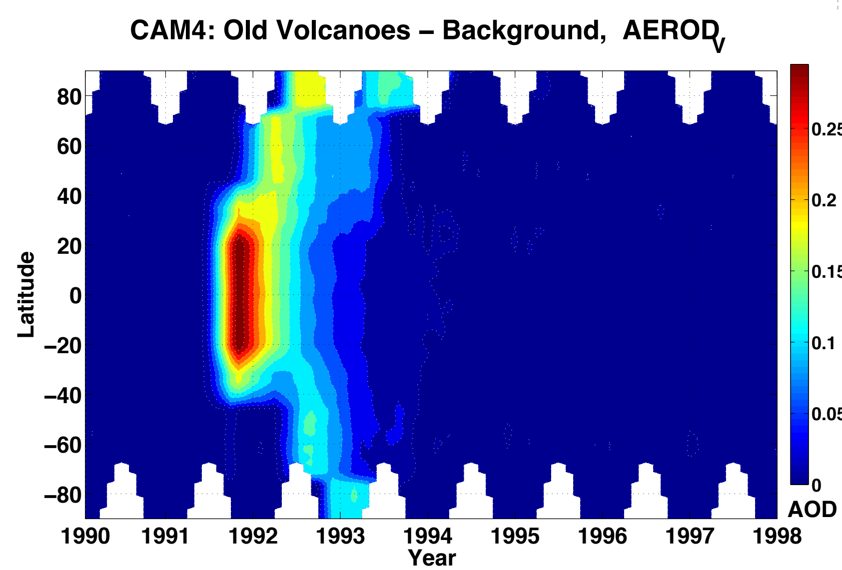


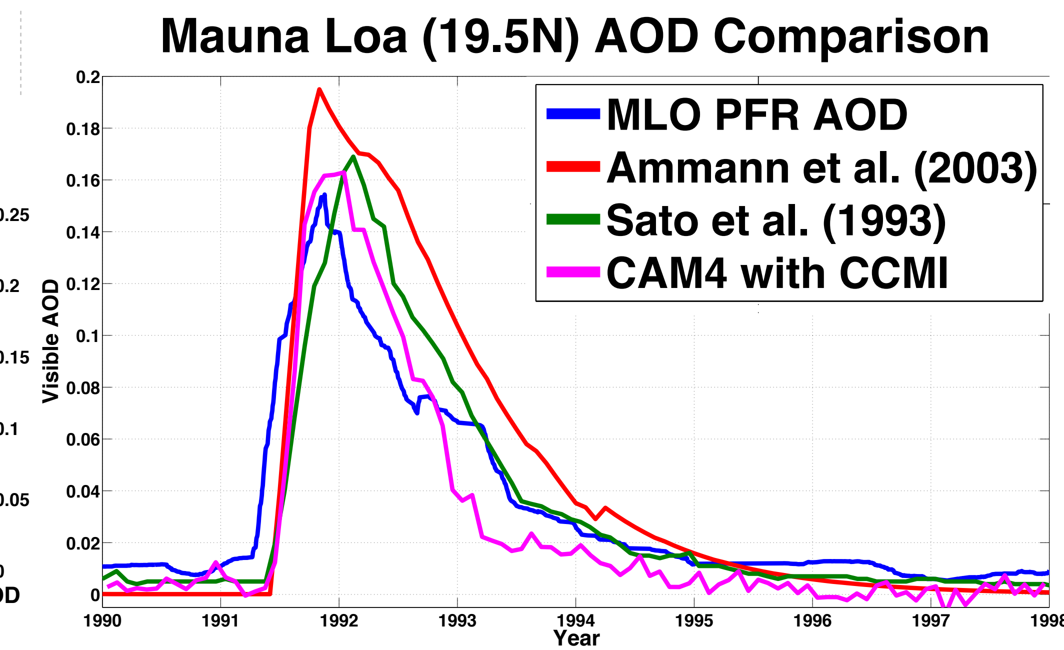
New 1960-2013 stratospheric sulfate dataset for CCMI (not yet released): significant improvements

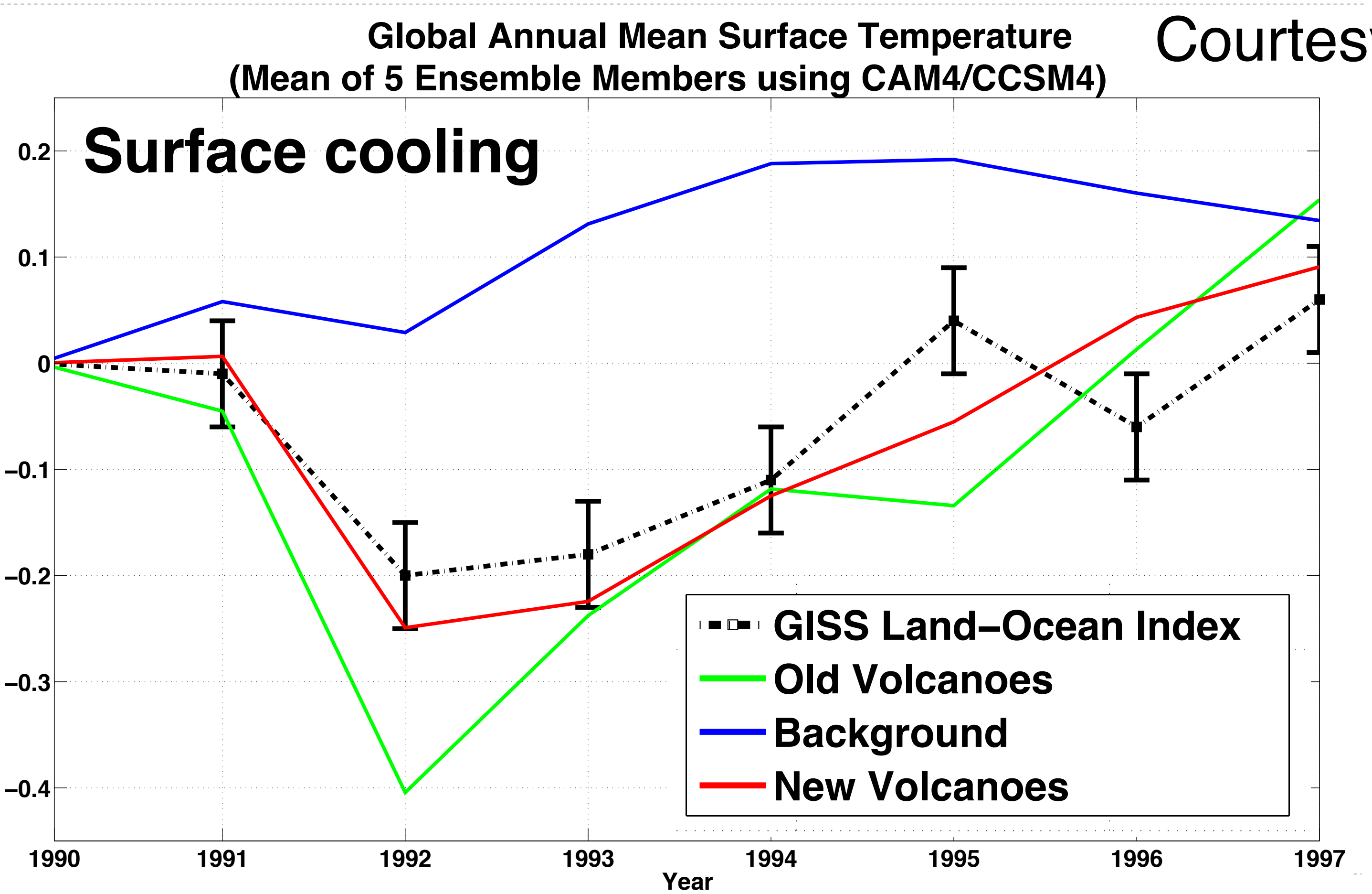
New/CCMI



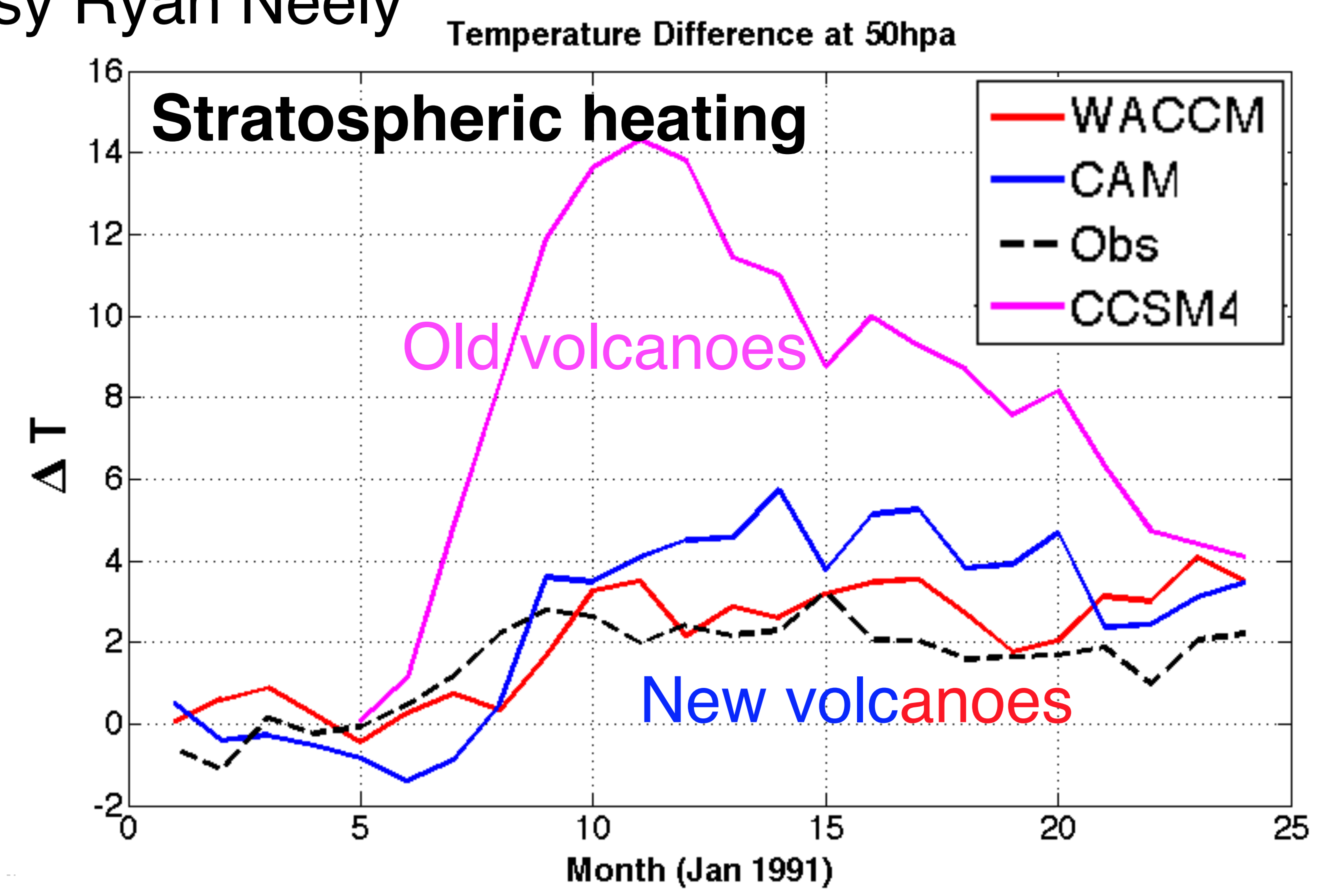
Old/CCSM4



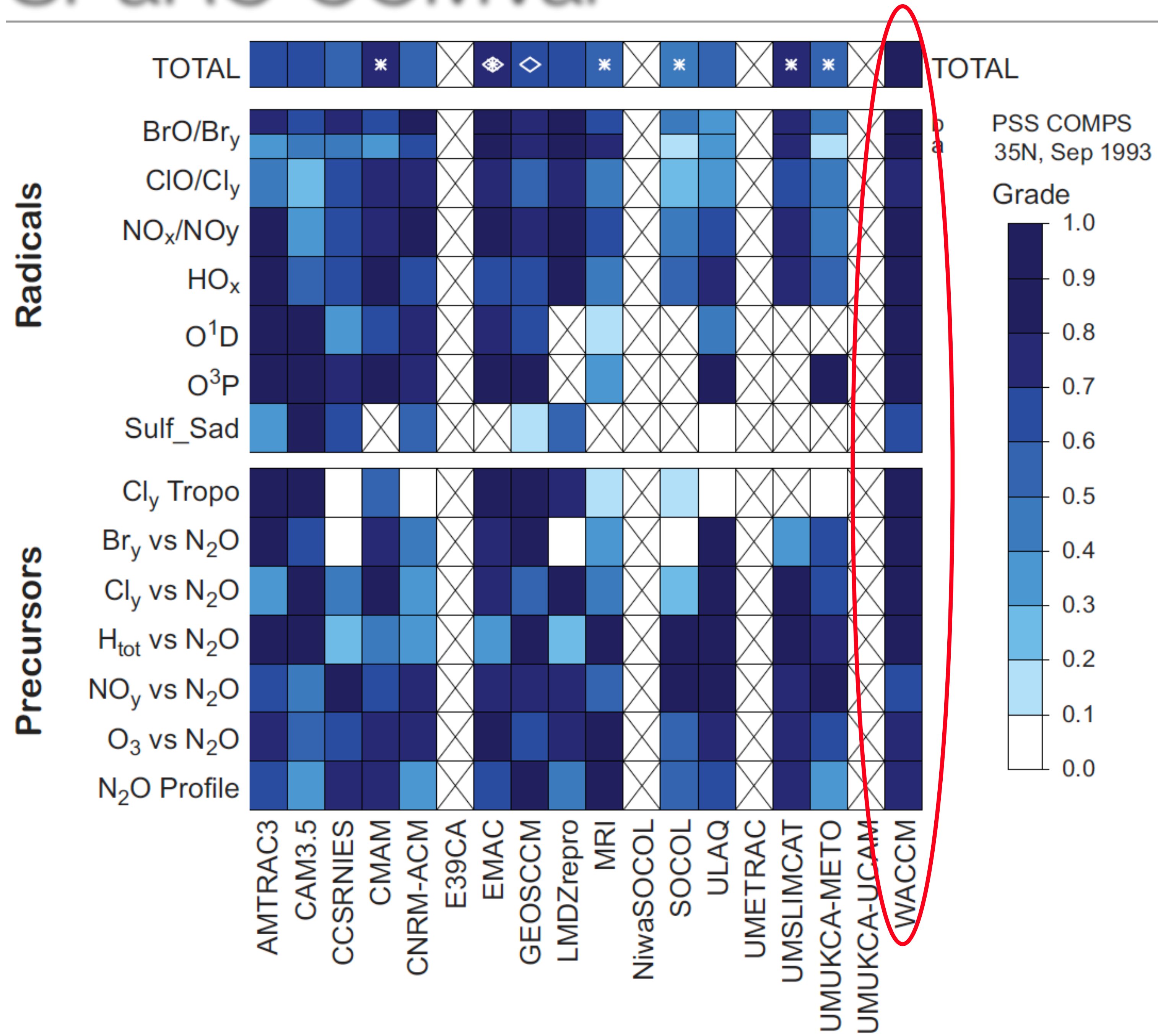




Courtesy Ryan Neely



Grading of Chemistry in CCMs: Chapter 6 of the SPaRC CCMVal



- CCMs were evaluated on their ability to represent long-lived constituents (**precursors**) and short-lived substances (**radicals**).
- WACCM graded out high in all categories (i.e., grade of 1 is the highest possible).
- This is a reflection of the:
  1) completeness of the chemical processes included
  2) accuracy of photolysis rates (J's)
  3) and accuracy of the numerical solution approach.

SPARC CCMVal, Report on the Evaluation of Chemistry-Climate Models, V.Eyring, T. G. Shepherd, D. W. Waugh (EDs.), SPARC Reprot No.4, WCRP-X, WMO/TD-No. X, http://www.atmosp.physics.utoronto.ca/SPARC, 2010.

WACCM Heterogeneous Chemistry Module

Temperature

Sulfate Aerosols (H_2O, H_2SO_4) - LBS

R_{lbs} = 0.1 μm

>200 K

Sulfate Aerosols (H_2O, HNO_3, H_2SO_4) - STS

R_{sts} = 0.5 μm

Thermo. Model (Tabazadeh)

?

Nitric Acid Hydrate (H_2O, HNO_3) – NAT

$R_{NAT} = f$ {Log Normal Size; # particles cc; width distribution; condensed phase HNO_3}

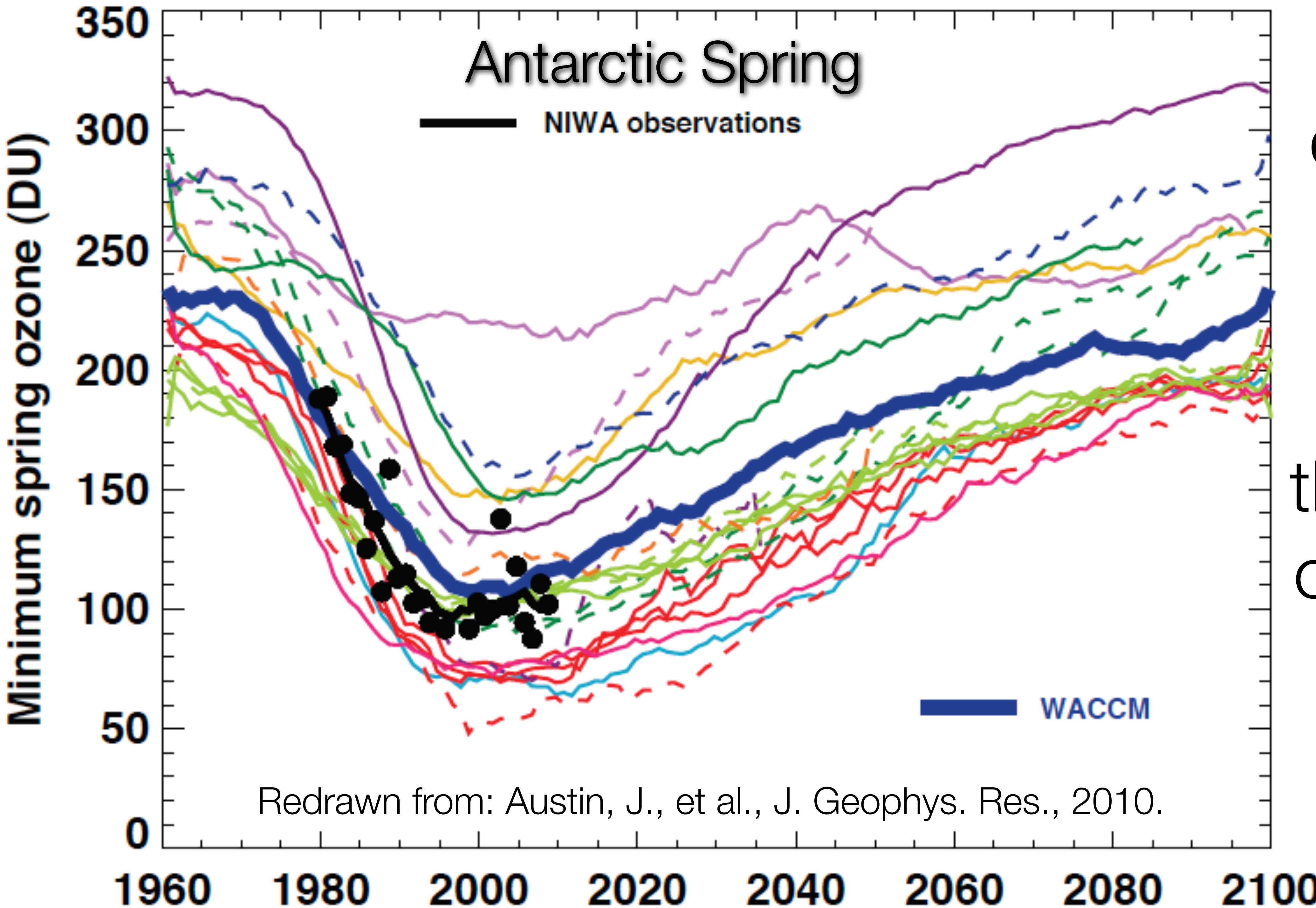
188 K (T_{sat})

ICE (H_2O, with NAT Coating)

R_{ice}= 10-30 μm

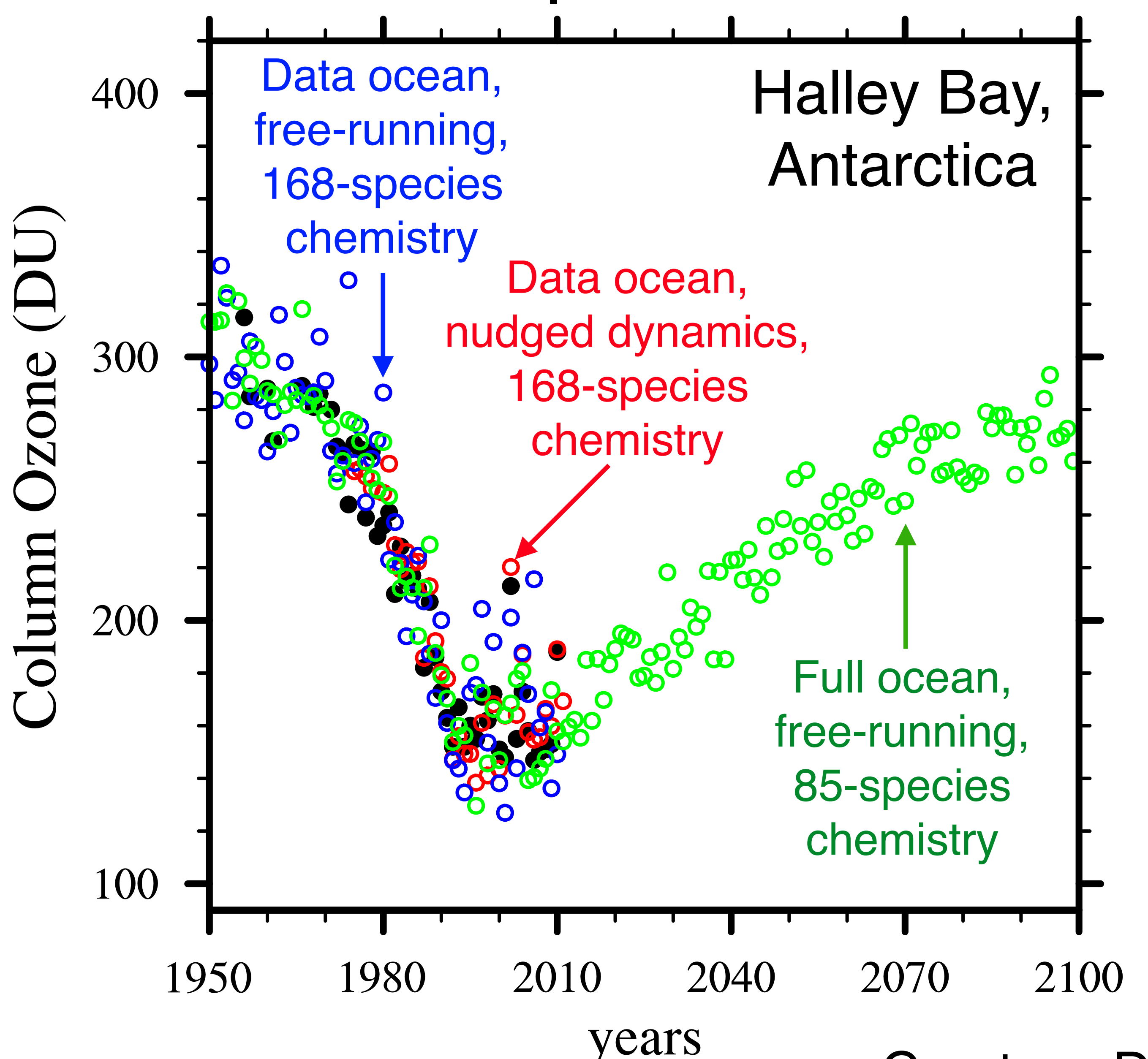
185 K (T_{nuc})

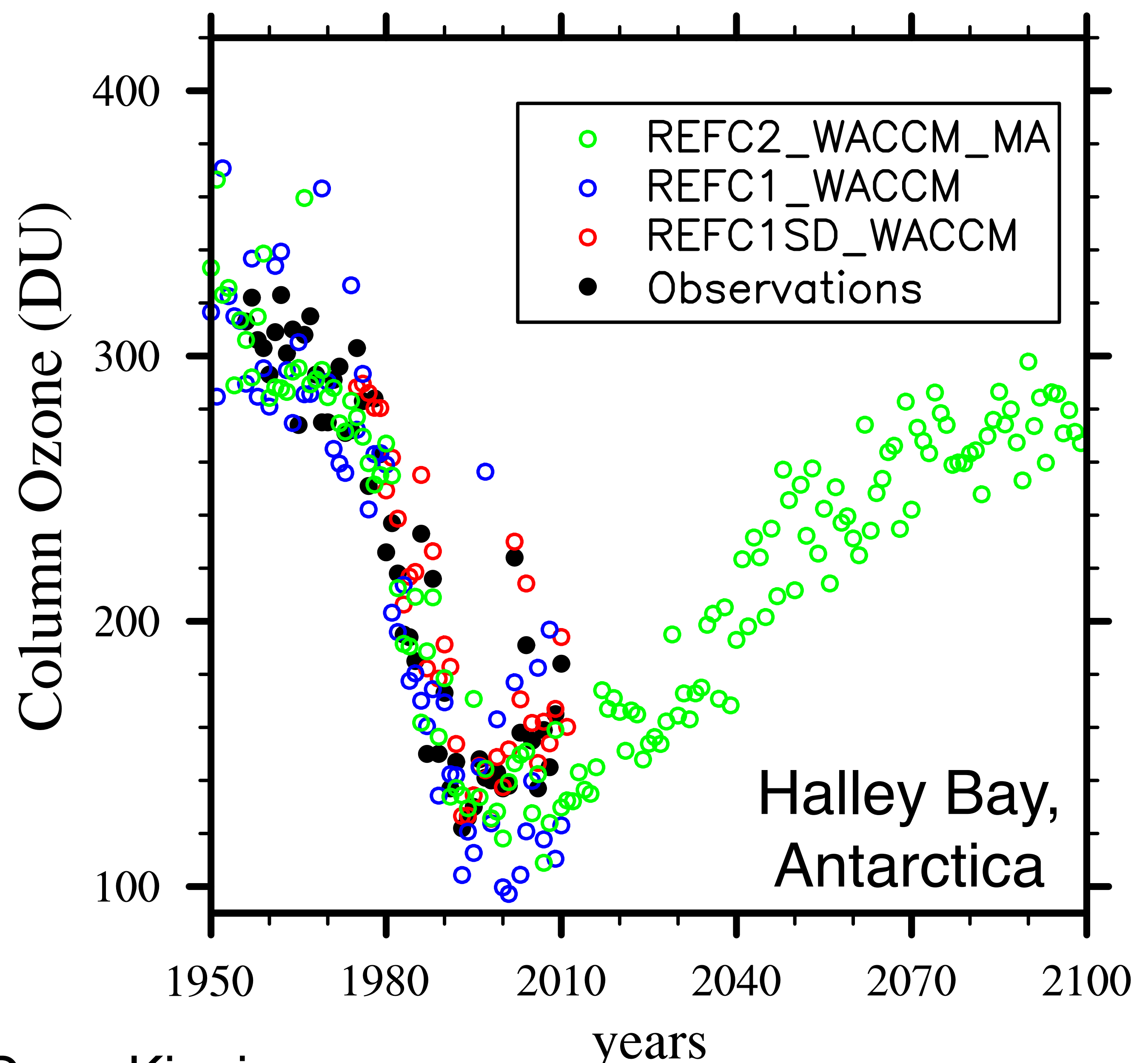
WACCM Ozone Trend: CCMVal and WMO (2010)



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

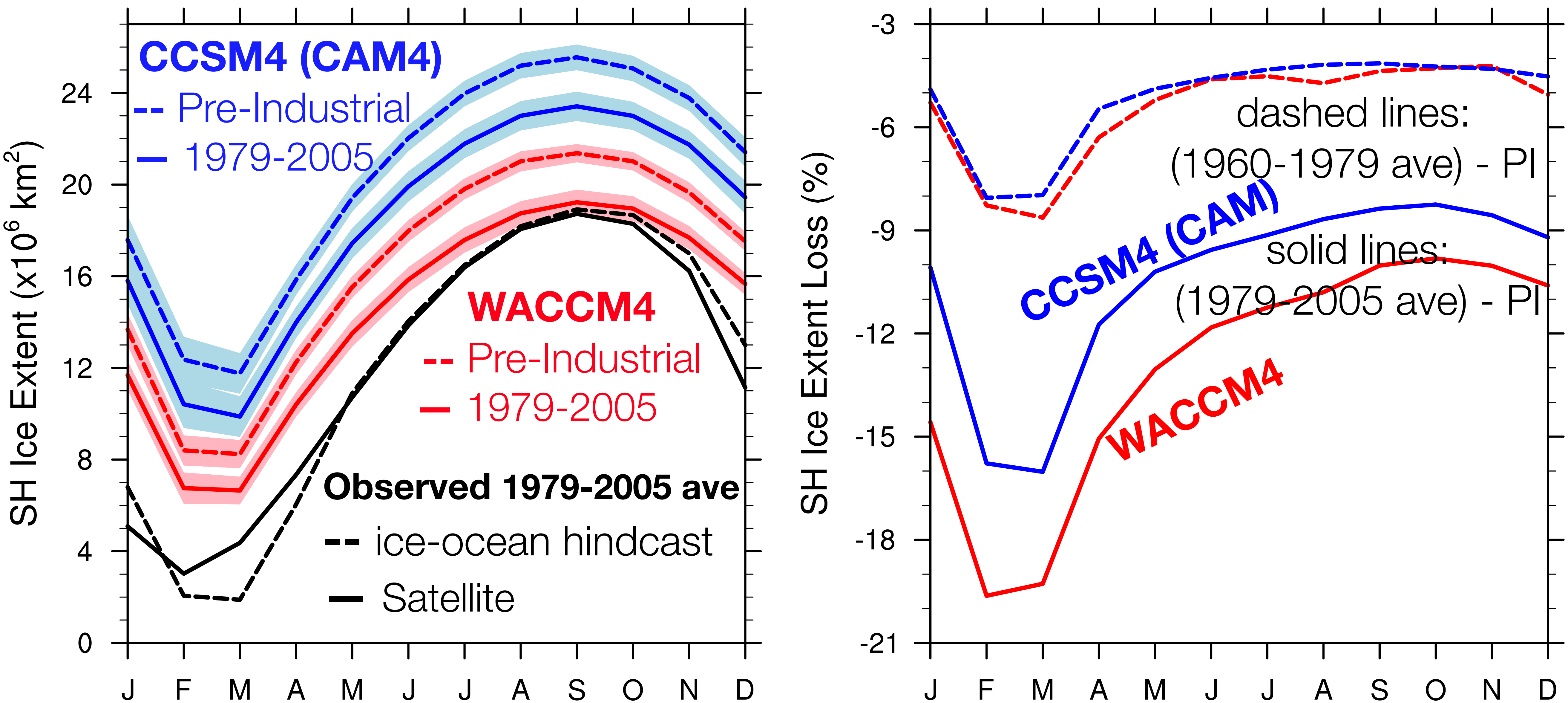
CCMI runs (2013/14): updated chemistry and dynamics (not yet released)





Courtesy Doug Kinnison

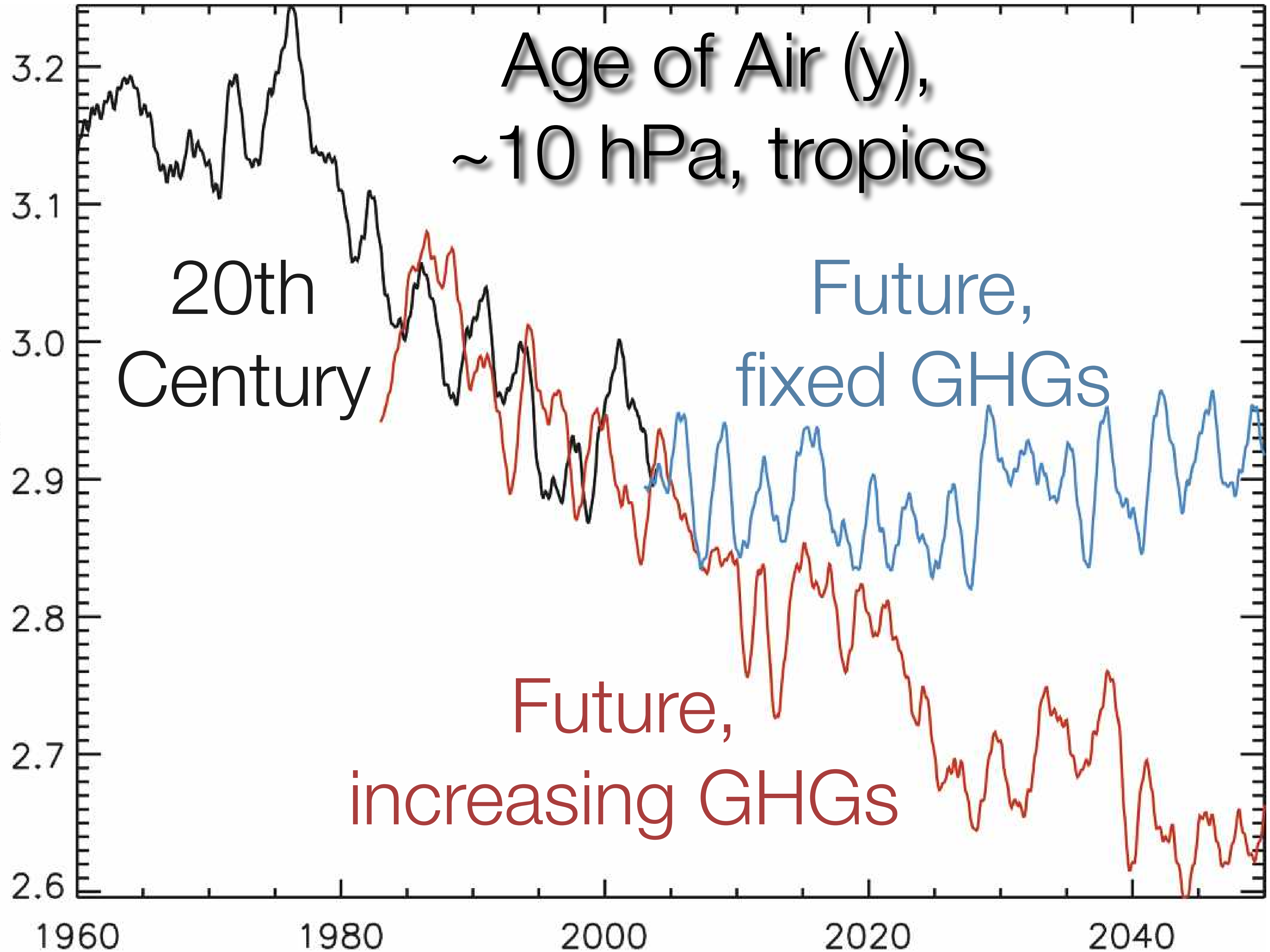
Antarctic sea-ice extent



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

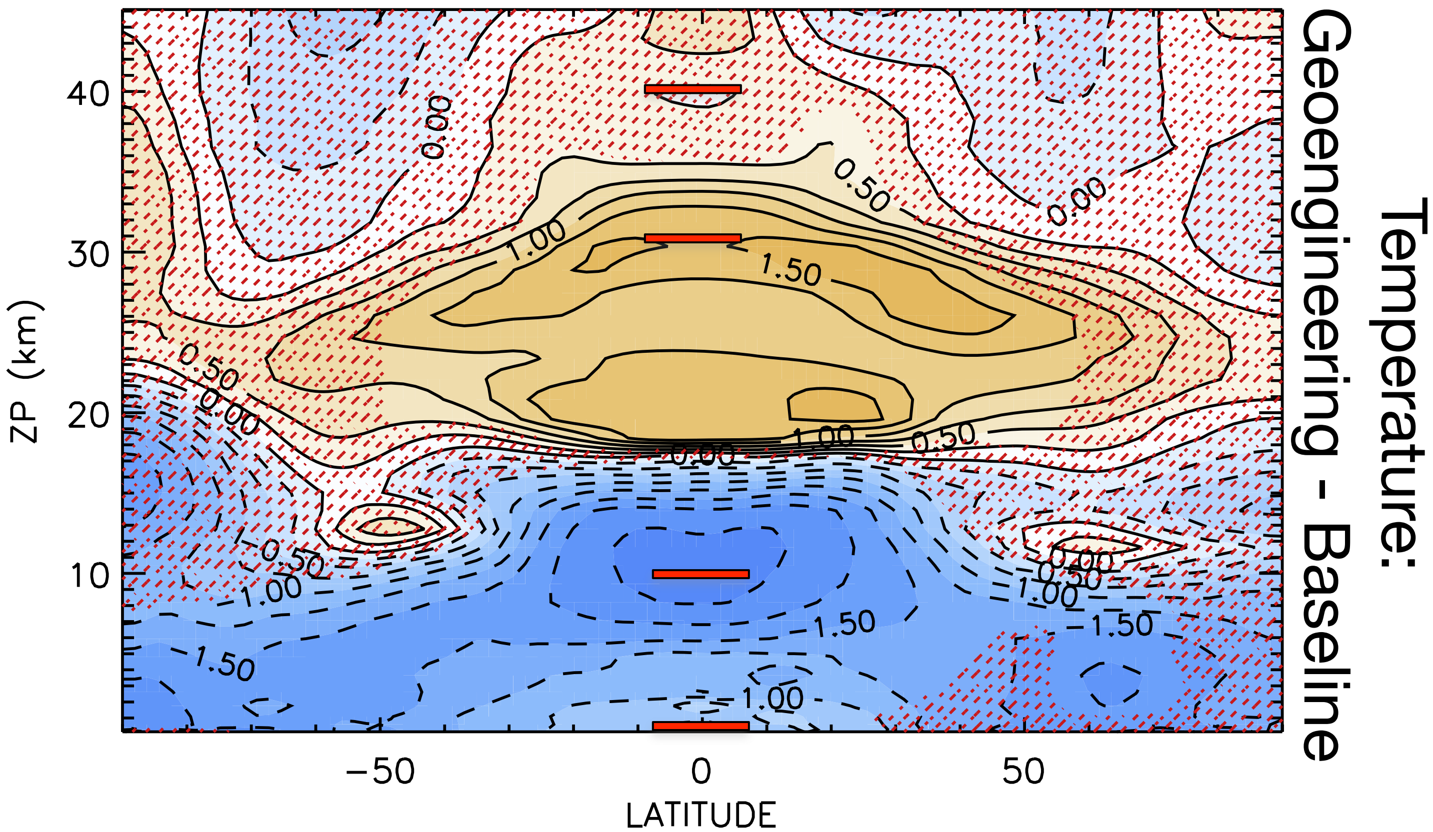
Acceleration of the Brewer–Dobson Circulation due to Increases in Greenhouse Gases
Garcia and Randel, *J. Atmos. Sci.*, vol. 65 (8), pp. 2731-2739, 2008.

- faster circulation in greenhouse world due to 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

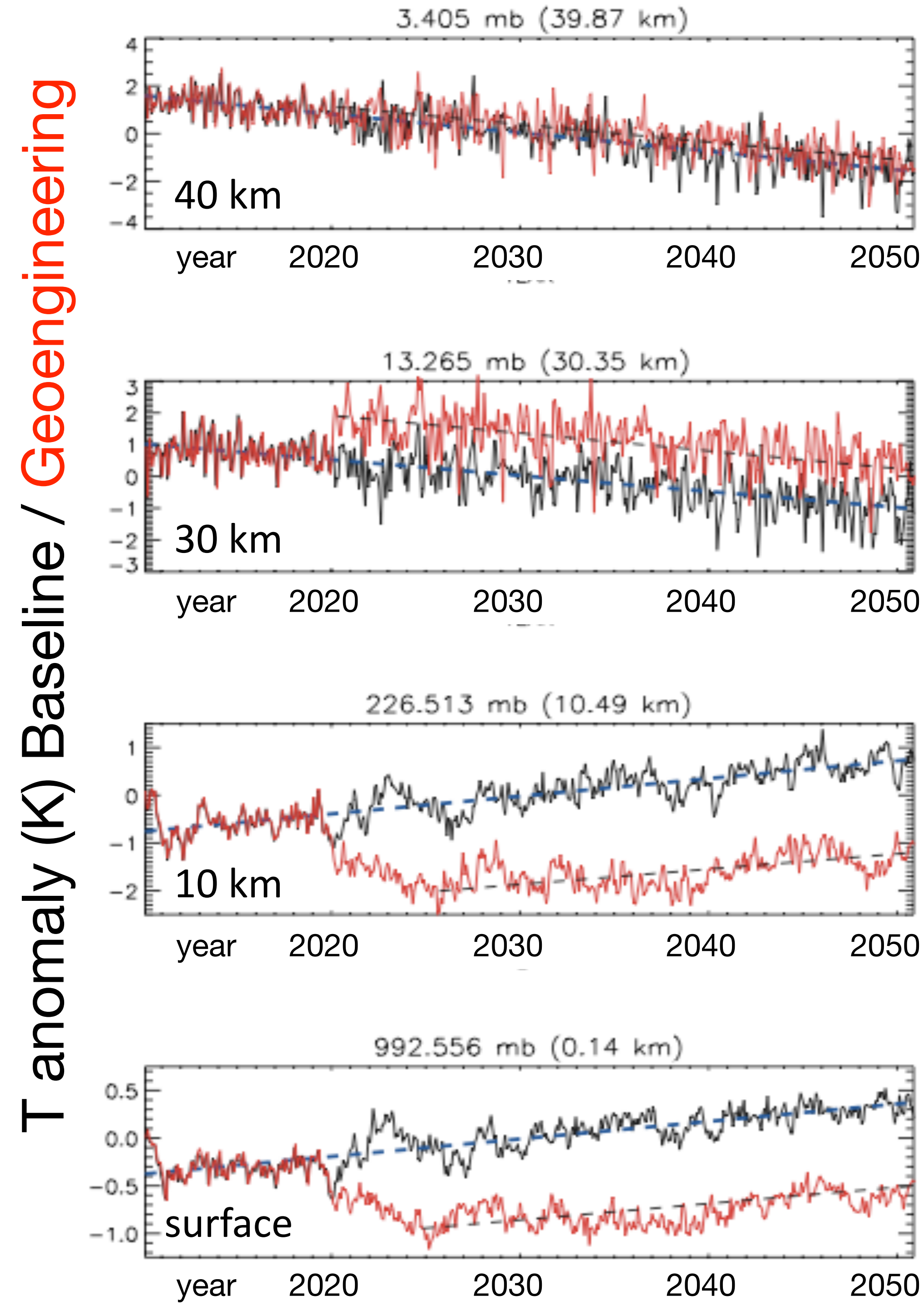


Impact of geoengineered aerosols on the troposphere and stratosphere

Tilmes *et al.*, J. Geophys. Res., vol. 114, pp. 12305, 2009.



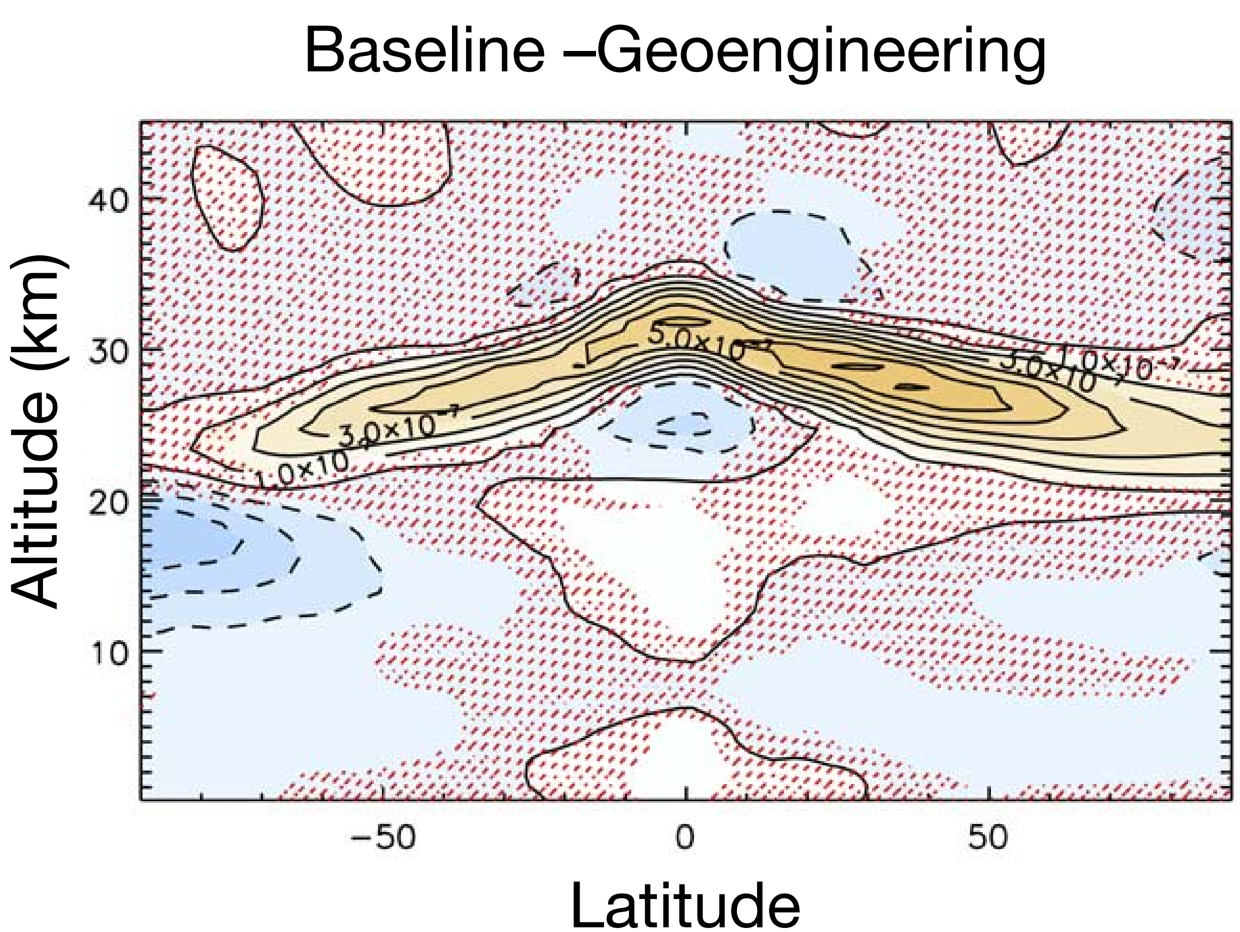
- ~5 years for adjustment of temperatures
- Constant temperature offset
- The fixed amount of sulfur cools the Earth's surface by ~0.9 K (Tropics), ~1.2 K (Global)
  - Delay of global warming by ~ 40 years
  - Dependence on continuous injection of sulfu

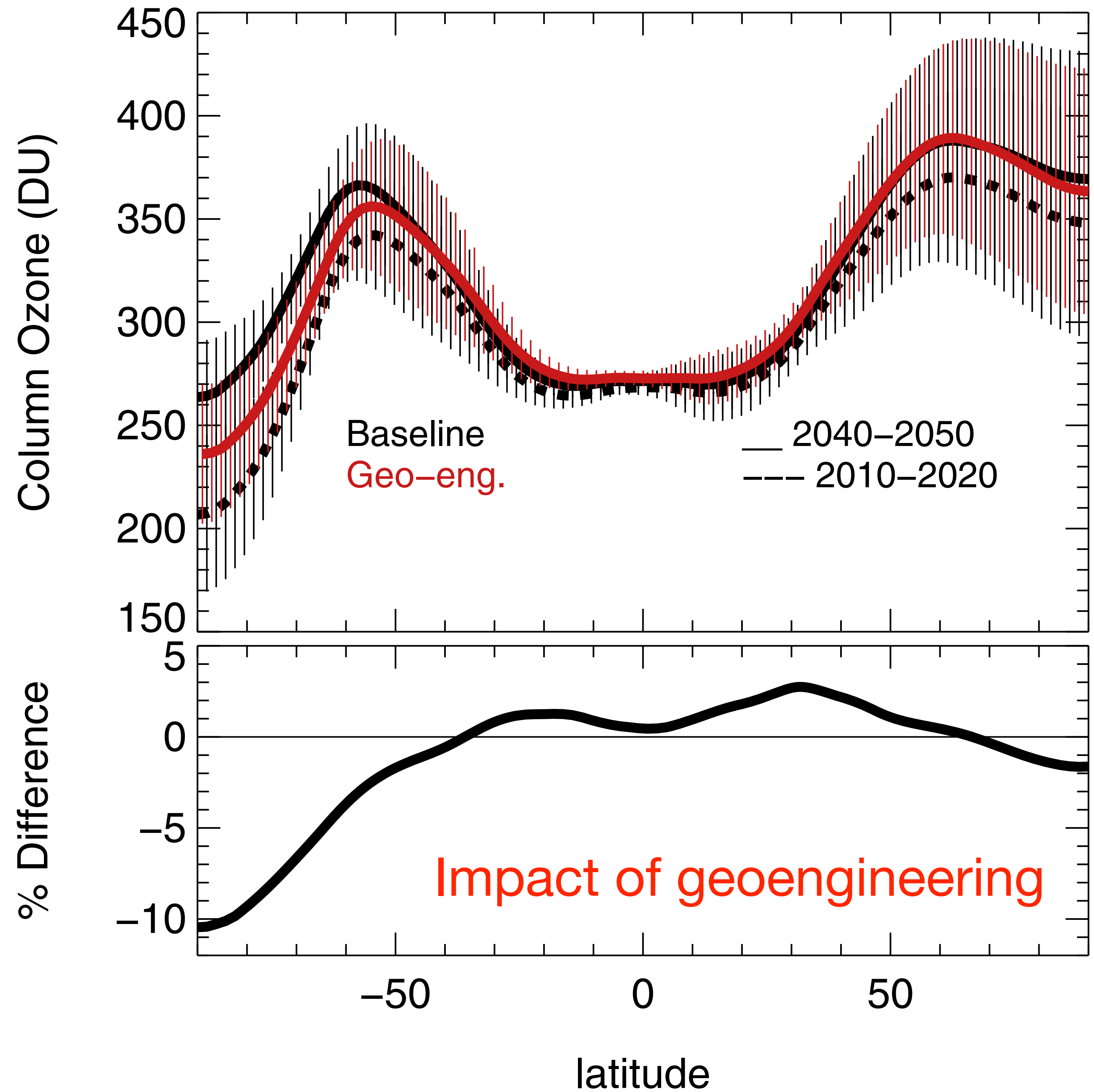


Impact of geoengineered aerosols on the troposphere and stratosphere

Tilmes *et al.*, J. Geophys. Res., vol. 114, pp. 12305, 2009.

Impacts of geoengineering on ozone



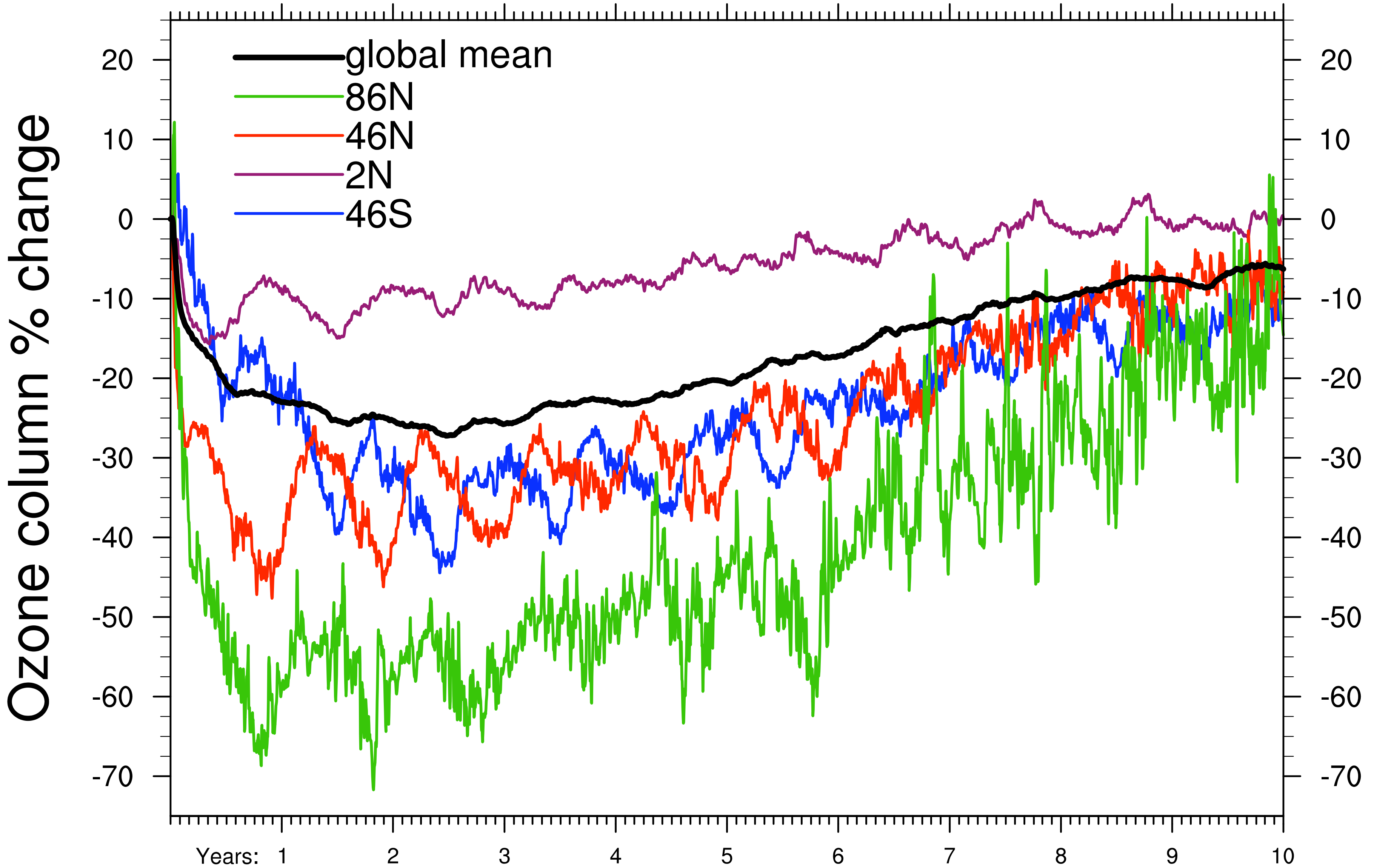


Massive global ozone loss predicted following regional nuclear conflict

Mills *et al.*, PNAS, vol. 105, pp. 5307, 2008

- WACCM input: new estimates of smoke produced by fires in contemporary cities following a regional nuclear war between India and Pakistan

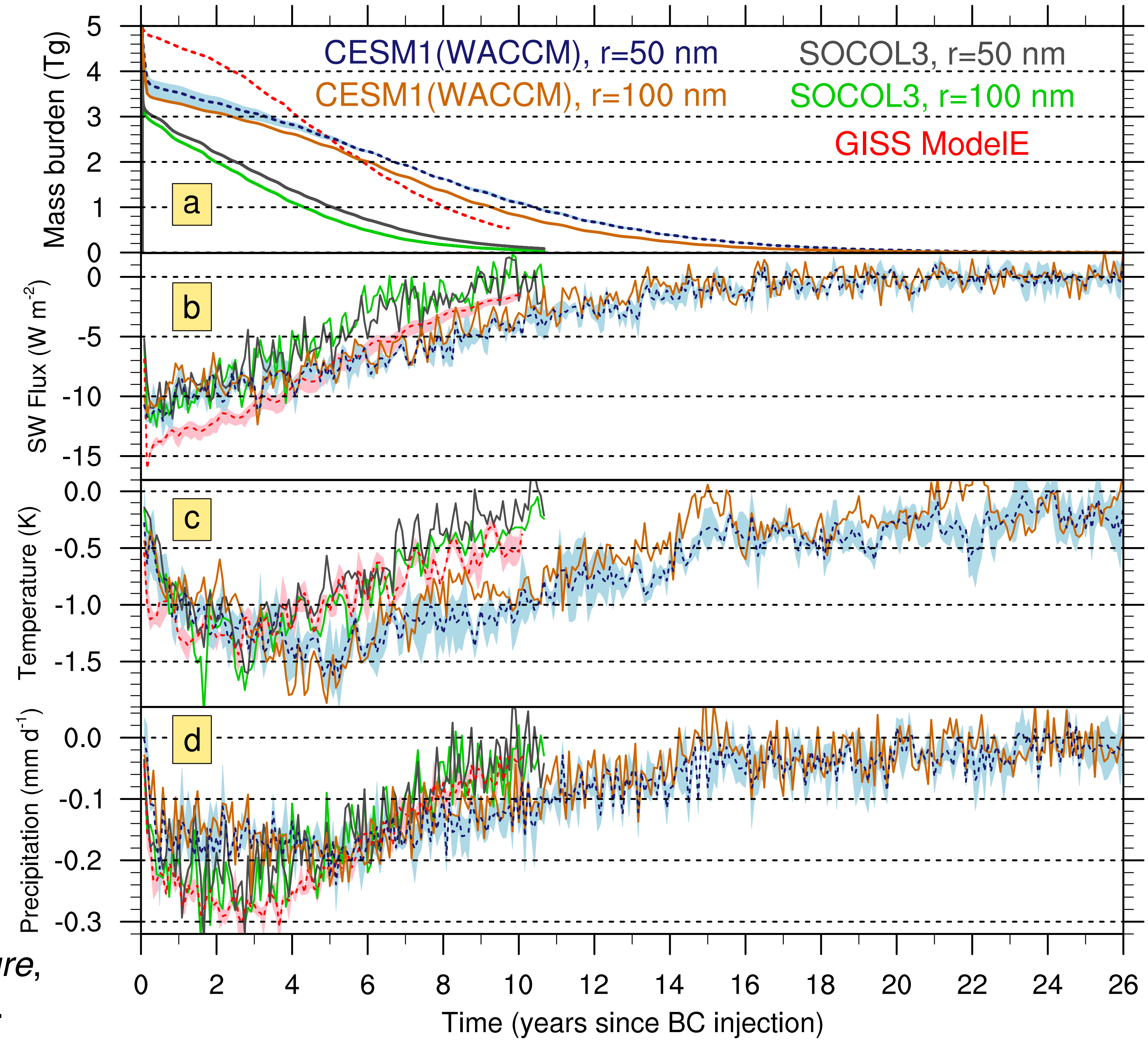
- Solar radiation heats the soot, lofting it to the stratopause, heating the the entire stratosphere for 10 years, altering reaction rates affecting ozone.



- Calculated ozone losses exceed 20% globally, 25-45% at midlatitudes, and 50-70% at northern high latitudes persisting for 5 years, with substantial losses continuing for 5 additional years. Column ozone amounts remain below that which defines the Antarctic ozone hole everywhere outside of the tropics.

Multidecadal global cooling and unprecedented ozone loss following a regional nuclear conflict

- CESM1(WACCM) with interactive chemistry, sea ice, full ocean, and land components
- Deep ocean cooling, sea ice expansion result in slower initial cooling, thermal inertia, increased albedo, more prolonged recovery: extended from 10 years in previous studies to more than 25 years
- Killing frosts would reduce growing seasons by 10 – 40 days per year for 5 years
- Summer enhancements in UV indices of 30% – 80% over midlatitudes
- Global nuclear famine?



Mills et al. (2014), *Earth's Future*, doi:10.1002/2013EF000205.

WACCM and CAM-Chem Customer Support

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

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