AEROSOL INTERACTIONS

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Thanks to Jerome Fast (PNNL) who provided material via the WRF-Chem tutorial lecture

OUTLINE

- Aerosol – Radiation Interactions
  - AOD measurements versus model prediction
- Aerosol – Cloud Interactions
- Processes that affect number of Aerosols
  - Aqueous phase chemistry
  - Heterogeneous chemistry
  - Lightning-NO_x

Throughout talk will note how different models represent aerosol properties or how they address a model process

CAM-Chem, GEOS-Chem, WRF-Chem, WRF-CMAQ

Thanks also to Andrew Gettelman (NCAR), Colette Heald (MIT), and Shawn Roselle (EPA) who ensured I say the right thing

3D ATMOSPHERIC CHEMISTRY MODELS CAN HAVE DIFFERENT AEROSOL CONFIGURATIONS

CAM-Chem with bulk aerosol

CAM-Chem with Modal Aerosol Module (MAM4)

GEOS-Chem with bulk aerosols

GEOS-Chem with sectional schemes (TOMAS, APM)

WRF-Chem with bulk aerosol scheme (GOCART)

WRF-Chem with modal scheme (MADE, MAM4)

WRF-Chem with sectional scheme (MOSAIC)

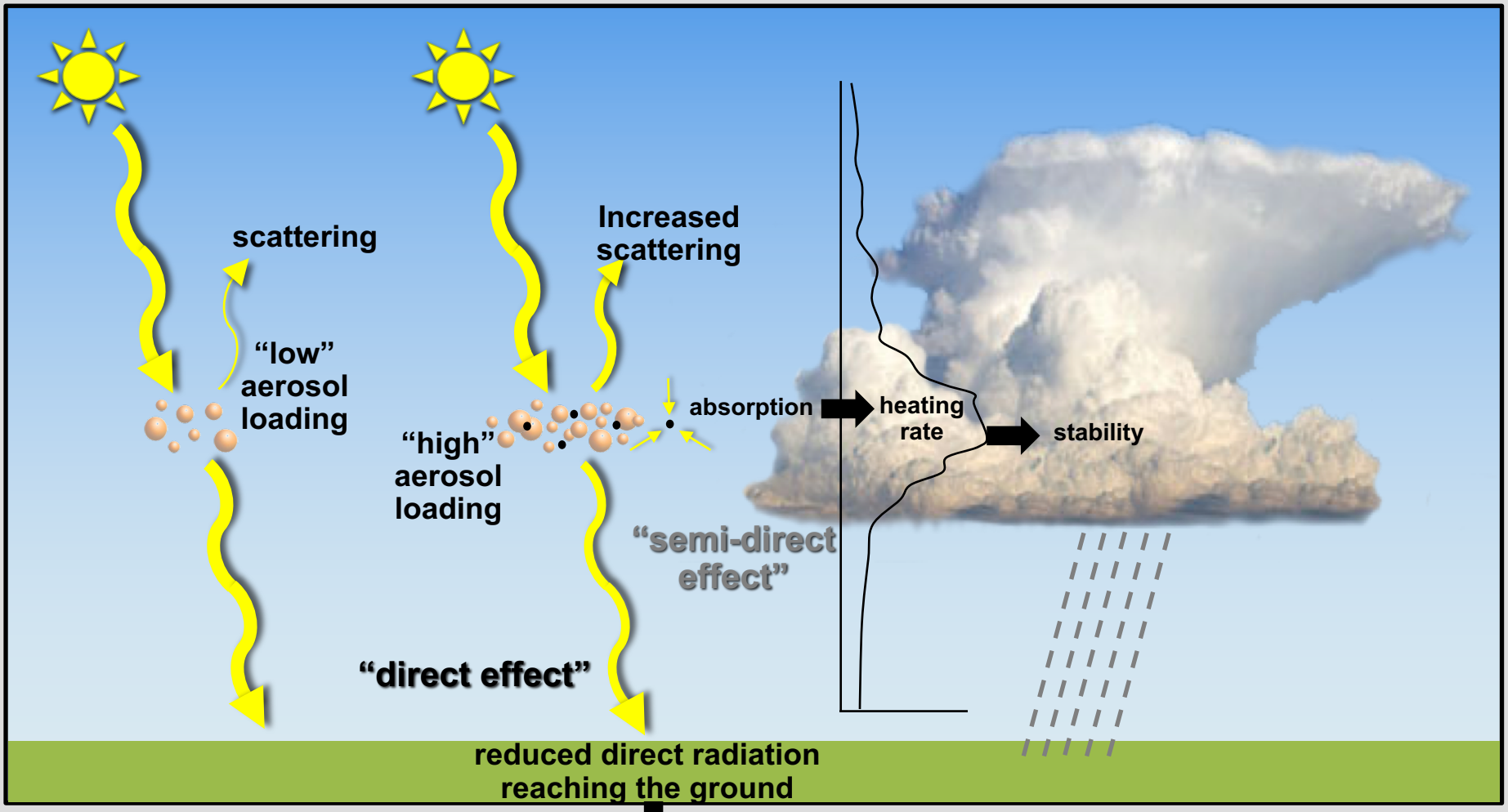
WRF-CMAQ with modal scheme

Why have different configurations of models?

Outline:

- Aerosol – Radiation Interactions

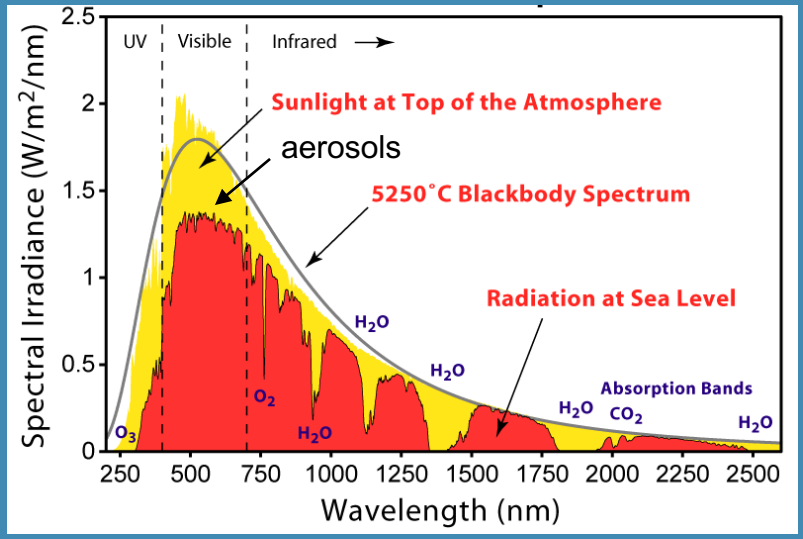
AEROSOL-RADIATION INTERACTIONS



In addition to water vapor, carbon dioxide, ozone, and other trace gases, aerosols can also affect the radiation budget, and atmospheric stability

surface heat and latent heat fluxes → boundary layer temperature and moisture → clouds

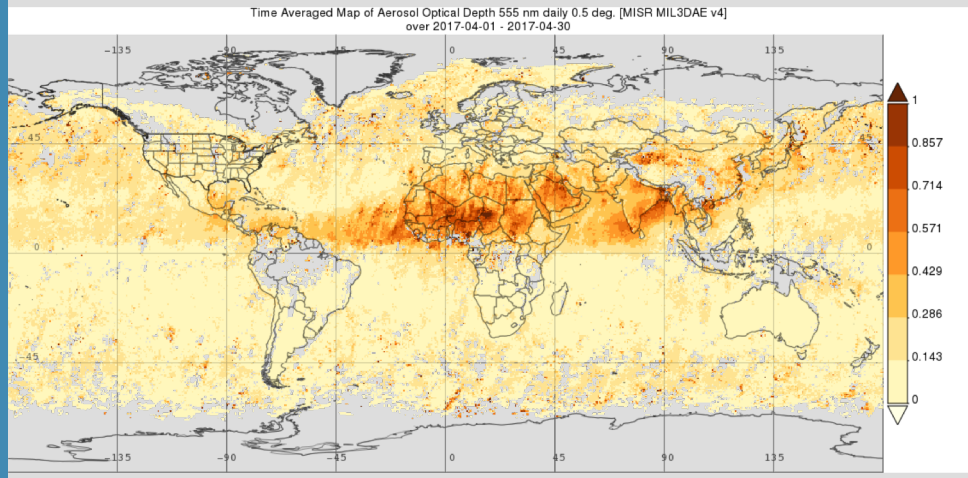
SOLAR RADIATION SPECTRUM



- Aerosols affect radiation in visible wavelengths

AEROSOLS IN THE TROPOSPHERE

- Aerosols are much more heterogeneous in nature compared to H_2O (v), CO_2
- Aerosols challenging to simulate
  - Episodic sources
  - Diverse composition: dust, sea salt, black carbon, brown carbon, sulfate, nitrate ….



Aerosol Optical Depth at 555 nm from MiSR, April 2017 monthly average

HOW DO WEATHER AND CLIMATE MODELS REPRESENT AEROSOL EFFECTS ON RADIATION?

1) Not at all: aerosols do not affect radiation and therefore surface and latent heat fluxes
2) Use aerosol climatological aerosol properties that can vary in space and seasonality WRF, NCEP models
3) Predict aerosol concentrations and optical properties

WRF-Chem, CAM-Chem, GEOS-Chem, WRF-CMAQ

Important aerosol properties to predict:

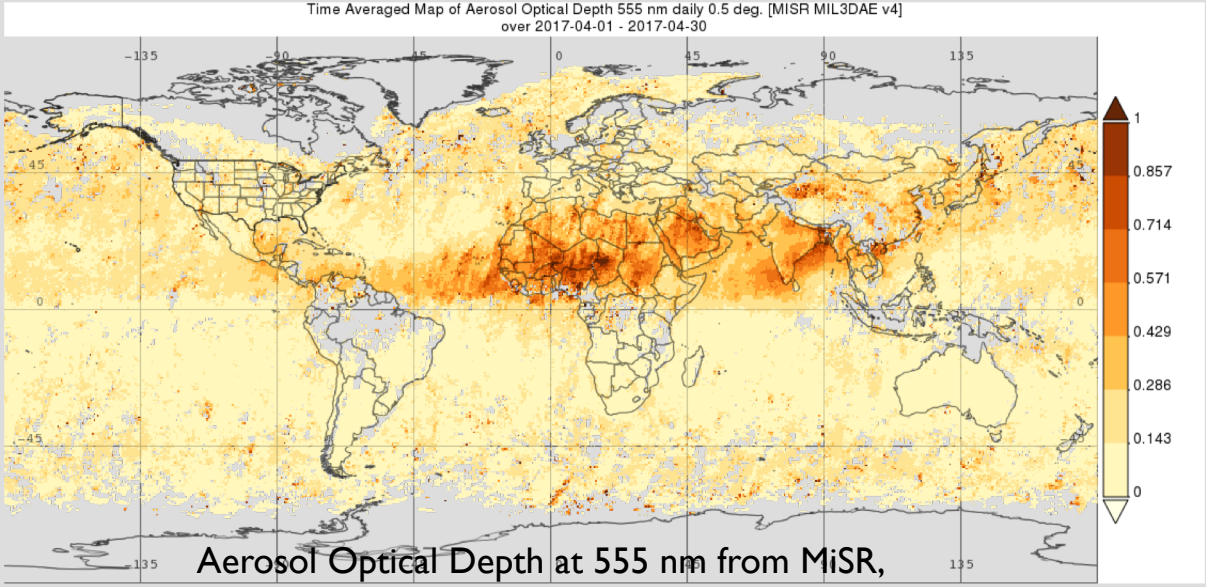
- Extinction coefficient, aerosol optical depth
- Single scattering albedo
- Asymmetry factor
- Aerosol water
- Refractive indices
- Mixing state of aerosols

EXTINCTION COEFFICIENT, AEROSOL OPTICAL DEPTH

Extinction coefficient: fractional depletion of radiance per unit path length (km^{-1}) due to scattering and absorption by aerosols

Aerosol optical depth (AOD) or **thickness** (AOT): integrated extinction coefficient over a vertical column, $I / I_o = e^{-AOD}$

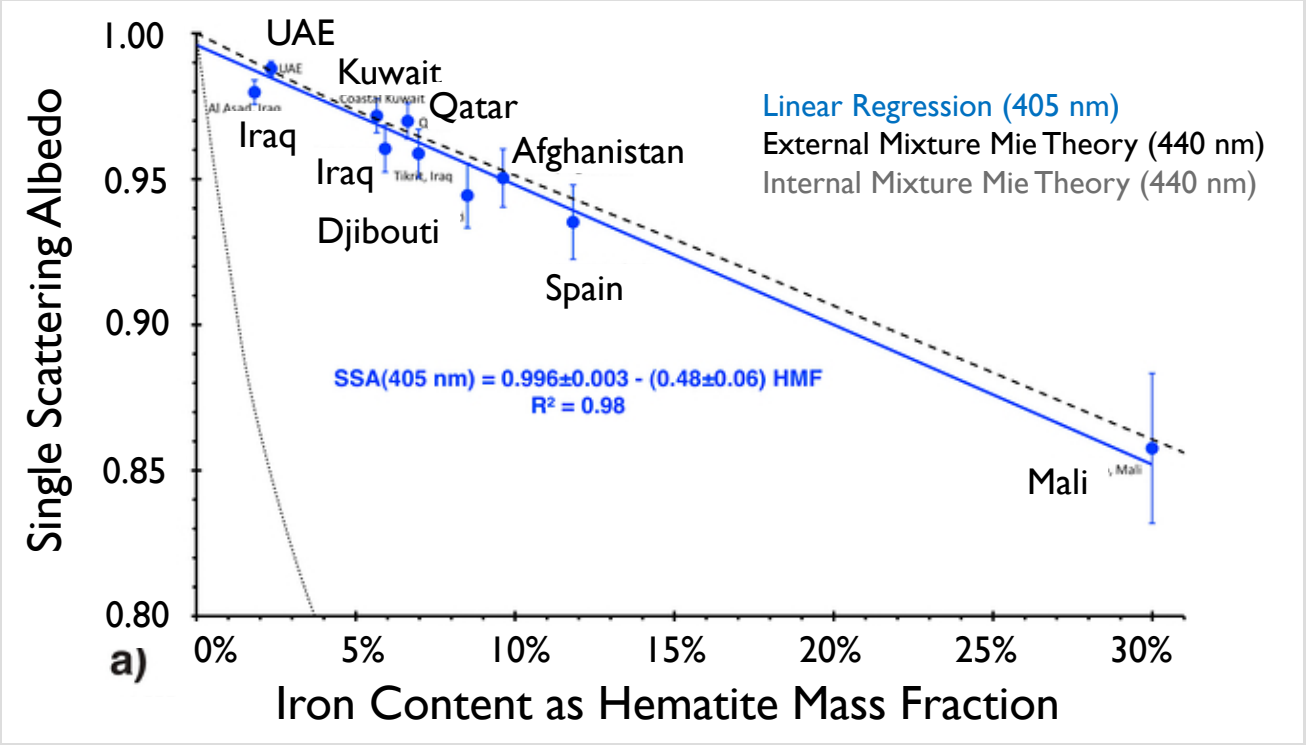
- AOD = 0 no aerosol effect
- AOD ~ 1 "large"
- AOD > 1 extremely high aerosol concentrations



Aerosol Optical Depth at 555 nm from MiSR, April 2017 monthly average

SINGLE SCATTERING ALBEDO (SSA)

- SSA is ratio of scattering to extinction efficiency, $\omega_o = k_s / (k_a + k_s)$
  - SSA = 1 all particle extinction due to scattering
  - SSA = 0 all particle extinction due to absorption (does not happen in reality)



Single scattering albedo of fine mineral dust aerosols is controlled by iron concentration

Moosmuller et al. (2012) *J. Geophys. Res.*

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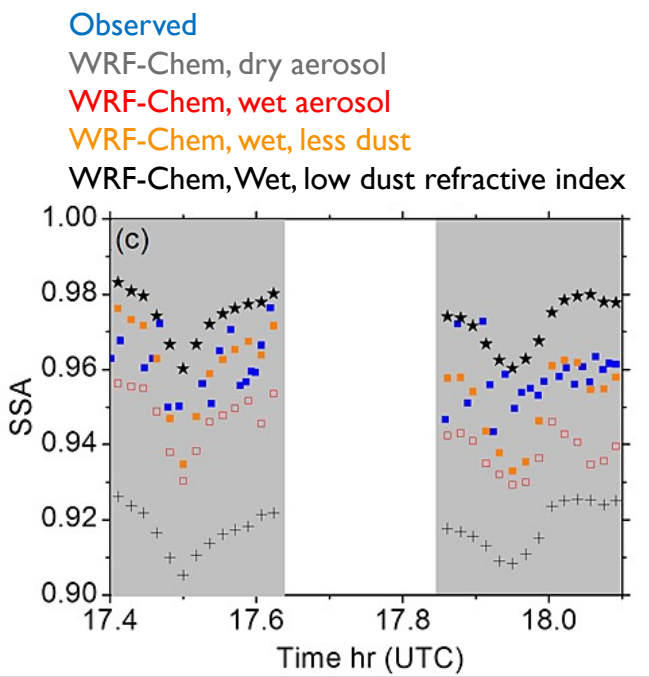


SSA is sensitive to assumptions in model configuration

- Aerosol water
- Aerosol mass concentrations
- Refractive index of dust

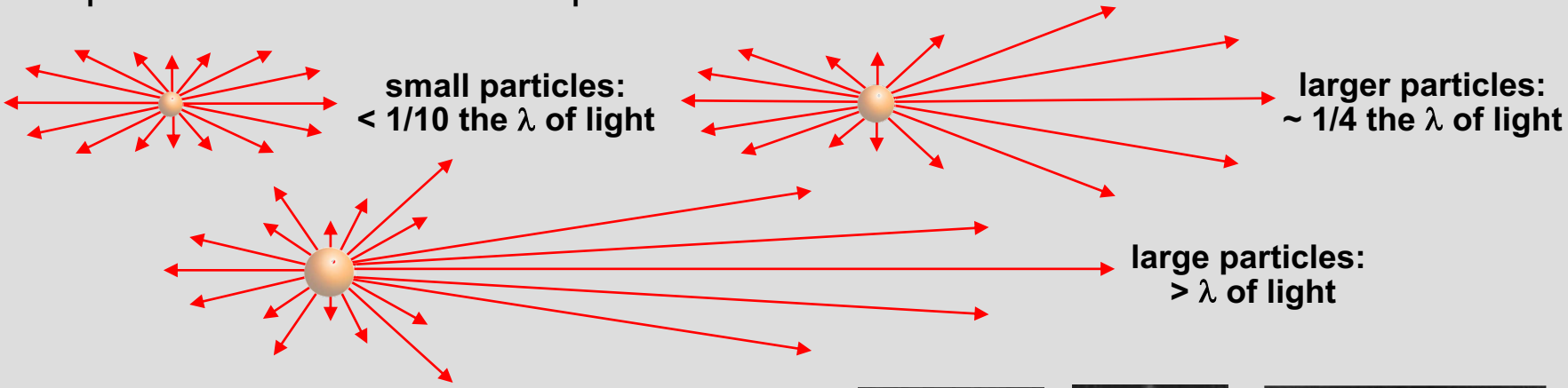
WRF-Chem simulation of aerosols
Sensitivity tests of optical properties use WRF-Chem aerosol concentration predictions in an offline radiation calculation.
Shrivastava et al. (2013) *J. Geophys. Res.*

ASYMMETRY FACTOR

Preferred scattering direction (forward or backward) for the light encountering the aerosol particles

- Approaches 1 for scattering strongly peaked in the forward direction
- Approaches -1 for scattering strongly peaked in the backward direction
- g = 0 means scattering evenly distributed between forward and backward scattering

Depends on both size and composition of aerosols



IMPORTANCE OF AEROSOL WATER

- Uptake of water by aerosols depends on relative humidity
- Composition affects water uptake (hydrophobic vs hydrophilic aerosol)
- Different ways to compute aerosol water
  - Specified growth factors from GADS — GEOS-Chem bulk
  - Hygroscopicity (κ value, Petters and Kreidenweis (2007)) — WRF-Chem bulk, GEOS-Chem TOMAS, WRF-CMAQ
  - Aerosol water explicitly predicted — WRF-Chem MOSAIC, WRF-CMAQ
  - Aerosol water determined from Kohler theory = f(RH, κ) — CAM-Chem

Smoky Mountains

REFRACTIVE INDICES

- Refractive index of a substance is a dimensionless number that describes how light propagates through a medium
- Refractive indices in models based on literature values derived from laboratory experiments, vary with wavelength for some aerosol compositions
- Challenge: brown carbon

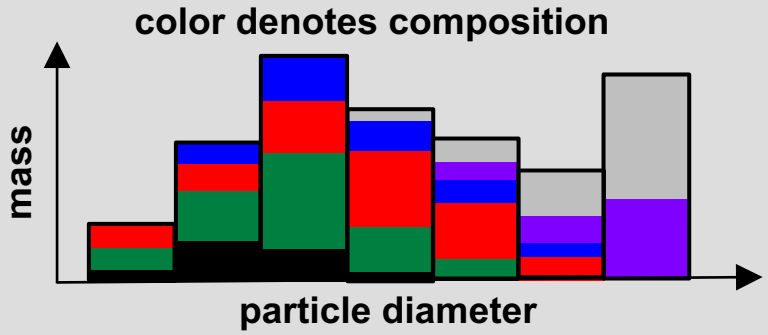
Default Values for SW Radiation in WRF

	real part		imaginary part
BC =	1.850	+	0.71i
OM =	1.450	+	0.00i
SO_4 =	1.468	+	1.0e-9i
NH_4NO_3 =	1.500	+	0.00i
NaCl =	1.510	+	0.866e-6i
dust =	1.550	+	0.003i
H_2O =	1.350	+	1.52e-8i

These values vary depending on wavelength and from one model to another

MIXING RULES FOR MIE CALCULATIONS

Often assumed that all particles within a size range have the same composition, although relative fraction can differ among size ranges.



Assumptions regarding mixing:

External Mixture of Aerosols bulk aerosol schemes

Internal Mixture of Aerosols sectional aerosols schemes

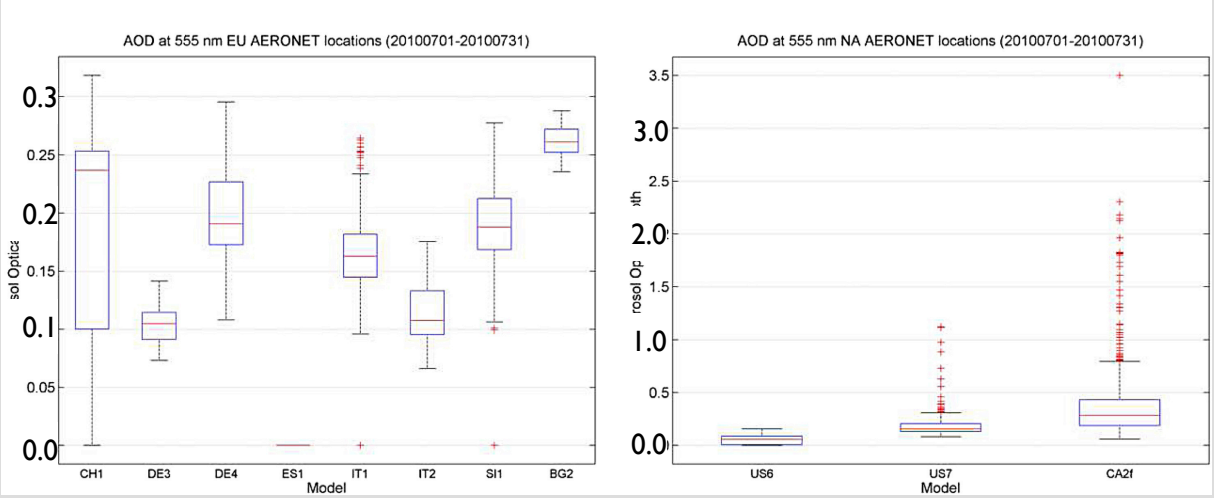
- **Volume Averaging:** averaging of refractive indices based on composition
- **Shell-Core**: black carbon core and average of other compositions in shell (Ackermann and Toon, 1983; Borhren and Huffman, 1983)
- **Maxwell-Garnett:** small spherical randomly distributed black carbon cores in particle (Borhren and Huffman, 1983)

Both External and Internal Mixture CAM-Chem MAM4, CMAQ

COMPARISON OF AEROSOL OPTICAL PROPERTIES AMONG REGIONAL MODELS

Uncertainties of simulated aerosol optical properties induced by assumptions on aerosol physical and chemical properties: An AQMEII-2 perspective

Curci et al. (2014) *Atmos. Environ.*



Aerosol optical depth over AERONET stations in July 2010 simulated by AQMEII-2 models (COSMO-ART, COSMO-MUSCAT, WRF-Chem, WRF-CMAQ, GEM-MACH)
Europe: 8 models, 85 AERONET stations
North America: 3 models, 77 AERONET stations

- Calculate optical properties from several aerosol models using same assumptions.
- Test choices on mixing state, refractive index, density and hygroscopicity.
- The most sensitive parameter is the aerosol mixing state.
- The related uncertainty on calculated AOD and SSA is 30–35%.

COMPARISON OF AEROSOL OPTICAL PROPERTIES AMONG REGIONAL MODELS

For each model and for different sensitivity tests:

AOD (440 nm)

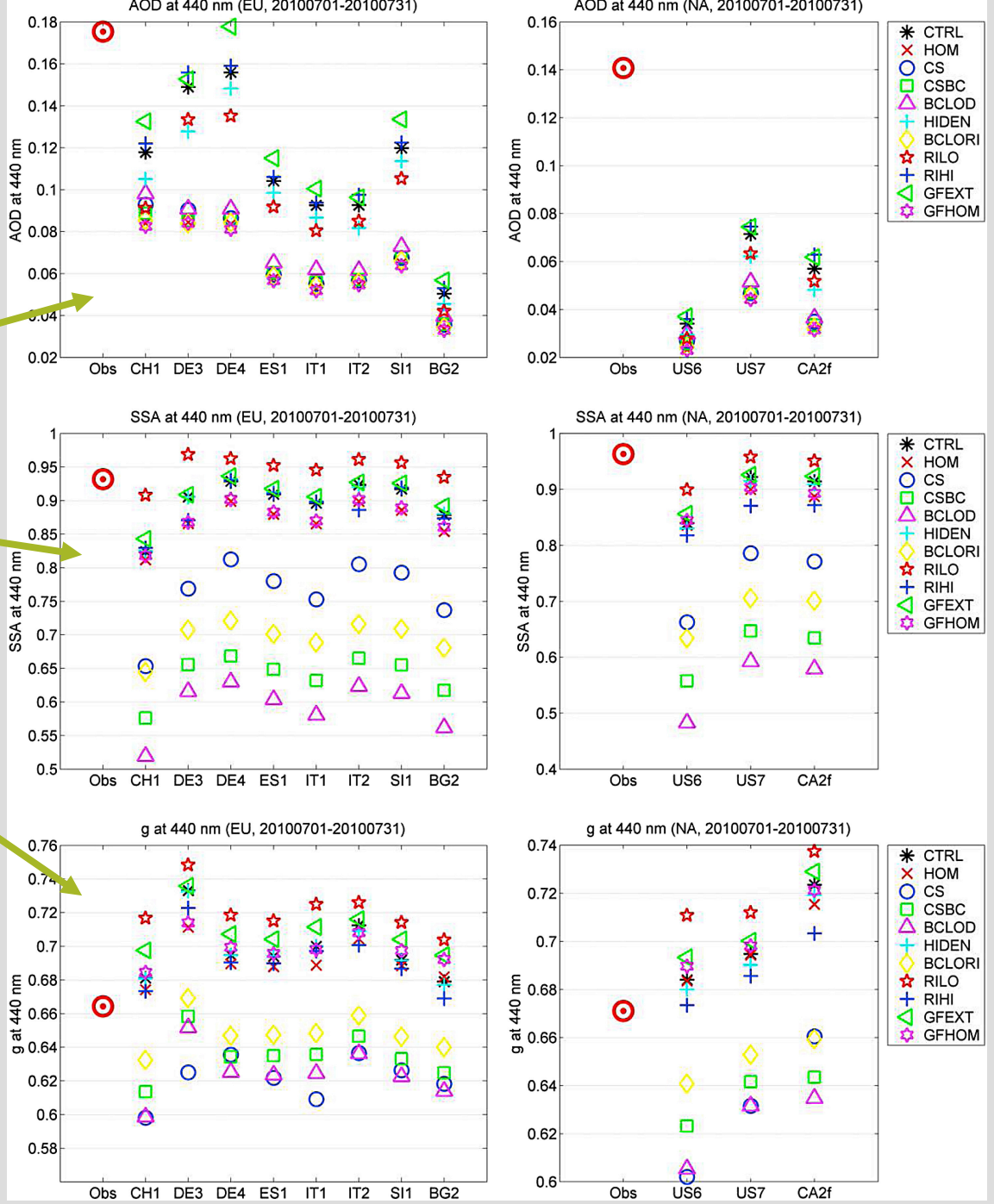
SSA (440 nm)

g (440 nm)

Over Europe (left) and N.America (right)

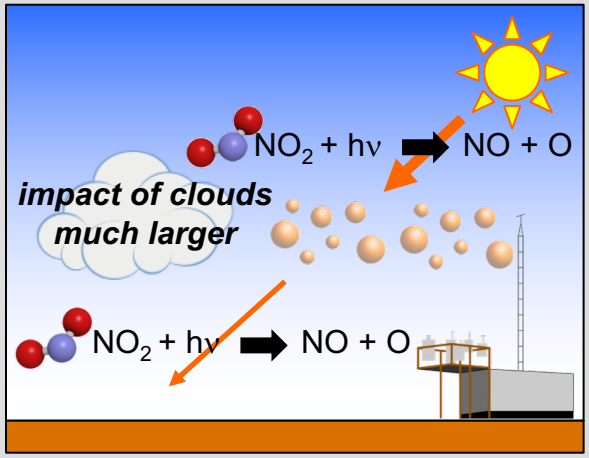
& compared to obs (red target)

Curci et al. (2014) *Atmos. Environ.*

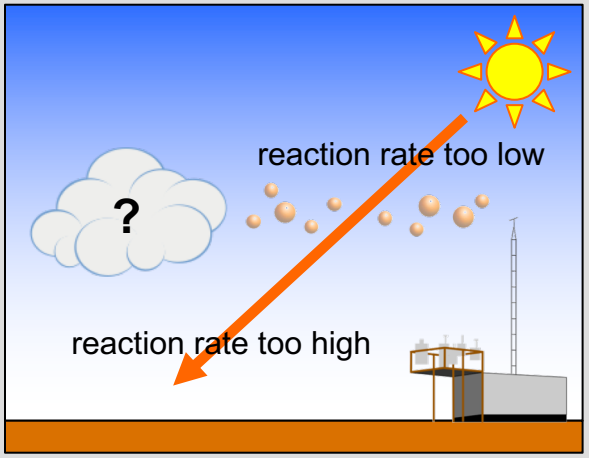


AEROSOL IMPACTS ON CHEMISTRY

Observed Aerosols



Simulated:
Too Few or Too Thin



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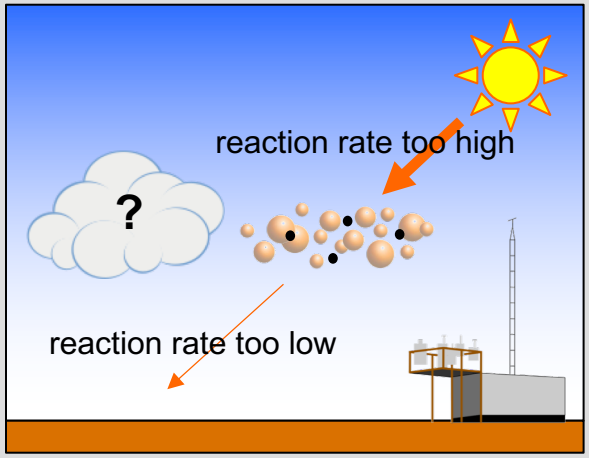
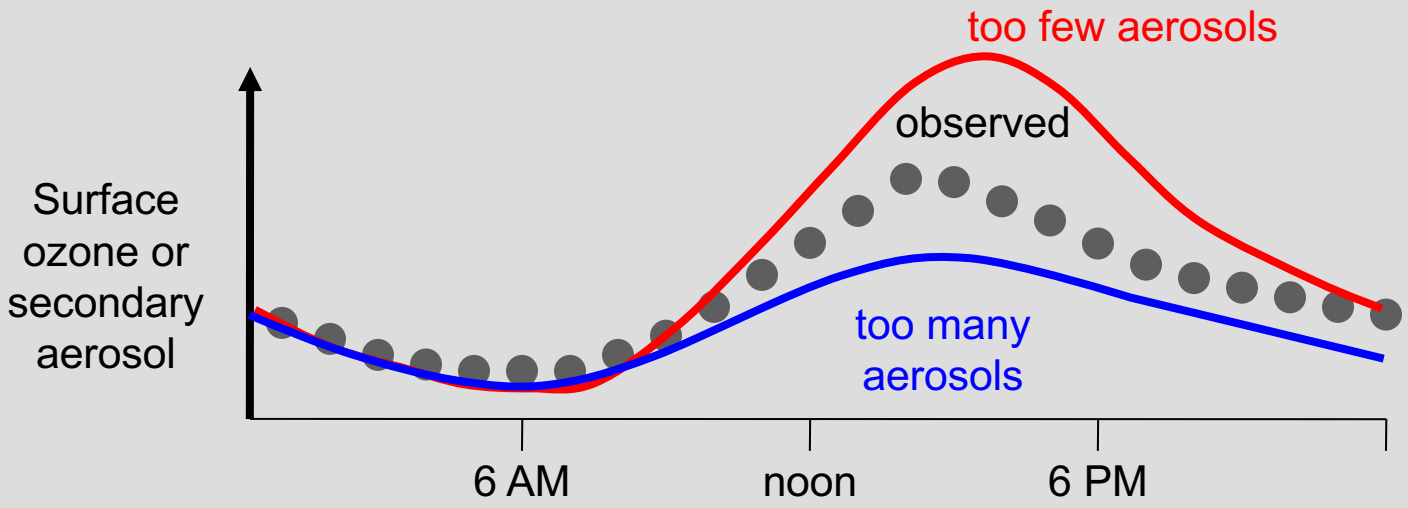
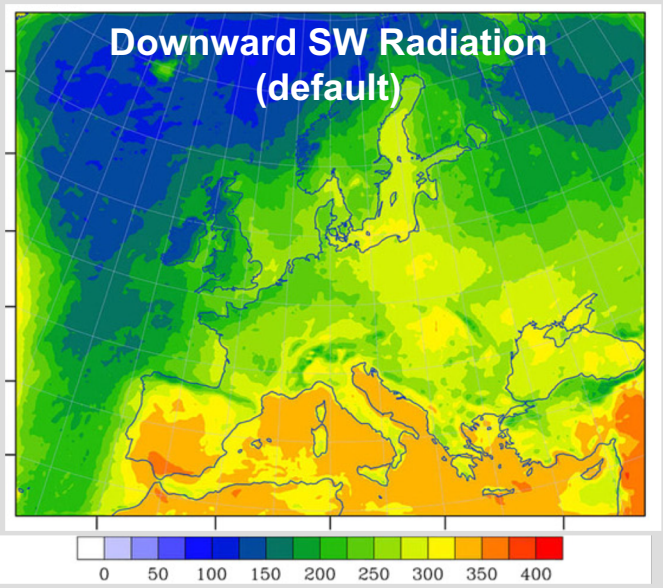
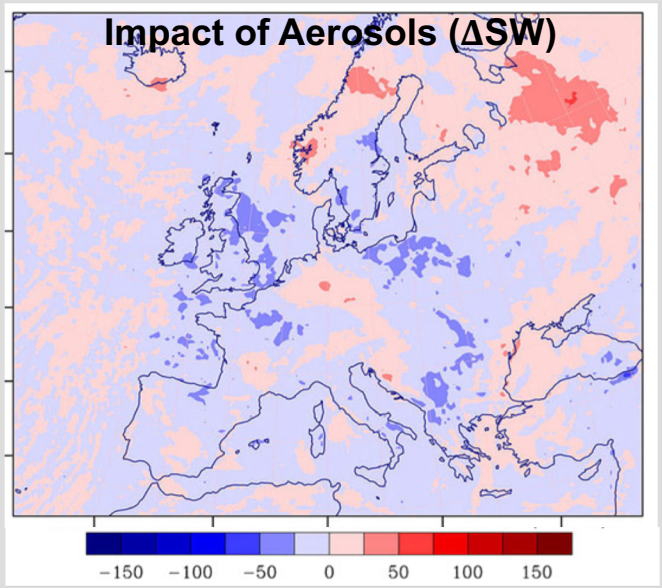


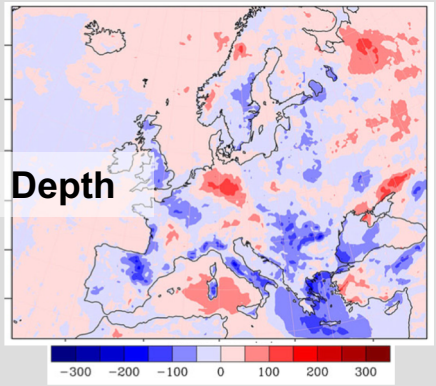


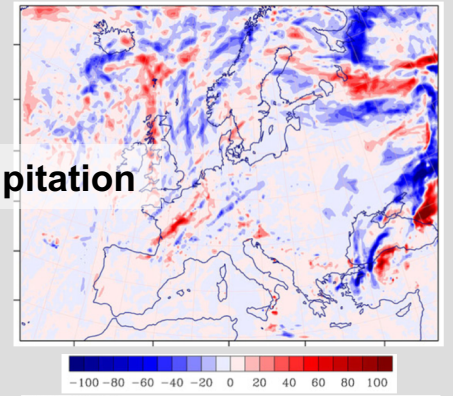
errors could impact simulated concentrations the next day

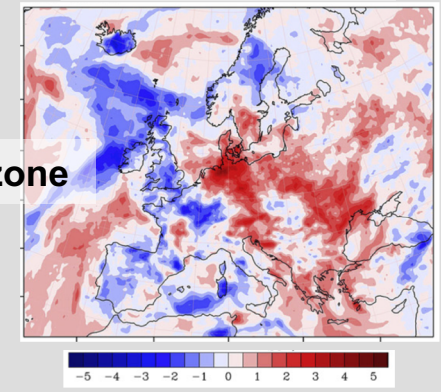
EFFECTS OF AEROSOL-RADIATION INTERACTIONS ON CHEMISTRY





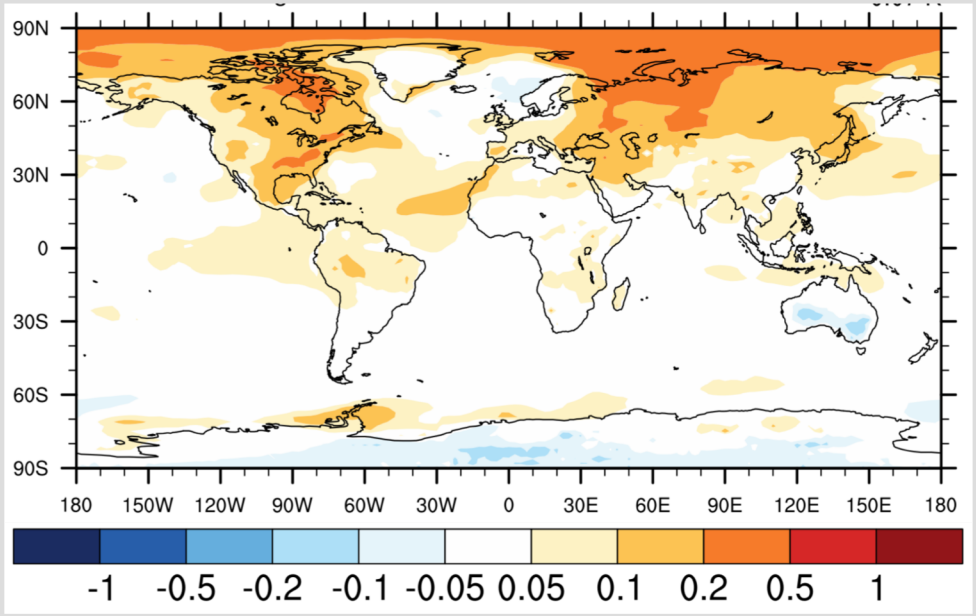






Forkel et al. (2012)
Atmos. Chem. Phys.

U.S. SO_2 EMISSIONS AFFECT TEMPERATURES OVER NORTHERN HEMISPHERE



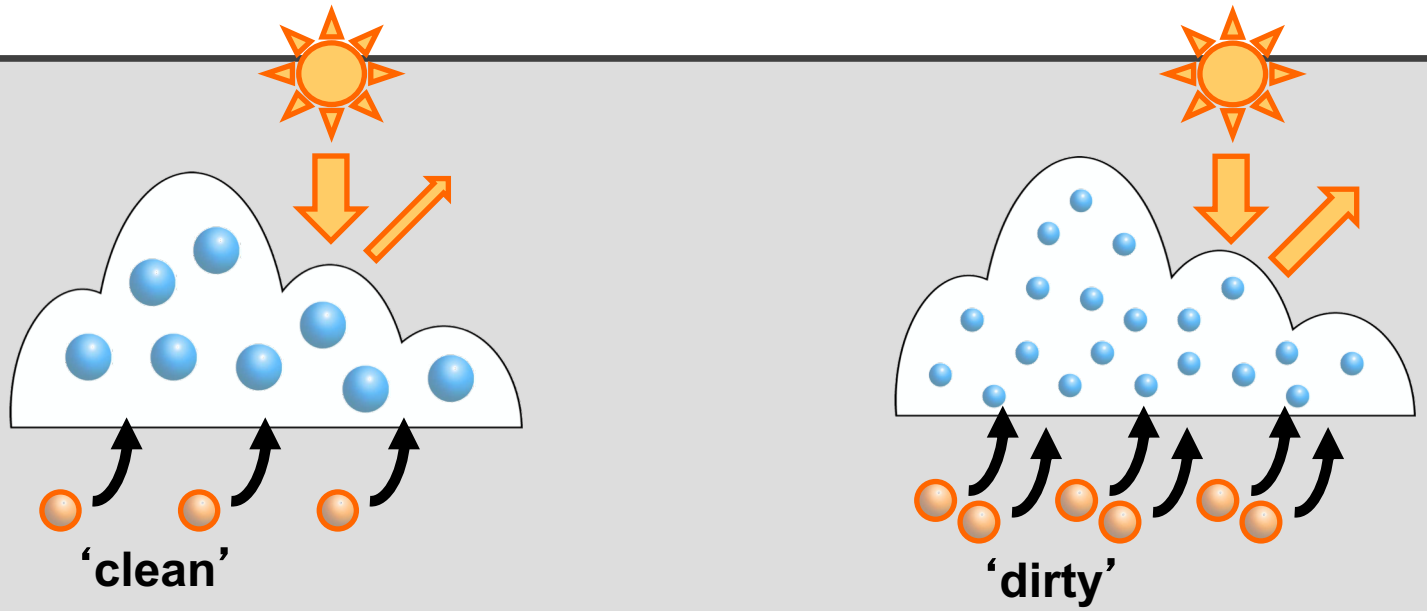
Conley et al. (2018) *J. Geophys. Res.*

Multi-model surface temperature response in K due to zeroing out US SO_2 emissions.

3 models participated in study: CESM, GFDL, and GISS

Both aerosol-radiation and aerosol-cloud interactions are represented

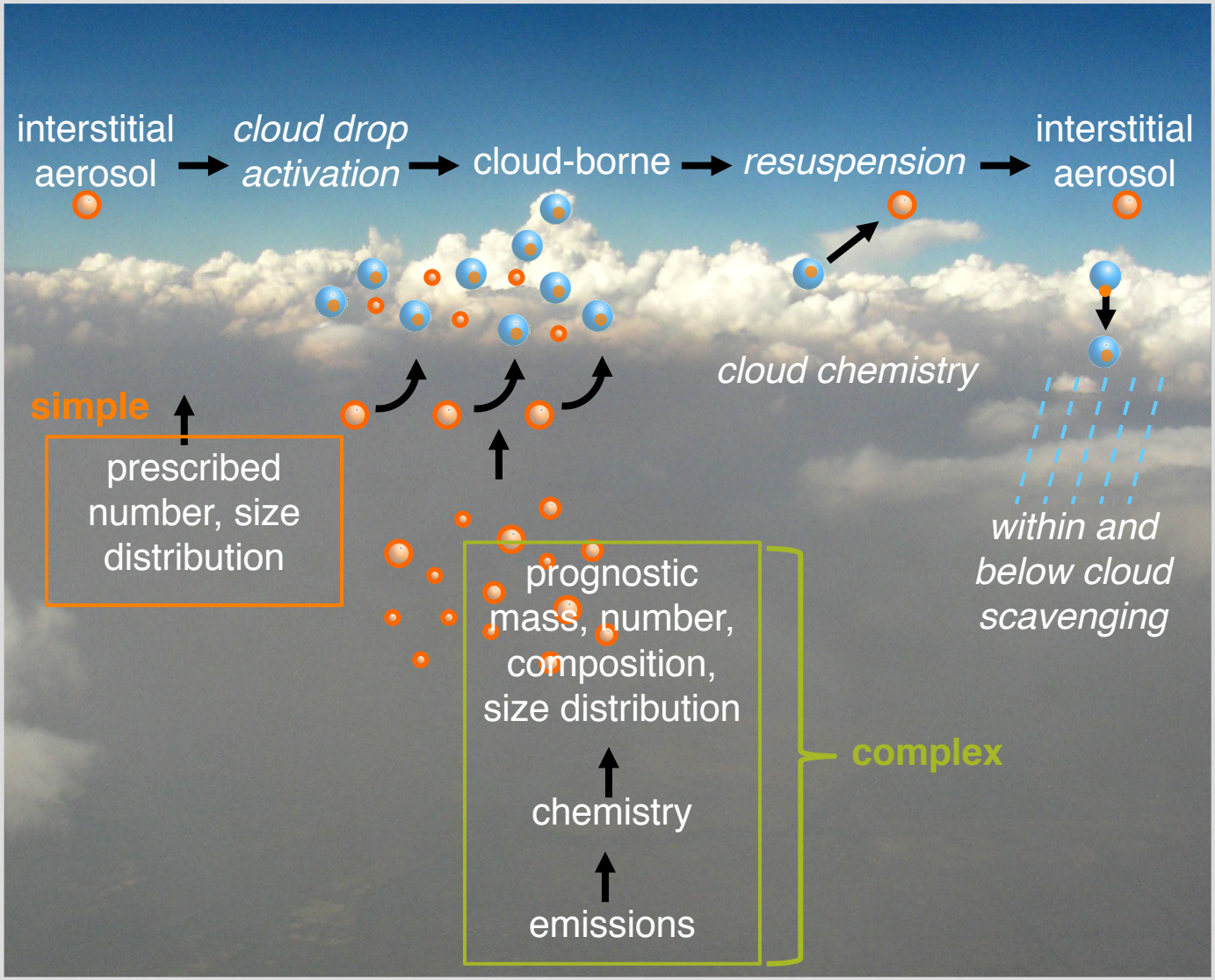
AEROSOL CLOUD INTERACTIONS



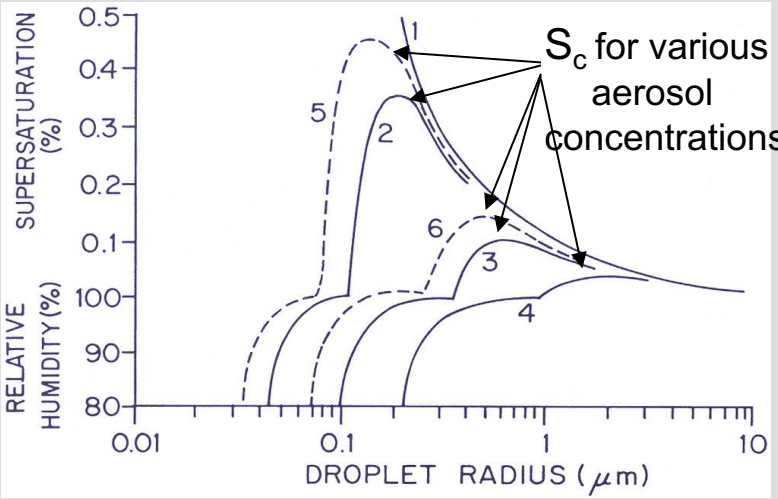
The number of cloud condensation nuclei affects the cloud drop size distribution, and consequently cloud albedo and radiation budget

AEROSOL CLOUD INTERACTIONS

General Description and Assumptions



CLOUD DROP ACTIVATION



1: pure H_2O
2: 10^{-19} kg NaCl
3: 10^{-18} kg NaCl
4: 10^{-17} kg NaCl
5: 10^{-19} kg $(NH_4)SO_4$
6: 10^{-18} kg $(NH_4)SO_4$

Köhler Theory
When a critical supersaturation (S_c) is reached, cloud drops form

→ Aerosols are activated when the air entering the cloud has maximum supersaturation $S_{max} > S_c$

Supersaturation (S) is a function of the vertical velocity

S_{max} depends on aerosol size and composition [*Abdul Razzak and Ghan,* 2000, 2002]

Number of cloud drops formed based on fraction of aerosol mass or number in the bin or mode having $\mathbf{S_{max} > S_c}$

Models that use this approach:
WRF-Chem MOSAIC aerosols
CAM-Chem
coupled WRF-CMAQ

MAXIMUM SUPERSATURATION DEPENDS ON AEROSOL COMPOSITION

Hygroscopicity
Tendency of aerosol to absorb water vapor from the surrounding atmosphere

Hygroscopicity depends on aerosol composition, Kappa (κ) values:
SO_4 = 0.5
NO_3 = 0.5
NH_4 = 0.5
OC = 0.14 (some OC may be more hydrophilic)
BC = 1.0e-6 *hydrophobic*
Dust = 0.10 – 0.14
NaCl = 1.16 *hydrophilic*

Mixture of aerosols (i.e. internal mixture) often uses a volume weighted bulk hygroscopicity

Petters and Kreidenweis (2007) *Atmos. Chem. Phys.* for more information on κ values

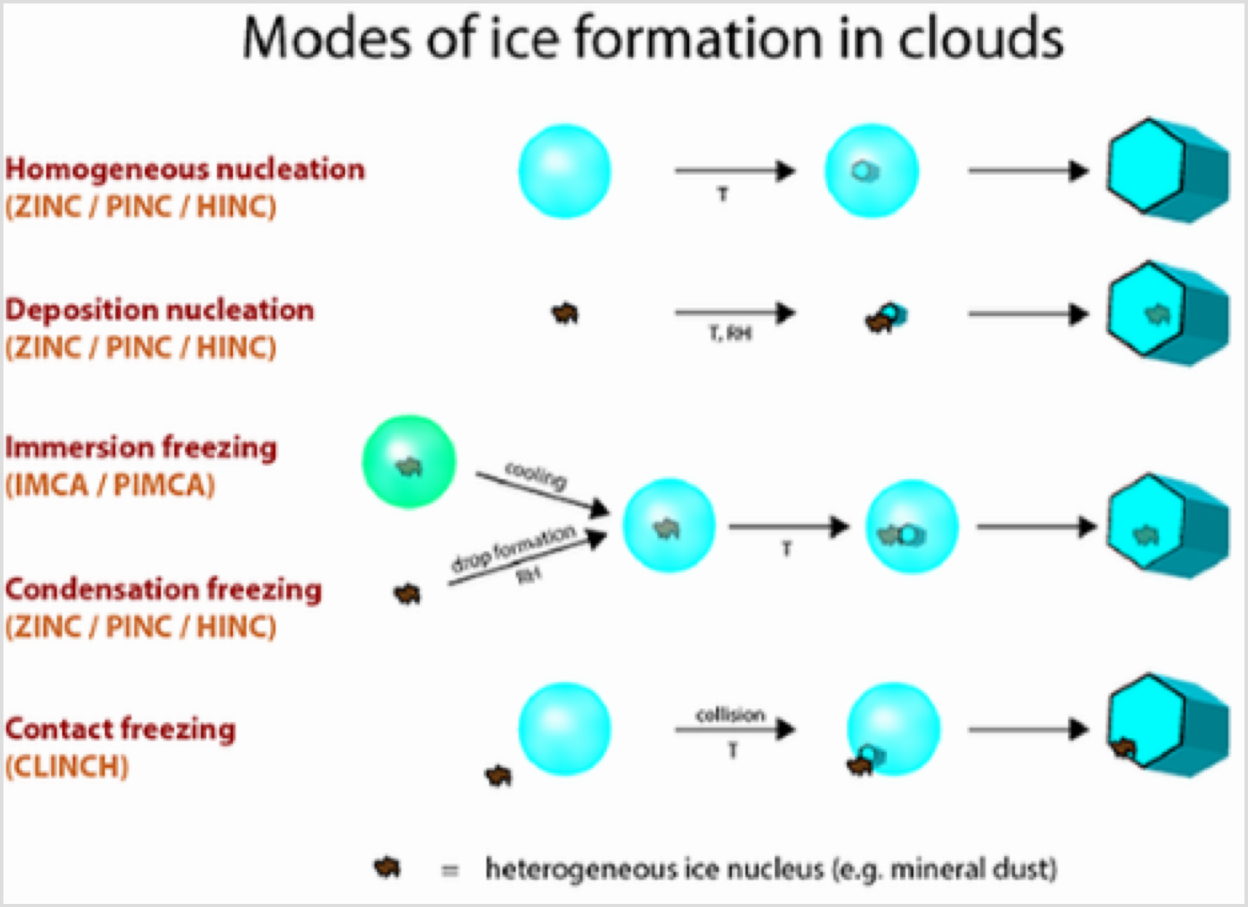
CLOUD DROP ACTIVATION

Köhler Theory
Cloud drop activation based on Kohler theory ($S_{max} > S_c$) occurs only at cloud base. Usually the case, but not always (e.g., deep convection).

Cloud drop activation dependent on vertical velocity in the grid cell. A coarse grid spacing will have smaller vertical velocities than a fine grid spacing. → Use a subgrid vertical velocity
CAM-Chem bases this on TKE (turbulent kinetic energy)
WRF-Chem and coupled WRF-CMAQ use "cloud resolving" grid spacings ($\Delta x < 5$ km) where this is less of an issue, yet vertical velocity still varies with grid sizes.

Models that use this approach:
WRF-Chem MOSAIC aerosols
CAM-Chem
coupled WRF-CMAQ

ICE NUCLEATION



Multiple ways ice nucleation can form.

Mineral dust serves as main form of ice nucleus (IN)

Models that allow predicted number of aerosols to affect ice crystal concentrations: CAM-Chem coupled WRF-CMAQ

From U. Lohmann (ETH) group web page:
http://www.iac.ethz.ch/group/atmospheric-physics/research/ice-nucleation.html

CLOUD CONDENSATION NUCLEI (CCN), ICE NUCLEI (IN)

CCN: Number concentration of aerosols that will be activated into cloud drops

IN: Number concentration of aerosols that will be nucleated into ice crystals

Note: Cloud physics models (or modules) are only concerned about the CCN and IN number. Thus, some of these cloud models predict only CCN and IN and not more explicit aerosol composition and sizes.

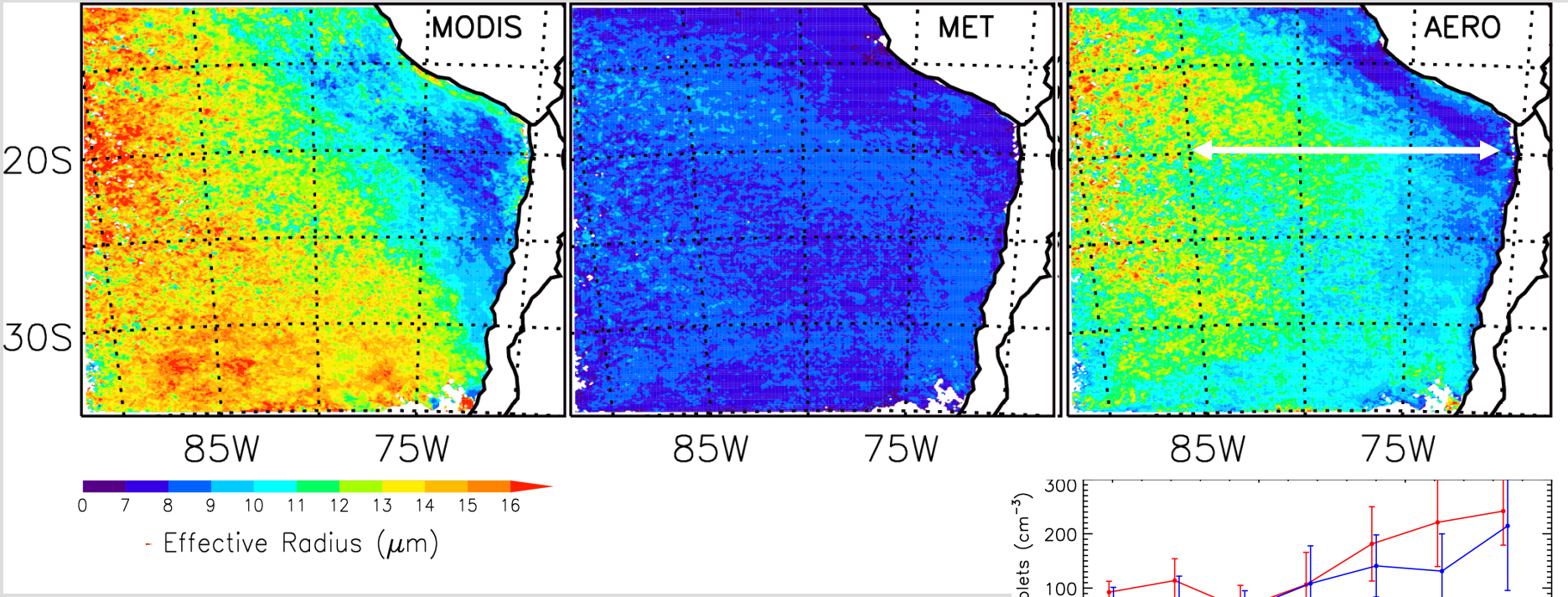
EXAMPLES OF AEROSOL-CLOUD INTERACTIONS

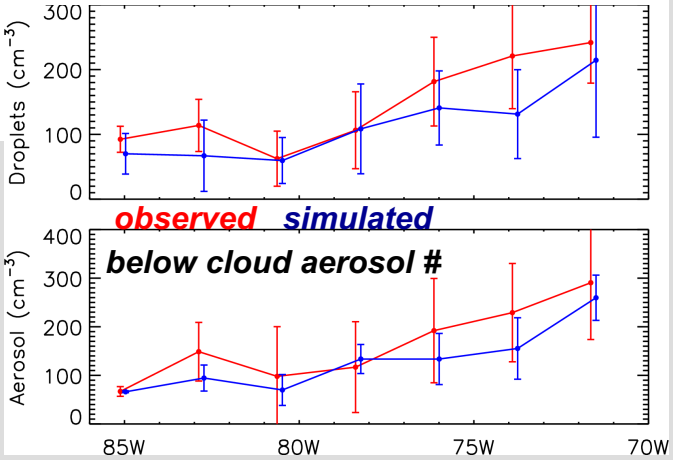
- Marine Stratocumulus (Yang et al., 2011)
  - Aerosol effects on cloud drop size
- Convective Clouds (several papers)
  - Aerosol effects on precipitation

MARINE STRATOCUMULUS

Average Effective Droplet Radius during 2008 VOCALS-REx

MODIS ***WRF no chemistry*** ***MOSAIC aerosols and Morrison microphysics***



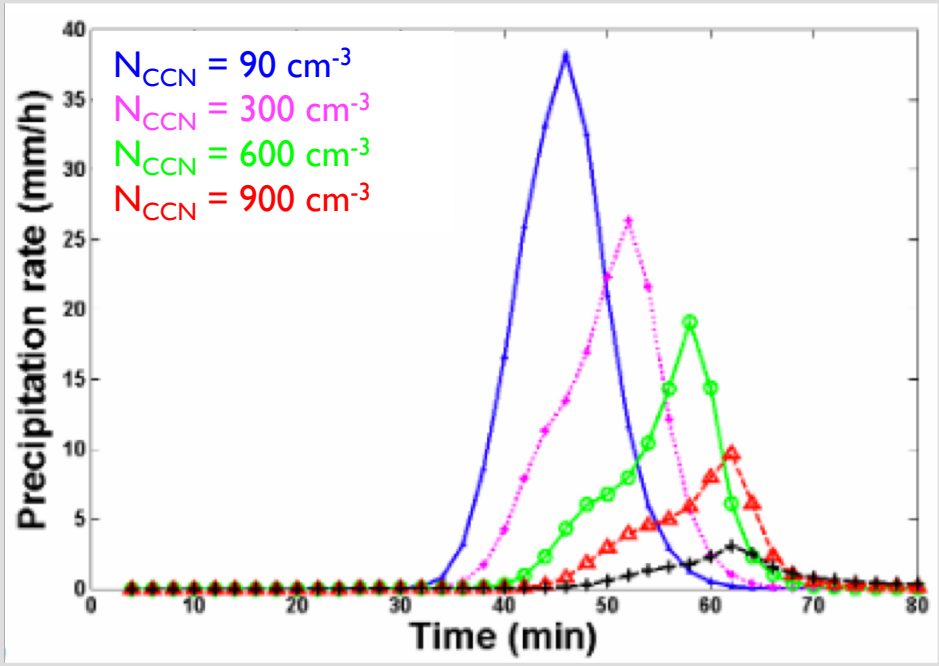


Yang et al. (2011) *Atmos. Chem. Phys.*

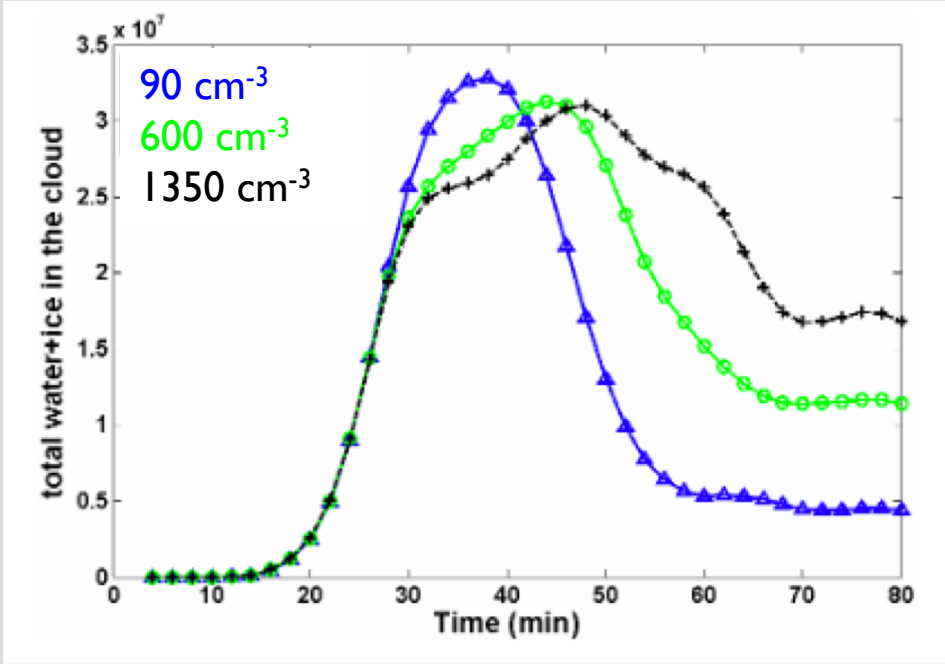
CONVECTIVE CLOUDS

Detailed cloud physics calculations for < 2 hour simulation

Precipitation rate as a function of time



Total condensed water



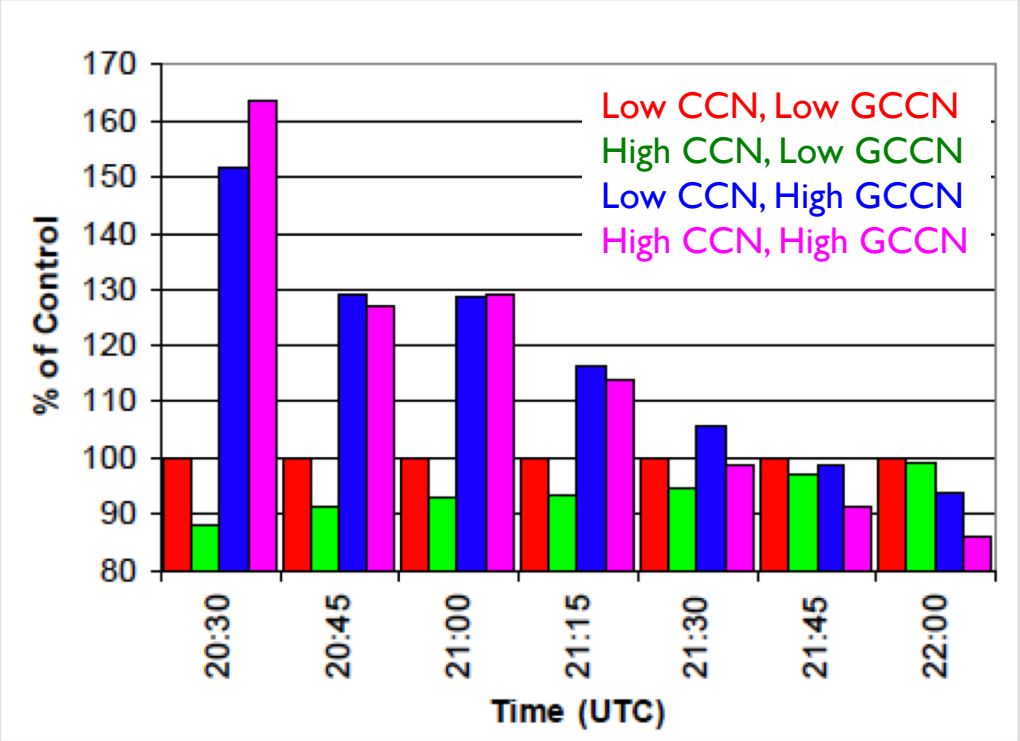
- Polluted clouds produce less precipitation, initiation of precipitation is delayed and lifetime of clouds is longer
- More water vapor transported to mid troposphere in polluted conditions

Teller and Levin (2006) *Atmos. Chem. Phys.*

CONVECTIVE CLOUDS

Detailed cloud physics calculations for several hour simulation

Percent Change in Total Precipitation



Change in CCN and Giant CCN concentrations sometimes lead to less precipitation sometimes more, and it changes with time

- Variations in aerosol concentration affect both physical and dynamical characteristics of storms
- Venting of aerosols actually cleans lower atmosphere consequently changing inflow aerosol concentrations
- Cold pools differ substantially between simulations altering storm dynamics

Van den Heever et al. (2006) *J. Atmos. Sci.*

Van den Heever and Cotton (2007) *J. Appl. Meteor.*

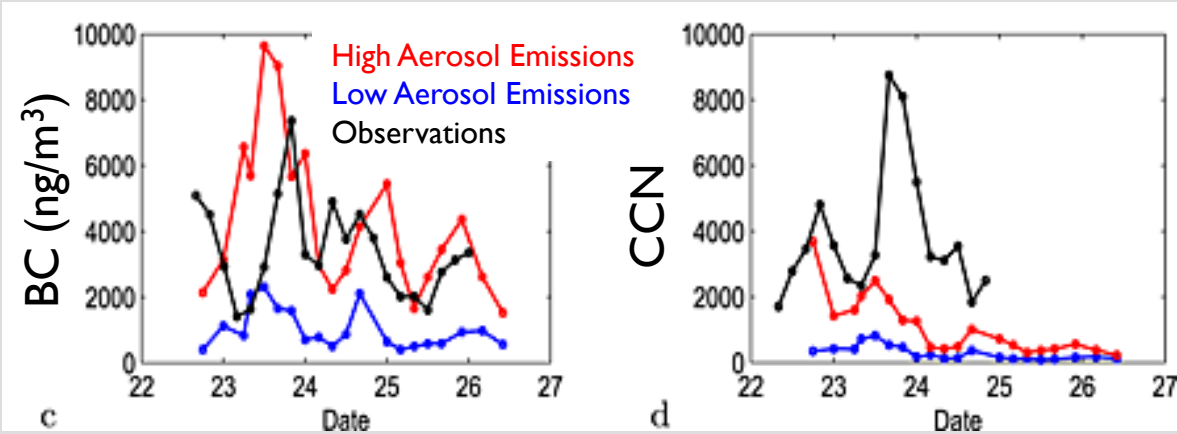
CONVECTIVE CLOUDS

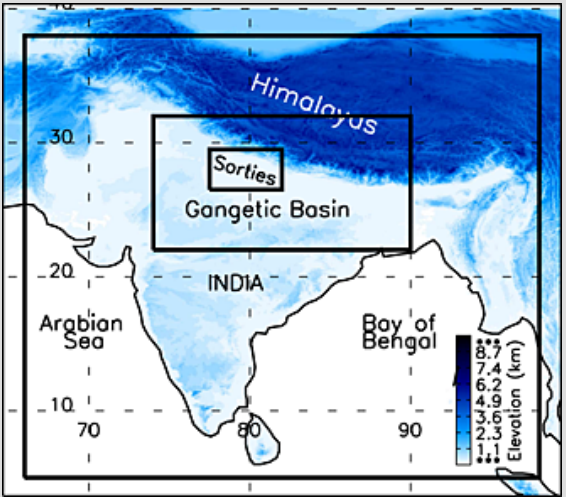
Aerosol-Cloud Interactions for a case in Gangetic Basin

Sarangi et al. (2015) *J. Geophys. Res.* allowed aerosols to affect both cloud physics and radiation

Aerosol anthropogenic emissions altered to evaluate changes

Aerosol concentration as f(date in August)





<u>Increasing Aerosol Concentrations via emissions:</u>

BC aerosol in PBL is absorbing radiation, heating PBL

→ Increase temperature and CAPE

→ Form more, smaller cloud drops near cloud base

→ Increase updraft velocities below the freezing level

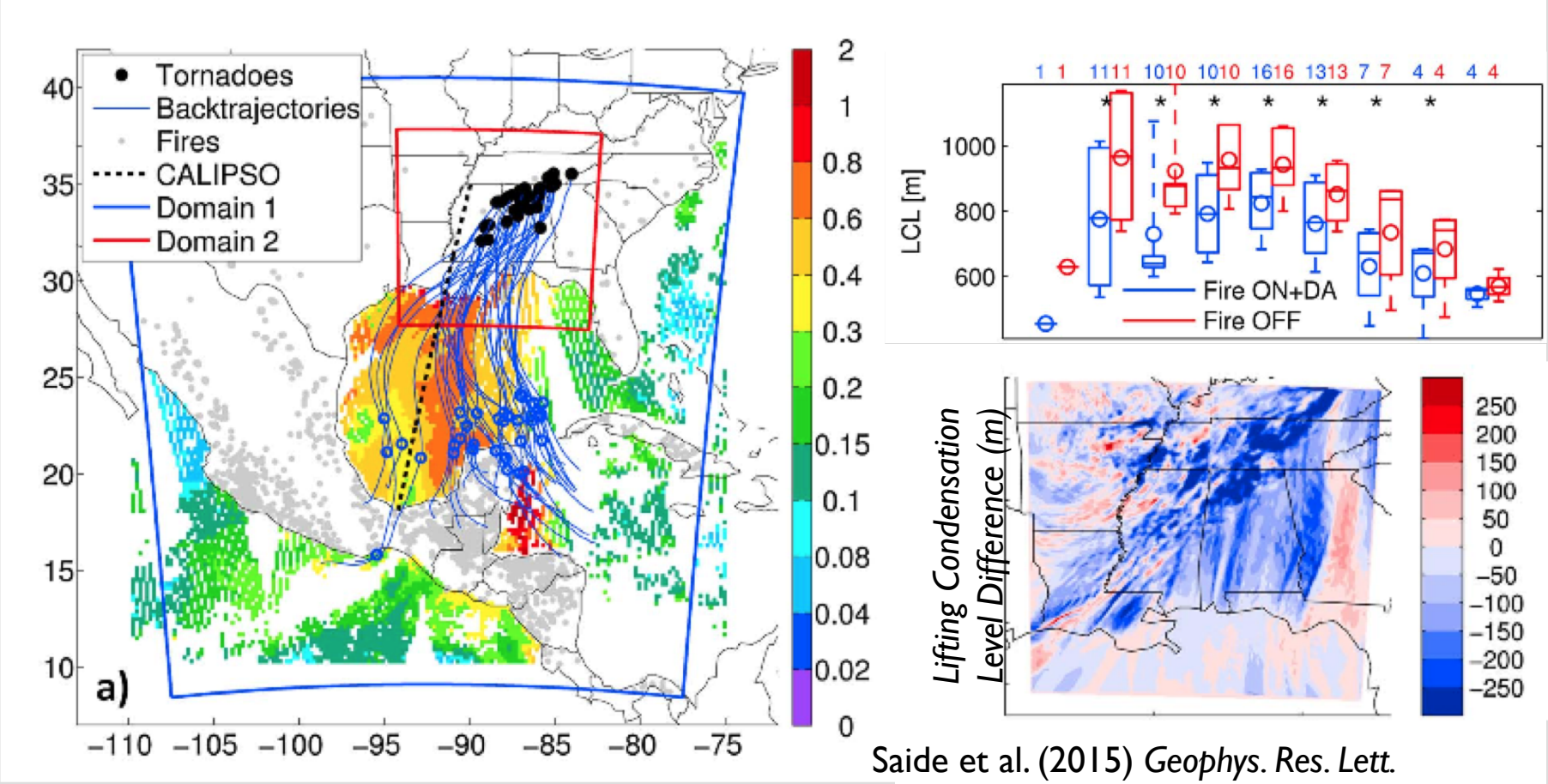
→ Increase cloud top height

→ Aerosol-induced cloud invigoration

Although aerosols were removed by precipitation during the first day (August 23), they were quickly replaced by the aerosol emissions

CONVECTIVE CLOUDS

Aerosols Affect Severe Storm Environment



Saide et al. (2015) *Geophys. Res. Lett.*

- Inclusion of smoke to an environment already conducive to severe thunderstorm development can increase the likelihood of significant tornado occurrence

PROCESSES THAT AFFECT NUMBER OF CCN AND IN

Ice Nuclei (i.e. dust concentrations)

Emissions, transport, dry deposition, wet deposition

- Coagulation or condensation of hygroscopic aerosols and HNO_3 makes the dust more hygroscopic and more likely to be removed by precipitation.

Cloud Condensation Nuclei (i.e. hygroscopic aerosols)

Emissions, transport, dry deposition, wet deposition

Chemistry: gas-phase, heterogeneous reactions, and aqueous-phase chemistry

Lightning-NO_x production

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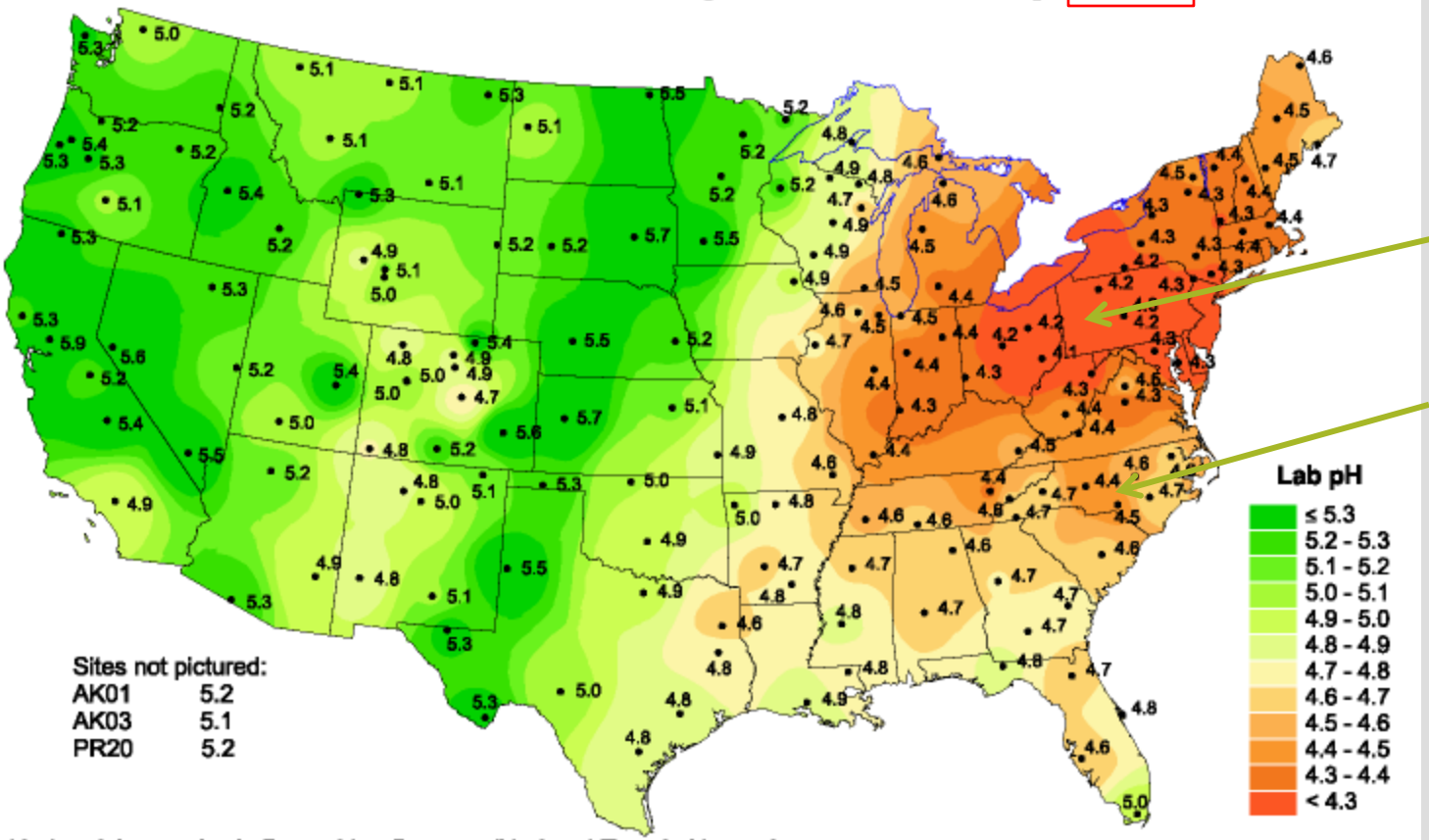
Lightning-NO_x production

EFFECTS OF ACID RAIN

Aqueous chemistry, SO_2 (aq) → sulfate, creates most of the acidity in the rain

Acidity is measured by the pH

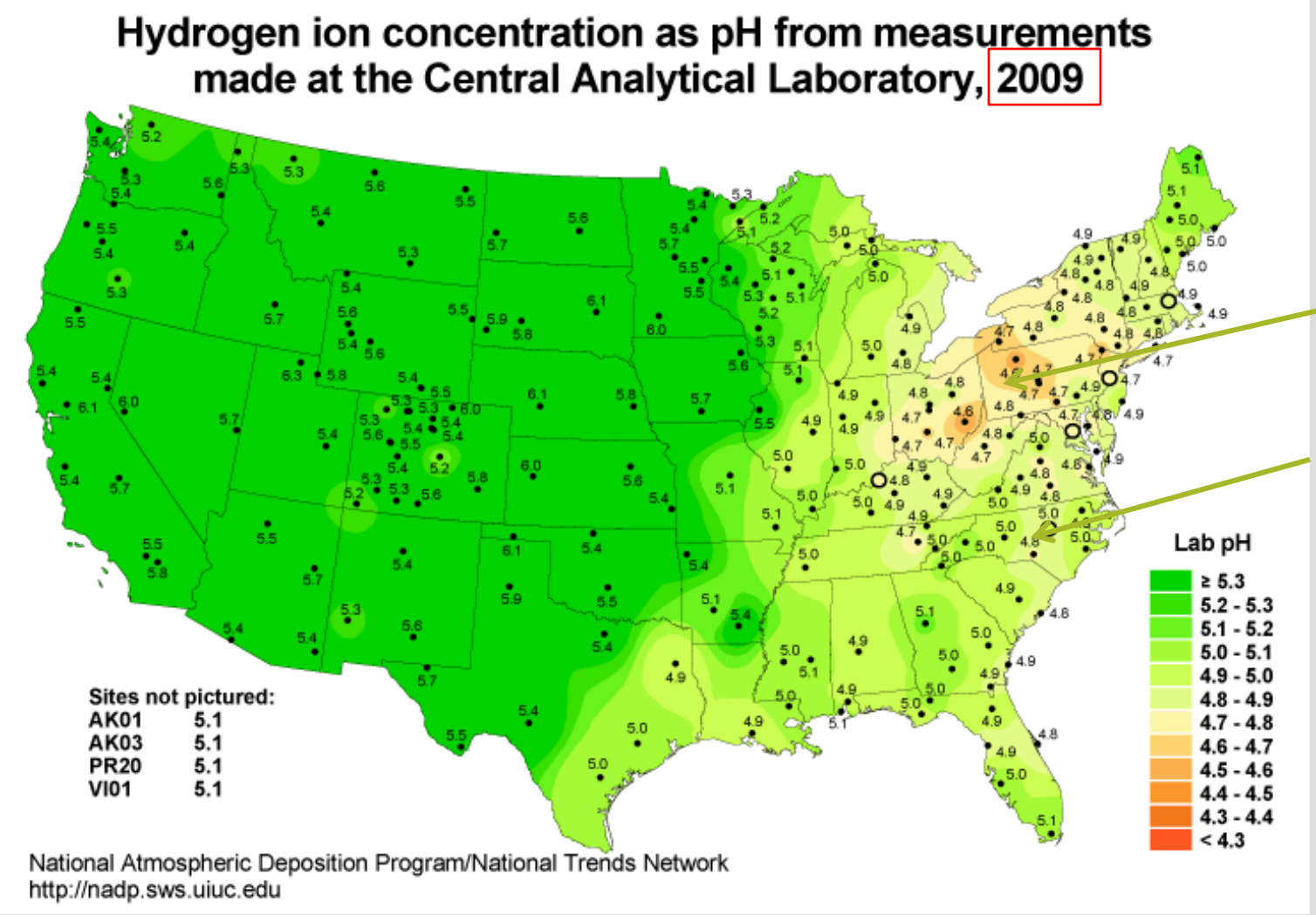
PH VALUES ACROSS U.S. IN 1994



pH=4.2

pH=4.5

PH VALUES ACROSS U.S. IN 2009

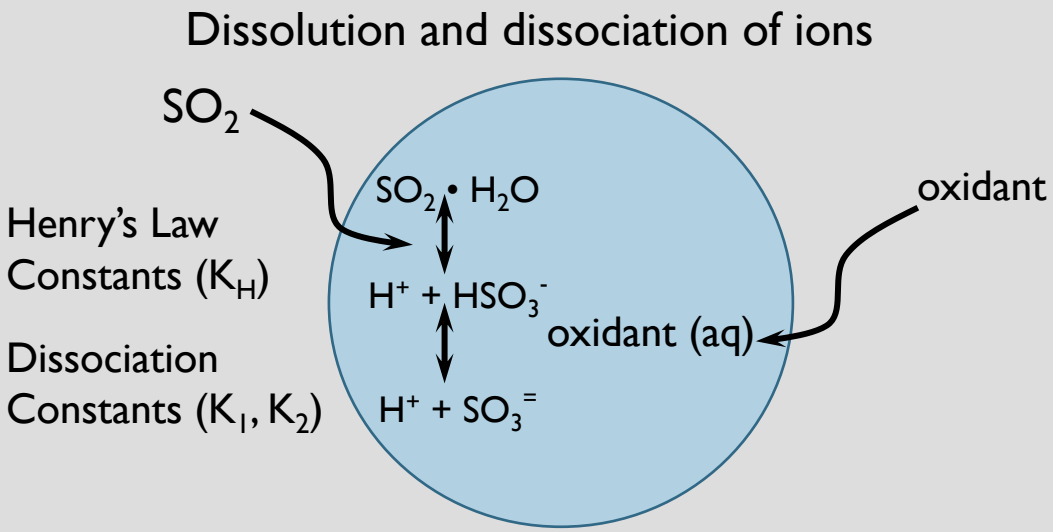


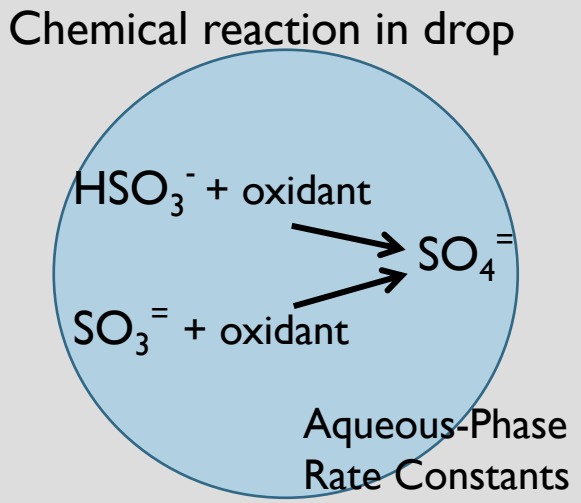
pH=4.6

pH=5.0

AQUEOUS PHASE CHEMISTRY

Sulfur Chemistry





- Dissociation of SO_2 makes SO_2 a soluble trace gas
- Effective Henry's Law for SO_2 depends on T and pH = $-\log_{10}[H^+]$

Key oxidants of aqueous-phase SO_2:

H_2O_2 at pH < 5, **O_3** at pH > 5

O_2 catalyzed by **transition metals** (Fe^{3+}, Mn^{2+})

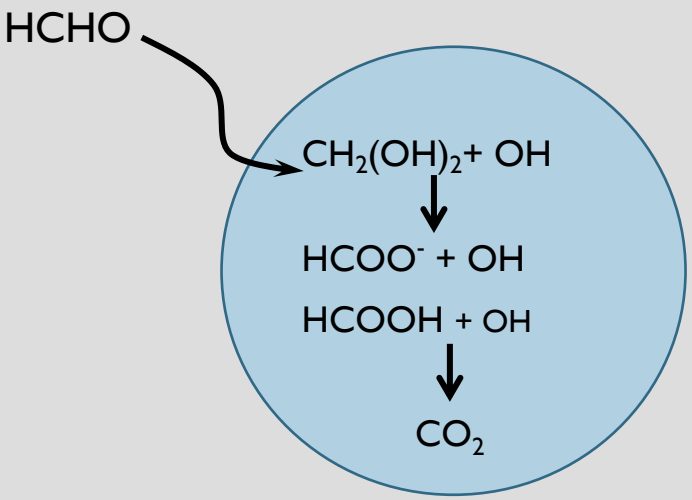
NO_2 at high pH and when NO_2 > 20 ppbv (fog in China)

HOBr

AQUEOUS PHASE CHEMISTRY

Organic Chemistry

Oxidation of Aldehydes by OH → organic acids



Models that include aqueous chemistry:

WRF-Chem (primarily SO_4 formation)

CAM-Chem (primarily SO_4 formation)

GEOS-Chem (SO_4 and organic acid formation)

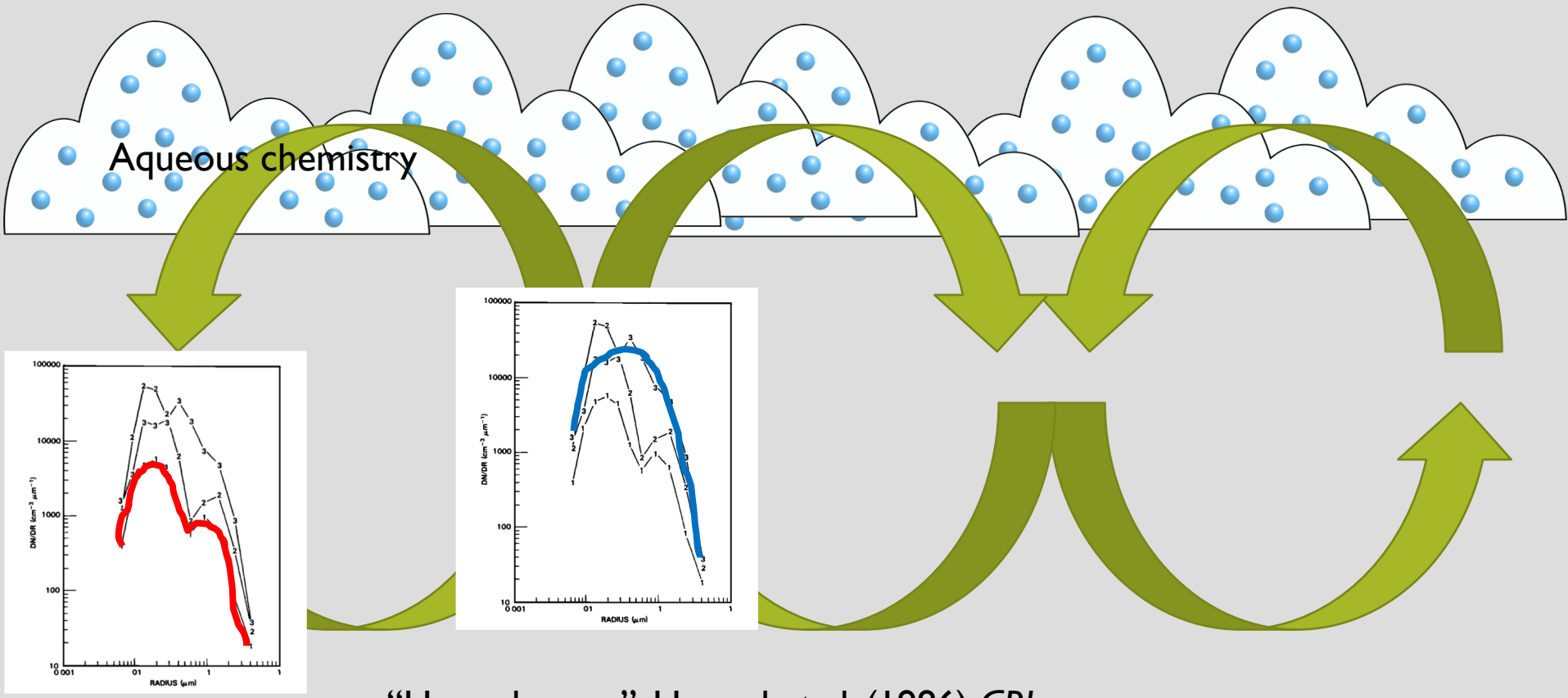
WRF-CMAQ (SO_4 and organic acid formation)

→ sulfate aerosols, SOA

- Other aldehydes (glyoxal, methyl glyoxal) undergo aqueous-phase oxidation too
- Form carboxylic and dicarboxylic acids
- Dicarboxylic acids → secondary organic acid production

Aqueous chemistry increases aerosol mass concentrations that can affect CCN concentrations and radiation

AQUEOUS PHASE CHEMISTRY



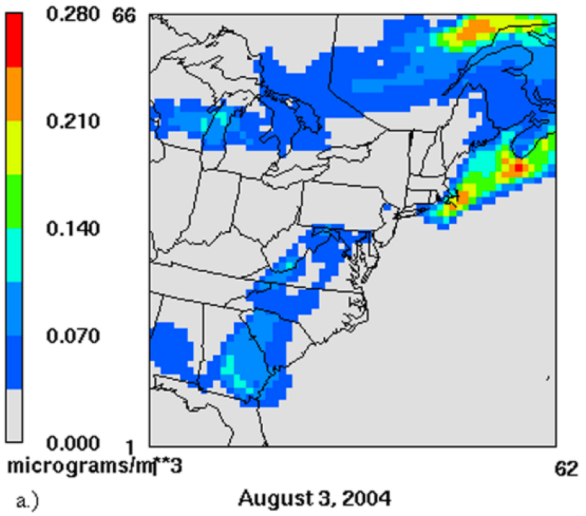
"Hoppel curve": Hoppel et al. (1986) *GRL*

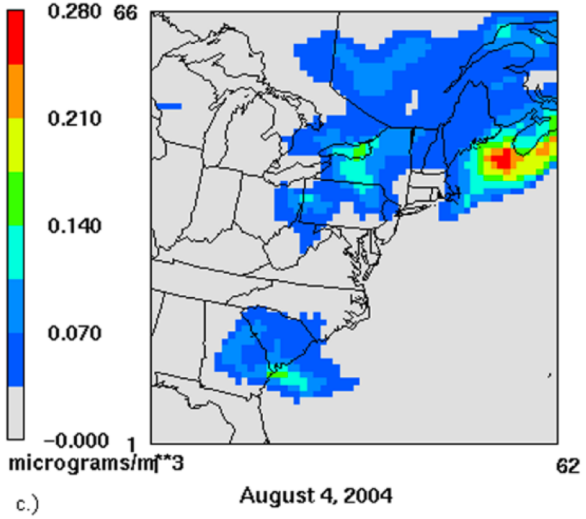
MODELING AQUEOUS PHASE CHEMISTRY

Methods vary among models

- Solve gas and aqueous chemistry together (or sequentially, e.g. WRF-Chem)
- Prescribe or diagnose **pH** (GEOS-Chem, WRF-CMAQ, WRF-Chem)
- Cloud physics prediction of **liquid water content** affects results
- Size of drops affects diffusion of gases into cloud drops

ORGANIC AQUEOUS CHEMISTRY



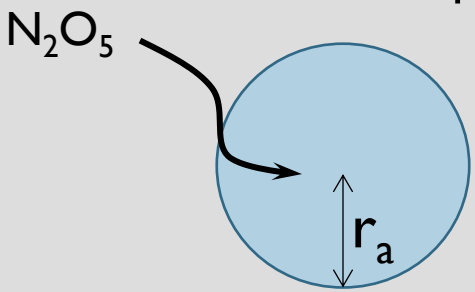


Chen et al. (2007) *Atmos. Chem. Phys.*
Show difference in SOA mass concentrations caused by aqueous-phase carbonyl oxidation, using WRF-CMAQ specialized to CACM chemistry and MPMPO.

Fahey et al. (2017) *Geophys. Model Develop.*
Significant advancements in representing organic aqueous chemistry in WRF-CMAQ making use of the Rosenbrock RODAS solver.

HETEROGENEOUS CHEMISTRY

Reaction between two species of different phases



$$N_2O_5(g) + H_2O(l) \rightarrow 2\ HNO_3$$

$$k = S_a/[r_a/D_g + 4/(w\ \gamma)]$$

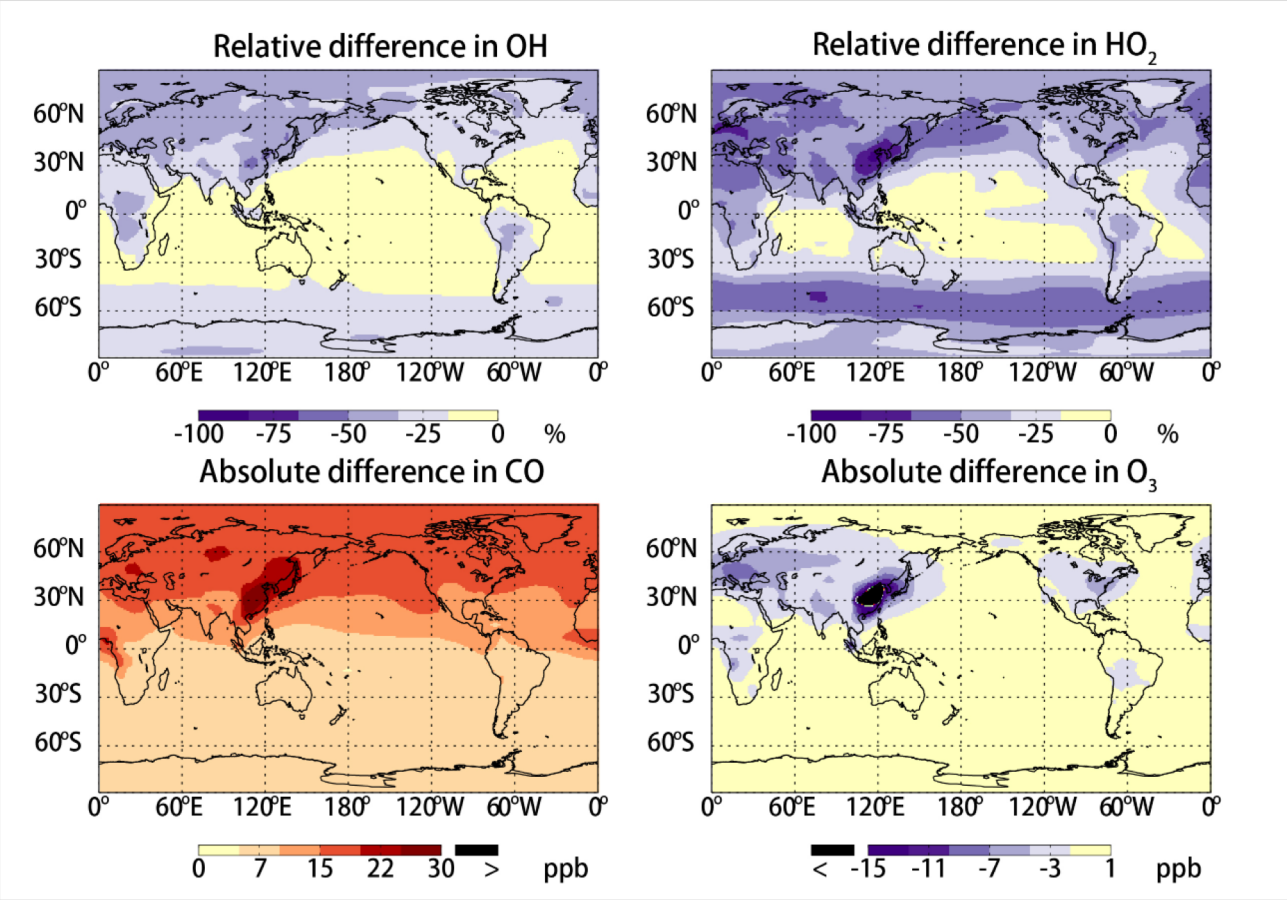
Reaction Rate controlled by surface area (S_a), diffusivity (D_g) into drop and uptake coefficient (γ)

Models include heterogeneous chemistry along with their gas-phase mechanism:

WRF-Chem T1 MOZCART (13 heterogeneous reactions)
CAM-Chem (4 tropospheric heterogeneous reactions)
GEOS-Chem (several tropospheric heterogeneous reactions)
WRF-CMAQ (several tropospheric heterogeneous reactions)

Predicted S_a and prescribed γ can vary among models

HETEROGENEOUS CHEMISTRY



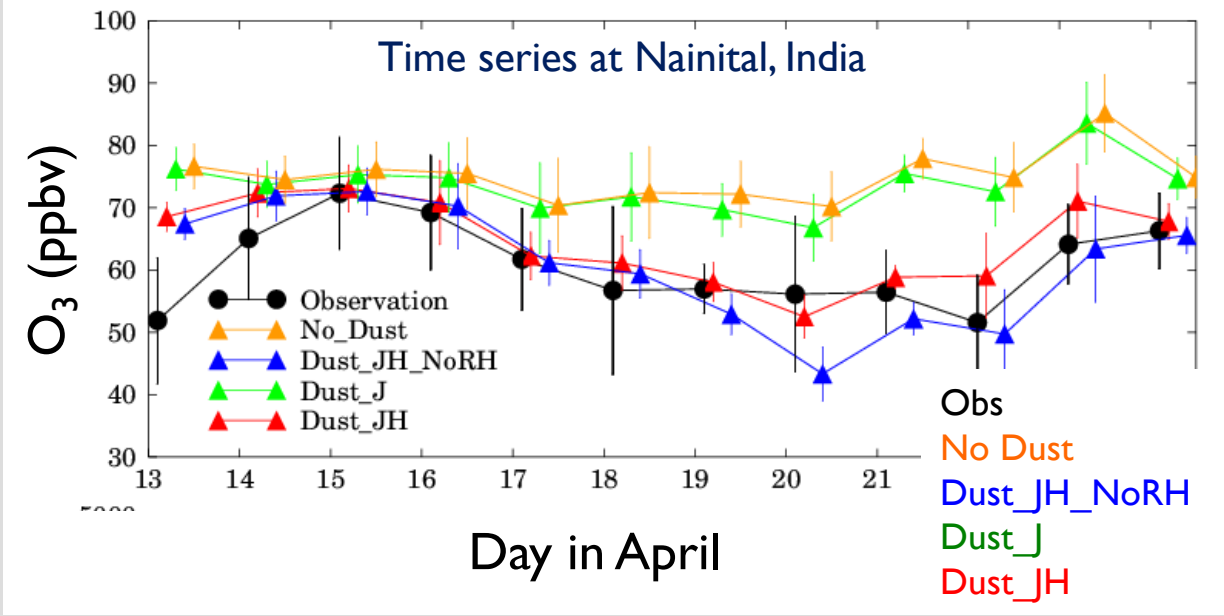
Mao et al. (2013) *Atmos. Chem. Phys.*
Show large effect (10 ppbv) on O_3 caused by HO_2 uptake onto aerosols catalyzed by transition metal ions, using GEOS-Chem.

HETEROGENEOUS CHEMISTRY

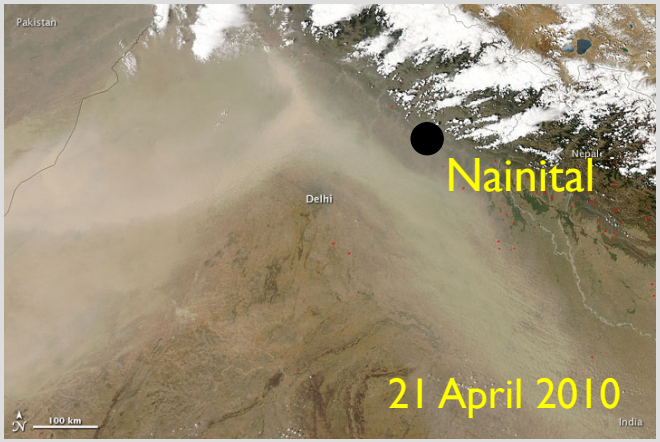
Dust storm over Indo-Gangetic Plain

Kumar et al. (2013) *Atmos. Chem. Phys.* show dust effects on O_3 using WRF-Chem.

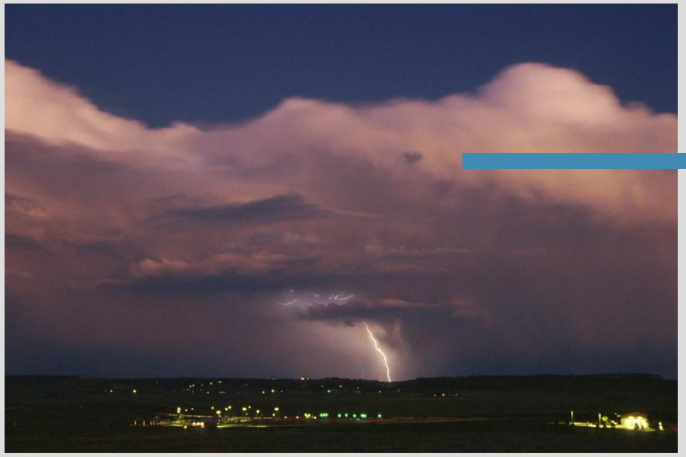
To match measurements, both the effect on photolysis and the heterogeneous reactions are needed.



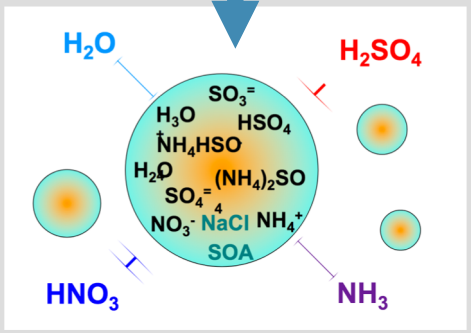
Dust storm over IGP



LIGHTNING-NO_X AFFECTS AEROSOLS



$NO_x = NO + NO_2$ → ($NO_2 + OH$) → HNO_3



- Lightning generates NO which quickly equilibrates with NO_2
- Then complex NO_x chemistry includes production of HNO_3
- HNO_3 adsorbs onto aerosols (aerosol thermodynamics)
- Aerosol concentrations increase which affects radiation and clouds

LIGHTNING FORMATION

Charge Separation

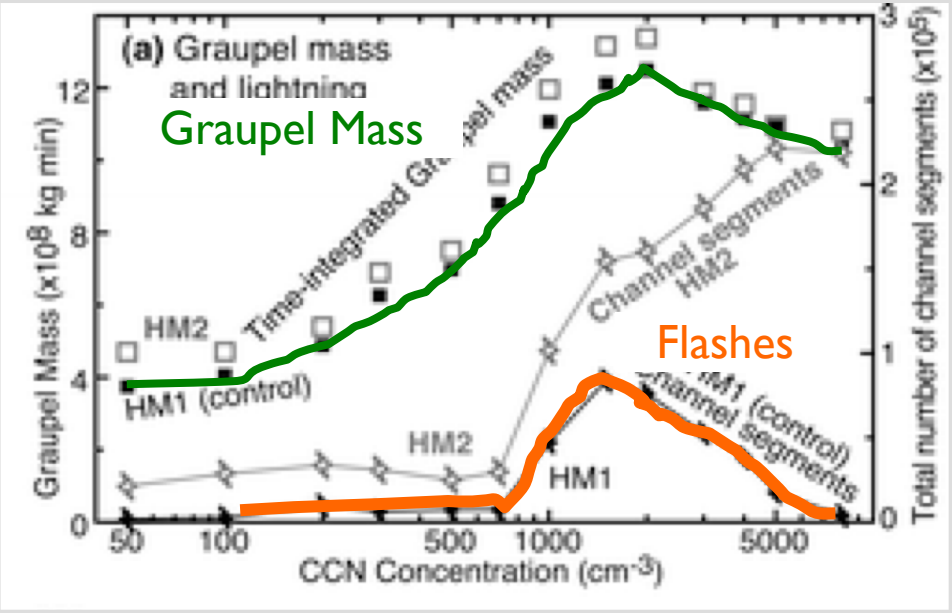
- Side-by-side updrafts and downdrafts
- Updrafts transport cloud droplets towards top of storm
- Downdrafts with falling hail and graupel
- **Graupel – water collisions creating a "soft shell" graupel or hail particle**
- **Further graupel – drop collisions cause electrons to shear off of the ascending water droplets and collect on the falling ice particles**

➢ Charge separation with negative charge in lower cloud and positive charge in upper part of storm

If aerosols affect the cloud physics and dynamics, then aerosols likely affect the lightning flash rate

http://www.srh.weather.gov/srh/jetstream/lightning/lightning.html

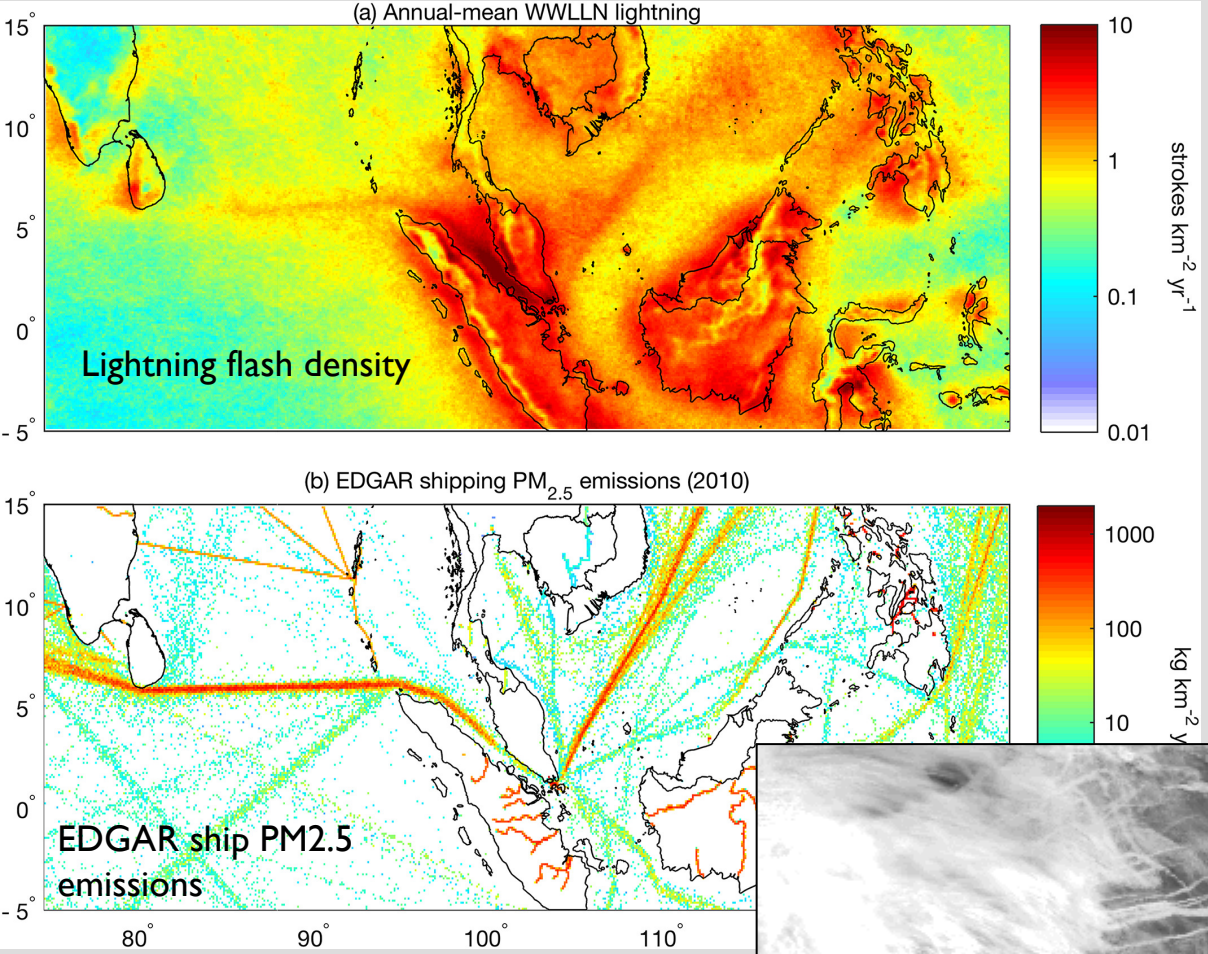
LIGHTNING FLASH RATES CORRELATED WITH UPDRAFT VOLUME, PRECIPITATION ICE MASS



- Cloud resolving model simulation of explicit prediction of charge examined effect of CCN on predicted lightning flash rate
- Graupel mass corresponds with lightning channel segments

Mansell and Ziegler (2013) *J. Atmos. Sci.*

AEROSOLS AFFECT LIGHTNING FLASH RATES

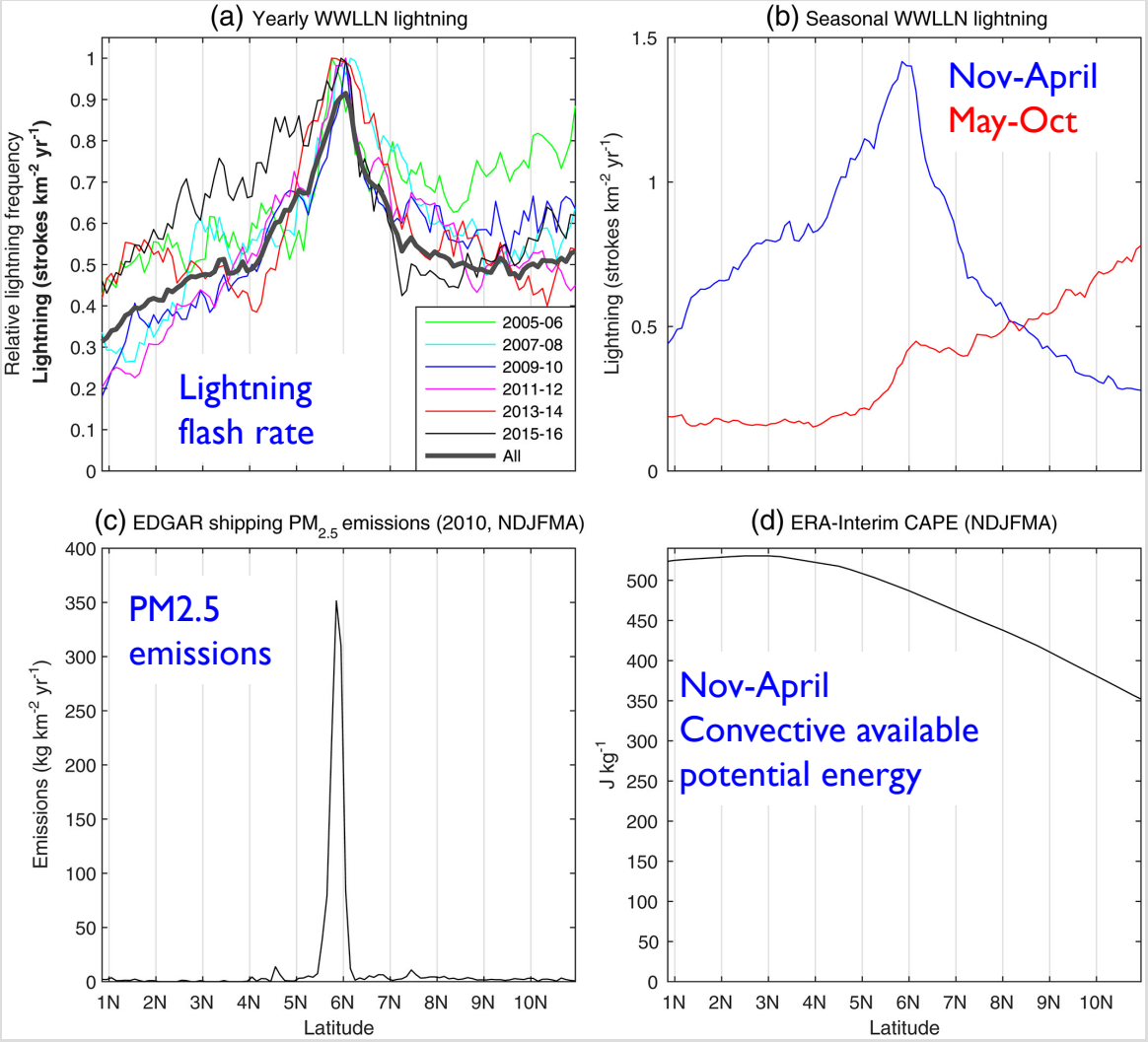


Thornton et al. (2017) *Geophys. Res. Lett.*

Found higher flash rates over shipping lane between Singapore and Sri Lanka

Signature of a ship track

AEROSOLS AFFECT LIGHTNING FLASH RATES



Thornton et al. (2017) *Geophys. Res. Lett.*

Shows that CAPE is not responsible for change in cloud properties and lightning flashes

PM2.5 emissions likely reason for higher flash rates

SUMMARY

- Aerosol – Radiation Interactions
  - AOD measurements versus model prediction from AQMEII activity: Mixing state of aerosols is big uncertainty
- Aerosol – Cloud Interactions
  - Chemistry – Aerosol models use Kohler theory to affect cloud drop activation at *cloud base*
  - Good parameterization for stratus & stratocumulus type clouds
  - Convective clouds are capable of drop activation above cloud base
  - Aerosol effects on ice nucleation is not represented in all models – important for cirrus clouds (less so for convection where other processes dominate)
- Processes that affect number of aerosols
  - Aerosols, clouds, and chemistry are intricately linked

Does it make sense?