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Aerosol Direct and Indirect Effects

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Background

First, A Brief History ...

- ▶ Gas-phase and aerosol models were implemented first in WRF-Chem
- ▶ Then, aerosol-radiation-cloud interactions were coupled to the MOSAIC aerosol model, adapted from those used in a global climate model
- ▶ Aerosol-radiation-cloud interactions have been expanded to handle more aerosol models (GOCART, MADE/SORGAM, MAM) and microphysics schemes (Lin, Morrison, Morrison-Gettelman)
- ▶ More capabilities are being added and tested, making modules more generic, and trying to follow WRF coding guidelines

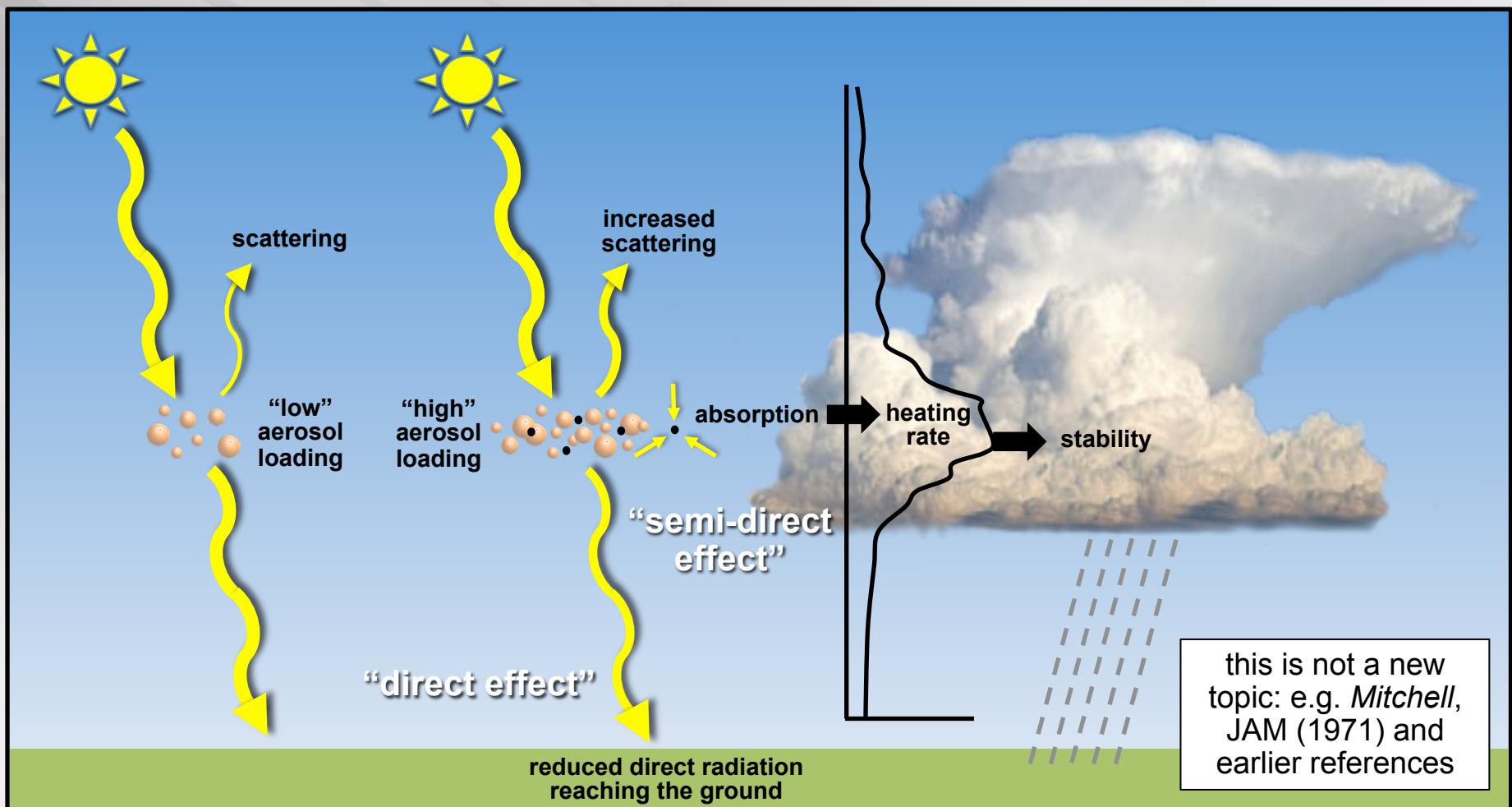
Overall motivation is to use the model to better understand and parameterize local to regional-scale evolution of particulates and their effect on radiation, clouds, and chemistry

Outline:

- ▶ Part 1: Direct Effects
- ▶ Part 2: Indirect Effects



Part 1: Aerosol Direct Effects



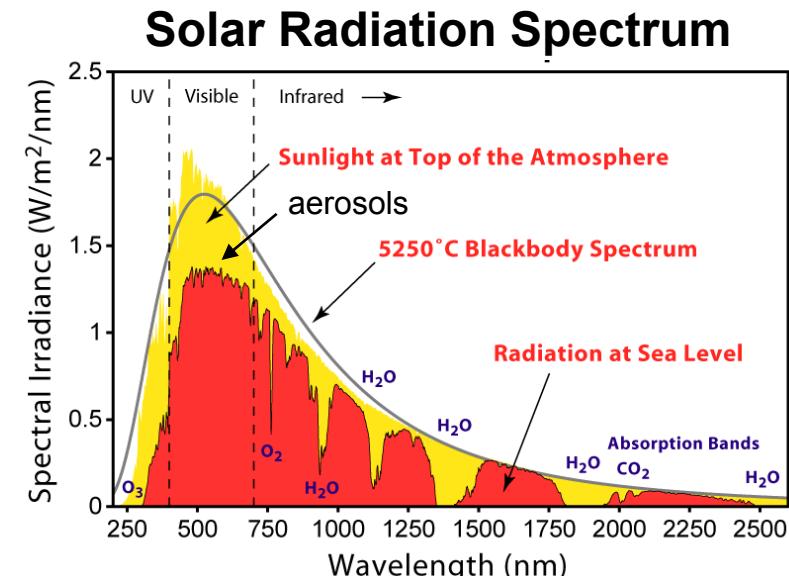
while water vapor, carbon dioxide, ozone, and other trace gases affect the radiation budget, aerosols can also play a role

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



Aerosols in Relation to Radiation Modules

- ▶ Aerosols affect radiation mostly in the visible wavelength region
- ▶ In contrast with water vapor, carbon dioxide, and ozone, the temporal and spatial variability of aerosols is much larger and difficult to simulate
 - **Episodic Sources:** dust, biomass burning, volcanic (potentially large concentrations)
 - **More “Continuous” Sources:** sea-salt, biogenic, anthropogenic (usually smaller concentrations)



How are aerosol effects accounted for in atmospheric models?

- ▶ **Ignored** - no effect of aerosols on radiation
- ▶ Use prescribed or **climatological** aerosol properties – that may vary in space and seasonally (not discussed in this presentation)
- ▶ Use **prognostic aerosols** (e.g. WRF-Chem)

Aerosol Optical Properties:

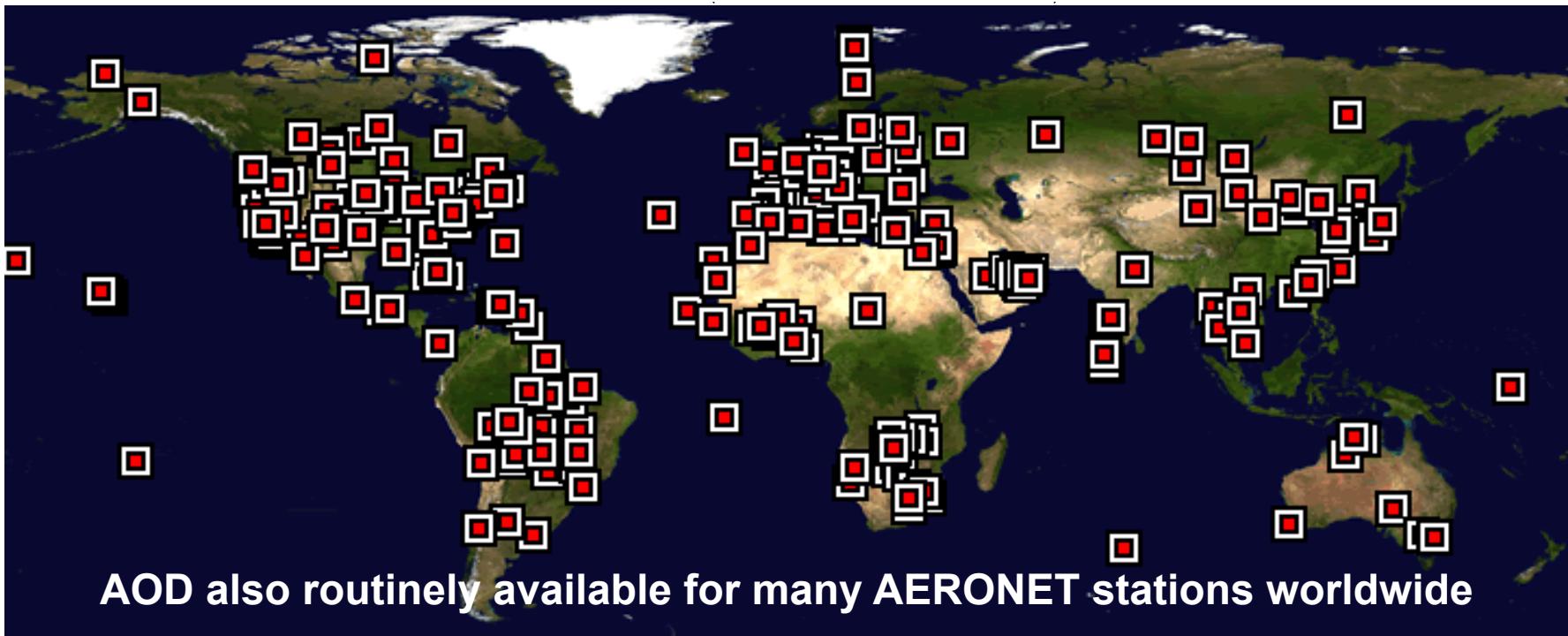
Aerosol Optical Depth, τ_λ



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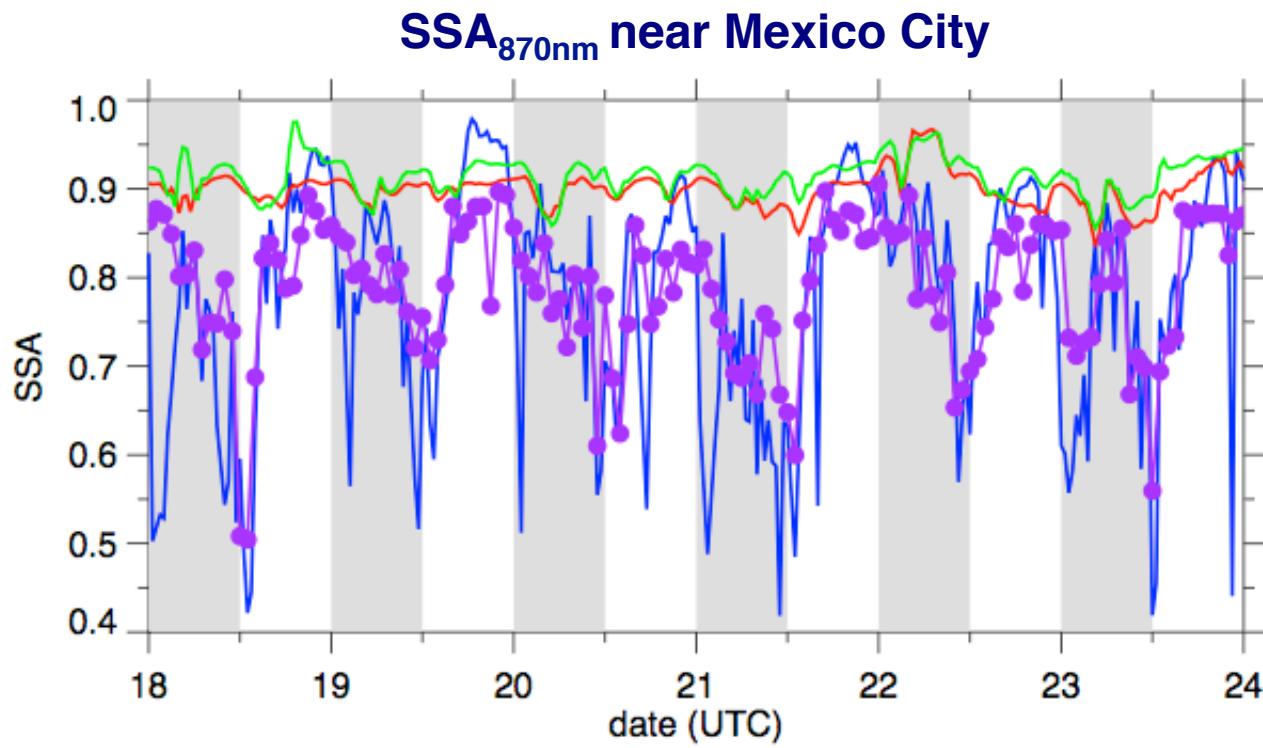
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- ▶ **Extinction coefficient:** fractional depletion of radiance per unit path length (km^{-1}) due to scattering and absorption by aerosols
- ▶ **Aerosol optical depth (AOD) or thickness (AOT):** integrated extinction coefficient over a vertical column, $I / I_0 = e^{-\tau}$
 - AOD = 0 no aerosol effect
 - AOD ~ 1 “large”
 - AOD > 1 extremely high aerosol concentrations

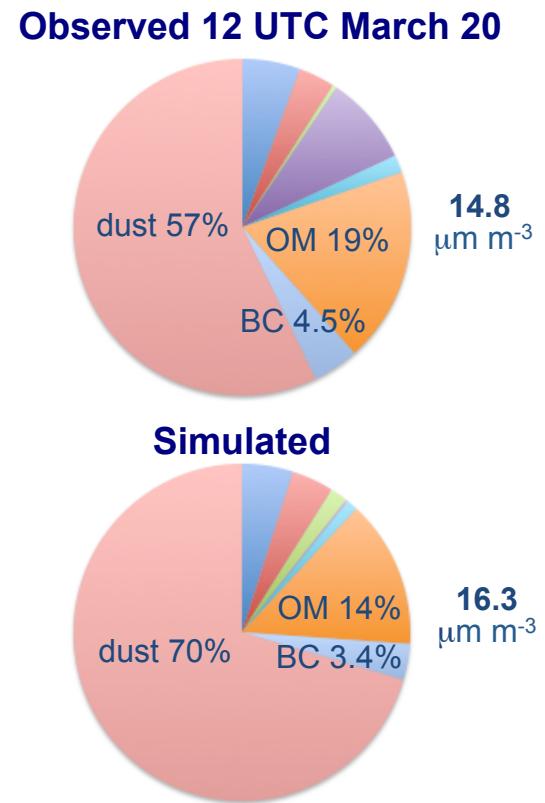


Aerosol Optical Properties: Single Scattering Albedo, ω_o

- ▶ SSA is ratio of scattering efficiency 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)
- ▶ Models simulate τ_λ “reasonably well”, but there are large uncertainties in ω_o



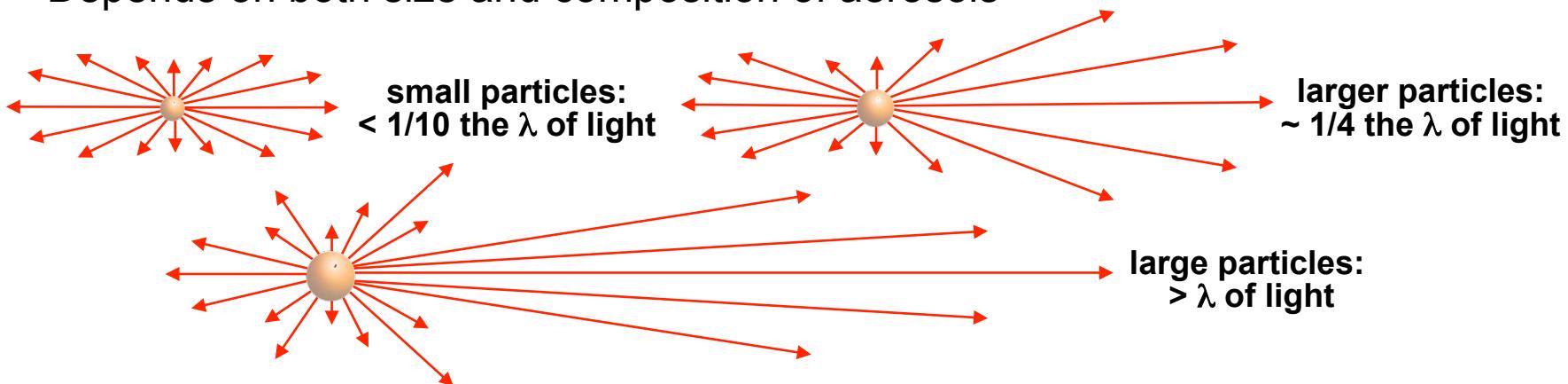
aerosol optical properties driven by measurements



Aerosol Optical Properties:

Asymmetry Factor, g

- ▶ 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 (isotropic scattering – such as from small particles)
- ▶ Depends on both size and composition of aerosols



- ▶ Theoretical relationships used to derive g from measurements





Methodology for Prognostic Aerosols

Generic Aerosol Optical Property Module



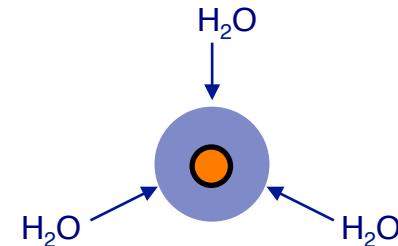
- ▶ τ , ω_o , and g computed at **4 wavelengths (300, 400, 600, and 1000 nm)** for shortwave radiation
- ▶ As of v3.3, τ , ω_o , and g computed at **16 wavelengths** for longwave radiation
- ▶ Compatible with GOCART (MOZCART), MADE/SORGAM, MOSAIC, and MAM aerosol models as of v3.5
- ▶ Compatible with Goddard shortwave scheme and RRTMG shortwave and longwave schemes
- ▶ There have been several papers evaluating simulated optical properties with measurements (in situ, satellite)
- ▶ If garbage is going into the module, then garbage will come out; therefore, evaluation of aerosol number, size, composition is warranted

Angstrom exponent used to convert to wavelengths needed by radiation schemes

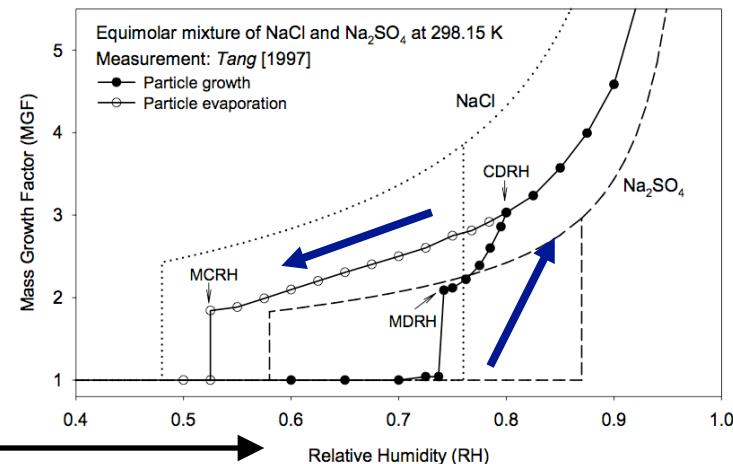


Importance of Aerosol Water

- ▶ Aerosol water will have a big impact on optical properties



- ▶ Aerosol water depends on relative humidity (RH); thus predictions of RH need to be examined when evaluating aerosol direct radiative effects
- ▶ Composition affects water uptake: hydrophobic vs hydrophilic aerosols
- ▶ Aerosols models have different methods of computing aerosol water
 - **GOCART:** Petters and Kreidenweiss (2007)
 - **MADE/SORGAM:** diagnosed
 - **MOSAIC:** prognostic specie that accounts for hysteresis effect
 - **MAM:** prognostic specie, Kohler theory





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

Default Values for SW Radiation in WRF (users can change)

	<u>real part</u>	<u>imaginary part</u>
--	------------------	-----------------------

BC = 1.850 + 0.71i (all λ)

OM = 1.450 + 0.00i (all λ)

SO₄ = 1.468 + 1.0e-9i (300 nm), small λ dependence

NH₄NO₃ = 1.500 + 0.00i (all λ)

NaCl = 1.510 + 0.866e-6i (300 nm), small λ dependence

dust = 1.550 + 0.003i (all λ), depends on type of dust

H₂O = 1.350 + 1.52e-8i (300 nm), small λ dependence

→ greater the # → more absorption

similar
relationships for
LW radiation

- On-going research:
 - secondary organic aerosols (SOA) may be absorbing at near-UV range
 - how to handle “brown carbon”



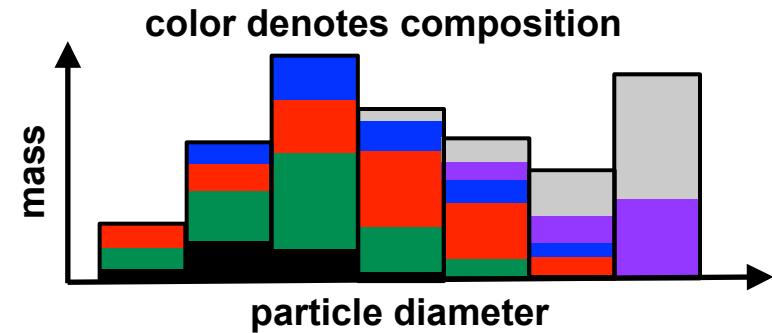
Mixing Rules for Mie Calculations

Prior to the Mie calculations, refractive indices need to be averaged among the compositions in some way for discrete size ranges of the aerosol size distribution.

All particles within a size range assumed to have the same composition, although relative fraction can differ among size ranges.

Currently three choices in WRF:

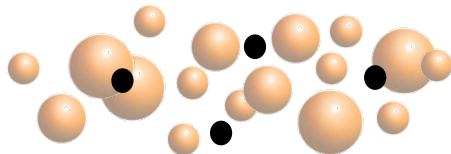
- ▶ **Volume Averaging:** averaging of refractive indices based on composition
- ▶ **Shell-Core:** black carbon core and average of other compositions in shell (Ackermann and Toon, 1983; Borhren and Huffman, 1983)
- ▶ **Maxwell-Garnett:** small spherical randomly distributed black carbon cores in particle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



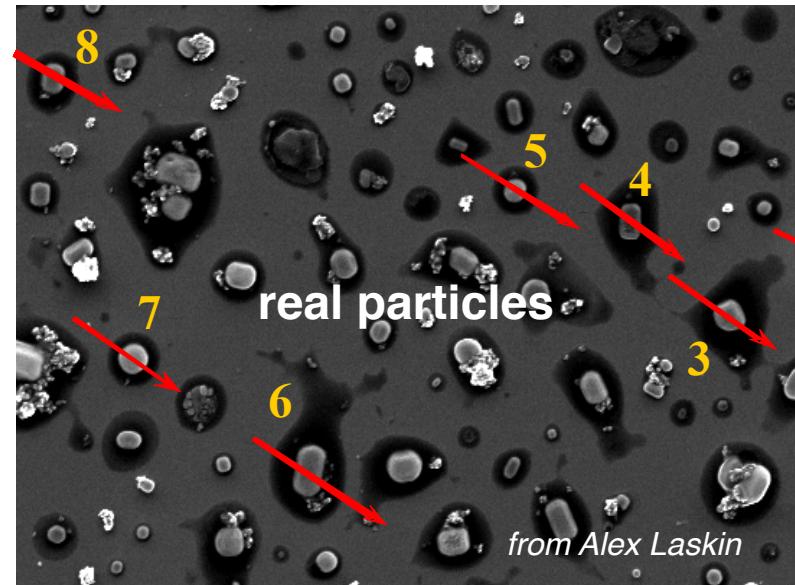


Mie Calculations

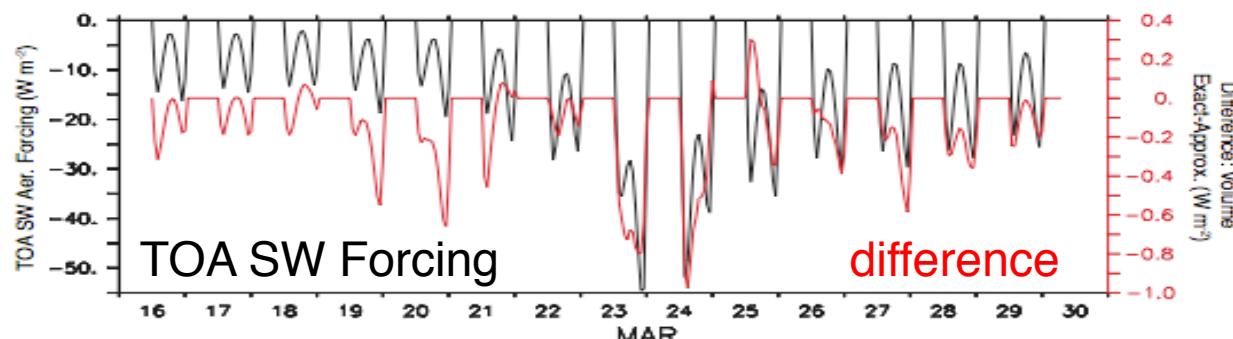
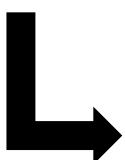
- The Mie solution to Maxwell's equations describes the scattering of radiation by a sphere, used to obtain τ_λ , ω_0 , and g



- Aerosols are rarely spheres; however, aged aerosols become more “sphere-like”
- Several “standard” codes available and one is included in WRF
- Mie codes can be computationally expensive, so an approximate version (*Ghan et al. JGR, 2001*) is also available



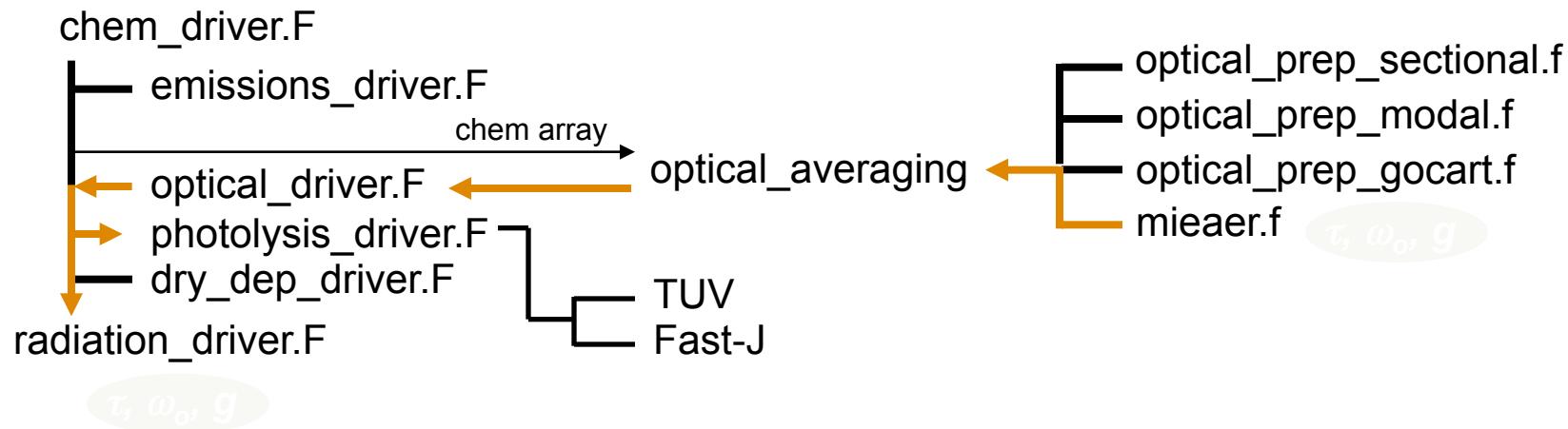
other codes available to handle more complex morphology, but not clear if it is really necessary





Coding Structure

Generic Aerosol Optical Properties Module for WRF-Chem

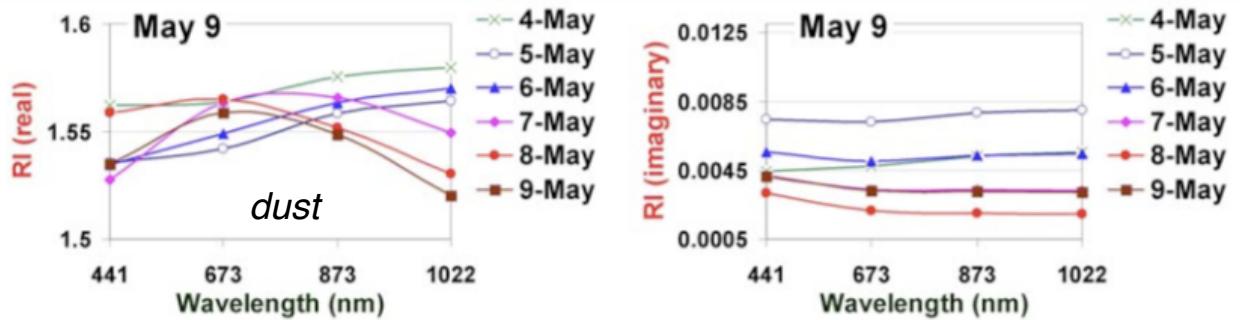
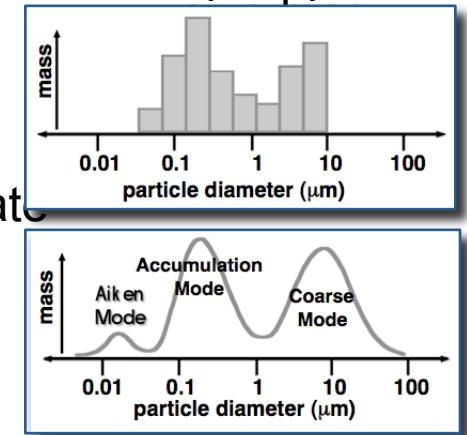


Example of making the code more generic and interoperable – want to have optical property computations in one place rather than in each aerosol model



Assumptions of Optical Property Module

- ▶ Interfaces with GOCART (MOZCART), MADE/SORGAM, MAM, and MOSAIC, but linking to other aerosol models should be relatively easy
- ▶ **Sectional** (MOSAIC): tested only with 4 and 8 should work if additional size bins are specified
- ▶ **Modal** (MADE/SORGAM, MAM): divides mass in modes sections, could divide into more sections to be more accurate
- ▶ **Bulk** (GOCART): converts bulk mass into assumed distribution, then divides mass into 8 sections
- ▶ Note: Refractive indices may need updating
 - Range of values reported in the literature
 - Wavelength dependence of refractive indices for some species

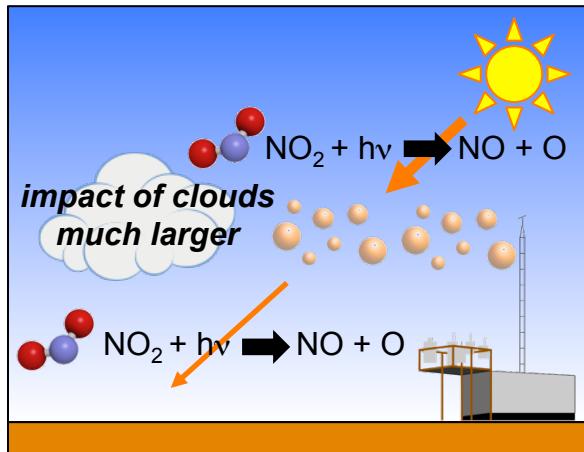


from Prasad and Singh, JGR, 2007

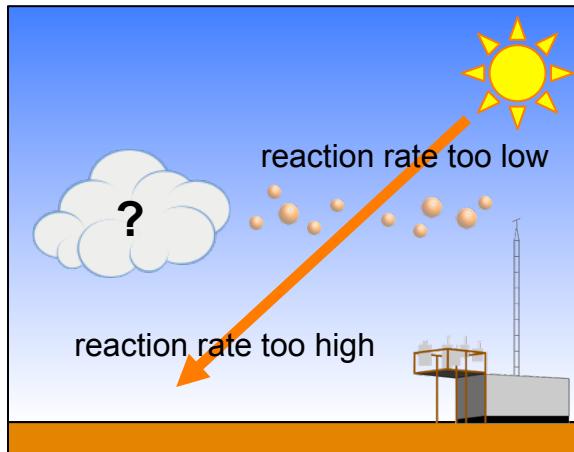
Dust refractive indices for SW constant by default – need to modify code to turn on

Impact of Aerosols on Chemistry

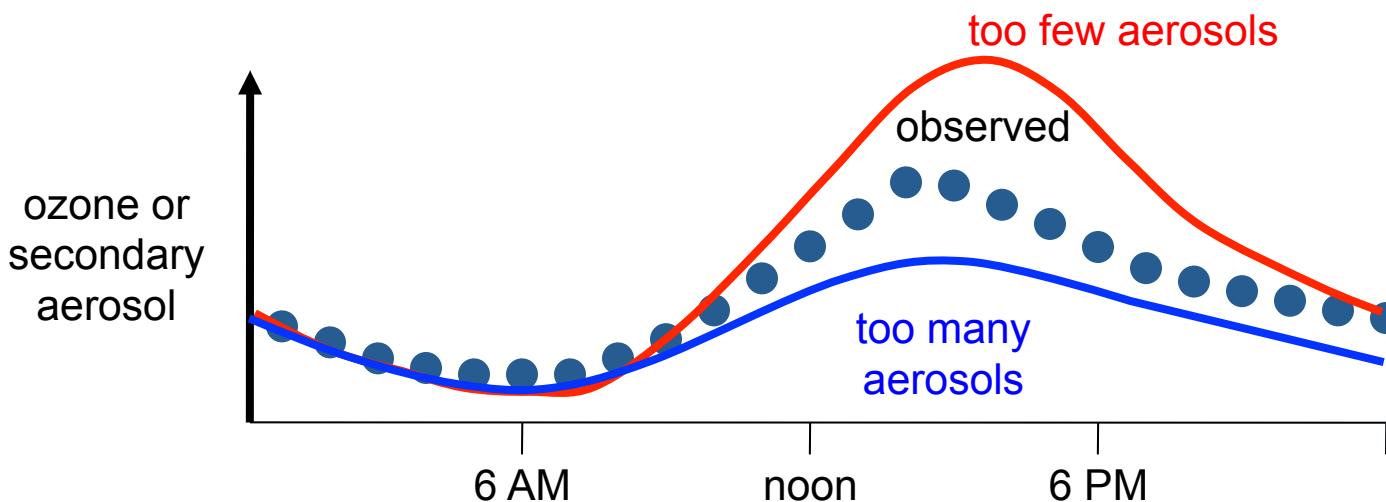
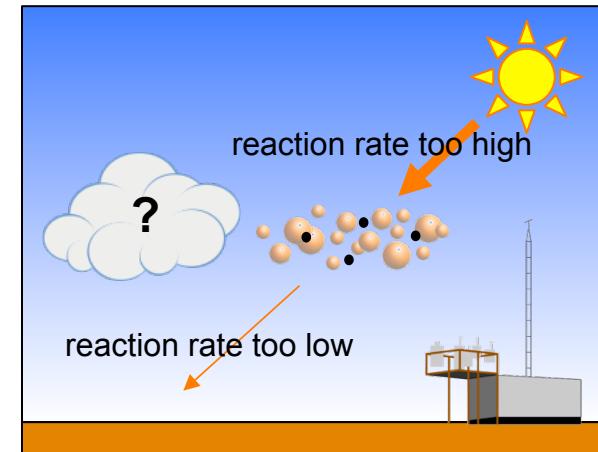
Observed Aerosols



Simulated: Too Few or Too Thin



Simulated: Too Many or Too Thick

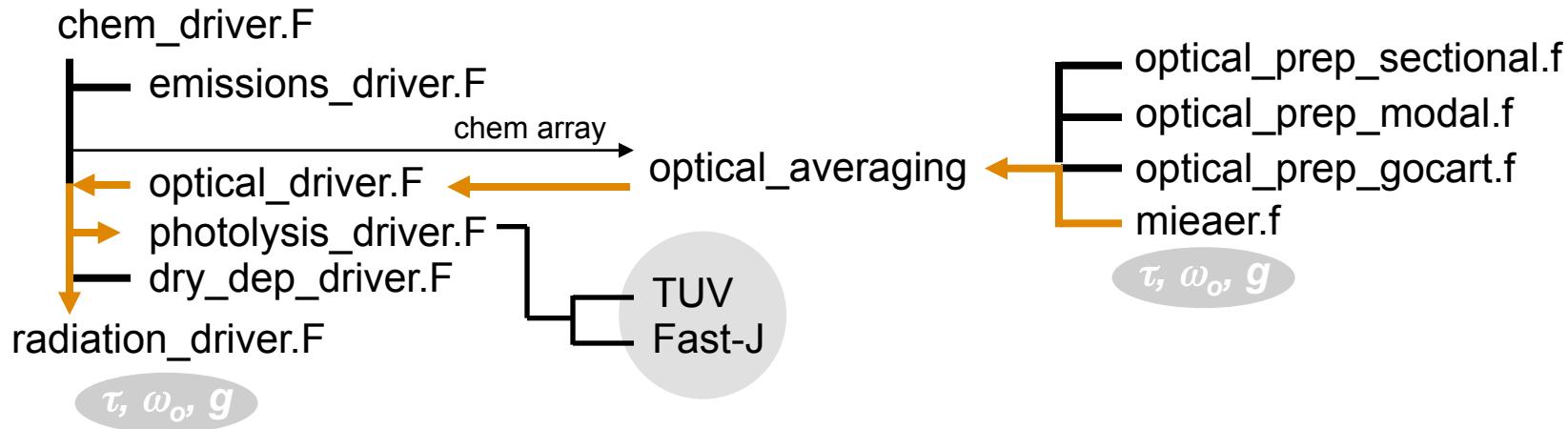




How Aerosols Affect Photolysis Rates

Aerosols → Photolysis Rates → Photochemistry

but clouds (if present) will have a bigger impact on photolysis rates than aerosols



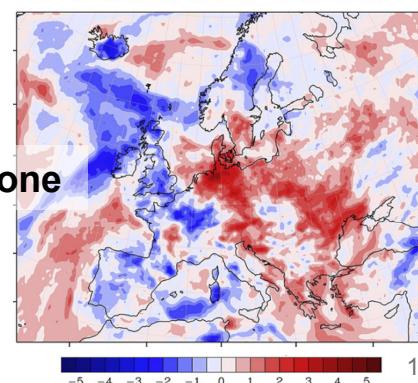
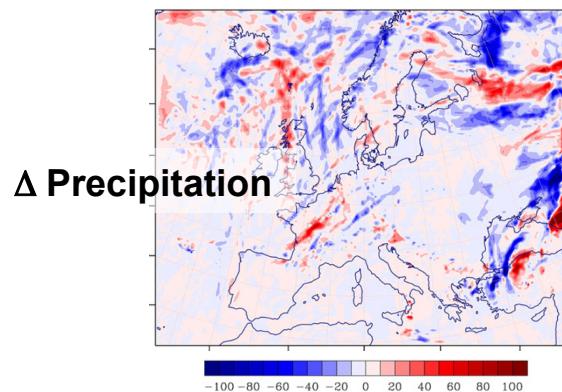
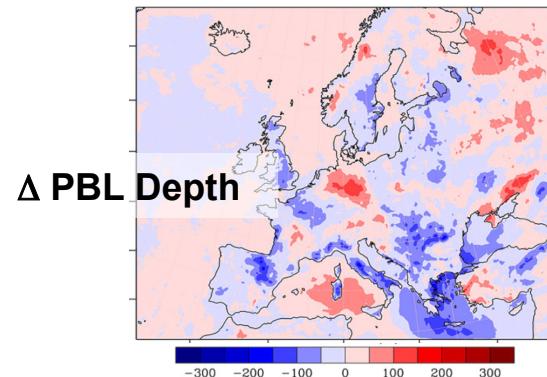
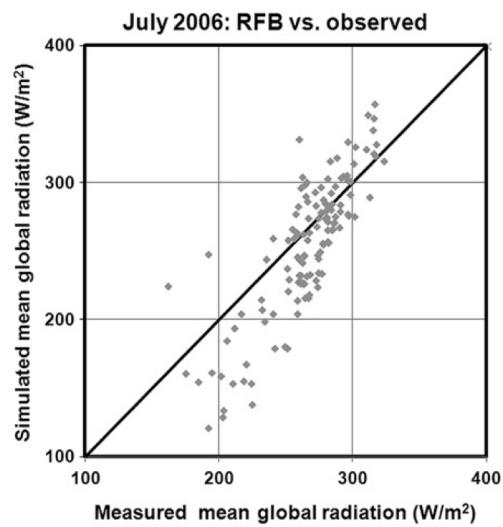
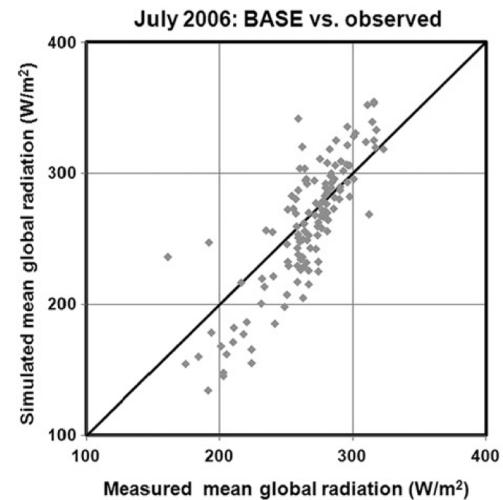
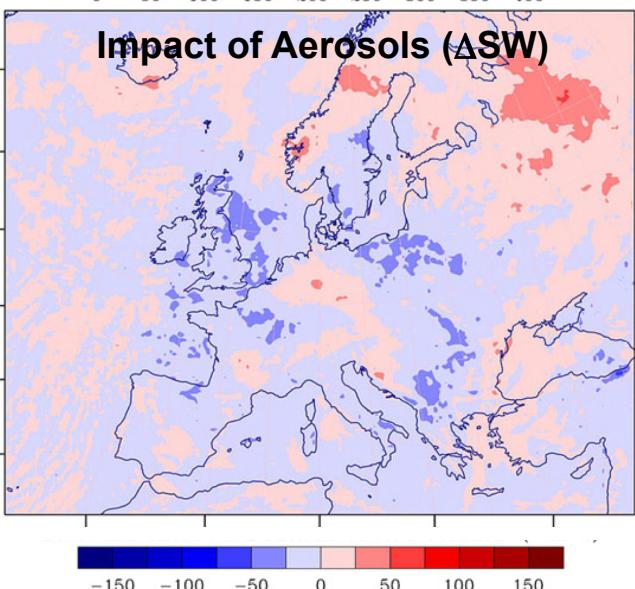
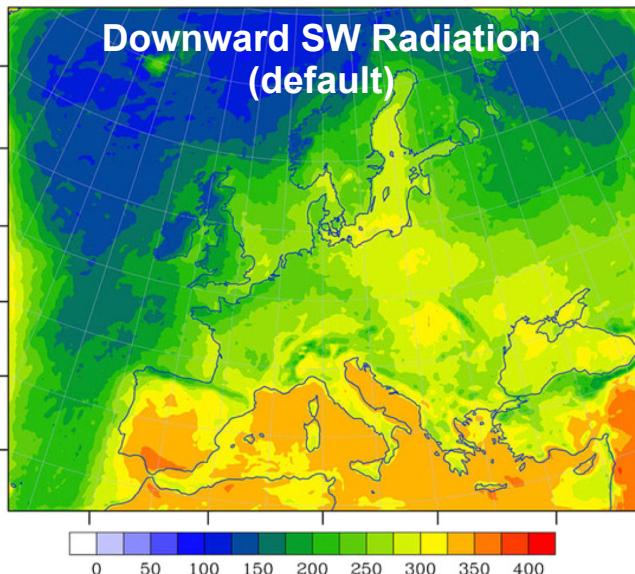
- ▶ Fast-J: uses τ , ω_o , and g computed by module_optical_averaging.F
 - Note: *limited testing of effect of aerosols on photolysis rates*
- ▶ FTUV: uses its own method of accounting for effects of aerosols on photolysis rates based on MADE/SORGAM species only
 - *other aerosol models will not affect photolysis rates when FTUV is used*



“fixing” this is not trivial – has not been a high priority

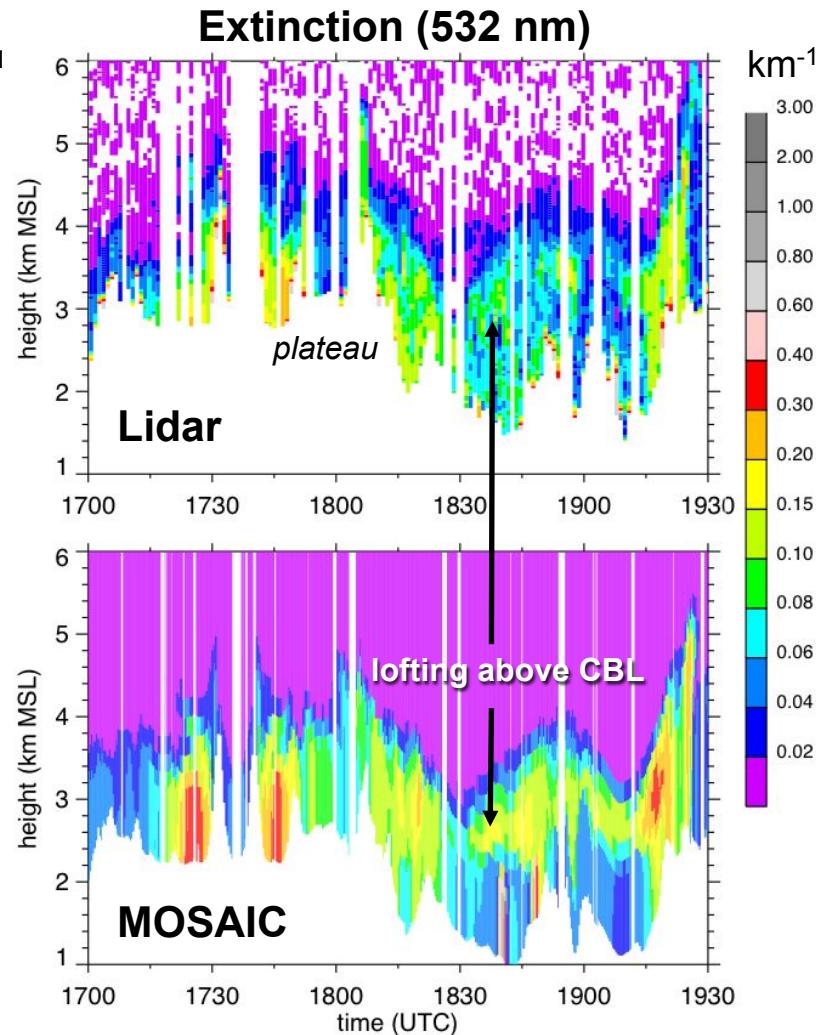
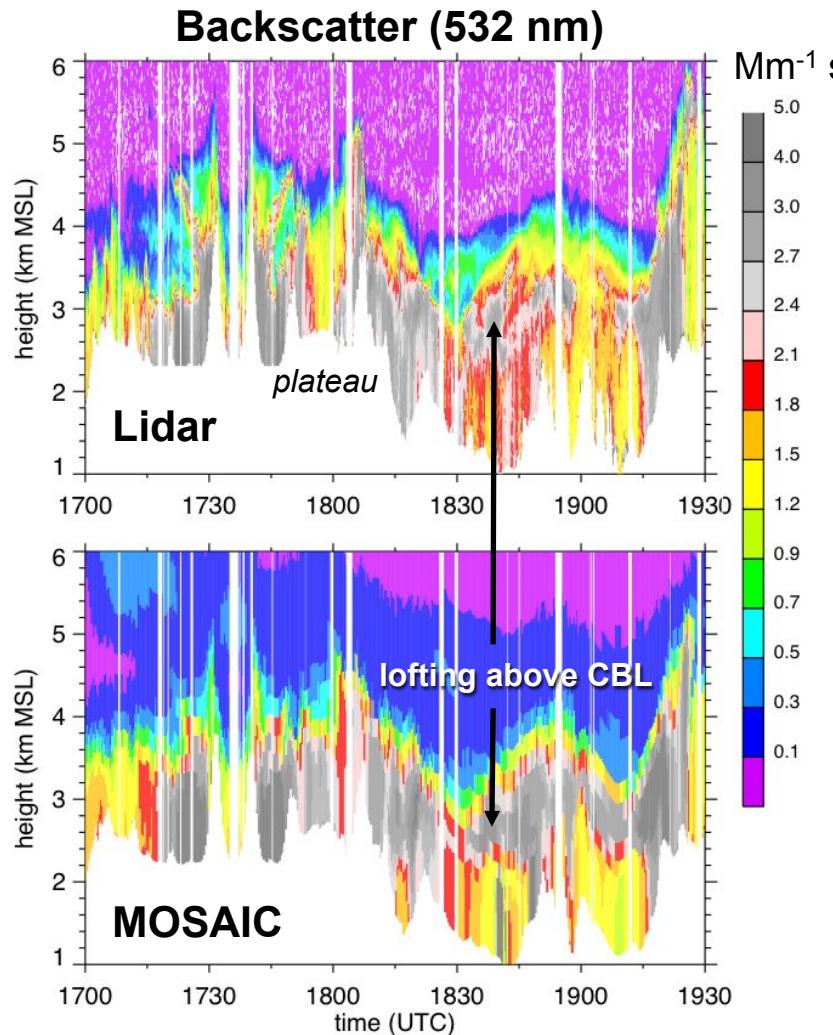
Example: Impact of Aerosols over Europe

from Forkel et al. ACP (2012)



Example: Evaluating Extinction Profiles from Fast et al. ACP (2009)

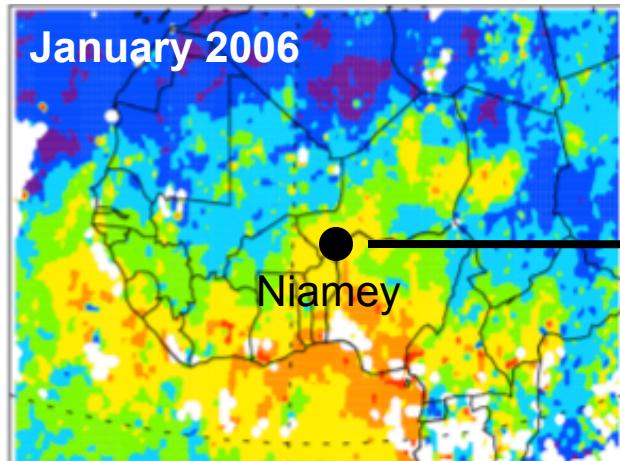
NASA B-200 Aircraft Flight Path 13 March 2006 over central Mexico



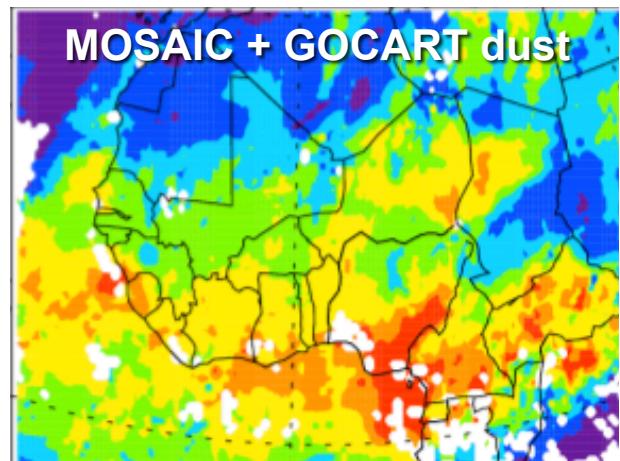
Example: Evaluating Heating Rate Profile

from Zhao et al. ACP (2010)

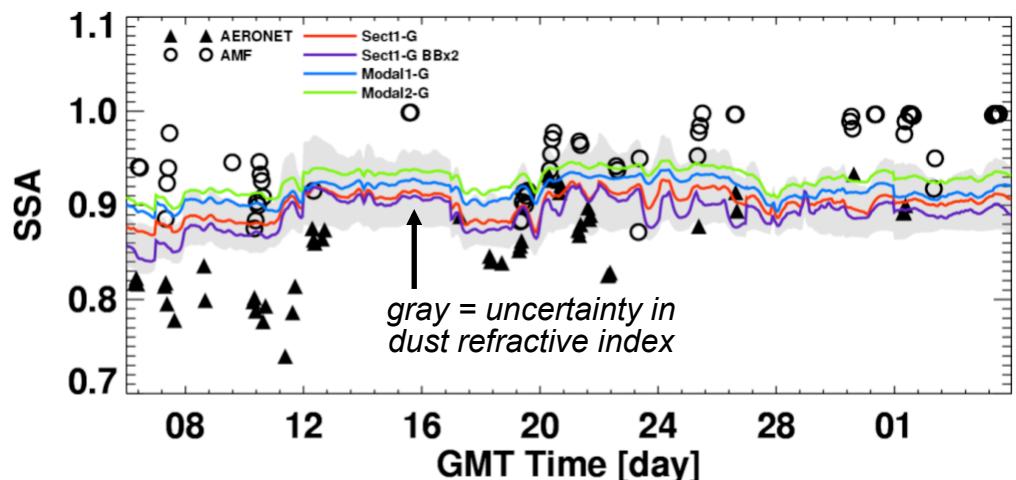
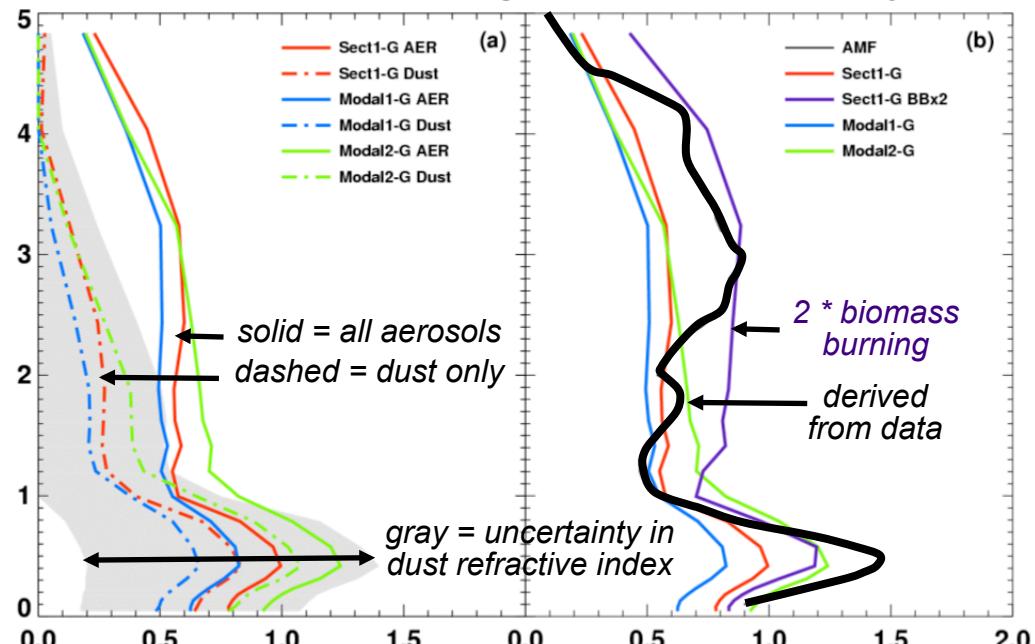
Averaged Terra-MISR AOD



Simulated AOD



SW Radiative Heating Rate Profile (K day^{-1})





Settings in namelist.input

Important Parameters:

- ▶ ra_sw_physics = 2 – aerosols affects radiation computed by Goddard scheme
- ▶ ra_sw_physics = 4] aerosols affects radiation computed by RRTMG scheme
- ▶ ra_lw_physics = 4]
- ▶ aer_ra_feedback = 1, turns on aerosol radiation feedback
- ▶ aer_op_opt = > 0, define the mixing rule for Mie calculations
- ▶ Works similarly for GOCART, MADE/SORGAM, MAM, and MOSAIC options

Direct Effects:

- ▶ Simulations with aer_ra_feedback = on or off can be used to quantify direct effects, but differences in clouds complicates interpretation
 - Useful to add code that computes radiation with and without aerosols and with and without clouds (either directly in the code or computed off-line)



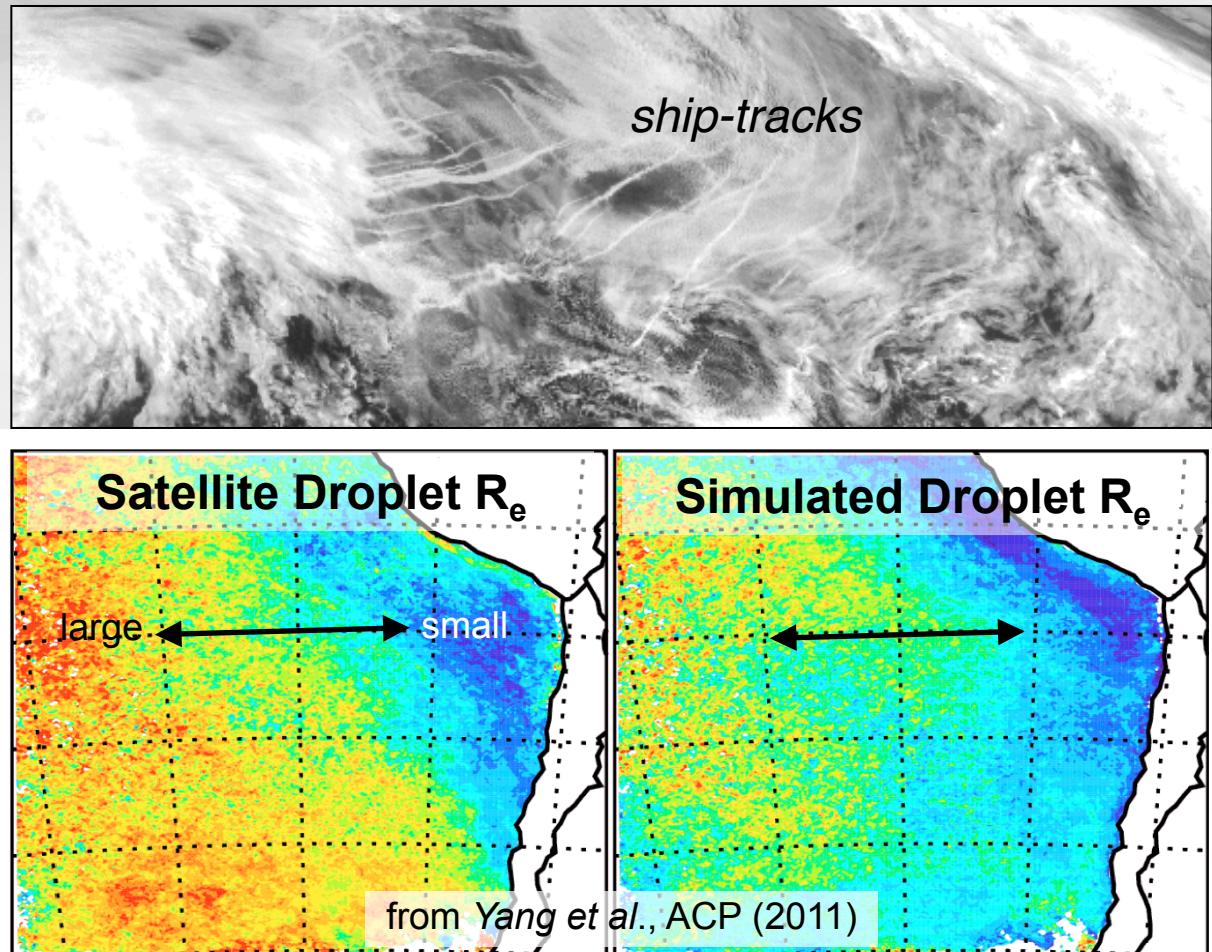
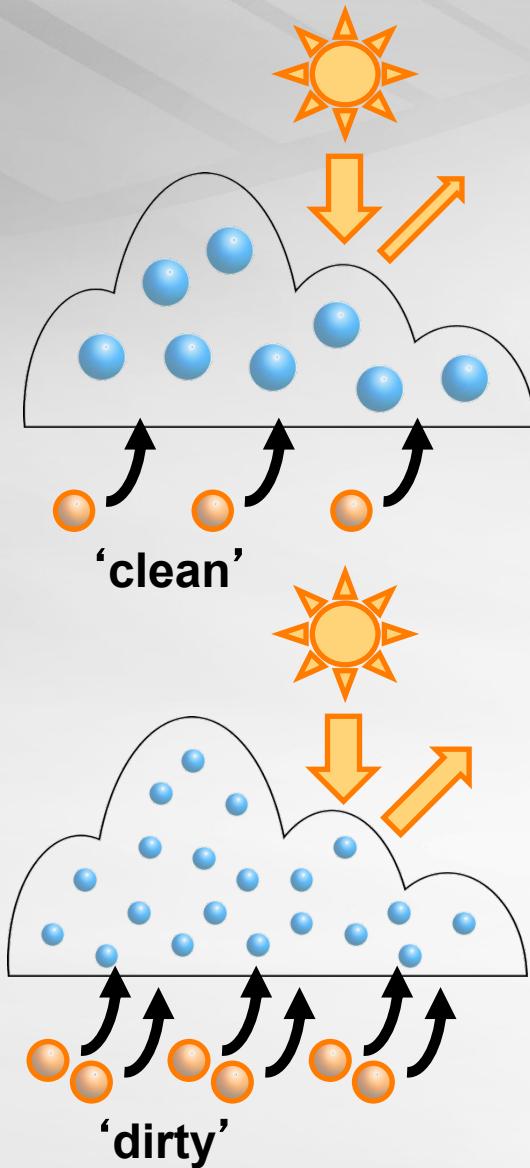
Future Capabilities

Research – Possibly in Upcoming Releases of WRF:

- ▶ Different refractive indices organic aerosol components
- ▶ More computationally efficient Mie calculations
- ▶ Consistency in aerosol optical properties for radiation and photolysis schemes
- ▶ Code to handle aerosol model with external mixtures
- ▶ Treatment for “brown carbon”



Part 2: Aerosol Indirect Effects

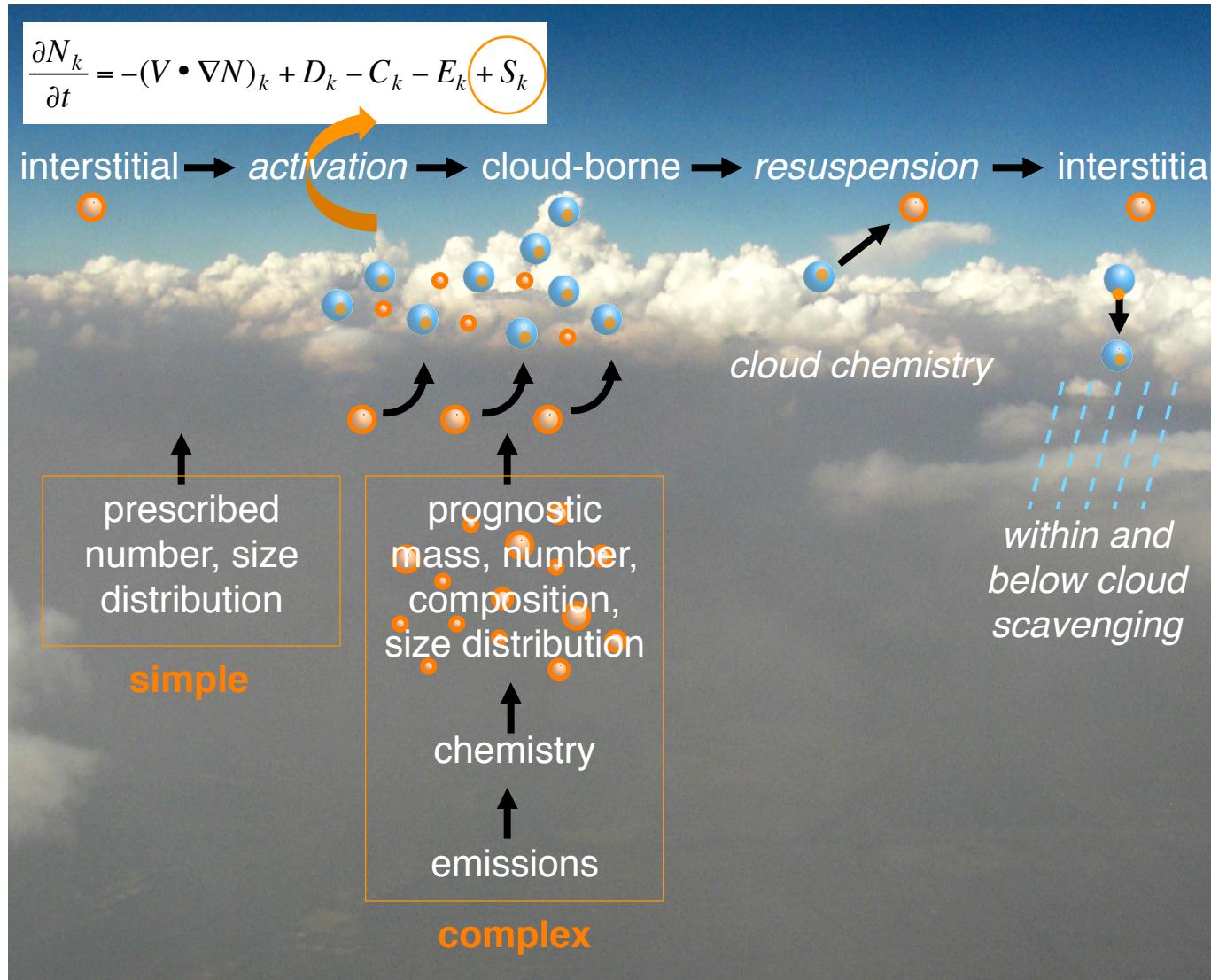


The number of activated aerosols affects the cloud drop size distribution, and consequently cloud albedo and radiation budget



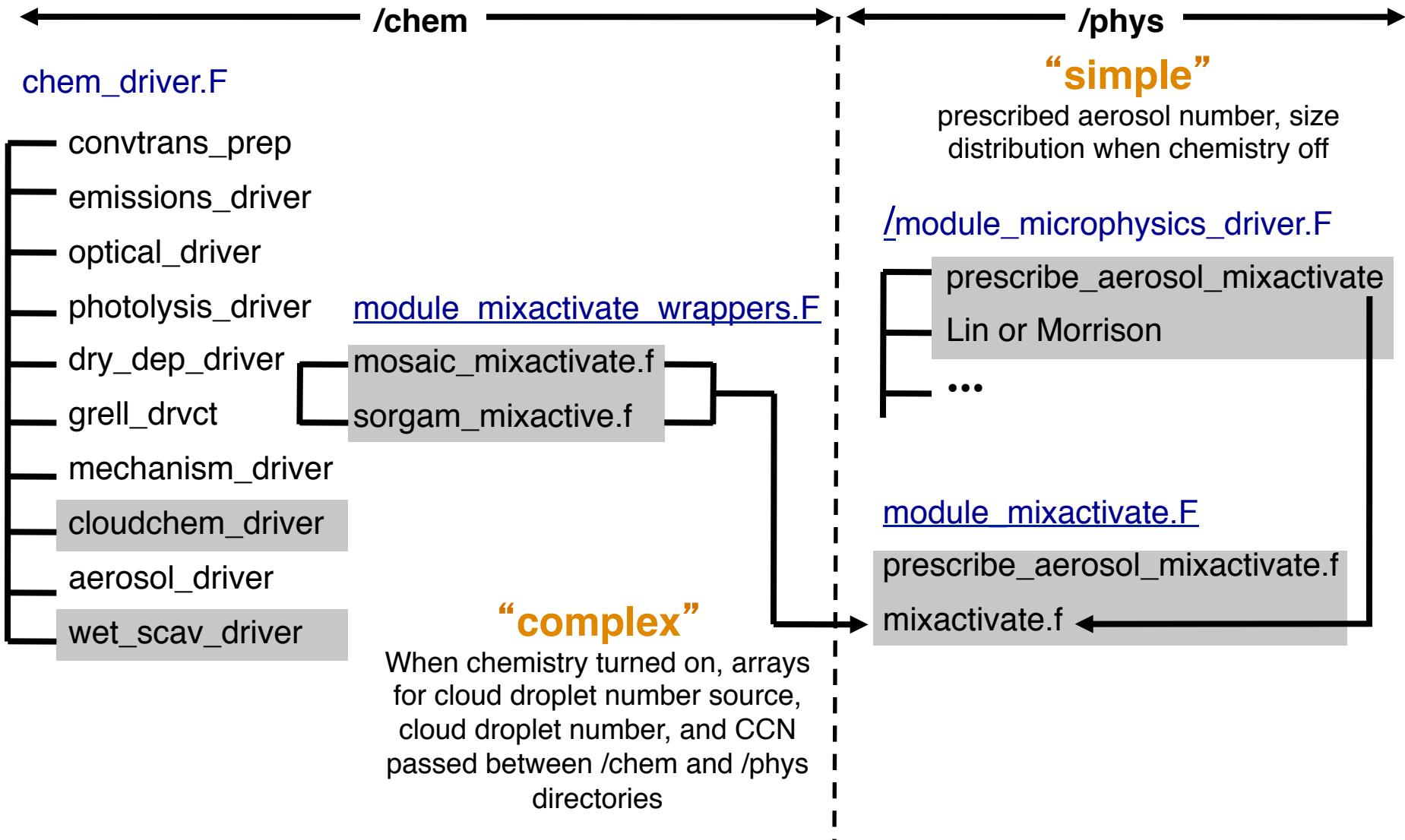
Cloud-Aerosol Interactions

General Description and Assumptions





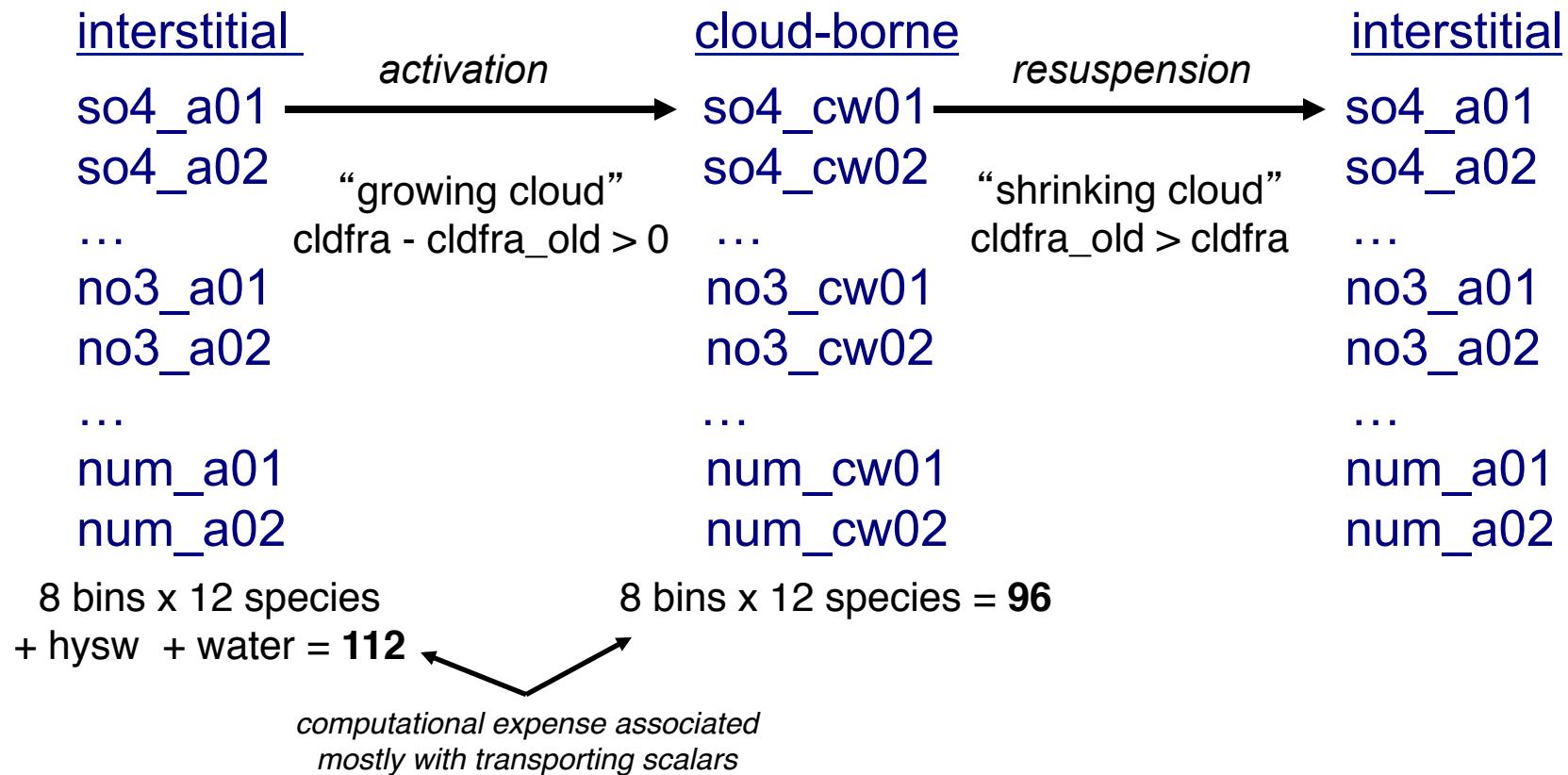
Flow Chart





Aerosol Species

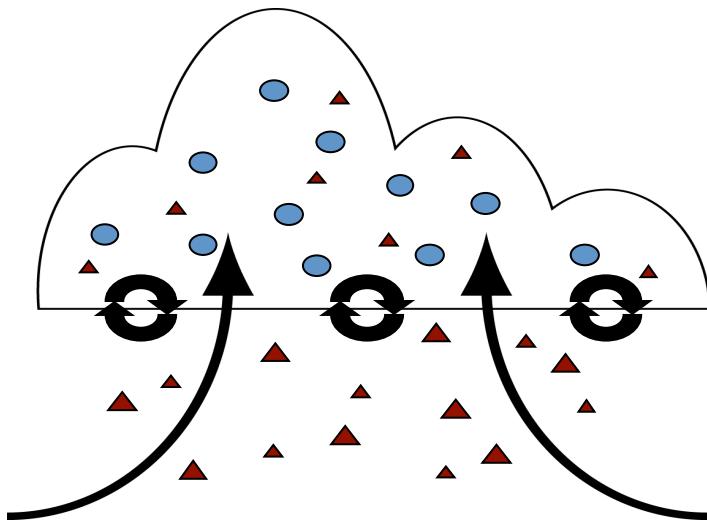
- ▶ Interstitial and cloud-borne aerosol particles treated explicitly, nearly doubling the number of transported species



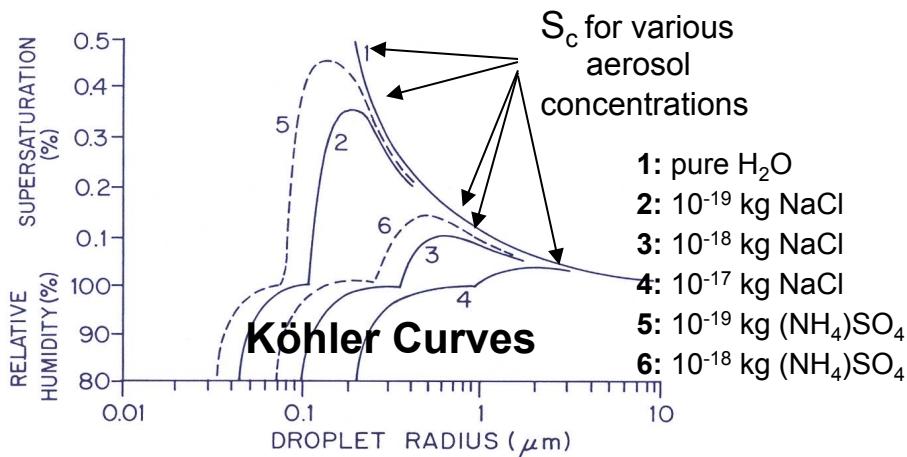
- ▶ Similar for MADE/SORGAM: $\text{so4aj} \longrightarrow \text{so4cwj} \longrightarrow \text{so4aj}$



Activation



Aerosols activated when the environmental supersaturation in the air “entering cloud”,
 S_{\max} > aerosols critical supersaturation, S_c



Activate.f computes activation fraction for mass and number for each bin/mode. Inputs include mean vertical velocity, $wbar$, and σ of the turbulent velocity spectrum, $sigw$.

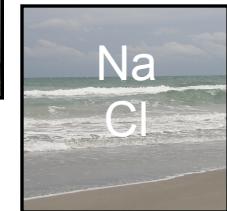
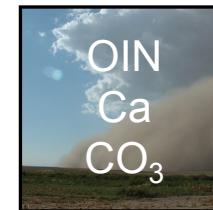
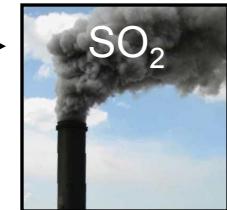
Note: $sigw$ based on $exch_h$, but some PBL options (ACM) do not have $exch_h$ passed out of the subroutine. Minimum $exch_h$ set to 0.2 m s^{-1} since predicted values may be too low in free atmosphere.

For each vertical velocity, peak S_{\max} depends on aerosol size and composition [Abdul Razzak and Ghan, 2000, 2002]. Activation fraction based distribution of S_c of the bin/mode - simply a fraction of aerosol mass or number in the bin/mode having $S_c < S_{\max}$



Hygroscopicity

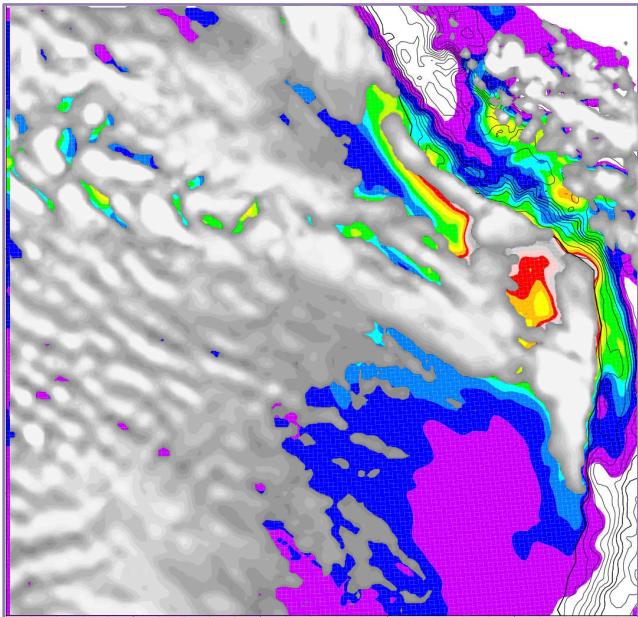
- ▶ Hygroscopic properties depend on particulate composition:
 - hygro_so4_aer = 0.5
 - hygro_no3_aer = 0.5
 - hygro_nh4_aer = 0.5
 - hygro_oc_aer = 0.14 (some OC may be hygroscopic – subject of research)
 - hygro_bc_aer = 1.0e-6 *hydrophobic*
 - hygro_oin_aer = 0.14
 - hygro_ca_aer = 0.1
 - hygro_co3_aer = 0.1
 - hygro_msa_aer = 0.58
 - hygro_cl_aer = 1.16
 - hygro_na_aer = 1.16 *hydrophilic*
- ▶ Activation depends on volume **weighted bulk hygroscopicity**, prior to call to mixactivate.f in module_mixactivate_wrappers.F
 - Not taken into account*  *What about coating?* ←
 - ▶ For chem_opt = 0 and nprog = 1, hygroscopicity set to 0.5



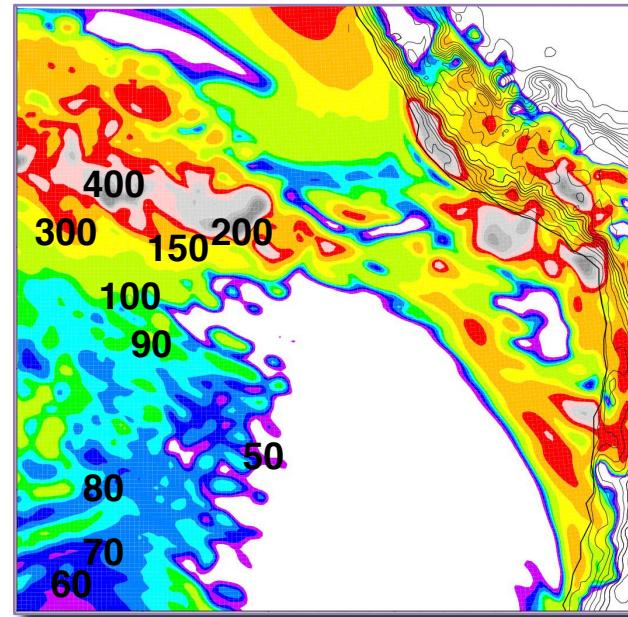
Cloud Condensation Nuclei

- ▶ CCN: number concentration of aerosols activated at a specified super-saturation → often have measured values to compare with
- ▶ Diagnostic quantity, varies in space and time
- ▶ Computed at 6 super-saturations (.02, .05, .1, .2, .5, and 1%) that correspond to CCN1, CCN2, CCN3, CCN4, CCN5, CCN6 in Registry
- ▶ Computed in module_mixactivate.F

AOD (600 nm) and COD



CCN at 0.1% SS (# cm⁻³)



*example from
VOCALS-Rex:
southeastern
Pacific marine
stratocumulus*

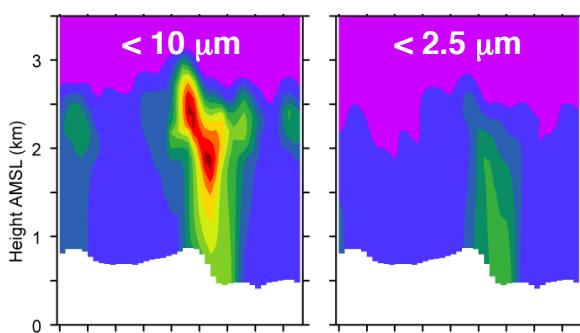


Aqueous Chemistry

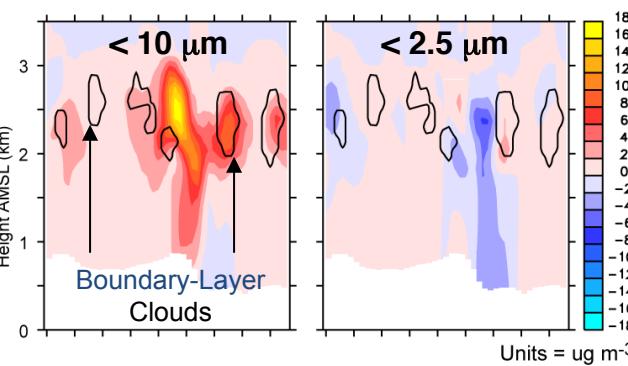
- ▶ Bulk cloud-chemistry module of Fahey and Pandis (2001) compatible with MOSAIC and MADE/SORGAM (cloudchem_driver.F)
- ▶ Chemistry in cloud drops, but not rain drops
- ▶ Oxidation of S(IV) by H_2O_2 , O_3 , trace metals, and radical species, as well as non-reactive uptake of HNO_3 , HCl , NH_3 , and other trace gases
- ▶ Bulk mass changes partitioned among cloud-borne aerosol size bins, followed by transfer of mass & number between bins due to growth; assumptions regarding the cloud water fraction for each bin mode

Vertical Cross-Section Through Power Plant SO_2 Plume

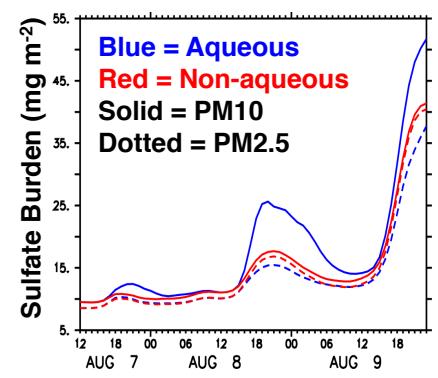
SO_4 , Aqueous Chemistry Simulation



SO_4 Difference (Aqueous - Non-Aqueous)



Sulfate Burden Over Domain

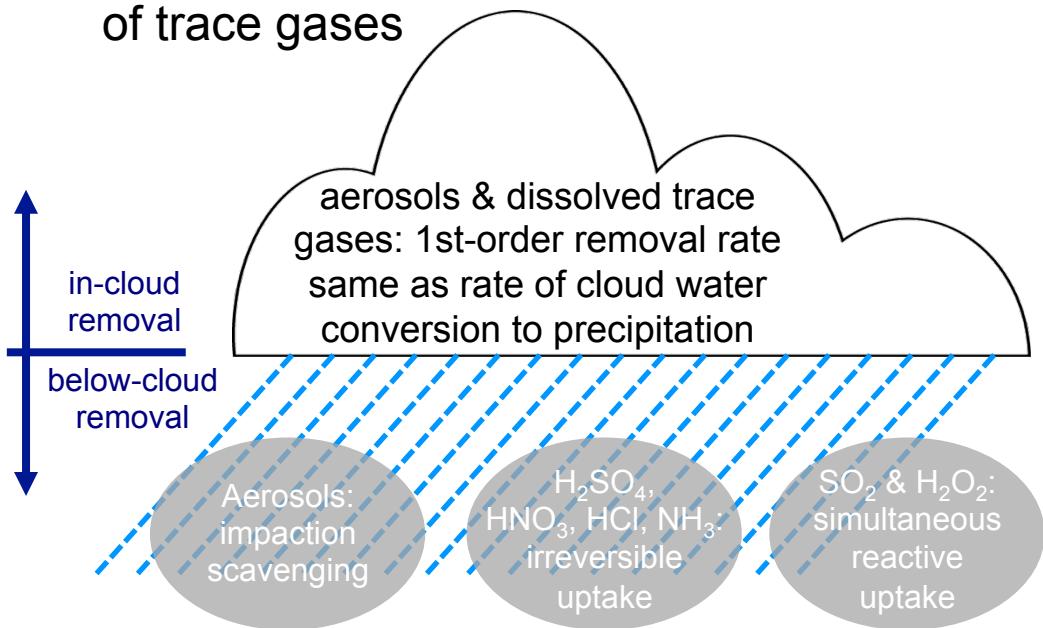


aqueous chemistry results in more SO_4 mass in coarse mode

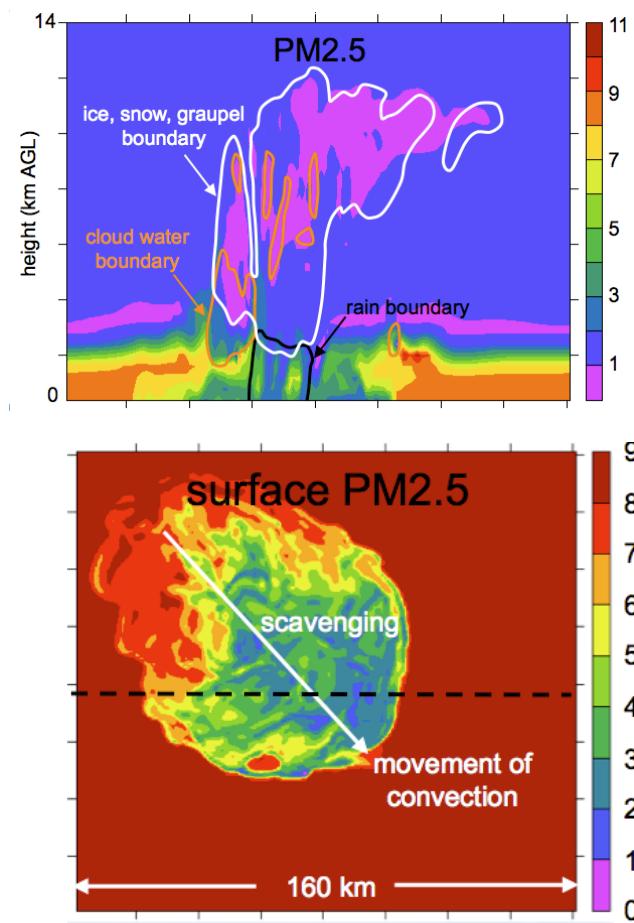


Wet Removal

- ▶ As cloud drops are collected by precipitation particles (rain, snow, graupel), cloud-borne aerosols and trace gases are also collected
- ▶ While cloud-borne aerosols are explicit, the cloud chemistry module provides the fraction of trace gas that is cloud-borne or dissolved in cloud water
 - MOZART has its own treatment of wet removal of trace gases



scavenged aerosols and gases instantly removed (and not saved) see Easter et al. (2004); aerosols are not resuspended for evaporating rain





Cloud Droplet Number

- ▶ converted Lin et al. microphysics scheme to a two-moment treatment (mass & number), in addition to adding impact of aerosols on droplet #
- ▶ Morrison microphysics is a two-moment treatment, so only needed to add code to include the impact of aerosols on droplet #

$$\frac{\partial N_k}{\partial t} = -(V \bullet \nabla N)_k + D_k - C_k - E_k + S_k$$

qndrop →

N_k - grid cell mean droplet number mixing ratio in layer k

D_k - vertical diffusion

C_k - droplet loss due to collision/coalescence & collection

E_k - droplet loss due to evaporation

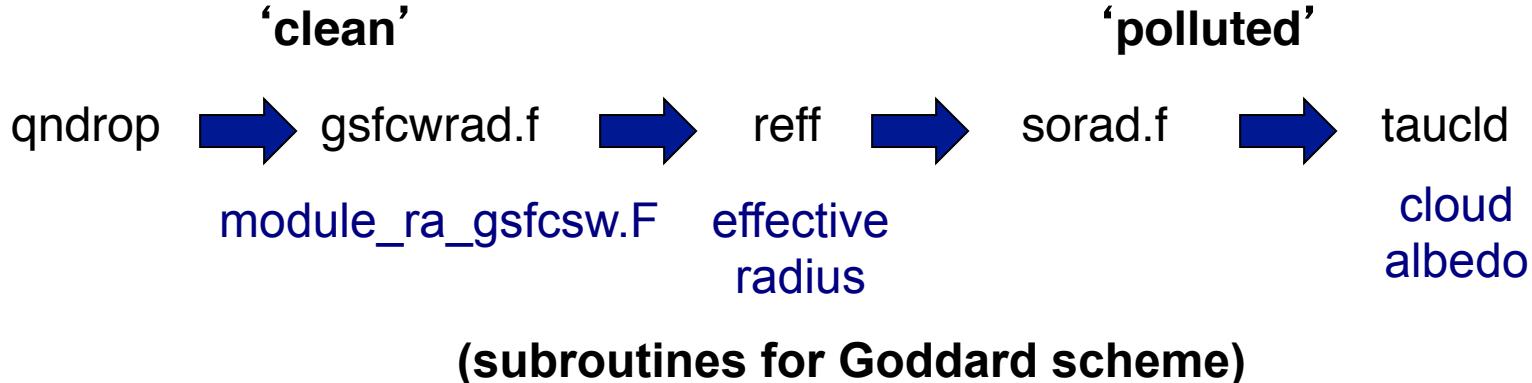
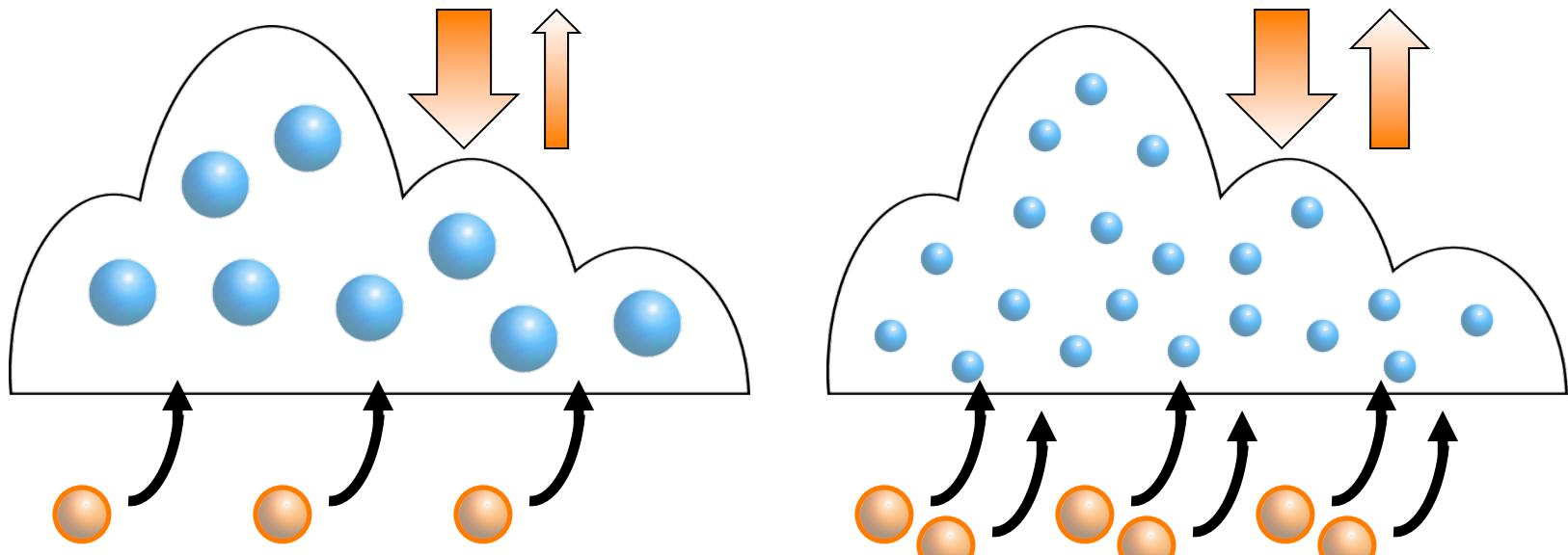
qndropsource
(nsource) → S_k - droplet source due to nucleation (determined in mixactivate.f)

- ▶ cloud droplet number source determined by aerosol activation (for meteorology-only runs a prescribed aerosol size distribution is used)
- ▶ droplet number and cloud water mixing ratio used to compute effective cloud-particle size for the cloud optical depth in Goddard or RRTMG shortwave radiation scheme (ra_sw_physics = 2 or 4)



First Indirect Effect

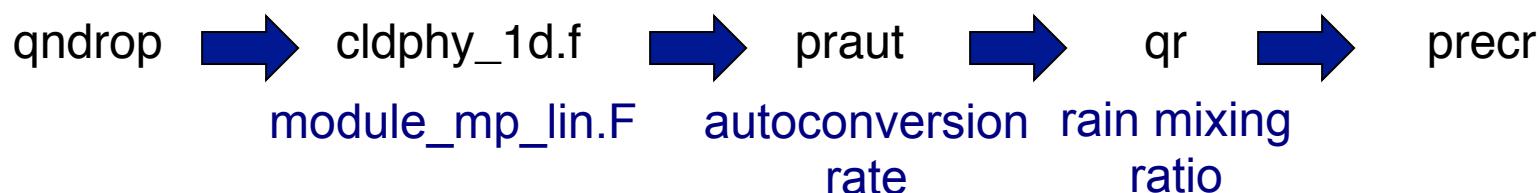
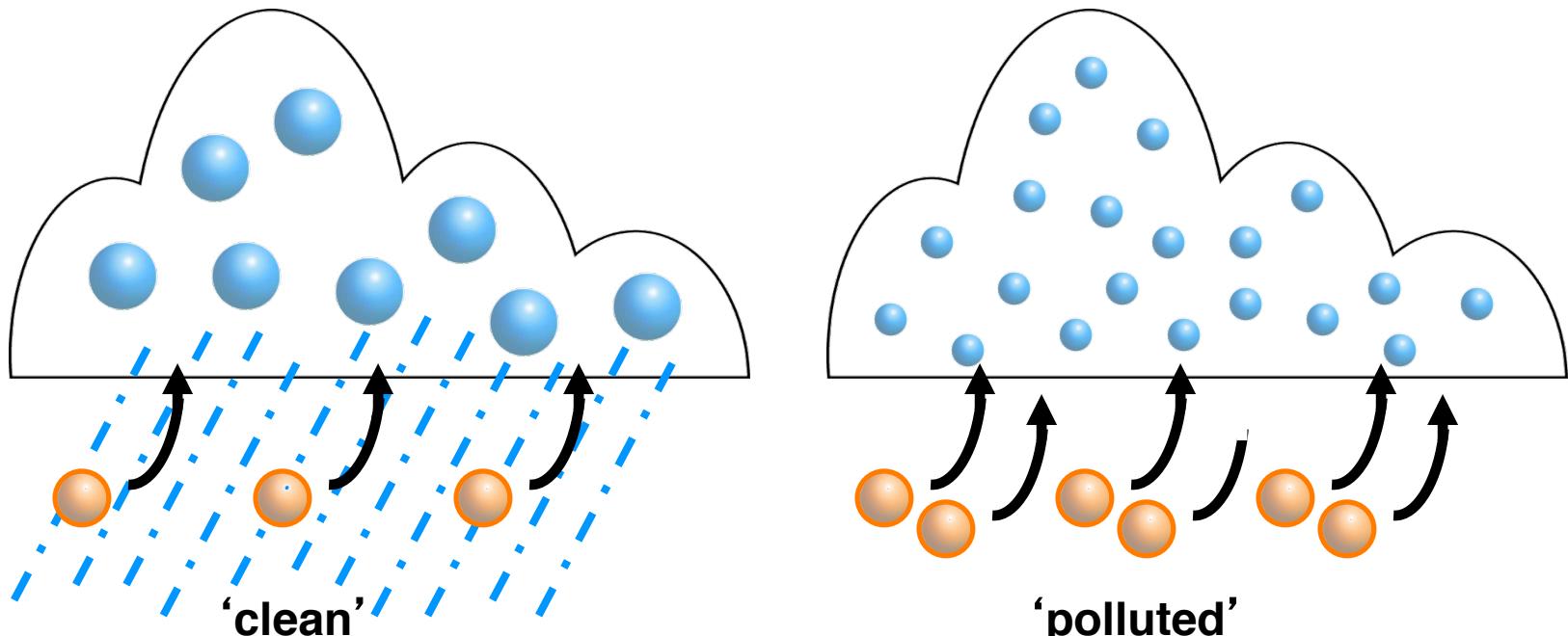
- Influence of cloud optical depth through impact on effective radius, with no change in water content of cloud





Second Indirect Effect

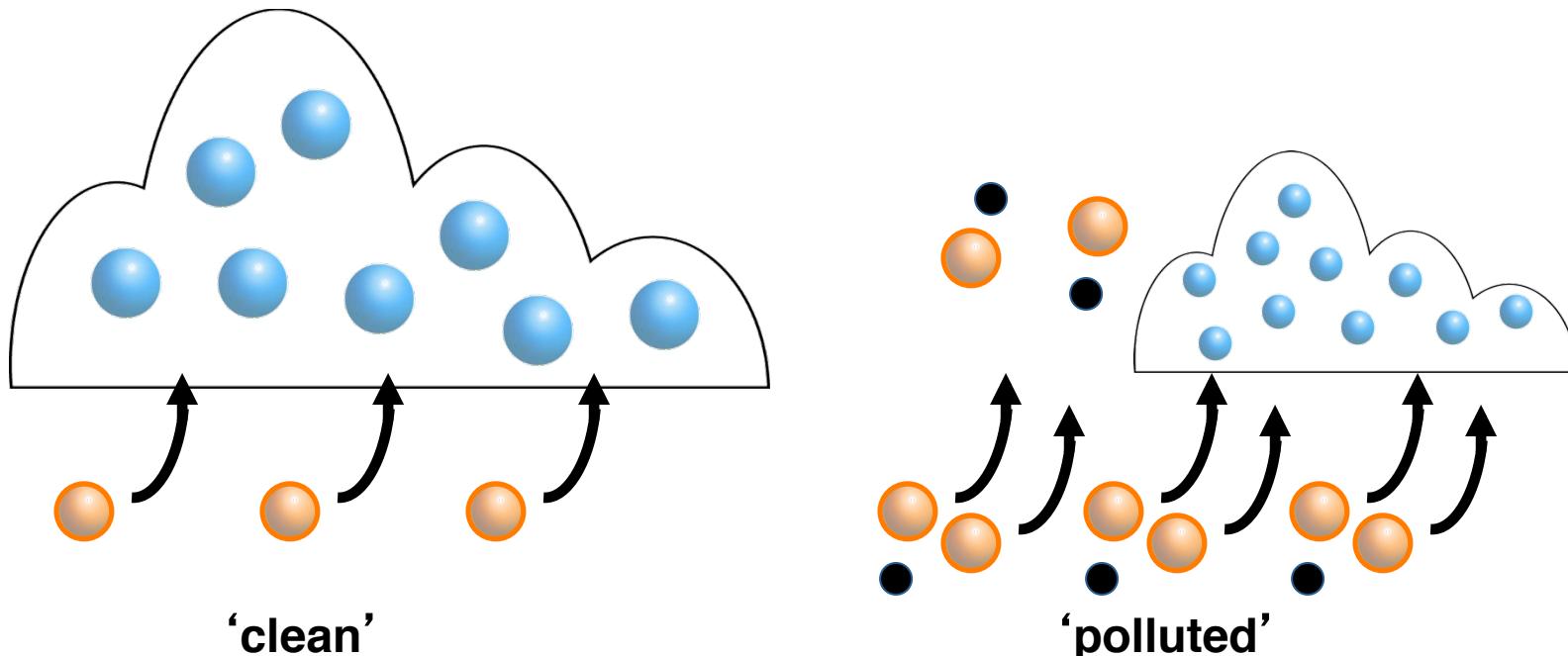
- Influence of cloud optical depth through influence of droplet number on mean droplet size and hence initiation of precipitation



(subroutines for Goddard scheme)

Semi-Direct Effect

- Influence of aerosol absorption of sunlight on cloud liquid water and hence cloud optical depth





Settings in namelist.input

Cloud-Aerosol Interactions for Lin and Morrison Microphysics

Simple:

- ▶ chem_opt = 0
- ▶ naer = specified value

Complex:

- ▶ chem_opt = 9 - 12, 32, 34, 35, 41, 42 - cloud-phase aerosols for MOSAIC and MADE/SORGAM
- ▶ cldchem_onoff = 1, turns on cloud chemistry
- ▶ wetscav_onoff = 1, turns on wet scavenging

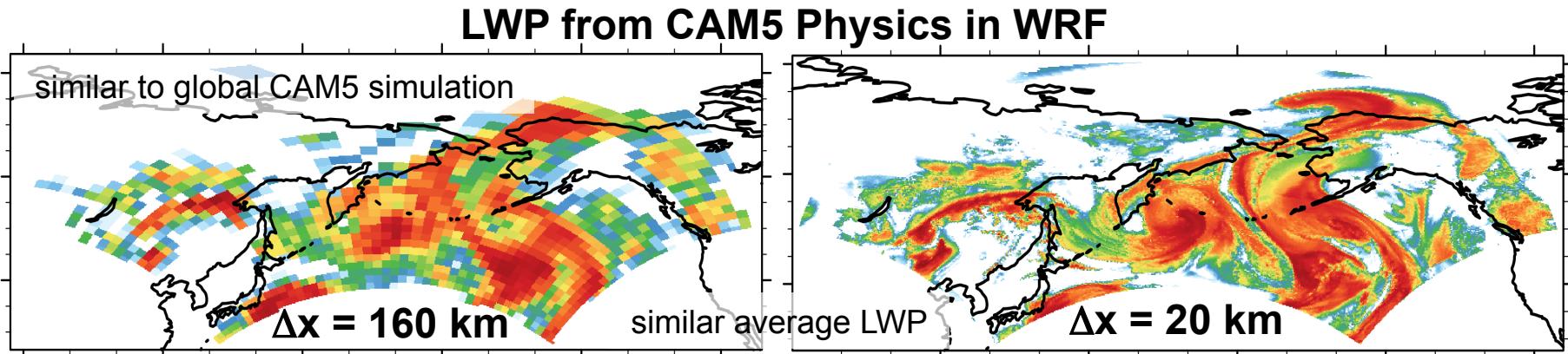
Both:

- ▶ mp_physics = 2, 10
- ▶ progn = 1, turns on prognostic cloud droplet number



CAM5 Physics is Different (1)

- ▶ Cloud-Aerosol Interactions for **Morrison and Gettelman microphysics handled separately**, because
 - CAM5 physics kept as same as possible as in the CESM climate model

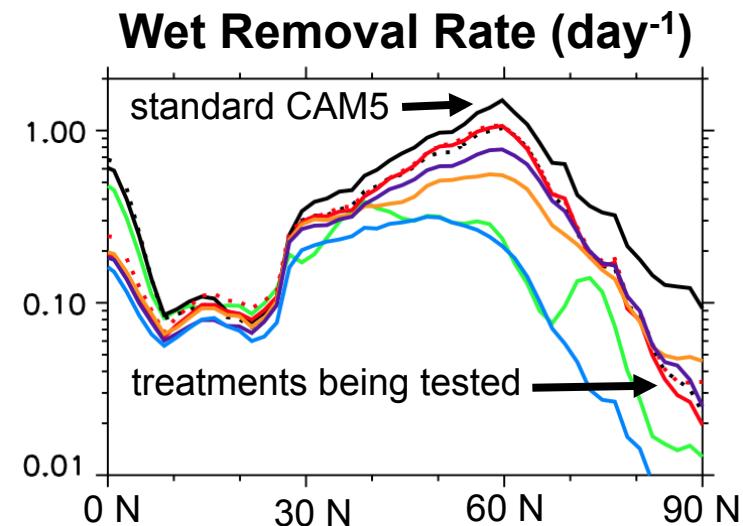


- ▶ Entire CAM5 physics suite must be used when simulating cloud-aerosol interactions in the Morrison and Gettelman microphysics scheme
 - `mp_physics=19, cu_physics=7, shcu=physics=2, bl_pbl_physics=9`
`chem_opt=503, cam_mam_mode=3, CAM_MP_MAM_cpled='true'`
- ▶ `/phys/module_mixactivate.F` is not used (activation is done elsewhere), but is conceptually very similar to how it has been handled in WRF previously



CAM5 Physics is Different (2)

- ▶ Morrison and Gettelman microphysics includes treatment of heterogeneous freezing on mineral dust
 - But, there are no ice-borne aerosols
 - Coupling of prognostic aerosols to ice nuclei (IN) not included for other microphysics scheme; the effect of aerosols on cloud droplets will affect ice processes indirectly however
- ▶ module_wetscav_driver.F modified to handle MAM aerosols
 - See Wang et al. GMD (2013) for a discussion on wet removal and its uncertainties
- ▶ Paper on CAM5 physics in WRF will be submitted soon

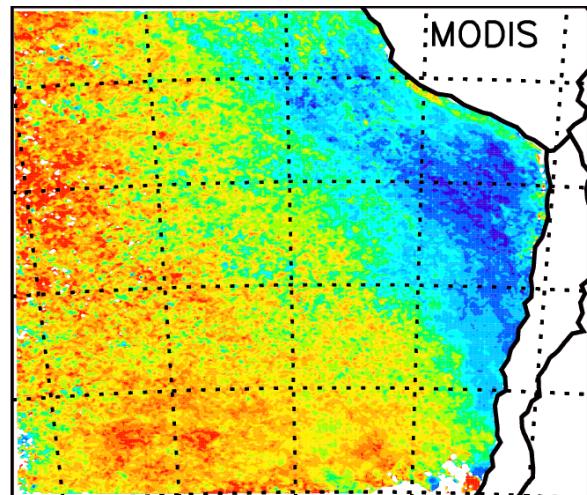


Example: Marine Stratocumulus

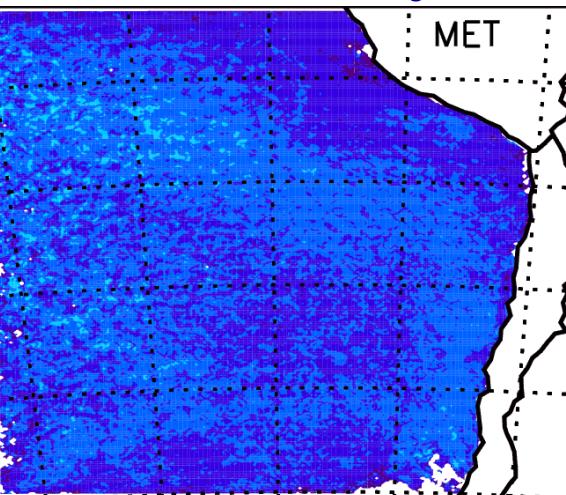
from Yang et al. ACP (2011)

Average Effective Droplet Radius during 2008 VOCALS-REx

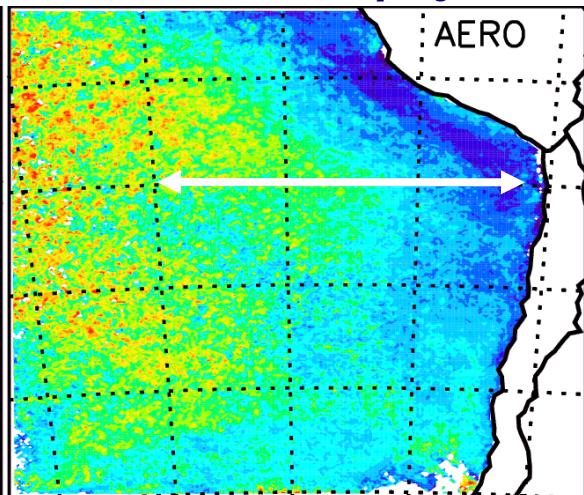
MODIS



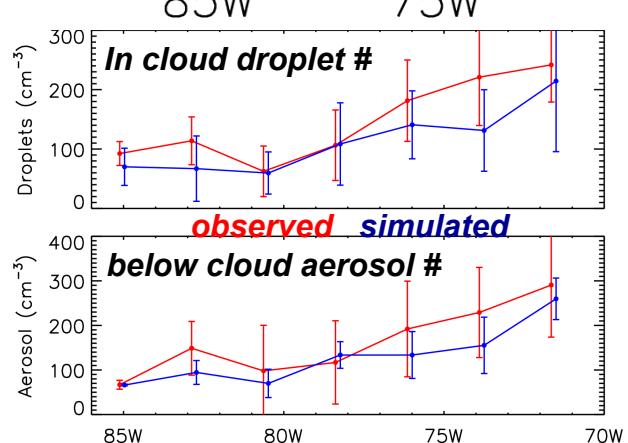
*WRF
no chemistry*



*MOSAIC aerosols and
Morrison microphysics*



- Yang et al. (2011) used the Morrison microphysics for this case, while Saide et al. ACP (2012) used the Lin microphysics to evaluate cloud-aerosol interactions



Example: Deep Convection

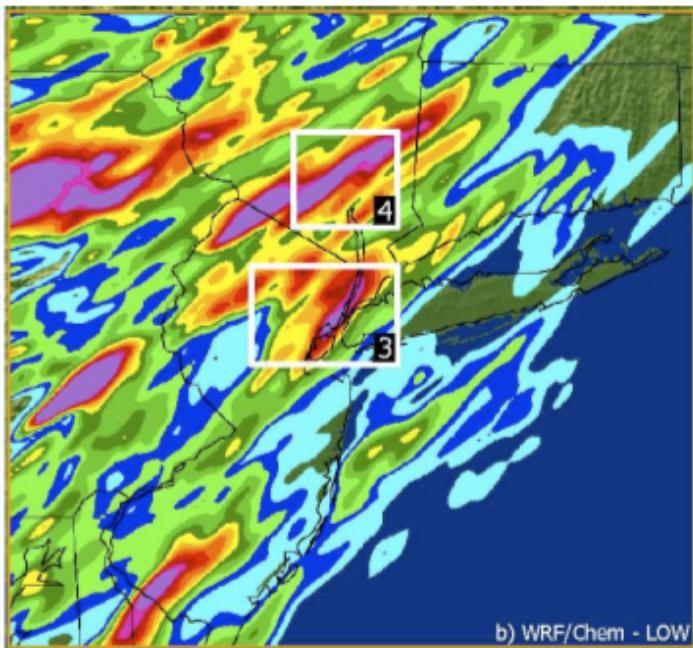
from Ntelekos et al. QJRMS (2008)



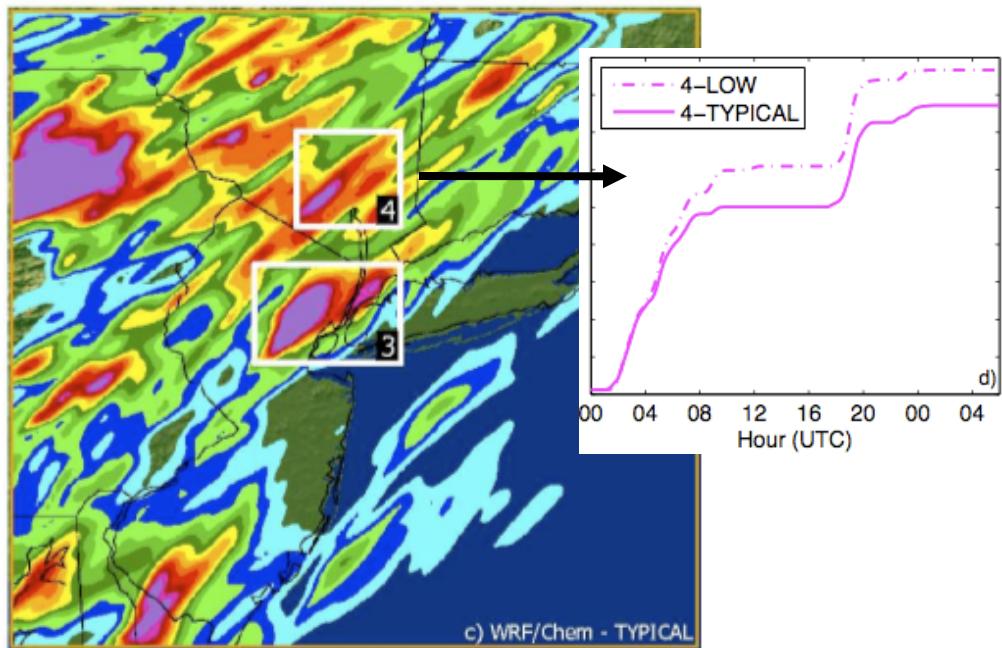
Pacific Northwest
NATIONAL LABORATORY
Proudly Operated by Battelle Since 1965

Impact of Particulates on Convective Precipitation Along the Urban East Coast Corridor

WRF-Chem: low emissions



WRF-Chem: typical emissions



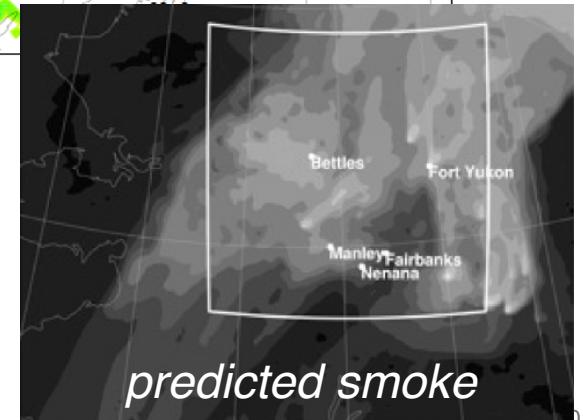
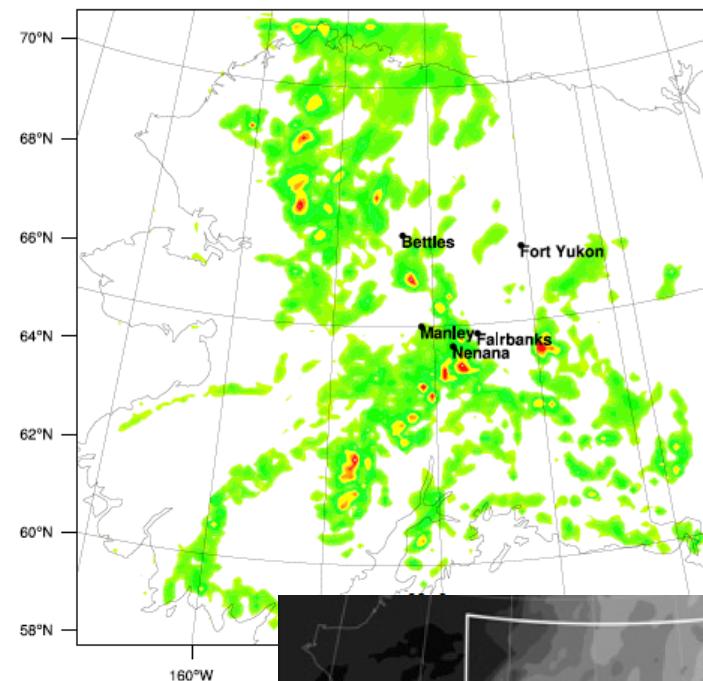
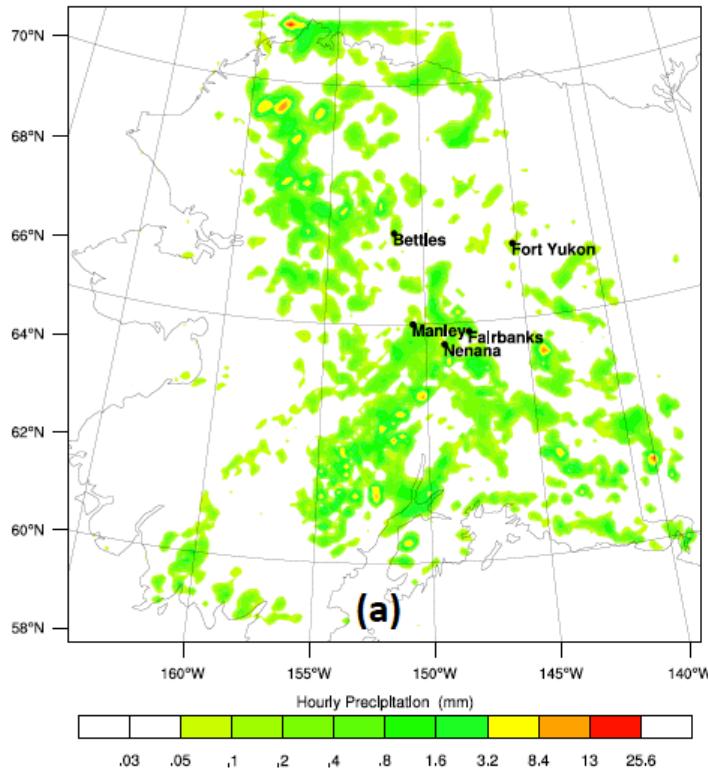
- ▶ Effects of aerosols leads to some cells having more precipitation, while other cells are less intense and have less precipitation

Example: Smoke Entrained into Clouds

from Grell et al. ACP (2011)

Precipitation over Alaska

Without aerosol-cloud interactions With aerosol-cloud interactions



- Overall pattern of precipitation unchanged, but more intense precipitation is simulated when biomass burning plumes interacting with cloud droplets are included



Comparing Options

Care Must be Taken in Quantifying Indirect Effects!

Indirect Effects:

- ▶ Comparing runs with `chem_opt = 8` (without cloud-borne aerosols) with `chem_opt = 10` (with cloud-borne aerosols) for MOSAIC coupled to Lin microphysics **does not** quantify the indirect effect
 - since the autoconversion scheme used in the Lin microphysics scheme will be different
 - Need to determine a prescribed aerosol scenario to compare with `chem_opt = 10` – see Gustafson et al., GRL, (2007)
 - An approach used with GCMs is to output “dirty-cloudy”, “dirty-clear”, “clean-cloudy”, and “clean-cloudy” radiation from the same run
- ▶ Often, it is just a matter of semantics. One should state how simulations are being compared and then stating the effects of aerosols on clouds rather than specifically calling them the indirect effect.

Indirect Effects Usage:

- ▶ Works with microphysics only – not cumulus parameterizations so users must be aware of issues associated with **spatial scale**
- ▶ In addition to Abdul-Razaak and Ghan (2000, 2002), other schemes have been used to compute aerosol activation (Fountoukis and Nenes, 2005)



Future Capabilities

Processes Under Development:

- ▶ Resuspension of aerosols from evaporating rain (PNNL)
- ▶ Aerosol-cloud interactions coupled with cumulus parameterizations for simulations $\Delta x > 10$ km (PNNL, NOAA); need to worry about double counting
- ▶ Ice-borne MOSAIC aerosols and IN treatment (PNNL)
- ▶ Wet removal scheme not coupled with aerosol indirect effect?
- ▶ Other users have tested other aerosol activation schemes (Fountoukis and Nenes, 2005) in WRF-Chem, in addition to the existing Abdul-Razaak and Ghan (2000, 2002) method, that could be made available in the public release
- ▶ Effects of aerosols (but not coupled to aerosol chemistry) in Thompson microphysics (NCAR)
- ▶ Other treatments are likely being developed by WRF-Chem users that are not known until they are published

difficult to evaluate directly with measurements

For more information and updates:

- ▶ PNNL modules: www.pnl.gov/atmospheric/research/wrf-chem
- ▶ See web page for list of papers on aerosol-cloud interactions