Aerosol-Radiation-Microphysics Interactions:
Aerosol Direct and Indirect Forcing

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Background

First A Brief History …

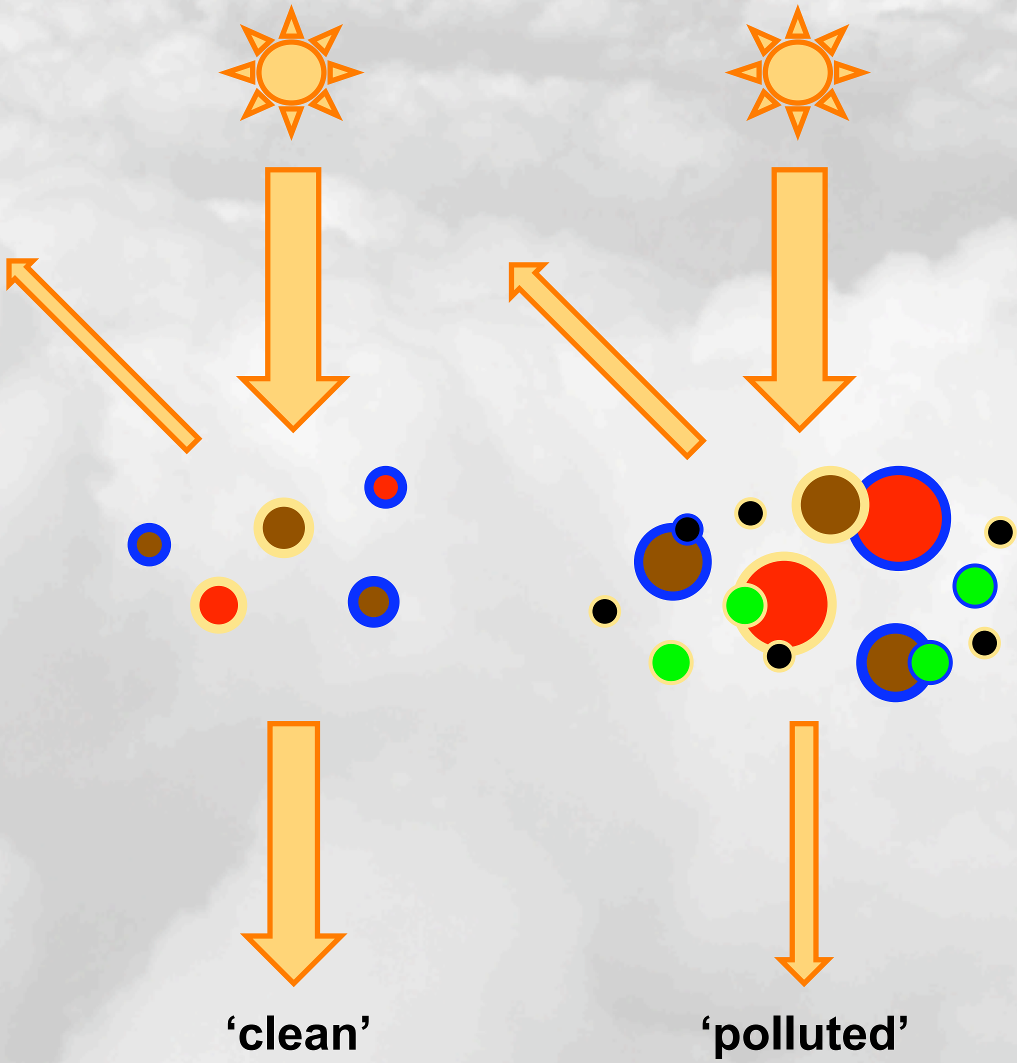
1) CBM-Z, Fast-J, and MOSAIC in WRF-Chem originated from a different off-line chemical transport model
2) Aerosol-radiation-cloud-chemistry interactions were added to MOSAIC, some adapted from MIRAGE global climate model
3) Aerosol-radiation-cloud-chemistry interactions subsequently coupled with MADE/SORGAM and GOCART
4) Currently adding more capabilities and making modules more generic

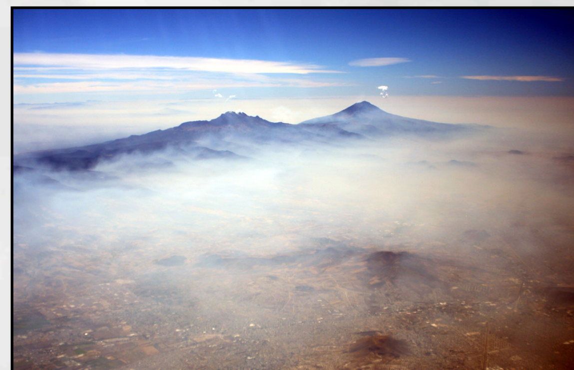
Our overall motivation is to use the model to better understand the local to regional-scale evolution of particulates and their effect on radiation, clouds, and chemistry

For more information and updates:

- PNNL modules: www.pnl.gov/atmospheric/research/wrf-chem

Part 1: Aerosol Direct Forcing





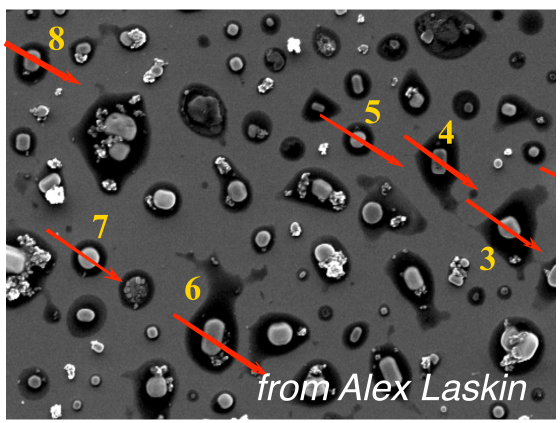
Aerosol Optical Properties

General Description and Assumptions

size and number distribution composition aerosol water ➡ refractive indices ➡ Mie theory ➡ layer optical depth, τ_λ single scattering albedo, ω_o asymmetry factor, g ➡ shortwave radiation

- τ, ω_o, and g function of wavelength, 300, 400, 600, 1000 nm
  - *τ = TAUAER1, TAUAER2, TAUAER3, TAUAER4 in Registry*
  - *ω_o = WAER1, WAER2, WAER3, WAER4 in Registry*
  - *g = GAER1, GAER2, GAER3, GAER4 in Registry*
- $\omega_o = k_s / (k_a + k_s)$, k_s and k_a = scattering and absorption coefficients
- various methods of obtaining refractive index, i.e. mixing rules

Mie theory assumes spheres



Mass, composition, and size distribution:

- *more mass ➡ bigger radiative impact*
- *amount of black carbon ➡ k_a*
- *aerosol size ➡ k_s*

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Aerosol Direct Radiative Effects

General Description and Assumptions

size and number distribution
composition
aerosol water → refractive indices → Mie theory → layer optical depth, τ_λ
single scattering albedo, ω_o
asymmetry factor, g → shortwave radiation

- Goddard shortwave scheme utilizes aerosol optical properties at 11 wavelengths, but the they are zero in default WRF / WRF-chem
- Use Angstrom relationship to interpolate between 4 wavelengths from optical property module to 11 wavelengths used in Goddard scheme
- Aerosols now account for scattering & absorption in Goddard scheme
- Effect of aerosols on longwave radiation not treated
- **New:** τ_λ, ω_o, and g also passed to RRTMG radiation scheme for both shortwave and longwave *(not in release version yet)*

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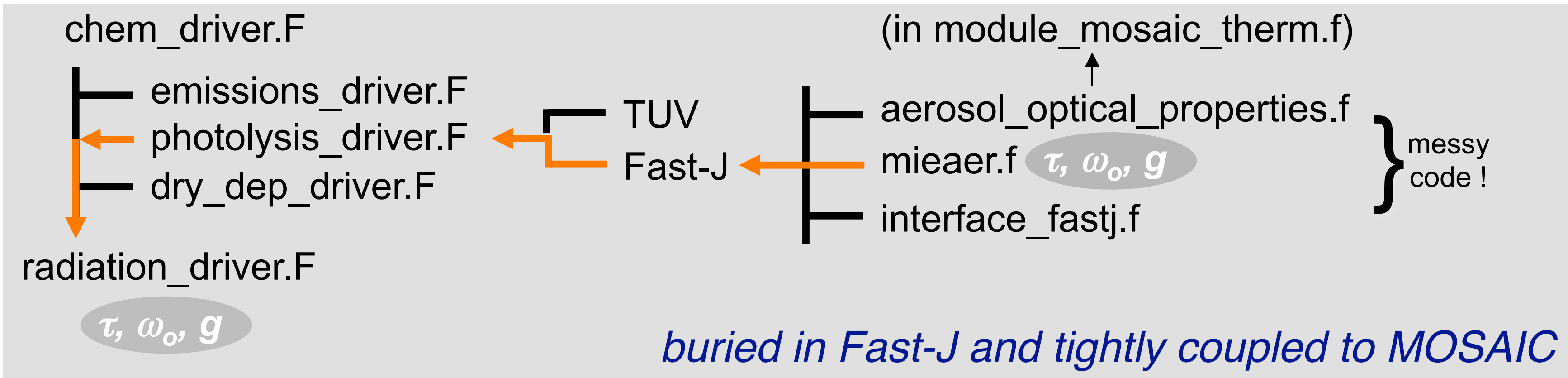
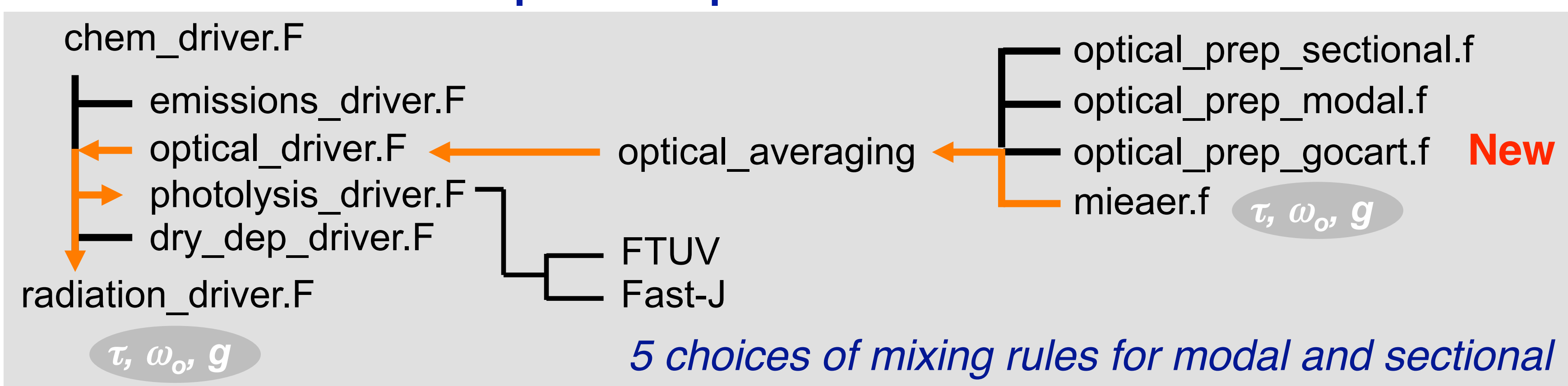
Coding Structure

Aerosol Optical Properties in WRF-chem v2.2



More Generic Aerosol Optical Properties for WRF-chem v3



Example of making the code more generic and interoperable

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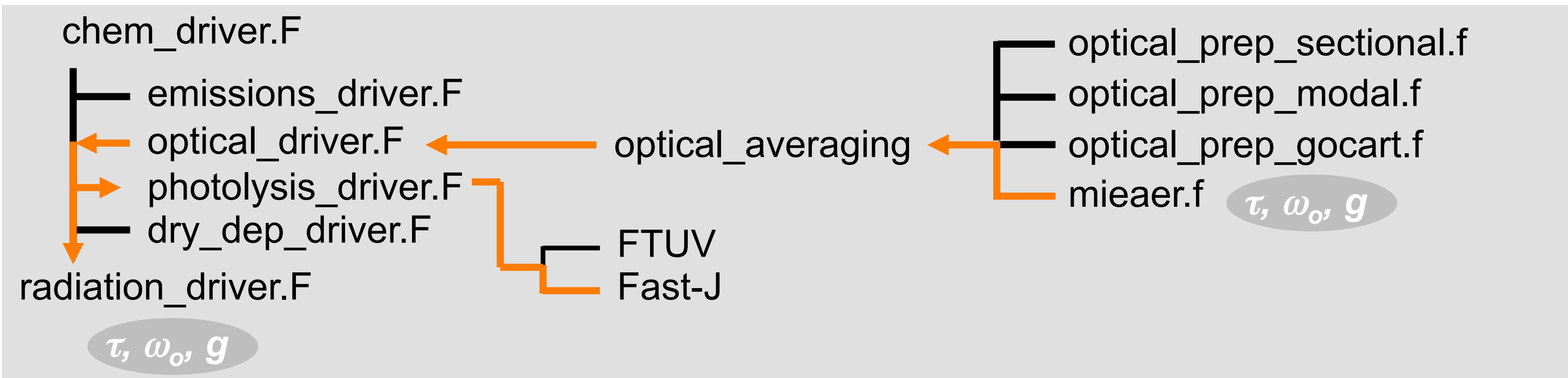
Aerosol Effects on Photolysis Rates

Aerosols → Photolysis Rates → Photochemistry

but clouds will usually have a bigger impact on photolysis rates overall than aerosols

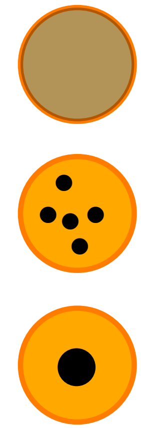


- Fast-J: uses (τ, ω_o, g) computed by module_optical_averaging.F
  - Not many tests done to evaluate impact of aerosols on photolysis rates
- FTUV: uses its own method to account for effects of aerosols on photolysis rates based on MADE/SORGAM species
  - ➢ MOSAIC aerosols will not affect photolysis rates when FTUV is used

not trivial – has not been a high priority yet

Choice of Mixing Rule

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



- Volume-Averaging and Maxwell-Garnett computed either exactly or approximately (faster), see *Ghan et al.* [2001] for approximate method
- Shell-core the most expensive computationally, but presumably the most accurate
- All very sensitive to changes in the amount of black carbon
- *aer_op_opt* in namelist.input:
  - *1 = Volume-Averaging approximate*
  - *2 = Maxwell-Garnett approximate*
  - *3 = Volume-Averaging exact*
  - *4 = Maxwell-Garnett exact*
  - *5 = Shell-Core (exact only)*

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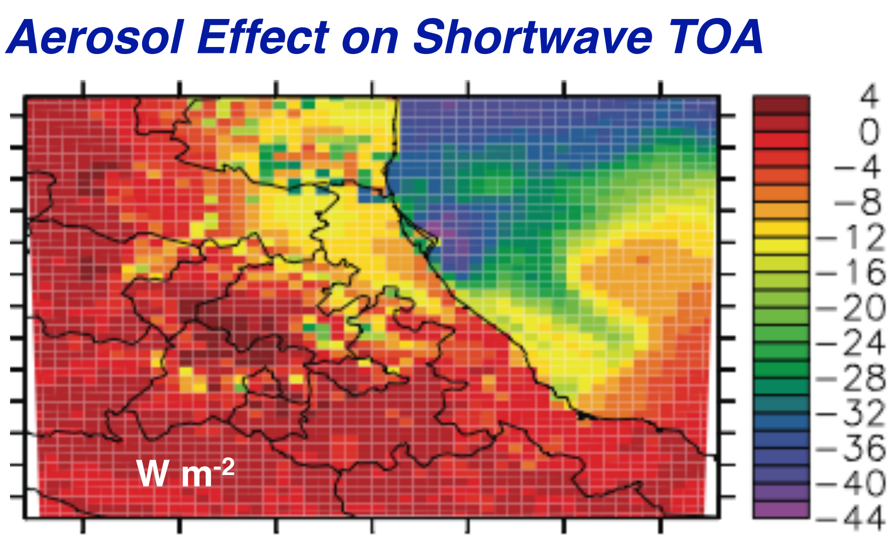
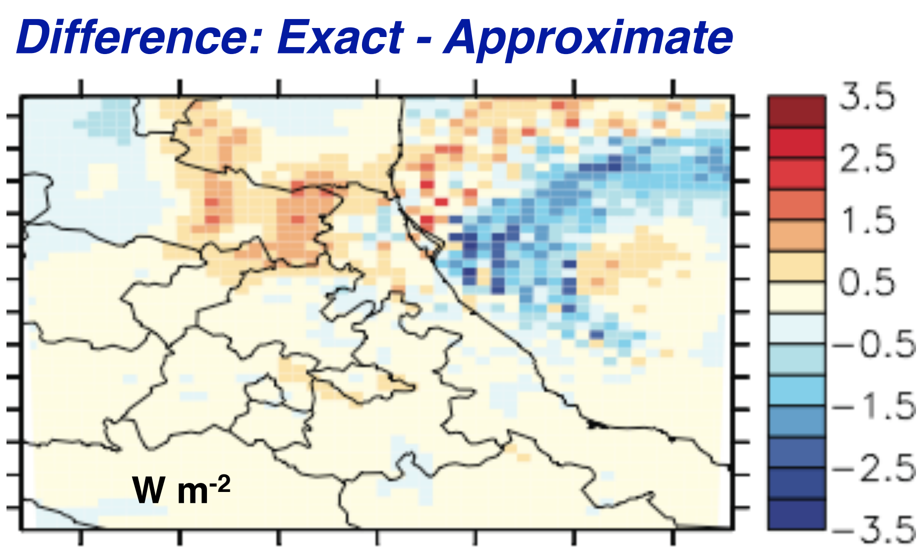
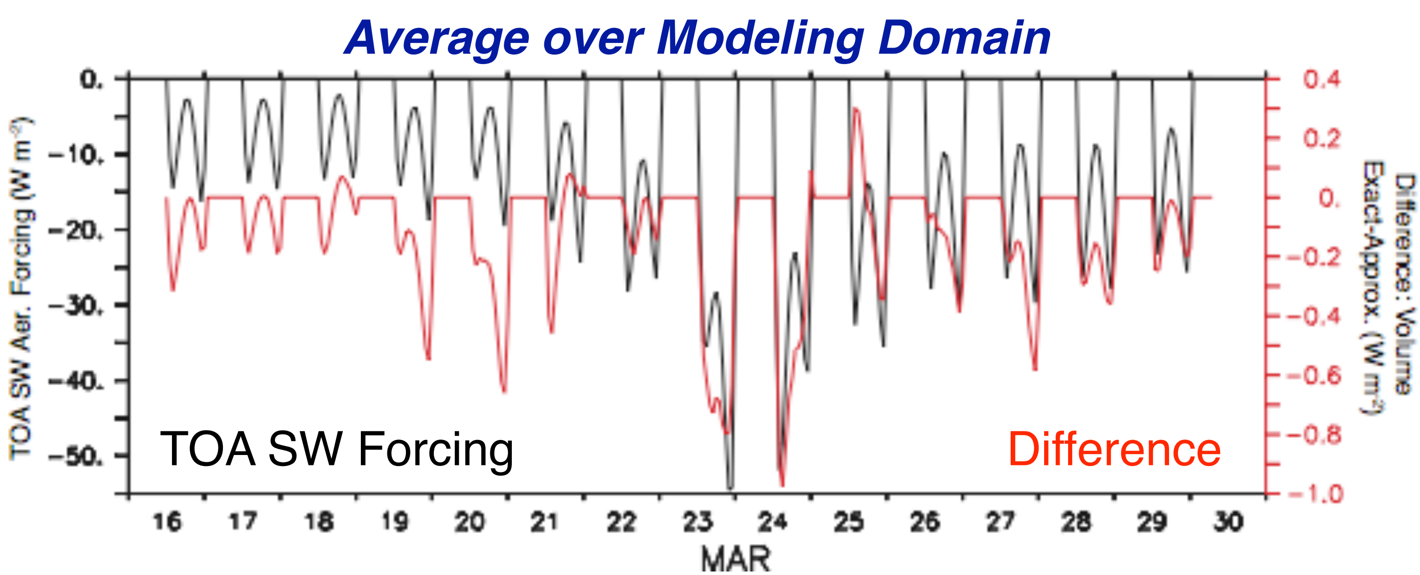
Mie Calculation Accuracy







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Assumptions

- Interfaces with GOCART, MADE/SORGAM, and MOSAIC, but linking to other aerosol models should be relatively easy
- **Sectional (MOSAIC):** tested only with 4 and 8 size bins – should work if additional size bins are specified

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- **Modal (MADE/SORGAM):** divides mass in modes into 8 sections - could divide into more sections to be more accurate

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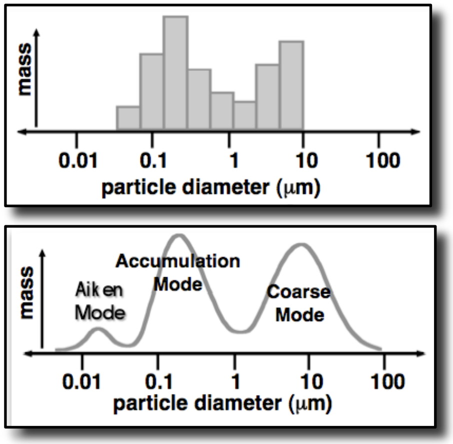
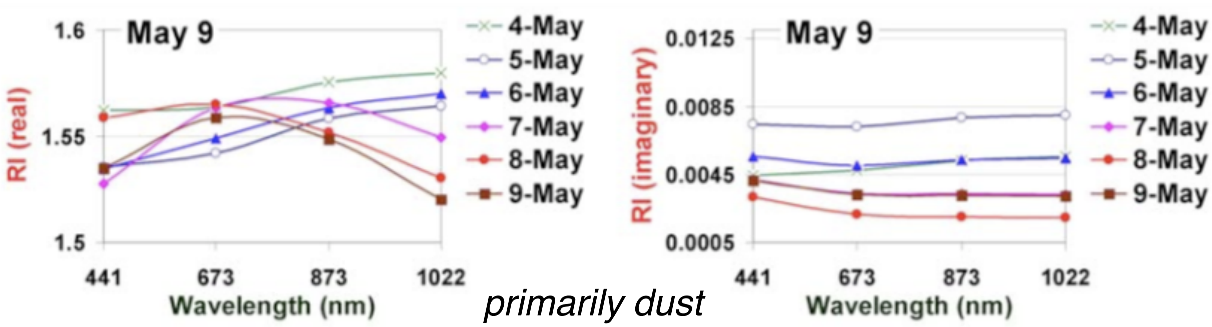
- **Bulk (GOCART):** converts bulk mass into assumed modal distribution, then divides mass into 8 sections

NOAA



- Refractive indices may need updating
  - Range of values reported in the literature
  - Do not assume wavelength dependence of refractive indices



primarily dust

from Prasad and Singh, JGR, 2007

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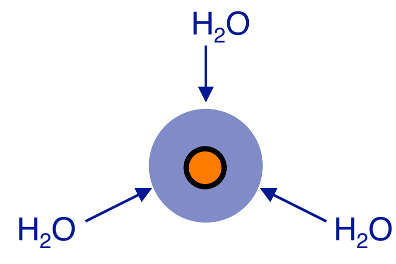
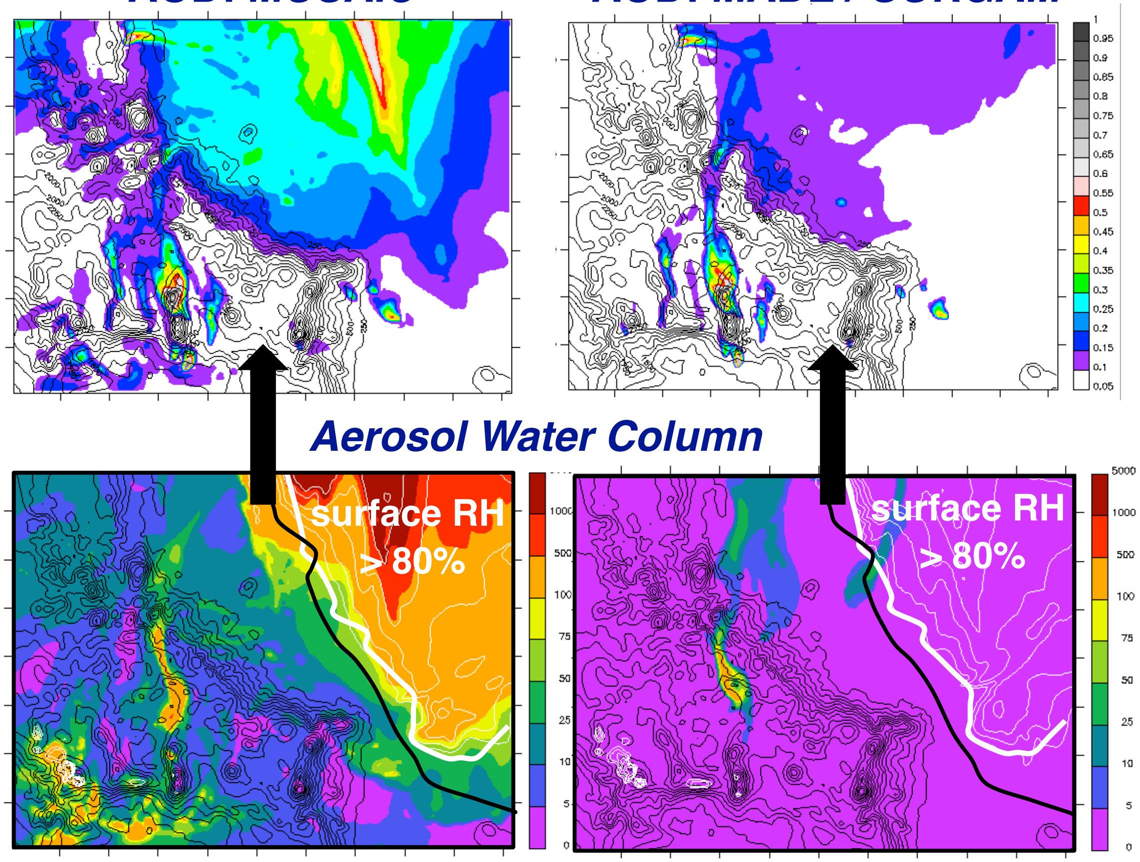
Importance of Aerosol Water

- Aerosol water will have a big impact on optical properties
- Aerosol water depends on relative humidity (RH); thus, predictions of RH need to be monitored when evaluating aerosol direct radiative forcing



AOD: *MOSAIC* **AOD: *MADE / SORGAM***

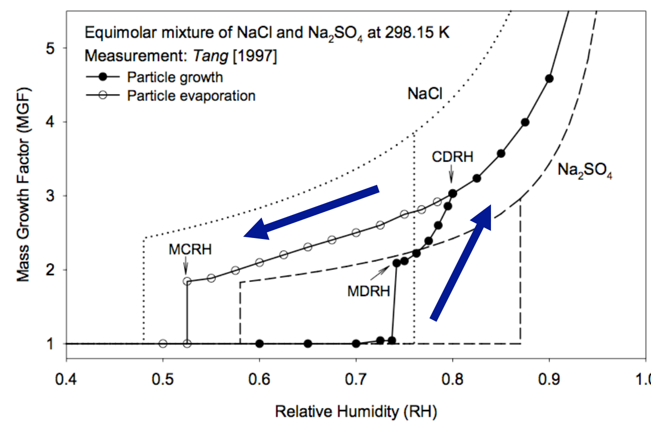


Aerosol Water

GOCART: diagnosed from RH using *Petters and Kreidenweiss* [2007]

MADE / SORGAM: diagnosed

MOSAIC: prognostic specie that accounts for hysteresis effect



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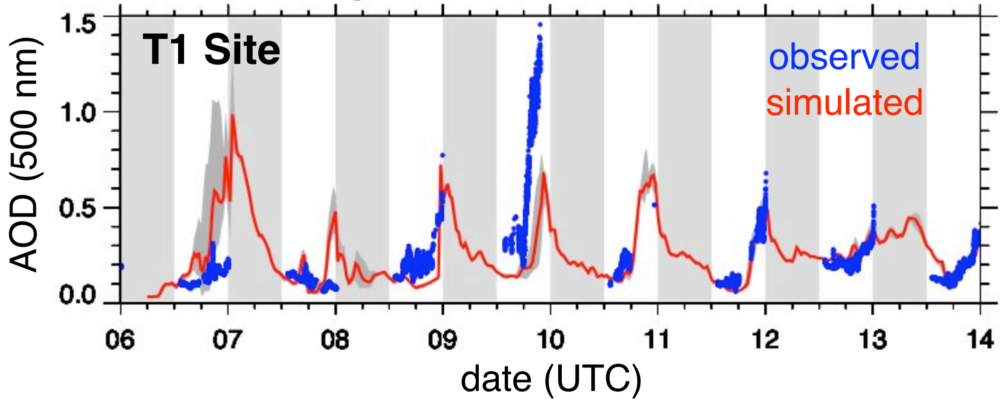
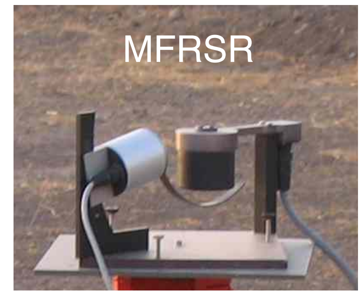
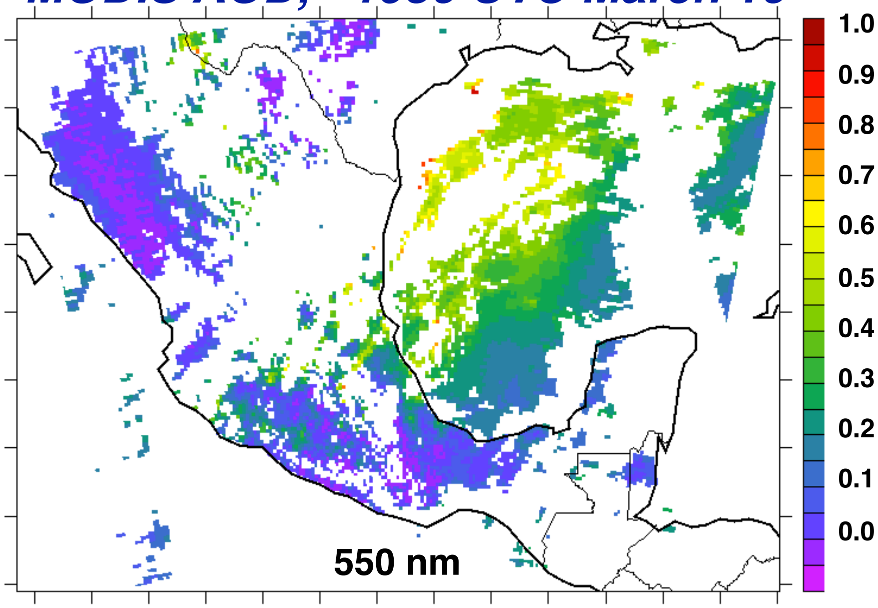
Example 1: *Aerosol Optical Depth*

AOD during March 2006 for MILAGRO Field Campaign

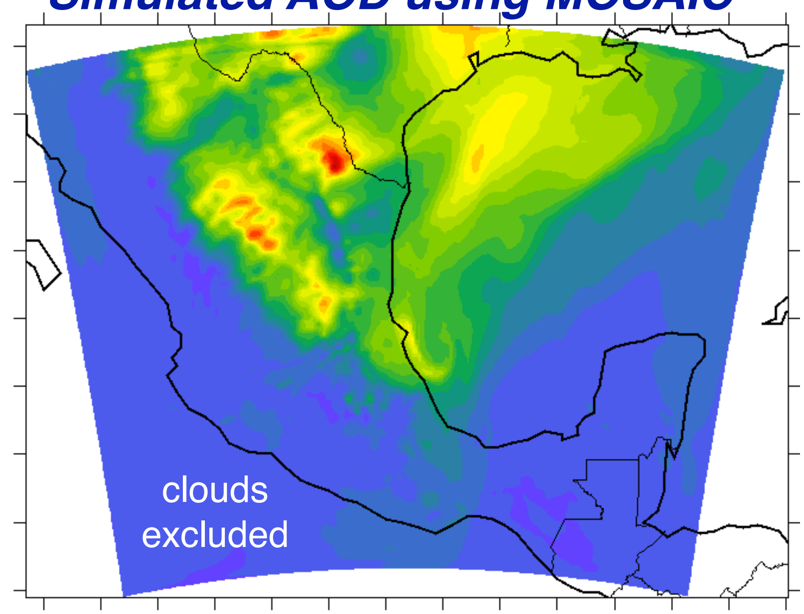




MODIS AOD, ~1930 UTC March 10



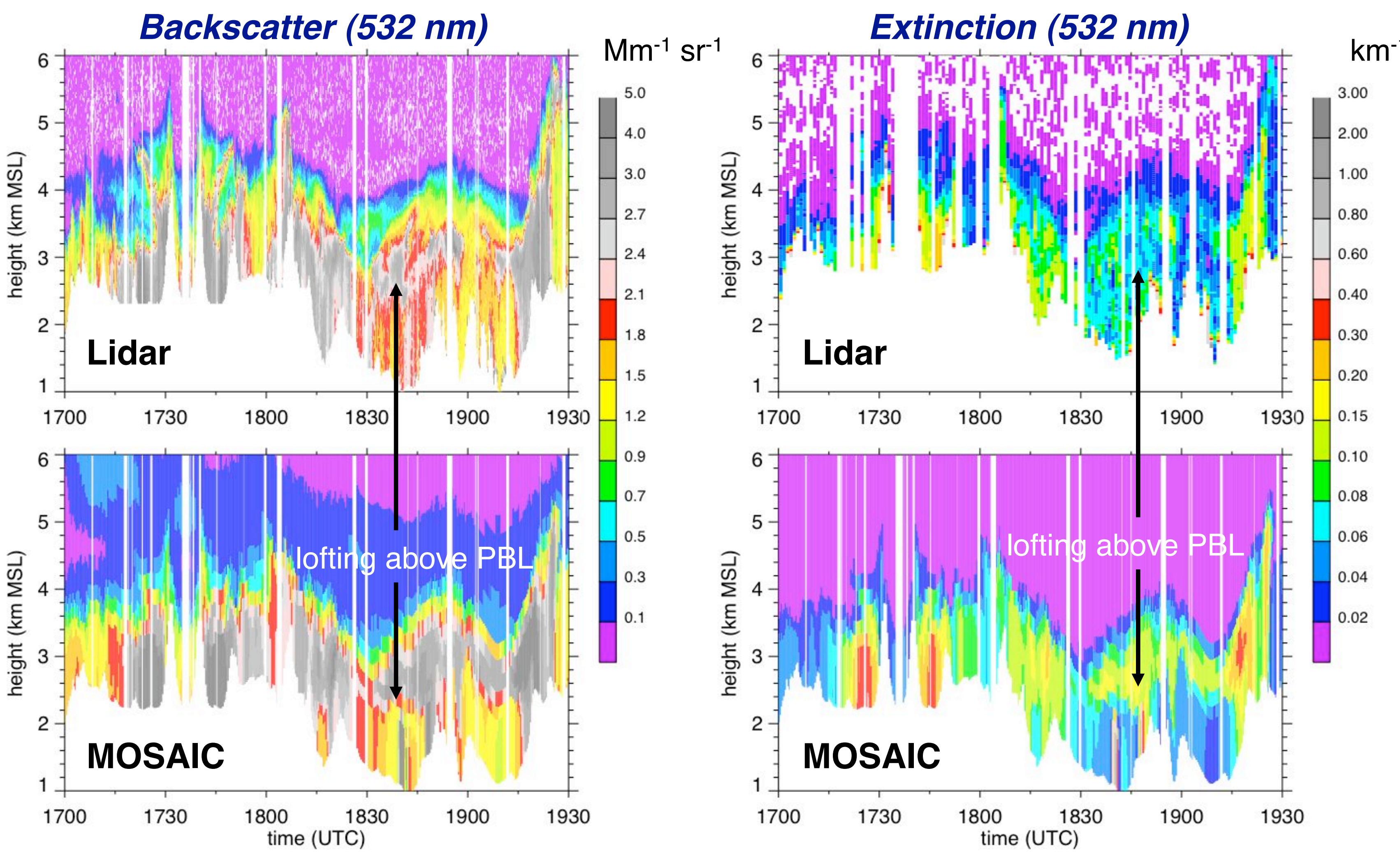
Simulated AOD using MOSAIC



Use Angstrom Exponent to get values at 550 nm from 500 and 600 nm computations

Example 2: *Backscatter and Extinction Profiles*

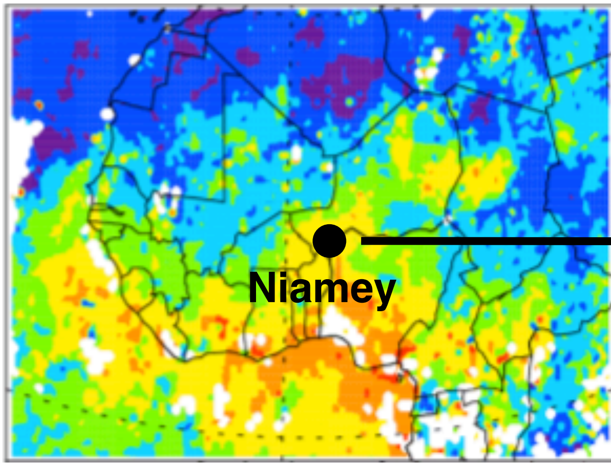
NASA B200 Aircraft Flight Path 13 March 2006 during MILAGRO



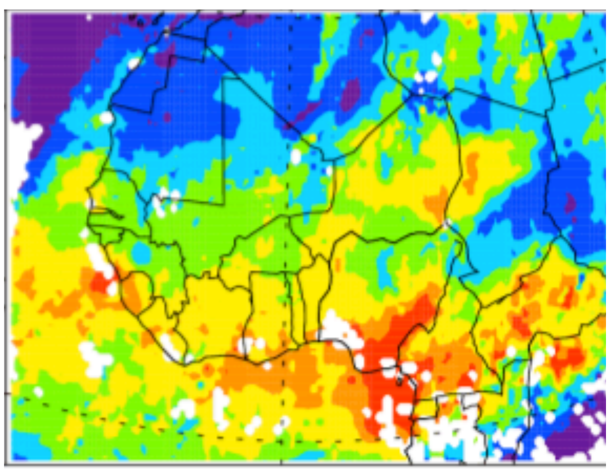
Use Angstrom Exponent to get values at 550 nm from 500 and 600 nm computations

Example 3: *Radiative Heating Rate*

Averaged Terra-MISR AOD
January 2006

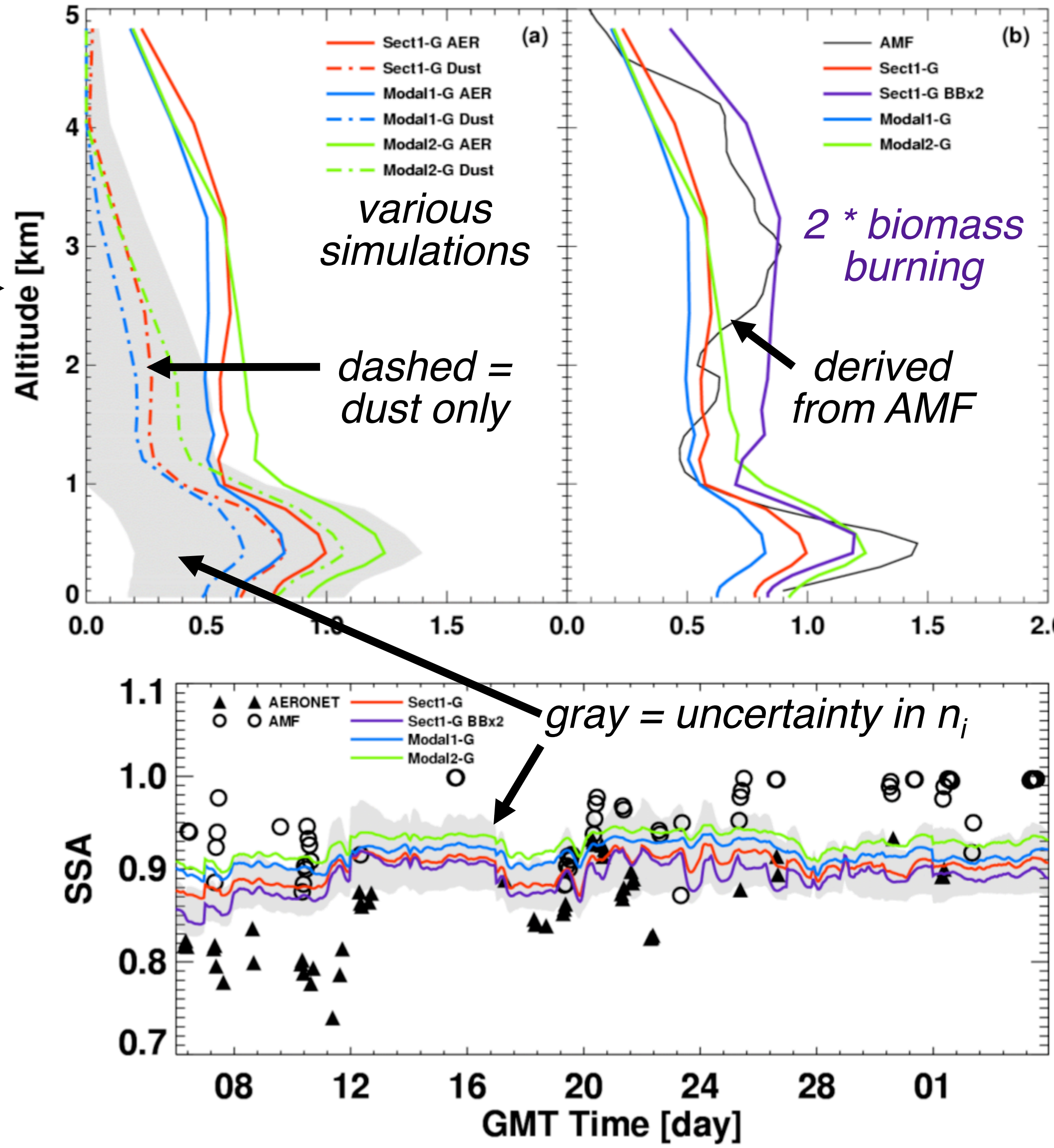


Simulated AOD
MOSAIC + GOCART dust



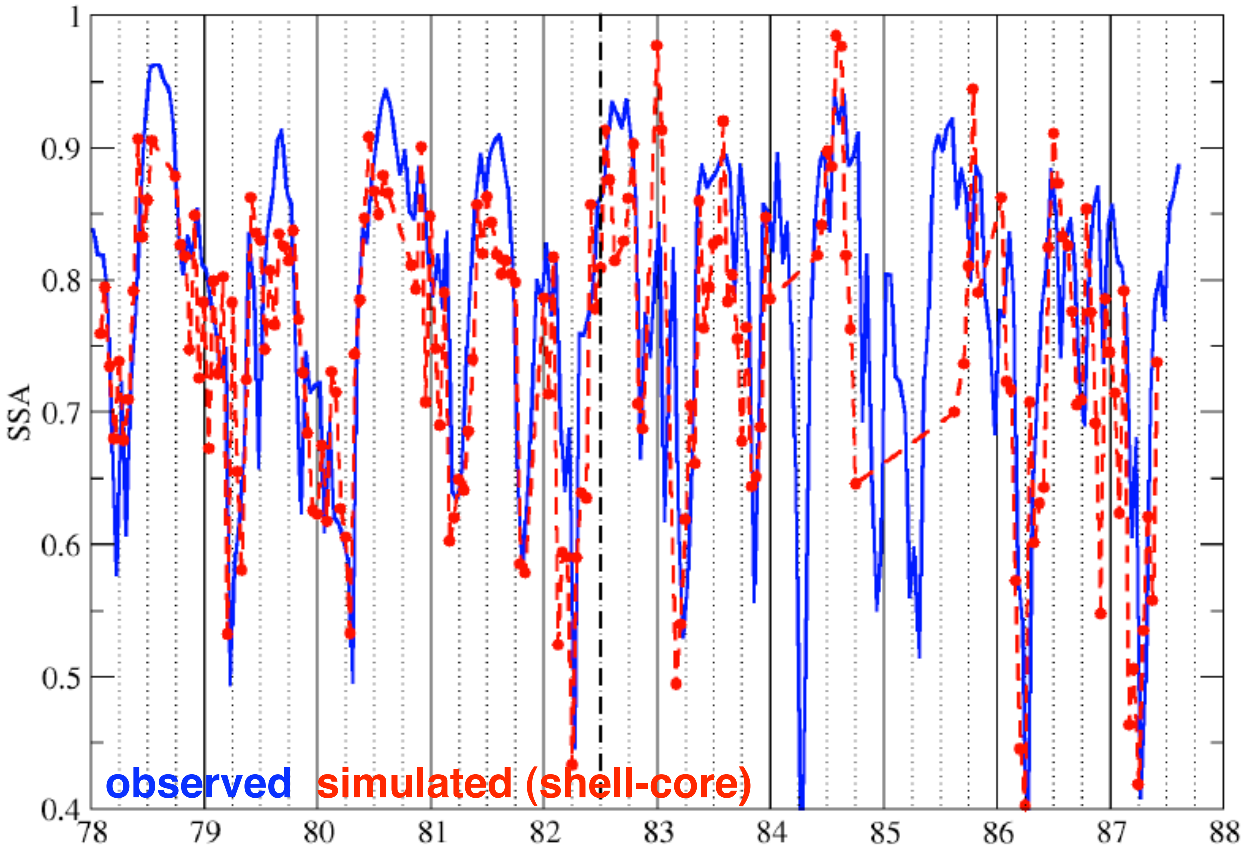
from Zhao et al., 2010

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



Example 4: *Single Scattering Albedo*

SSA during March 2006 MILAGRO Field Campaign



Aerosol optical property modules driven by measurements of particulate mass, composition, and size distribution (some uncertainties in data)

Most of the error in scattering

Other mixing rules obtain similar results

From offline version of aerosol optical property modules in WRF-chem, Barnard et al. 2010

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Settings in namelist.input

- *ra_physics* = 2, affects only radiation computed by Goddard scheme
- *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, and MOSAIC options

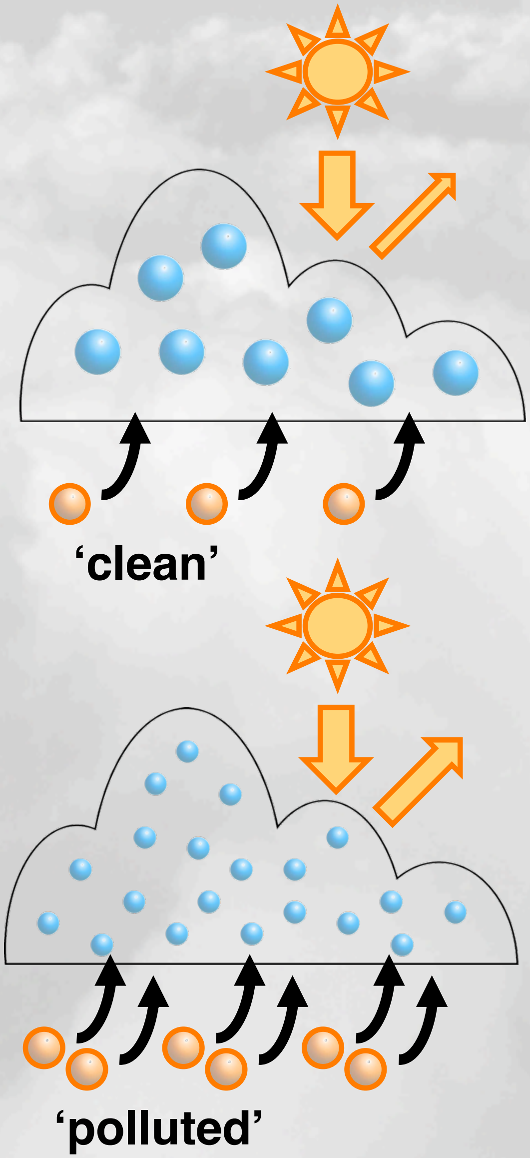
Coming Soon (next release):

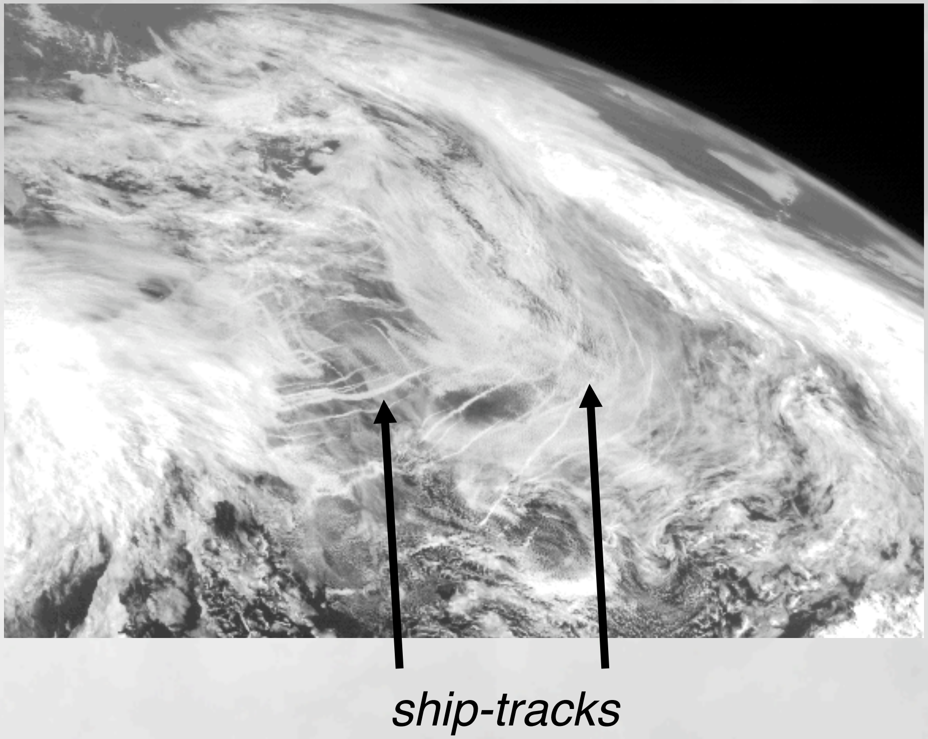
- Coupling aerosol effects to RRTMG
- Mie subroutine that is computationally more efficient

Research:

- Mie subroutine that handles non-spherical particles
- Wavelength dependence of refractive indices
- Refractive indices for organic aerosol species (primary vs secondary)

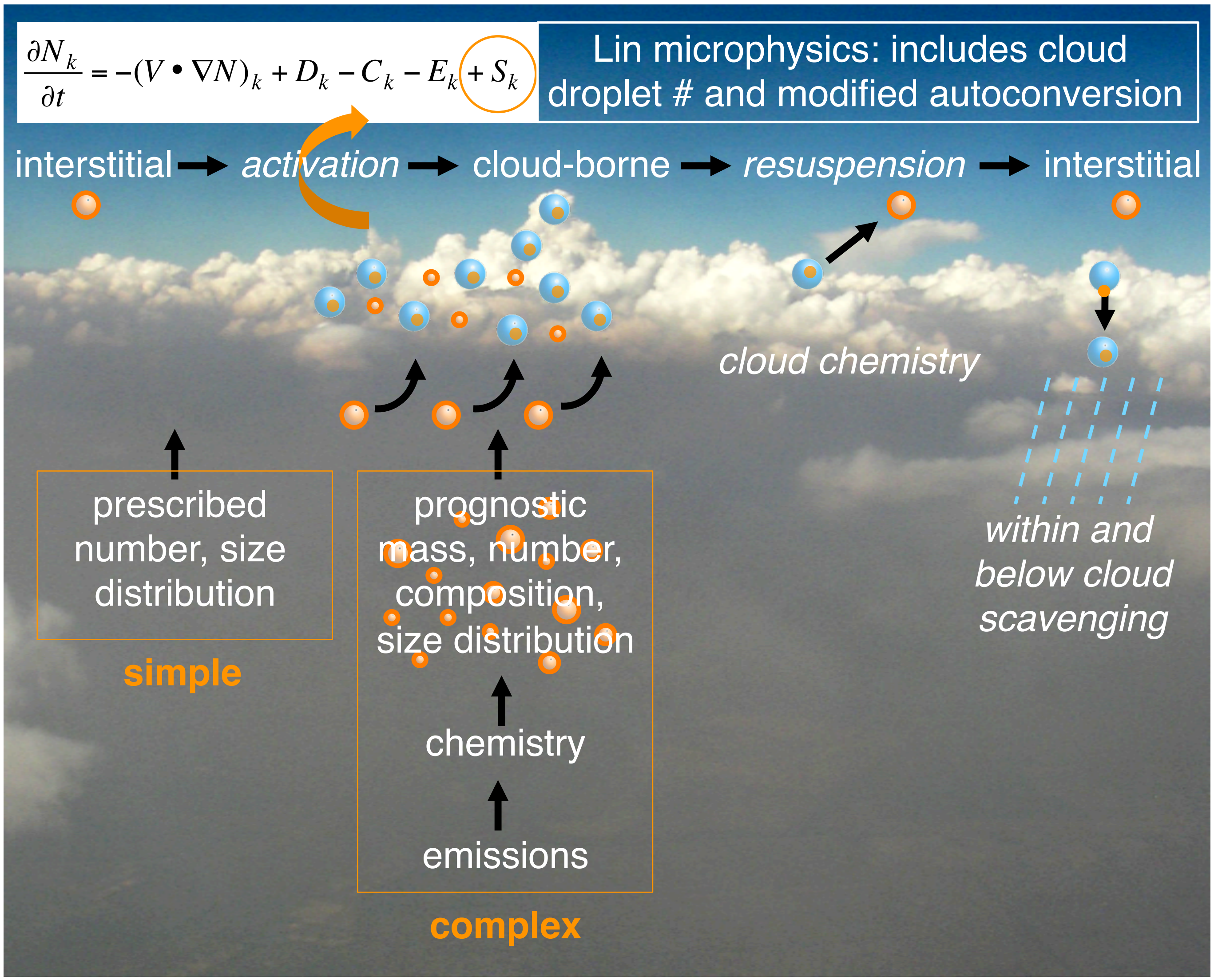
Part 2: Aerosol Indirect Forcing





Cloud-Aerosol Interactions

General Description and Assumptions



Simple:

- chem_opt = 0
- progn = 1
- naer = specified
- mp_physics = 2
- ra_sw_physics = 2

Complex:

- chem_opt = 9 - 12
- progn=1
- naer = ignored
- mp_physics = 2
- ra_sw_physics = 2

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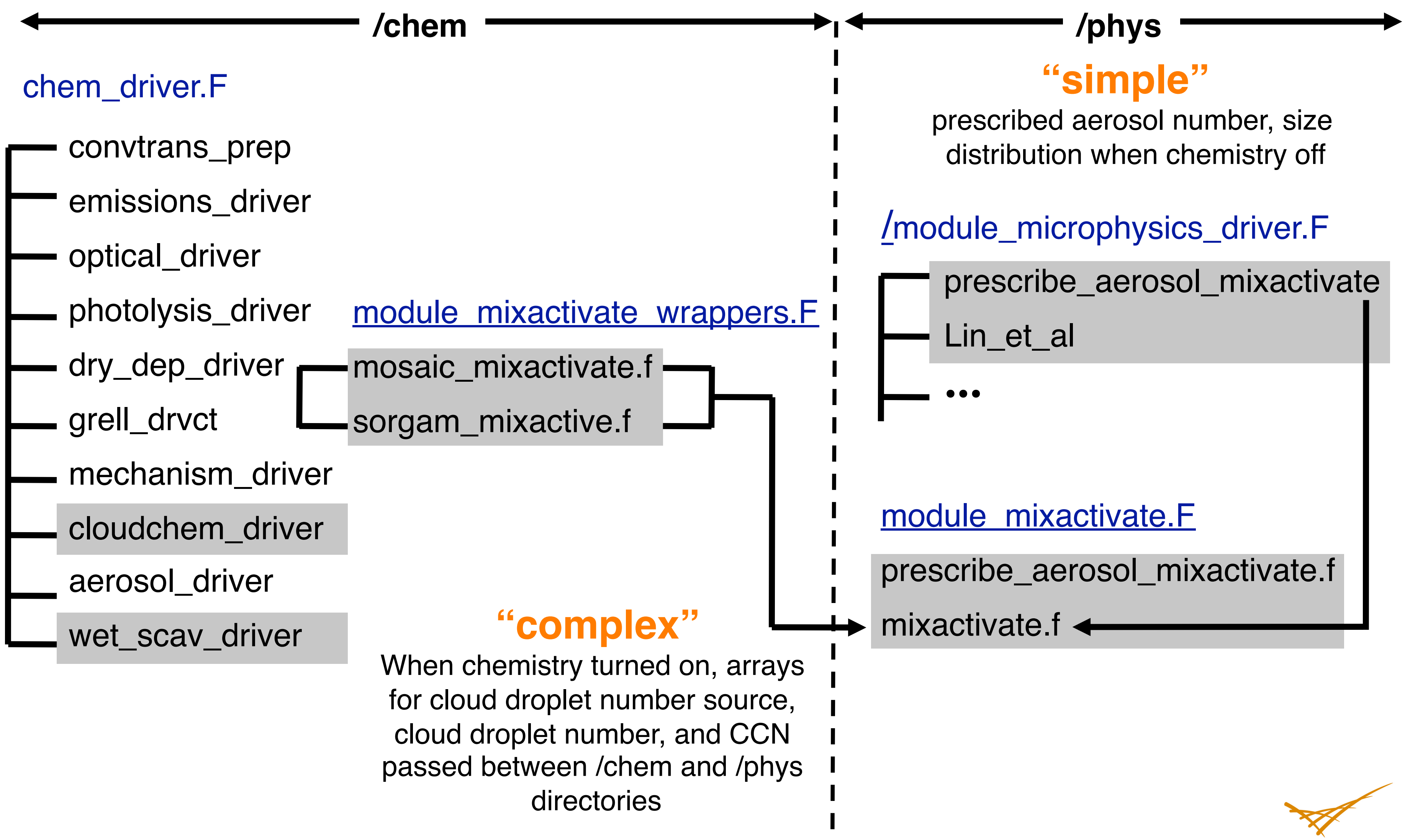
Flow Chart



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Aerosol Species

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

interstitial		cloud-borne		interstitial
so4_a01	*activation* →	so4_cw01	*resuspension* →	so4_a01
so4_a02	"growing cloud"	so4_cw02	"shrinking cloud"	so4_a02
…	cldfra - cldfra_old > 0	…	cldfra_old > cldfra	…
no3_a01		no3_cw01		no3_a01
no3_a02		no3_cw02		no3_a02
…		…		…
num_a01		num_cw01		num_a01
num_a02		num_cw02		num_a02

8 bins x 12 species + hysw + water = **112**

8 bins x 12 species = **96**

computational expense associated mostly with transporting scalars

- similar for MADE/SORGAM: so4aj → so4cwj → so4aj

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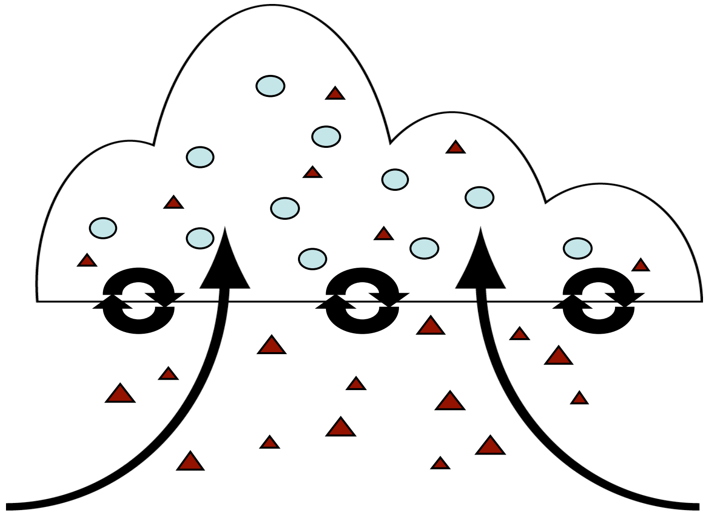
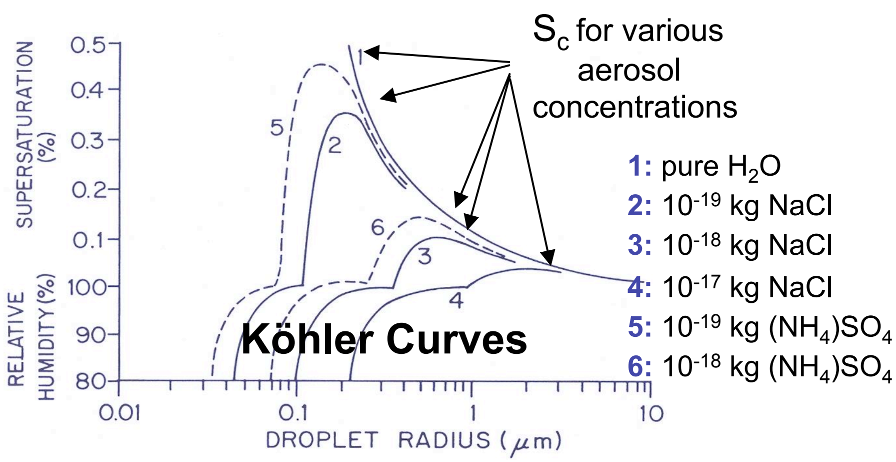
Activation



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



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.

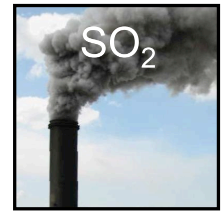
For each vertical velocity, peak S_{max} depends on aerosol size and composition [*Abdul Razzak and Ghan,* 2000, 2002]. Activation fraction based distribution of $\mathbf{S_c}$ of the bin/mode - simply a fraction of aerosol mass or number in the bin/mode having $\mathbf{S_c} < \mathbf{S_{max}}$

Hygroscopicity

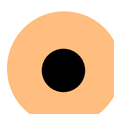
- Hygroscopic properties depend on particulate composition:
  - hygro_so4_aer = 0.5
  - hygro_no3_aer = 0.5
  - hygro_nh4_aer = 0.5
  - hygro_oc_aer = 0.14 (*some OC may be hygrophilic – subject of research)*
  - hygro_bc_aer = 1.0e-6
  - hygro_oin_aer = 0.14
  - hygro_ca_aer = 0.1
  - hygro_co3_aer = 0.1
  - hygro_msa_aer = 0.58
  - hygro_cl_aer = 1.16
  - hygro_na_aer = 1.16

hygrophobic

hygrophilic



- Activation depends on volume weighted bulk hygroscopicity, prior to call to mixactivate.f in module_mixactivate_wrappers.F



What about coating?

- For *chem_opt* = 0 and *nprog* = 1, hygroscopicity set to 0.5

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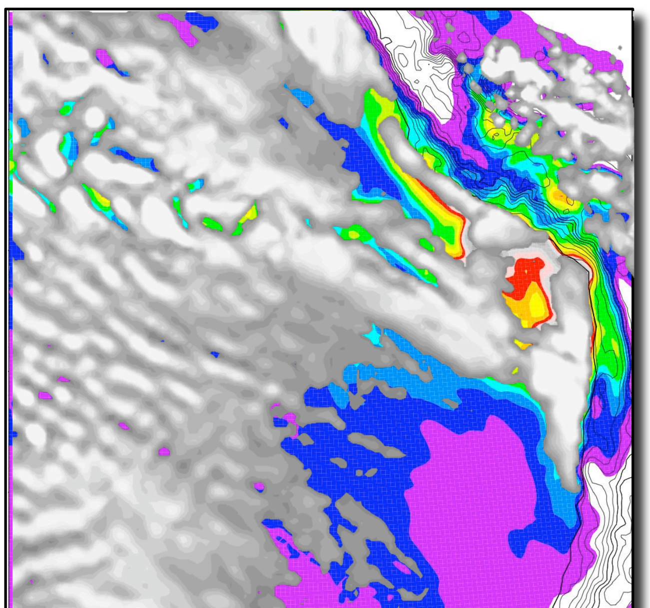
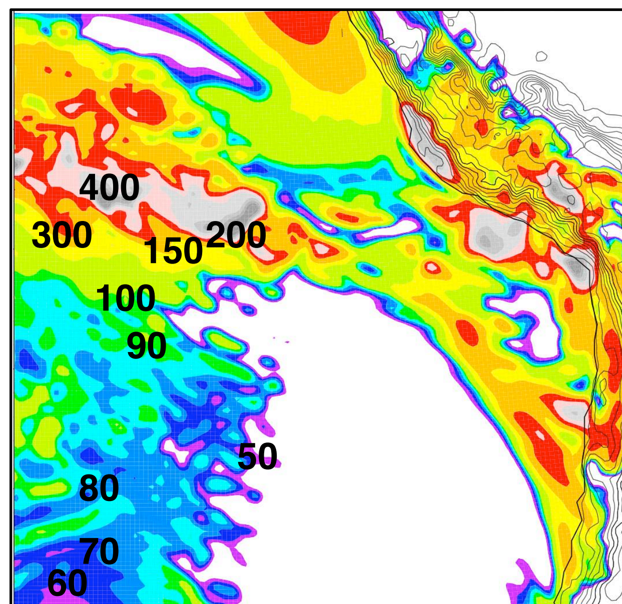
Cloud Condensation Nuclei

- 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})



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Cloud Droplet Number

- converted Lin et al. microphysics scheme (*mp_physics* = 2) to a two-moment treatment (mass & number)

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

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

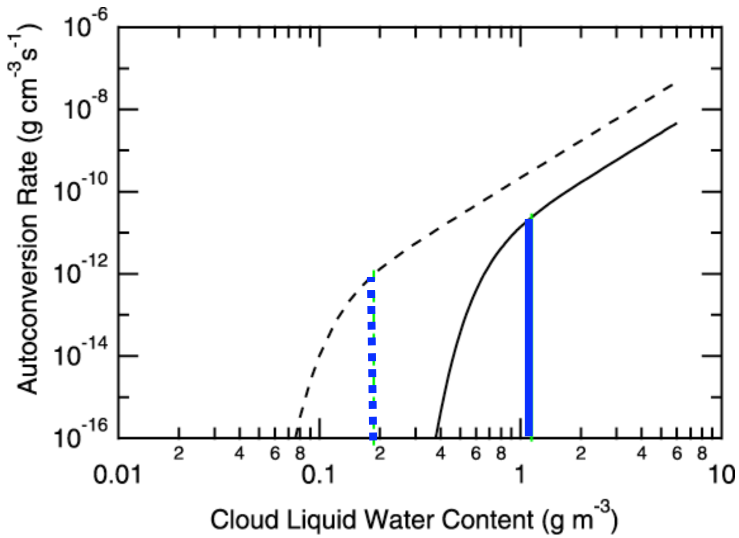
E_k - droplet loss due to evaporation

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 shortwave radiation scheme (*ra_sw_physics* = 2)

Autoconversion

- autoconversion = coalescence of cloud droplets to form embryonic rain drops
- replaced autoconversion parameterization employed by Lin et al. microphysics (*mp_physics* = 2) with *Liu et al.* [2005] parameterization
  - adds droplet number dependence
  - physically based w/o tunable parameters



Black: New Liu et al. parameterization
Blue: Kessler-type parameterization, similar to default Lin et al. scheme

Dashed: N = 50 cm^{-3}
Solid: N = 500 cm^{-3}

Adapted from Fig. 3, Liu et al., 2005, GRL.

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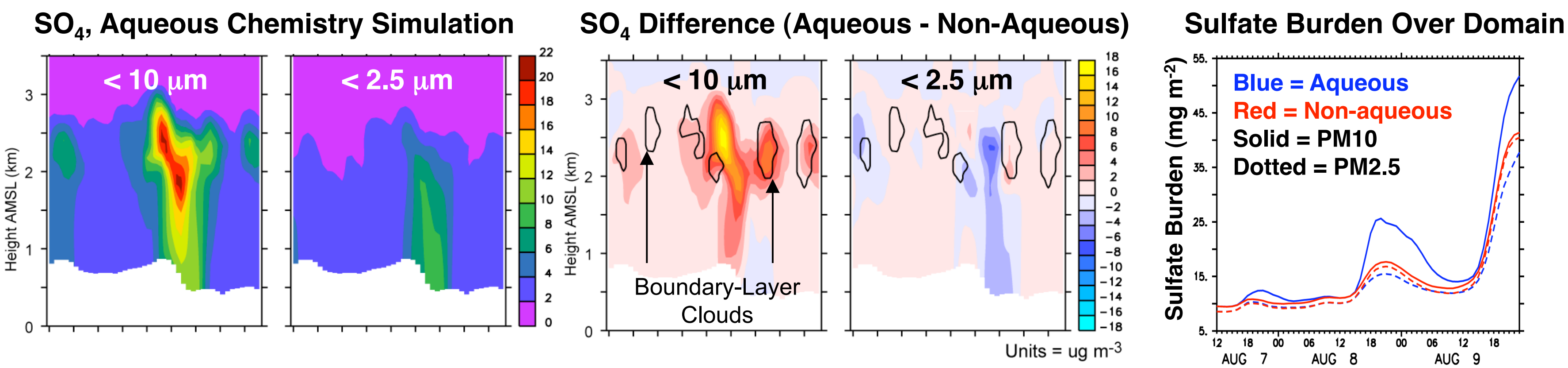
Aqueous Chemistry

- Bulk cloud-chemistry module of *Fahey and Pandis* [2001]
  - compatible with MOSAIC and MADE/SORGAM
  - cloudchem_driver.F, module_cmu_*
- 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

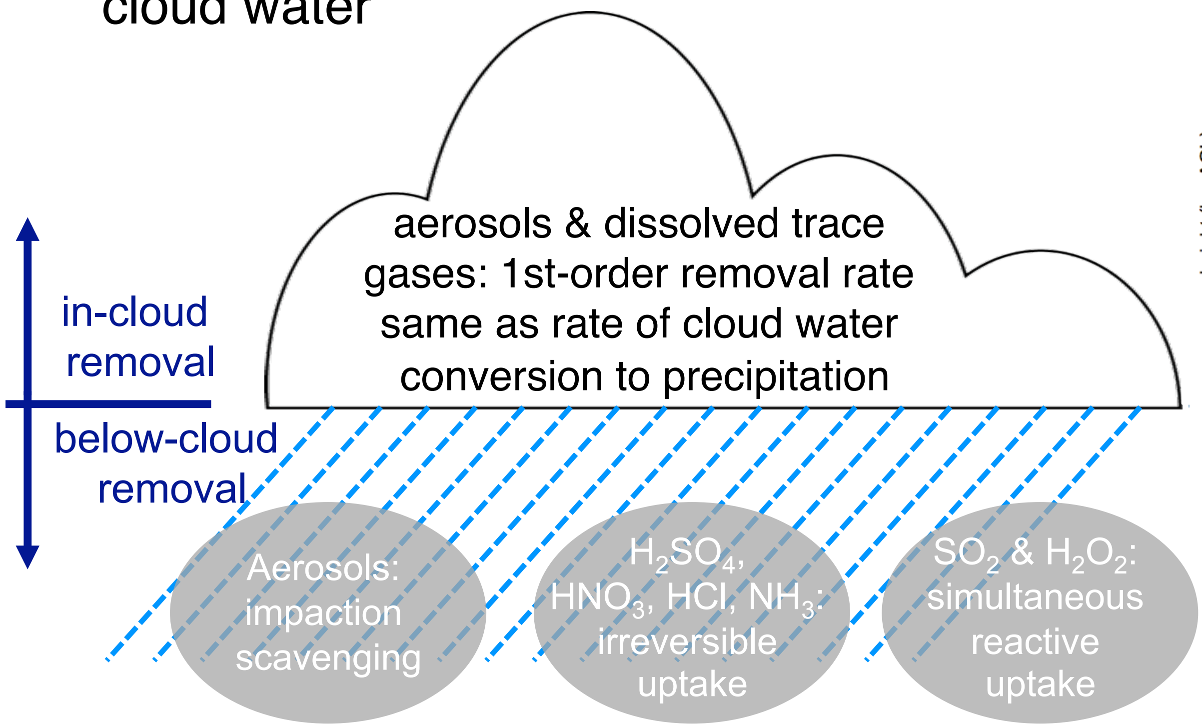
Vertical Cross-Section Though Power Plant SO_2 Plume



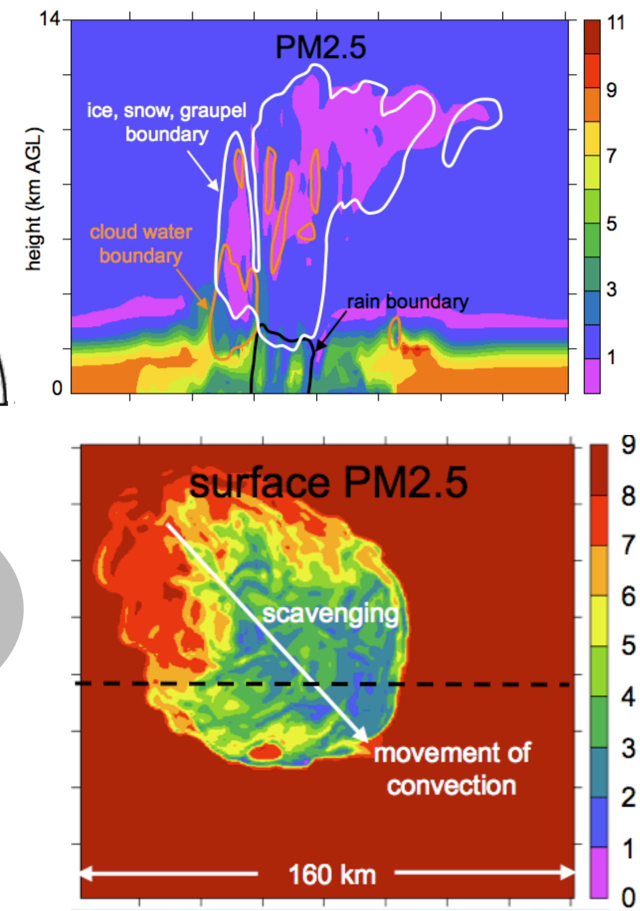
- Aqueous chemistry in module_ctrans_grelldrct.F being developed

Wet Removal - Scavenging

- 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

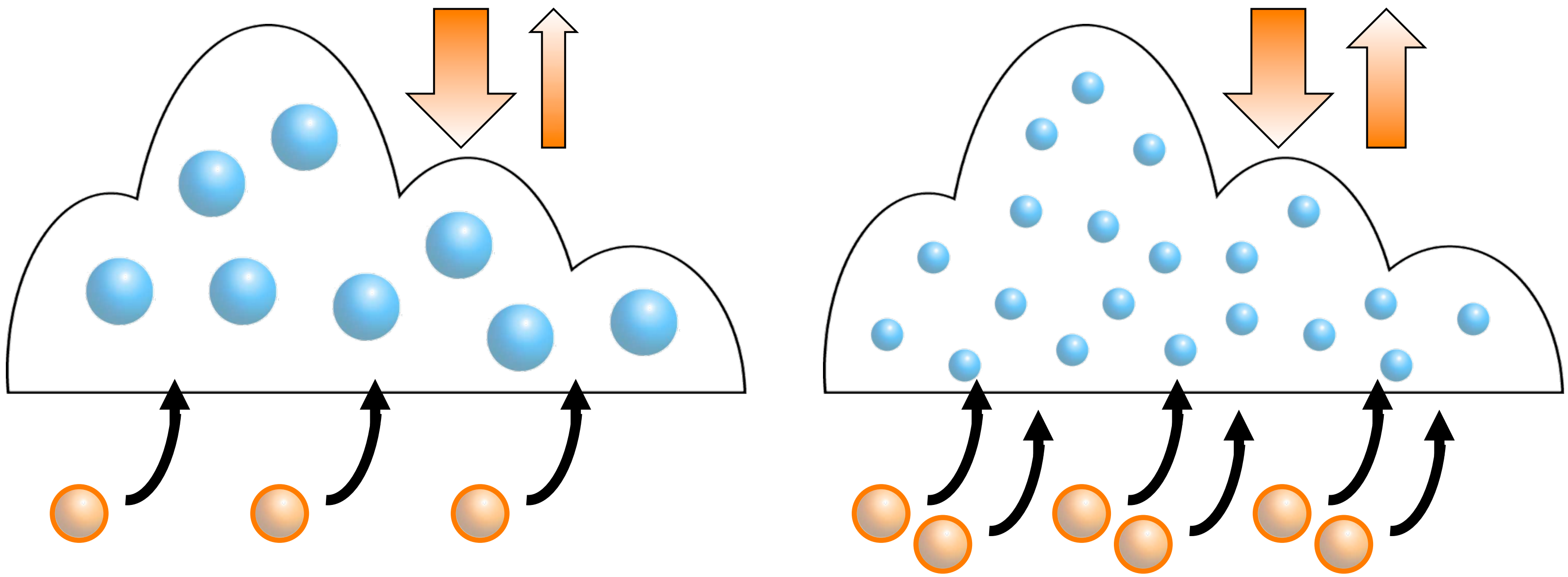


scavenged aerosols and gases instantly removed (but not saved) see *Easter et al.* [2004], also aerosols are not resuspended for evaporating rain



First Indirect Effect

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



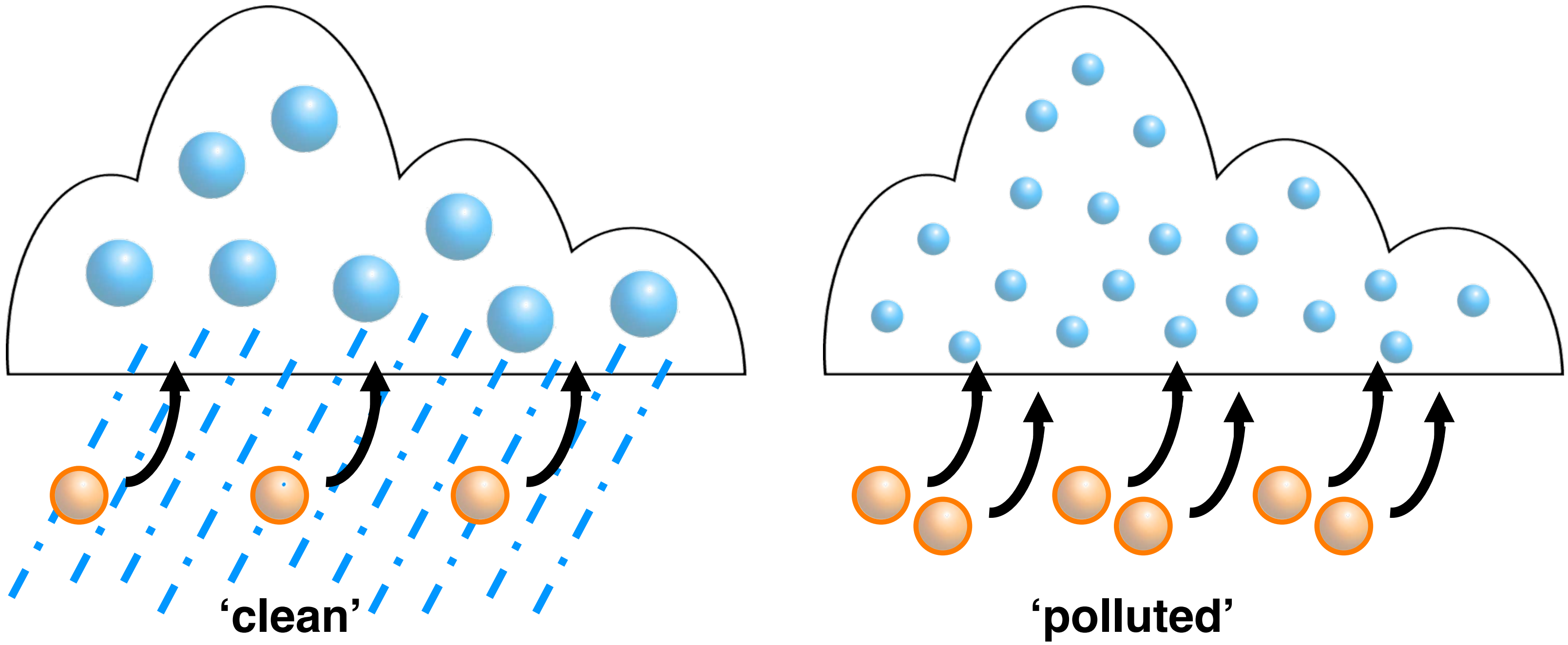
‘clean’ **‘polluted’**

qndrop → gsfcwrad.f → reff → sorad.f → taucld

module_ra_gsfcsw.F effective radius cloud albedo

Second Indirect Effect

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

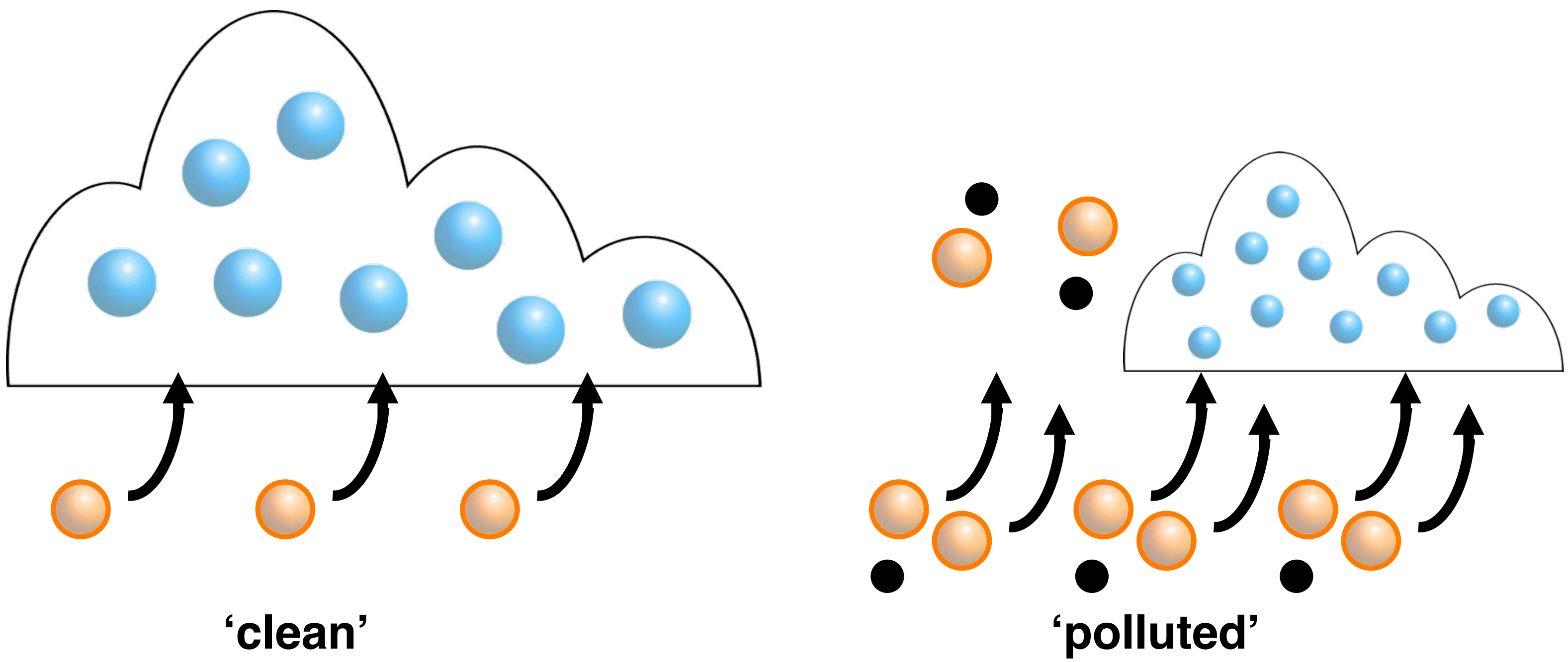


qndrop → cldphy_1d.f → praut → qr → precr

module_mp_lin.F | autoconversion rate | rain mixing ratio | precipitation rate

Semi-Direct Effect

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



τ, ω_o, g → gsfcwrad.f → sorad.f → flx → ttend2d → θ tendency

module_ra_gsfcsw.F

solar uv and ir fluxes

heating rate

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Interactions not Treated

- **First Dispersion Effect:** Affects cloud optical depth via the influence of aerosols on the width of the droplet size distribution, with no change in water content of cloud
- **Second Dispersion Effect:** Affects cloud optical depth via the influence of aerosols on the width of the droplet size distribution and hence initiation of precipitation
- **Glaciation Indirect Effect:** Influence of aerosol on conversion of haze and droplets to ice crystals, and hence on cloud optical depth and initiation of precipitation

(Ice processes are a current research topic for PNNL, NCAR, others)

pointer system already in place to handle ice-borne species

so4_a01	→	so4_cw01	→	so4_ci01
so4_a02		so4_cw02		so4_ci02
…		…		…
num_a01		num_cw01		num_ci01
num_a02		num_cw02		num_ci02

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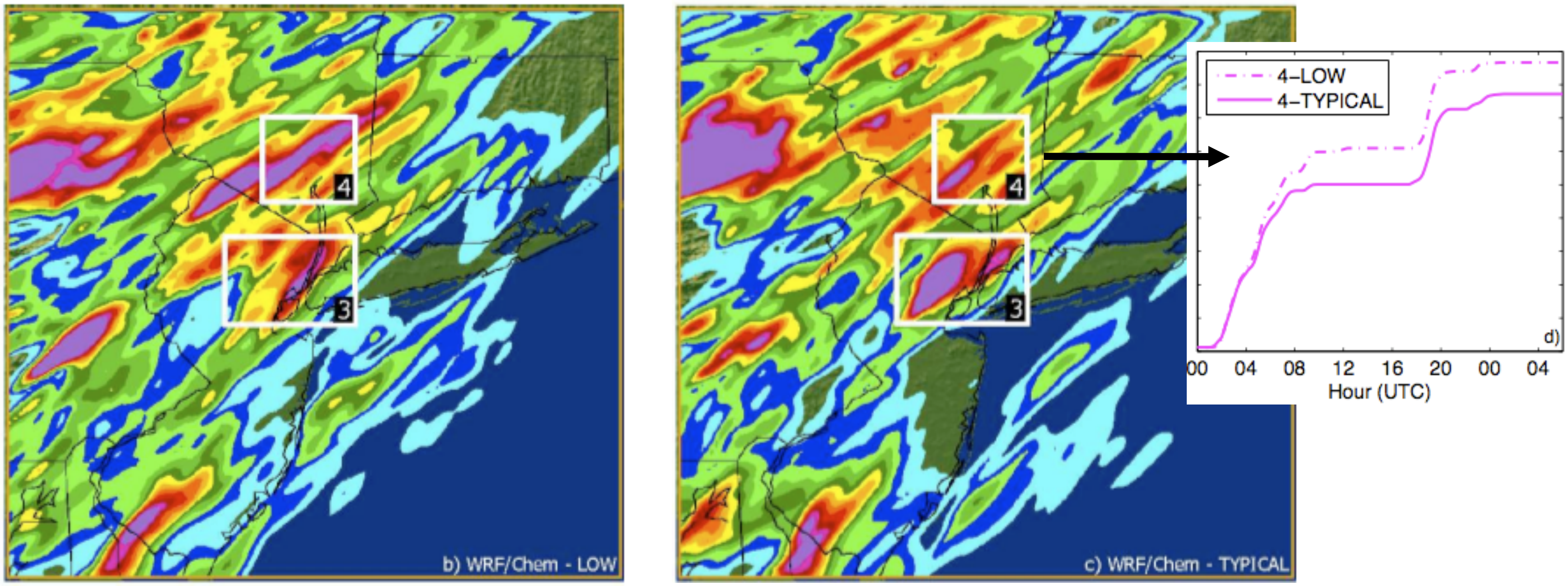
Example 1: *Deep Convection*

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

WRF-chem: low emissions ***WRF-chem: typical emissions***



- Ntelekos, A., J.A. Smith, L. Donner, J.D. Fast, E.G. Chapman, W.I. Gustafson Jr., and W.F. Krajewski, 2008: The Effects of aerosols on intense convective precipitation in the northeastern U.S. *Q. J. Roy. Meteor. Soc.,* 135, 1367-1391.

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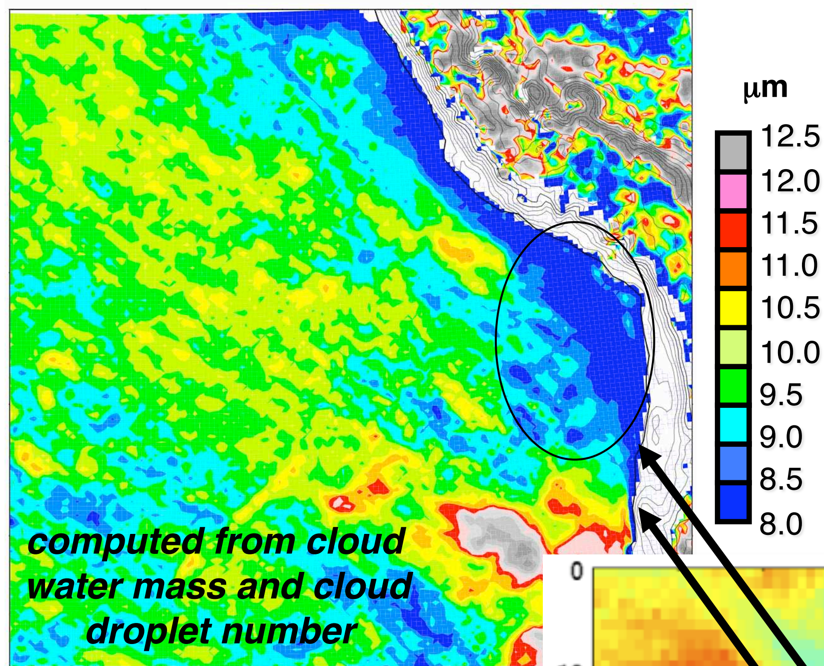
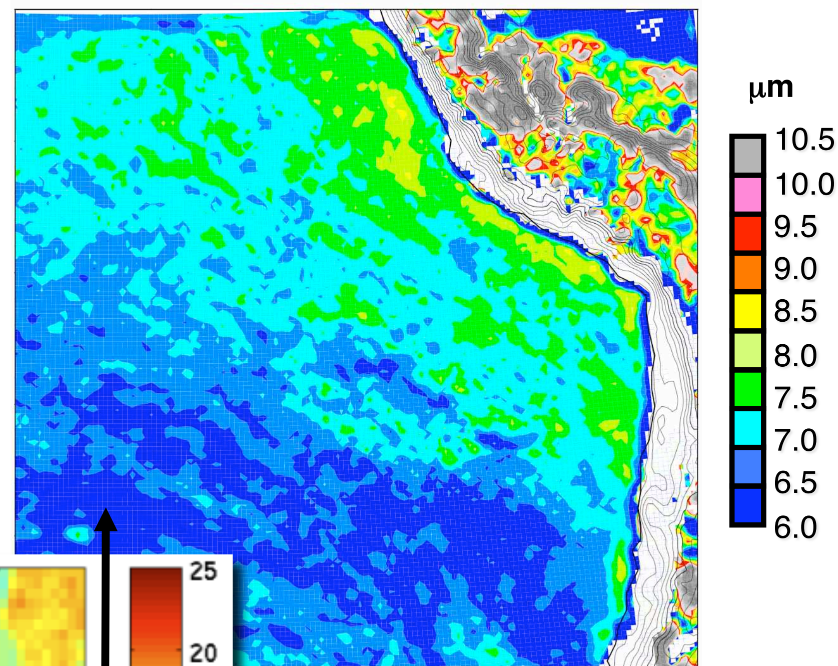
Example 2: *Droplet Effective Radius*

October 2006 Average at 12 UTC

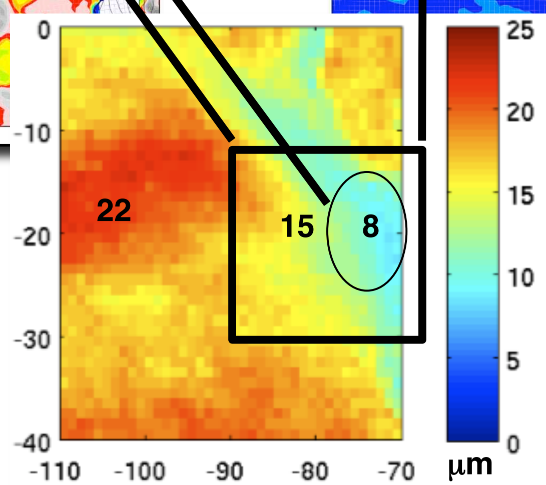
Full Chemistry Simulation



Prescribed Aerosol # Simulation



depends on # chosen !



Mean MODIS Cloud Droplet Effective Radius October 2006

from Matthew Wyant (UW)

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Settings in namelist.input

Simple:

- *chem_opt* = 0
- *naer* = specified value

Complex:

- *chem_opt* = 9 - 12, cloud-phase aerosols for MOSAIC and MADE/SORGAM
- *cldchem_onoff* = 1, turns on cloud chemistry
- *wetscav_onoff* = 1, turns on wet scavenging

Both:

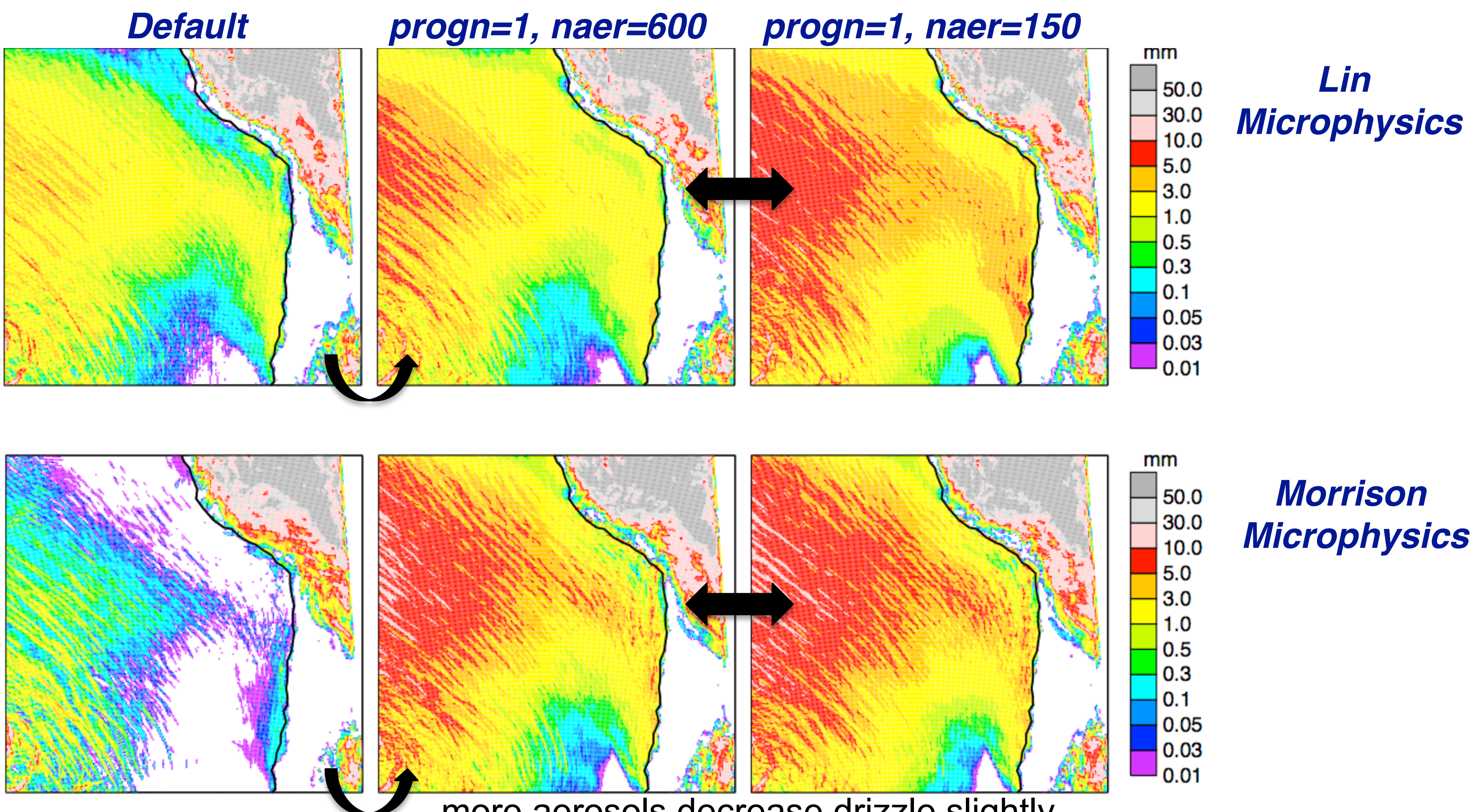
- *mp_physics* = 2, cloud-aerosol interactions only for Lin scheme
- *progn* = 1, turns on prognostic cloud droplet number

Coming Soon:

- *mp_physics* = 8, cloud-aerosol interactions for Thompson scheme
- *mp_physics* = 10, cloud-aerosol interactions for Morrison scheme

Morrison and Thompson Schemes

3-Day Accumulated Precipitation During VOCALS Field Campaign



more aerosols decrease drizzle slightly

Need to redo Thompson tests with v3.2, some of the differences in precipitation due to differences in autoconversion

Comparing Options

Care must be taken in quantifying direct and indirect effects!

- **Direct Effect:**
  - Run with *aer_ra_feedback* on versus off, or
  - Add code to output clean-sky and dirty-sky from the same run
- **Indirect Effects:**
  - Comparing a *chem_opt* = 8 with a *chem_opt* = 10 for MOSAIC run **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
- **Indirect Effects Usage:**
  - Works with microphysics only – not cumulus parameterizations
  - There are proposed efforts to extend cloud-aerosol interactions to cumulus parameterizations (for Δx > 10 km); need to worry about double counting
  - In addition to *Abdul-Razaak and Ghan* [2000, 2002], other schemes have been used to compute aerosol activation [*Foutoukis and Nenes*, 2005]

References

WRF-chem Papers Describing Aerosol-Radiation-Cloud Interactions (see our web site)

- Fast, J.D, W.I. Gustafson, Jr., R.C. Easter, R.A. Zaveri, J.C. Barnard, E.G. Chapman, and G.A. Grell, 2006: Evolution of ozone, particulates, and aerosol direct forcing in an urban area using a new fully-coupled meteorology, chemistry, and aerosol model. *J. Geophys. Res.,* 111, doi:10.1029/2005JD006721.
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- Barnard, J.C., J.D. Fast, G.L. Paredes-Miranda, P.W. Arnott, and A. Laskin, 2010: Technical Note: Evaluation of the WRF-Chem “aerosol chemical to aerosol optical properties” module using data from the MILAGRO campaign. *Atmos. Chem. Phys*. *Discuss.*, 10, 8927-8961.
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- Matsui, H., M. Koike, Y. Kondo, N. Takegawa, Y. Miyazaki, J.D. Fast, U. Poschl, R.M. Garland, A. Wiedensohler, N. Sugimoto, and T. Zhu, 2010: Spatial and temporal variations of aerosols around Beijing in summer 2006: 2. Local and column aerosol optical properties. In press, *J. Geophys. Res*.

More on the way evaluating performance and developing new techniques using MILAGRO, VOCALS, CHAPS, ISDAC/ARTAS, CARES/CalNex field campaign data

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