### **Aerosol Direct and Indirect Forcing**

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## Background

#### First A Brief History ...

- CBM-Z, Fast-J, and an early version of MOSAIC in WRF-chem originated from an off-line chemical transport model
- Then, aerosol-radiation-cloud-chemistry interactions were added
- We are currently adding more capabilities, making modules more generic, and trying to follow WRF coding guidelines
- 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 atmospheric sciences: www.pnl.gov/atmospheric
- PNNL modules: www.pnl.gov/atmospheric/research/wrf-chem, including capabilities, current research, and publications



## **Part 1: Aerosol Direct Forcing**

Aerosols - Radiation

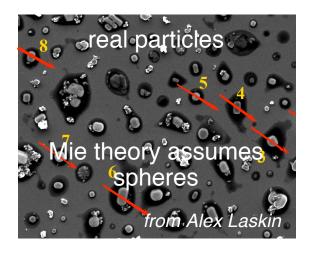


## **Aerosol Properties**

#### **General Description and Assumptions**



- $\tau$ ,  $\omega_0$ , and g function of wavelength, 300, 400, 600, 1000 nm
  - $\tau$  = TAUAER1, TAUAER2, TAUAER3, TAUAER4 in Registry
  - $\omega_o = WAER1$ , WAER2, WAER3, WAER4 in Registry
  - ➤ g = GAER1, GAER2, GAER3, GAER4 in Registry
- $\omega_0 = k_s / (k_a + k_s)$ ,  $k_s$  and  $k_a = scattering$  and absorption coefficients
- various methods of obtaining refractive index



#### Mass, composition, and size distribution:

- more mass → bigger radiative impact
- amount of black carbon → k<sub>a</sub>
- aerosol size **→** k<sub>s</sub>



### **Aerosol Direct Radiative Effects**

#### **General Description and Assumptions**



- 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



# **Coding Structure**

#### **Aerosol Optical Properties in WRF-chem v2.2**

```
chem_driver.F

emissions_driver.F

photolysis_driver.F

dry_dep_driver.F

\tau, \omega_o, g

buried in Fast-J and tightly coupled to MOSAIC
```

### More Generic Aerosol Optical Properties for WRF-chem v3

```
chem_driver.F

emissions_driver.F

optical_prep_sectional.f

optical_prep_modal.f

photolysis_driver.F

dry_dep_driver.F

TUV

radiation_driver.F

\sigma_{o}, \sigma_{o}, \sigma_{o}

\sigma_{o}
```

Example of making the code more generic and interoperable



## **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)
- 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
- 4 = Maxwell-Garnett exact
- 2 = Maxwell-Garnett approximate
- > 5 = Shell-Core

➤ 3 = Volume-Averaging exact

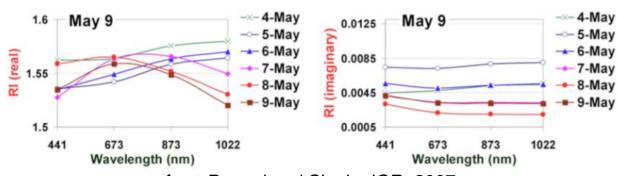


## **Assumptions**

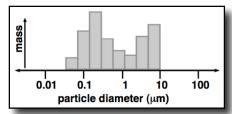
- Interfaces with MADE/SORGAM and MOSAIC only, but modifications needed for other aerosol models should be minor
- Sectional (MOSAIC): tested only with 4 and 8 size bins – should work if additional size bins are specified
- Modal (MADE/SORGAM): divides mass in modes into 8 sections - could divide into more sections to be more accurate

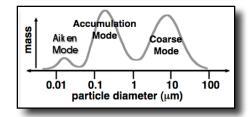


- Range of values reported in the literature
- Do not assume wavelength dependence of refractive indices





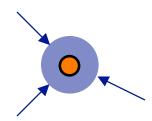


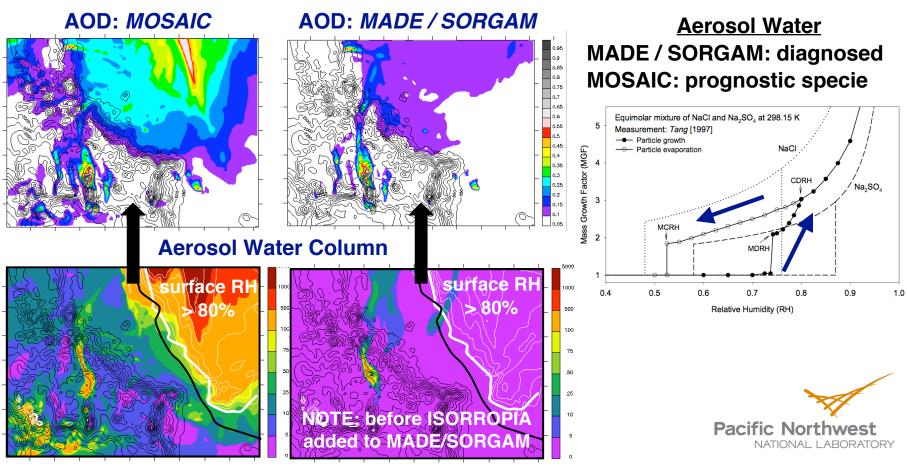




## **Importance of Aerosol Water**

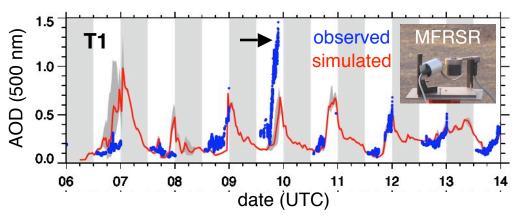
- Amount of aerosol water will have a big impact on  $\tau$ ,  $\omega_o$ , and g that depends on the relative humidity (RH)
- Therefore, predictions of RH need to be monitored when evaluating aerosol direct radiative forcing





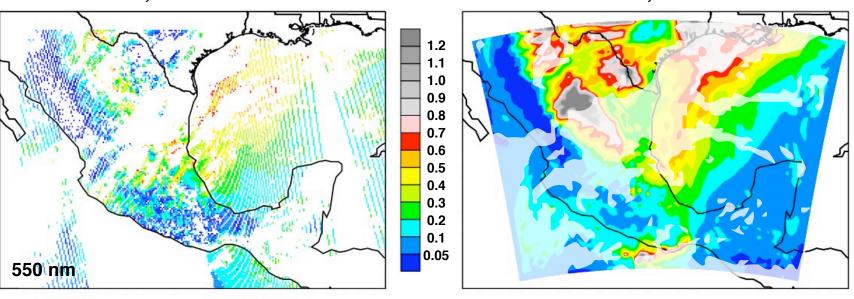
# **Example 1: AOD**

### **AOD during March 2006 for MILAGRO Field Campaign**



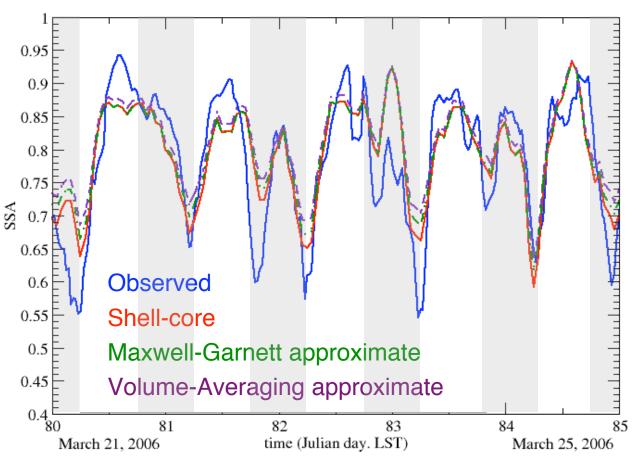
MODIS AOD, ~1930 UTC March 10

Simulated AOD, 20 UTC March 10



## **Example 2: SSA**

#### SSA during March 2006 MILAGRO Field Campaign



Aerosol optical property modules driven by measurements of particulate mass, composition, and size distribution

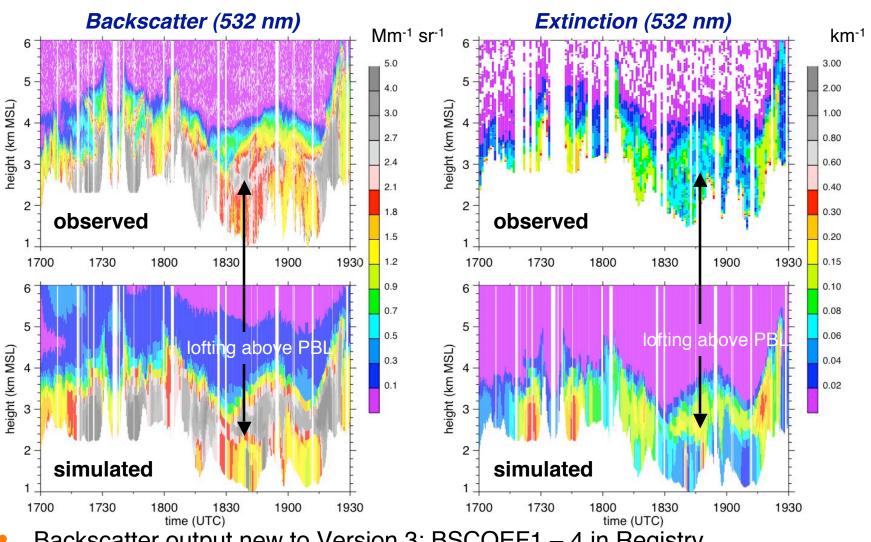
Some uncertainties in measured mass

Offline Version of Aerosol Optical Property
Modules in WRF-chem



## **Example 3: Backscatter**

### NASA B200 Aircraft Flight Path 13 March 2006 during MILAGRO



Backscatter output new to Version 3: BSCOEF1 – 4 in Registry

## 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 for either MADE/SORGAM and MOSAIC options

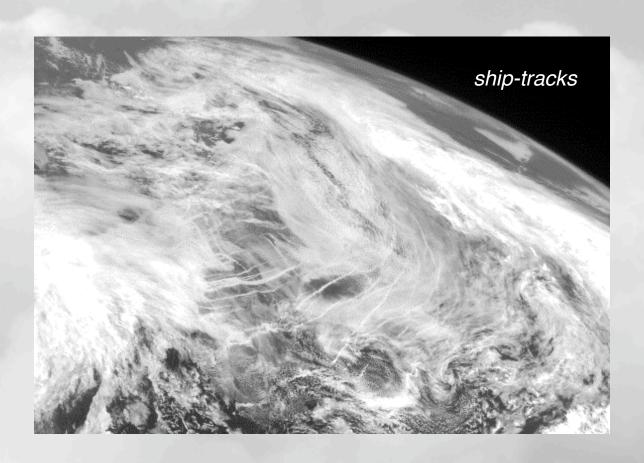
#### **Coming Soon:**

- Adapt CAM scheme ( $ra\_sw\_physics = 3$ ) by replacing  $\tau$ ,  $\omega_o$ , and g computed from off-line monthly averaged aerosols with on-line values
- Wavelength dependence of refractive indices



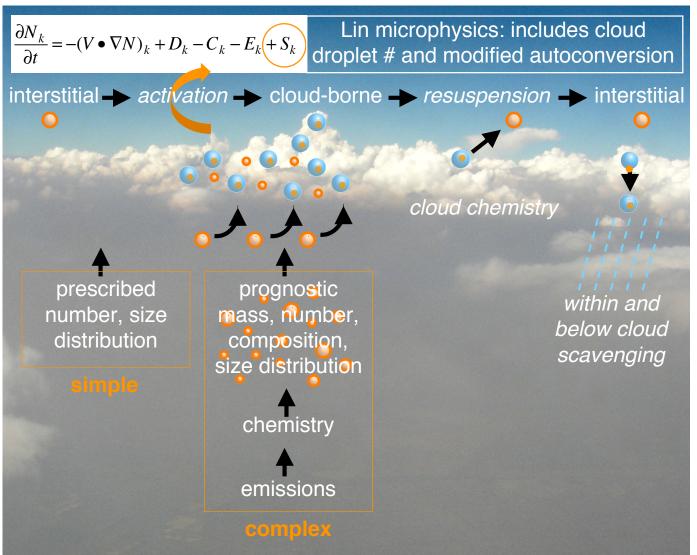
### **Part 2: Aerosol Indirect Forcing**

### Aerosols - Clouds - Radiation



### **Cloud-Aerosol Interactions**

### **General Description and Assumptions**



#### Simple:

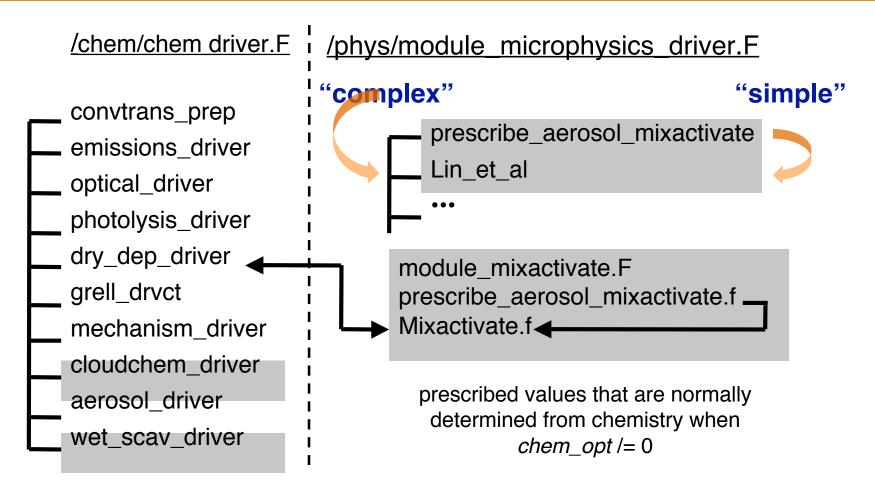
- chem\_opt = 0
- progn = 1
- naer = specified
- mp\_physics = 2
- ra\_sw\_physics = 2

#### **Complex:**

- chem\_opt = 9 or 10
- progn=1
- naer = ignored
- mp\_physics = 2
- ra\_sw\_phycis = 2



### Flow chart



When chemistry turned on, arrays for cloud droplet number, cloud droplet number source, and CCN passed between /chem and /phys directories



## **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 
$$\longrightarrow N_k \quad \text{grid cell mean droplet number mixing ratio in layer } k$$
 
$$D_k \quad \text{- vertical diffusion}$$
 
$$C_k \quad \text{- droplet loss due to collision/coalescence \& collection}$$
 
$$E_k \quad \text{- droplet loss due to evaporation}$$
 
$$S_k \quad \text{- droplet source due to nucleation (determined in mixactivate.f)}$$

- cloud droplet number source determined by aerosol activation (for meteorology-only runs a prescribed background 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)

### **Activation**

- 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 \_\_\_\_\_
  - hygro\_bc\_aer = 1.0e-6
    hygrophobic
  - 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

hygrophilic

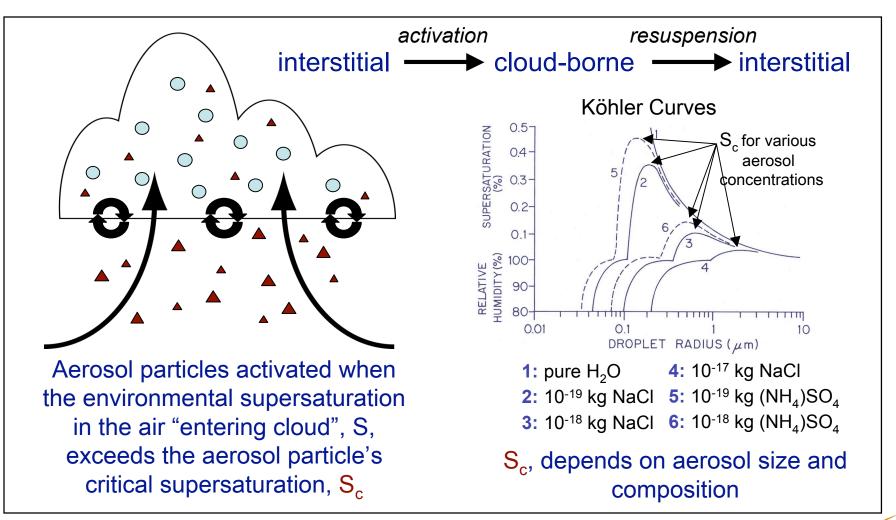
- Hygroscopicity used in activation determined by volume weighted bulk hygroscopicity, prior to the call to mixactivate.f, and total aerosol number
- For chem\_opt = 0 and nprog = 1, hygroscopicity set to 0.5



some OC is hygrophilic

- subject of research

# **Activation and Resuspension (1)**





# **Activation and Resuspension (2)**

#### **Aerosol Species**

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

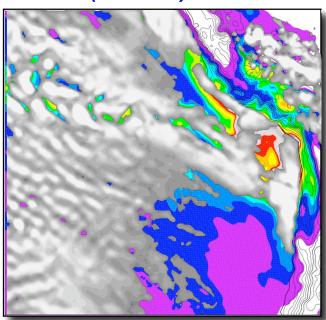
	activation r	resuspension
interstitial		interstitial
so4_a01	so4_cw01	so4_a01
so4_a02	so4_cw02	so4_a02
no3_a01	no3_cw01	no3_a01
no3_a02	no3_cw02	no3_a02
num_a01	num_cw01	num_a01
num_a02	num_cw02	num_a02
8 bins x 12 species	8 bins x 12 species	
+ hysw + wat		
= 112	_ 00	Paci



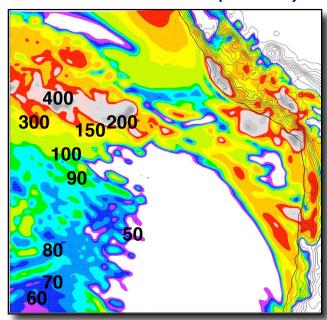
### **Cloud Condensation Nuclei**

- CCN: number concentration of aerosols activated at a specified supersaturation
- Diagnostic quantity, but varies in space and time since particulates and humidity varies
- 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



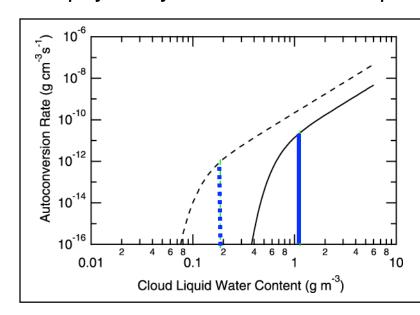
CCN at 0.1% SS (# cm<sup>-3</sup>)





### 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:  $N = 50 \text{ cm}^{-3}$ Solid:  $N = 500 \text{ cm}^{-3}$ 

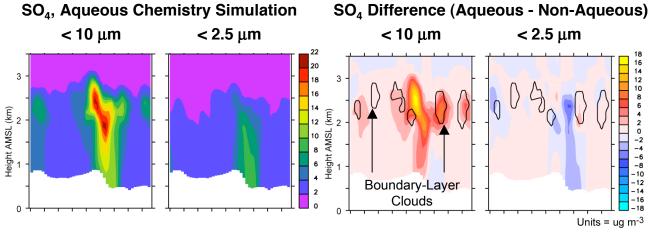
Adapted from Fig. 3, Liu et al., 2005, GRL.

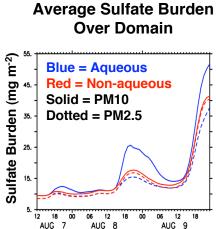


## **Aqueous Chemistry**

- Bulk cloud-chemistry module of Fahey and Pandis [2001]
- Oxidation of S(IV) by H<sub>2</sub>O<sub>2</sub>, O<sub>3</sub>, trace metals, and radical species
- Non-reactive uptake of HNO<sub>3</sub>, HCl, NH<sub>3</sub>, 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

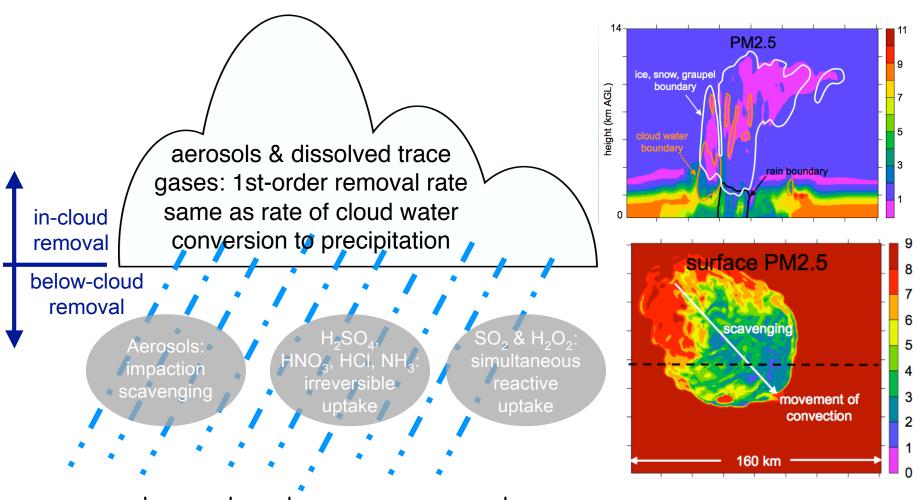
#### Vertical Cross-Section Though Power Plant SO<sub>2</sub> Plume







## Scavenging

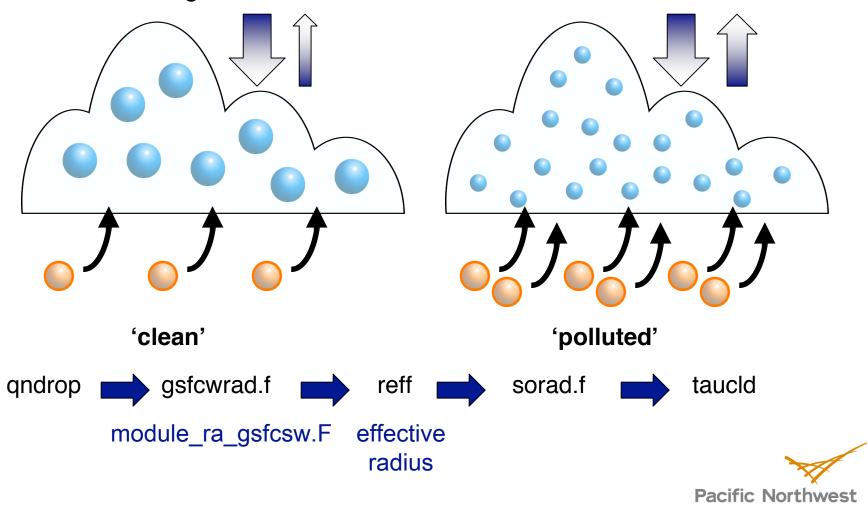


scavenged aerosols and gases are assumed to be instantly removed (wet deposited)



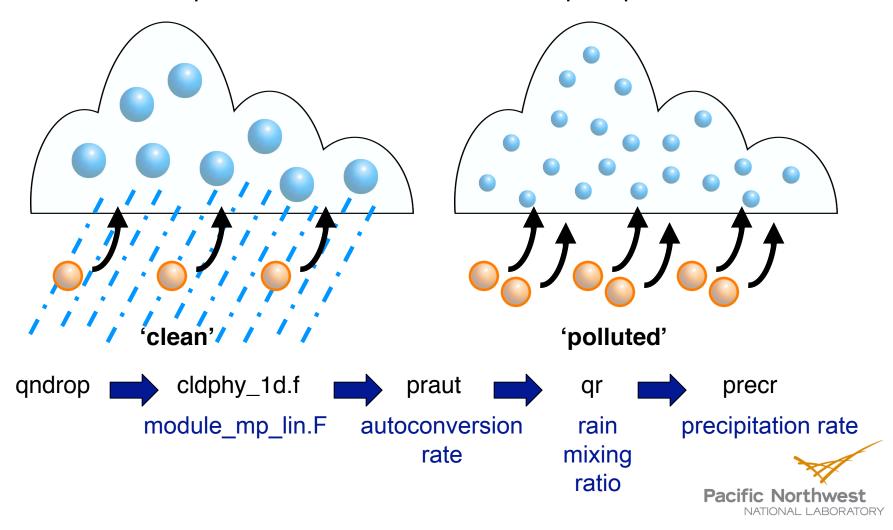
### **First Indirect Effect**

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



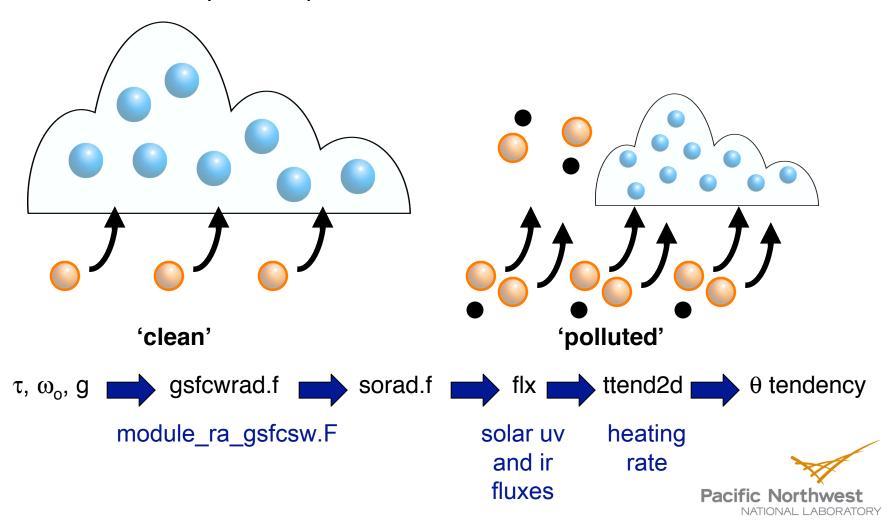
### **Second Indirect Effect**

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



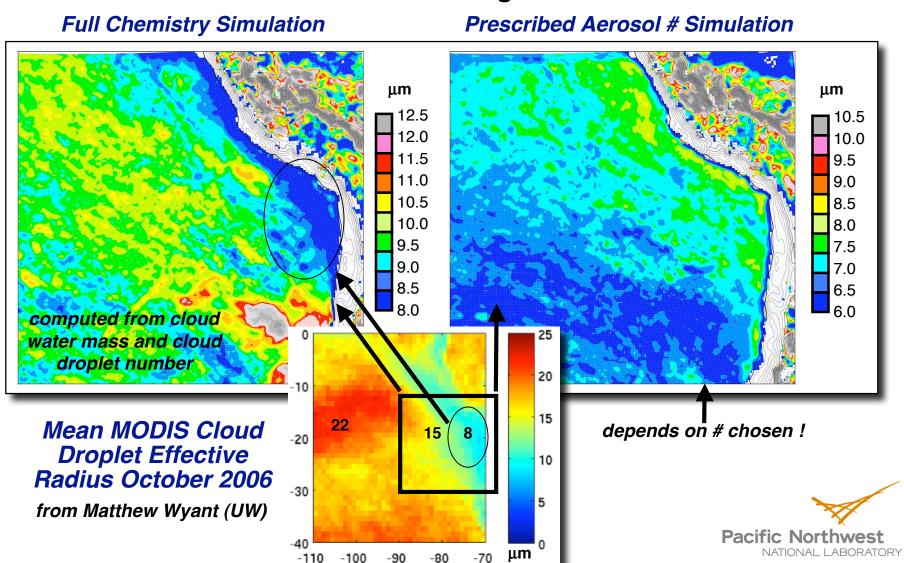
### **Semi-Direct Effect**

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



## **Example: Effective Radius**

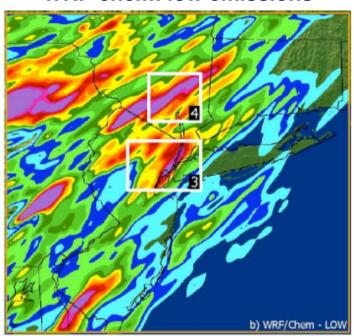
#### October 2006 Average at 12 UTC



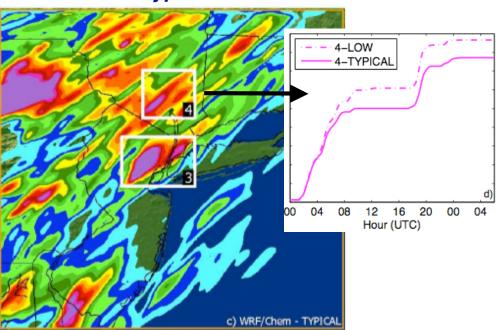
# **Example: Deep Convection**

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

WRF-chem: low emissions



WRF-chem: typical emissions



Pacific Northwest

 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. Submitted to Q. J. Roy. Meteor. Soc.

### **Interactions not Treated**

- First Dispersion Effect: Influence on cloud optical depth through influence of aerosol on dispersion of droplet size distribution, with no change in water content of cloud
- Second Dispersion Effect: Influence on cloud optical depth through influence of aerosol on dispersion 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 team)

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



## Settings in namelist.input

#### Simple:

- chem\_opt = 0
- naer = specified value

### **Complex:**

- chem\_opt = 9 or 10, cloud-phase aerosols only for MOSAIC
- 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:**

- chem\_opt = 11 or 12, cloud-phase aerosols for MADE/SORGAM
- mp\_physics = 8, cloud-aerosol interactions for Thompson scheme



## **Comparing Options**

#### Care must be taken in quantifying direct and indirect effects

#### Direct effect:

- Run with aer\_ra\_feedback on versus off
- 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 run does not quantify the indirect effect since the autoconversion scheme used in the Lin scheme will be different
- ➤ Have 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



### References

### WRF-chem Papers Describing Aerosol-Radiation-Cloud Interactions

- 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.
- Gustafson Jr., W.I., E.G. Chapman, S.J. Ghan, and J.D. Fast, 2007: Impact on modeled cloud characteristics due to simplified treatment of uniform cloud condensation nuclei during NEAQS 2004. *Geophys. Res. Lett.*, 34, L19809
- Chapman, E.G., W.I. Gustafson Jr., R.C. Easter, J.C. Barnard, S.J. Ghan, M.S. Pekour, and J.D. Fast, 2008: Coupling aerosols-cloud-radiative processes in the WRF-chem model: Investigating the radiative impact of large point sources. Submitted to *Atmos. Chem. Phys.*

#### Additional details included in cited papers

(More on the way regarding performance during MILAGRO field campaign)

