Regional and Global Modeling Needs from MILAGRO

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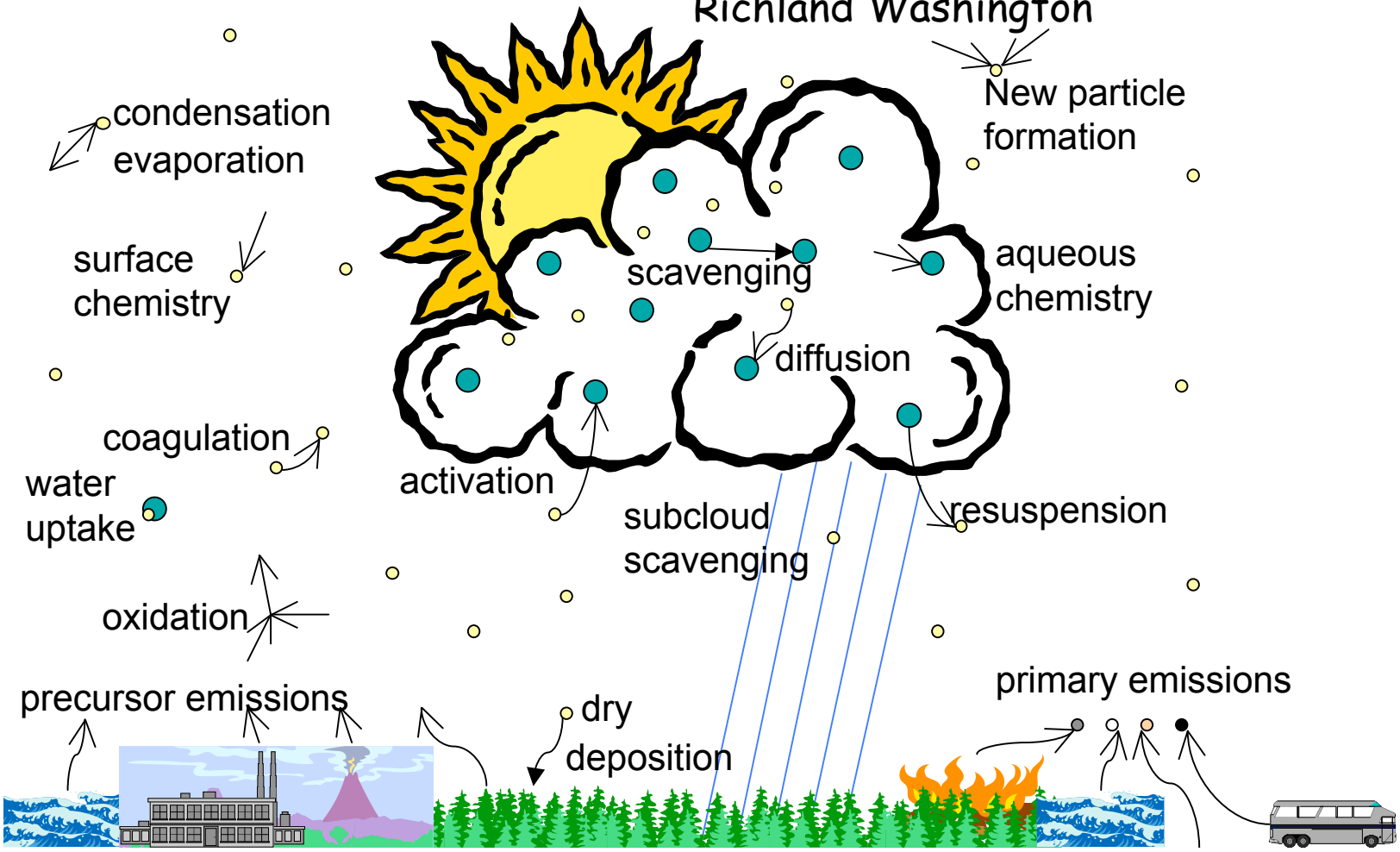


Outline

- Definitions
- Aerosol Properties
- Chemistry and Aerosol Processes
- Closure Studies

Experiments

- Closure Studies
- Parcel Modeling
- Chemical Transport Modeling

Parcel Modeling

- Focus on chemistry and physics rather than transport
- Requires coordinated measurements upwind and downwind.

Experiment	Input Data	Validation Data
Organic chemistry		
Nitrogen chemistry		
Oxidant chemistry		
Mass transfer to aerosol		
Heterogeous chemistry		

Definitions

Current treatment	Target treatment	How MILAGRO can help
Global Climate Models used for IPCC AR4 simulations: NCAR CCSM3 GFDL AM2 GISS Model E	Global Climate Models used for IPCC AR5 simulations (2009). No more than 3x slower *Global Climate Models used for IPCC AR6 simulations (2014).* *No more than 6x slower*	MILAGRO measurements, analysis, and modeling

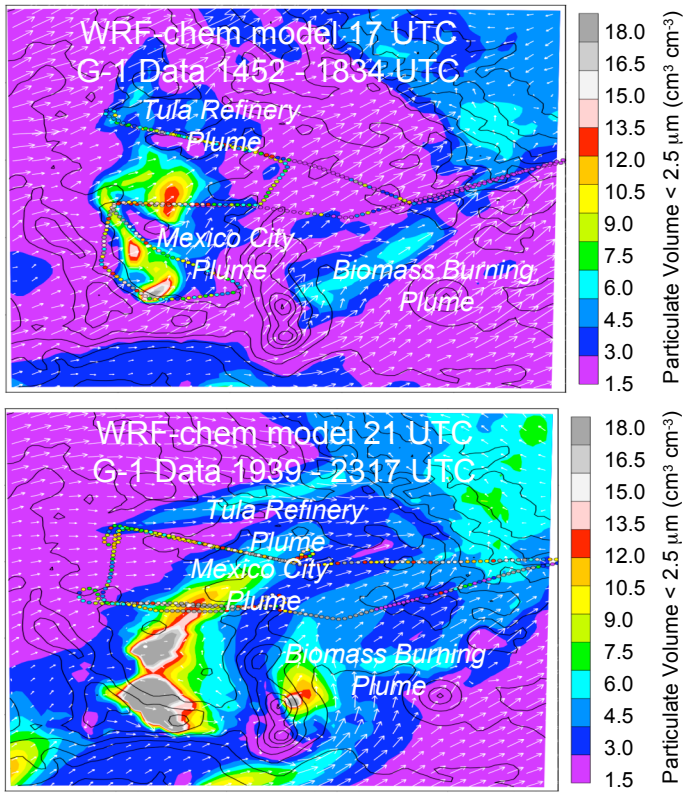
Aerosol Properties

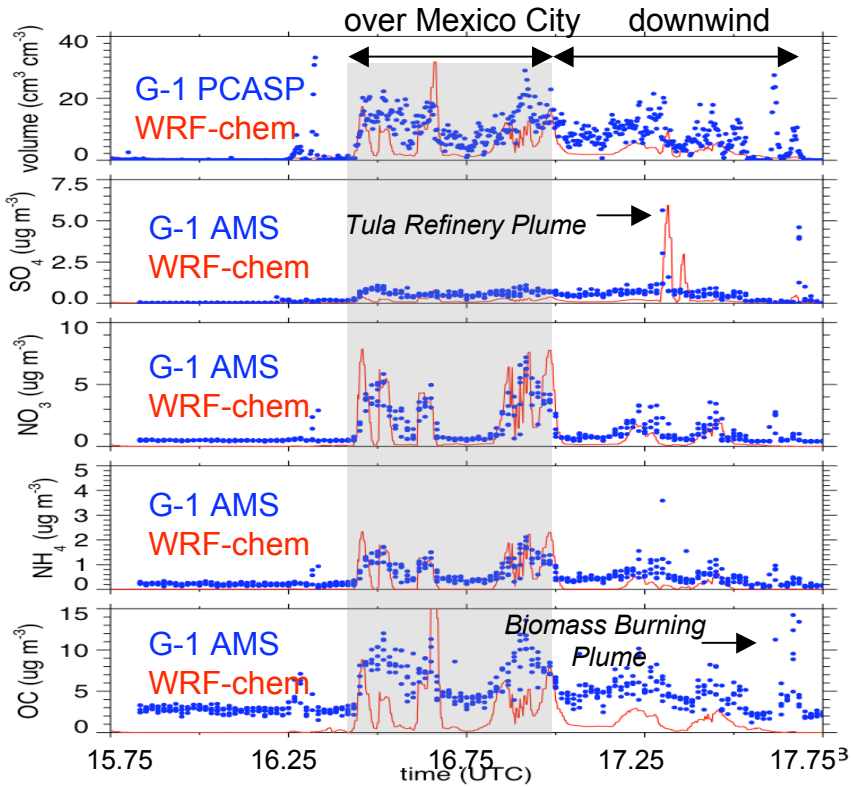
Aerosol Properties

Property	Current status	Target treatment	How MILAGRO can help
Components	Sulfate, dust, seasalt, OC, BC	Add nitrate, ammonia	Evaluation of integrated models
Size distribution	Prescribed	Predicted •modal •sectional •moments	New particle formation Evaluation of integrated models
Mixing state	External	Internal and external	Evaluation of models
Cloud-phase	Prescribed	Predicted	Evaluation of integrated models
Refractive index	For external mixtures only	Internal and external	Local radiative closure
Hygroscopicity	For external mixtures only	Internal and external	Evaluation of diameter growth factor
CCN Activity	Externally mixed inorganic salts	External mixtures of internally-mixed inorganic and soluble organic salts.	CCN closure .

Aerosol Properties: Mass Concentration and Composition

Current treatment	Target treatment	How MILAGRO can help
Interactive sulfate. Ammonia proportionate to sulfate. Prescribed dust, sea salt, hydrophilic and hydrophobic OC & BC.	All species interactive. Add nitrate, ammonia.	AMS, PILS, thermal TDMA measurements. Evaluation of integrated models

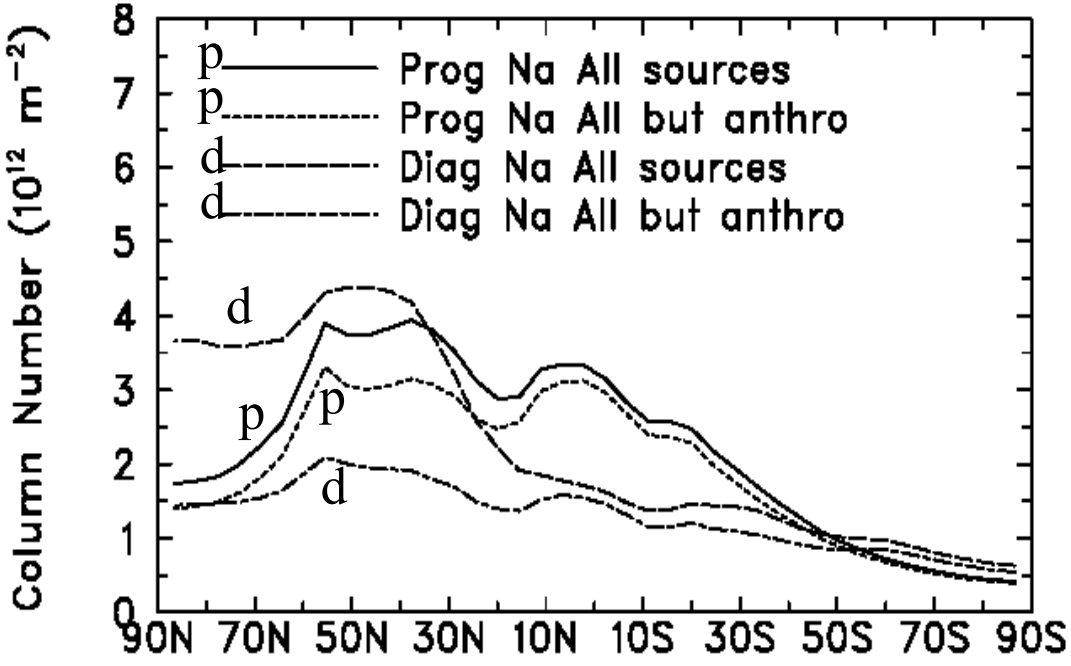




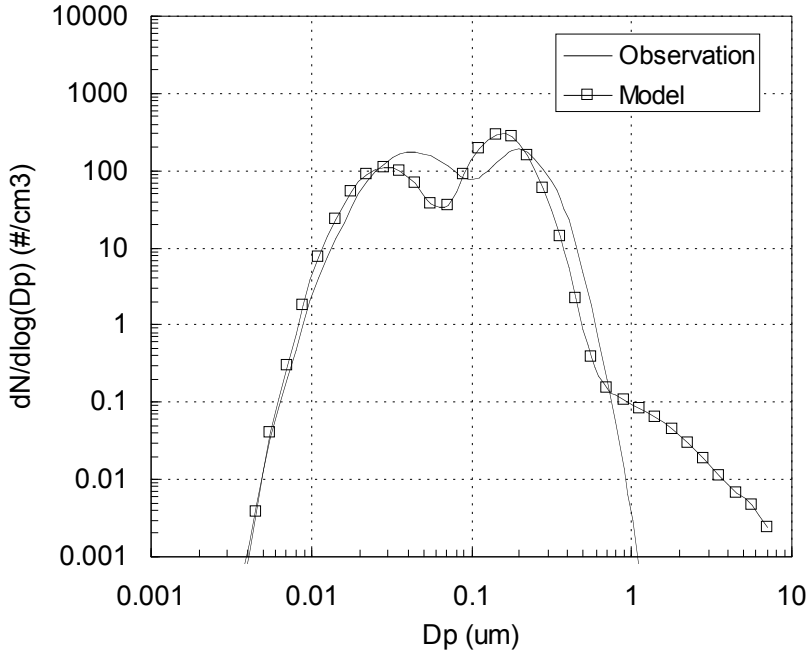
MAX-MEX.
Jerome Fast

Aerosol Properties: Size Distribution

Current treatment	Target treatment	How MILAGRO can help
Prescribed for each aerosol type except dust. Multiple sizes for dust and sea salt.	Predicted •modal (Easter 2004; Liu 2005) •*sectional (Adams & Seinfeld 2002)* •*moments (Wright 2001)*	DMA+PCASP measurements: 1-20,000 nm. Evaluation of distribution simulated by integrated models.



Ghan, Easter, Chapman, Abdul-Razzak, Zhang, Leung, Laulainen, Saylor and Zaveri: A physically-based estimate of radiative forcing by anthropogenic sulfate aerosol *JGR 2001.*



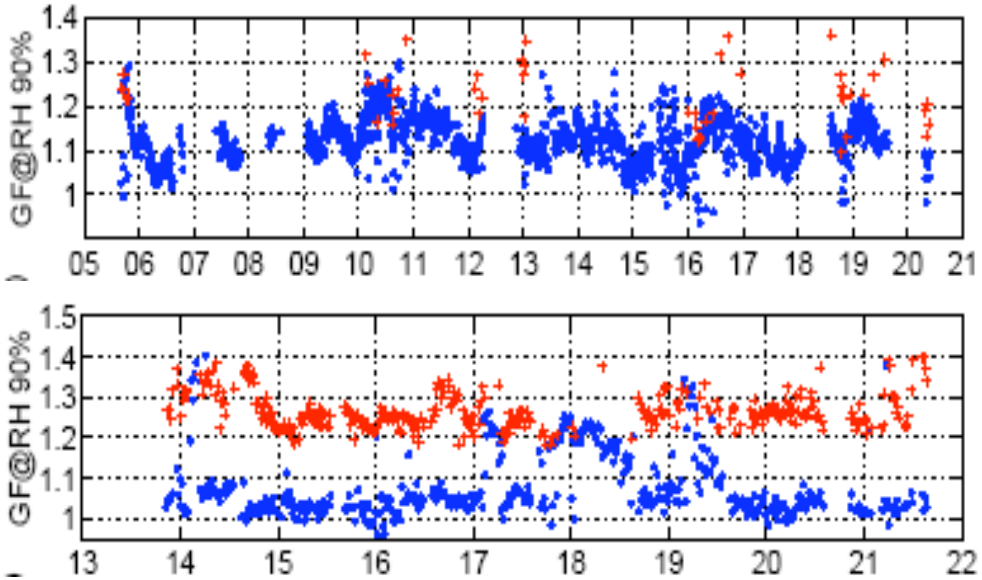
Liu, Penner, and Herzog: Global modeling of aerosol dynamics: Model description, evaluation, and interactions between sulfate and nonsulfate aerosols. *JGR 2005*

Aerosol Properties: Mixing State

Current treatment	Target treatment	How MILAGRO can help
External	Internal and external (Wilson 2001; Easter 2004; Stier 2005).	H-TDMA, AMS, PILS, single particle measurements. Evaluation of models.

Hygroscopic growth factor of 10 nm particles at RH=90%

Helsinki urban site



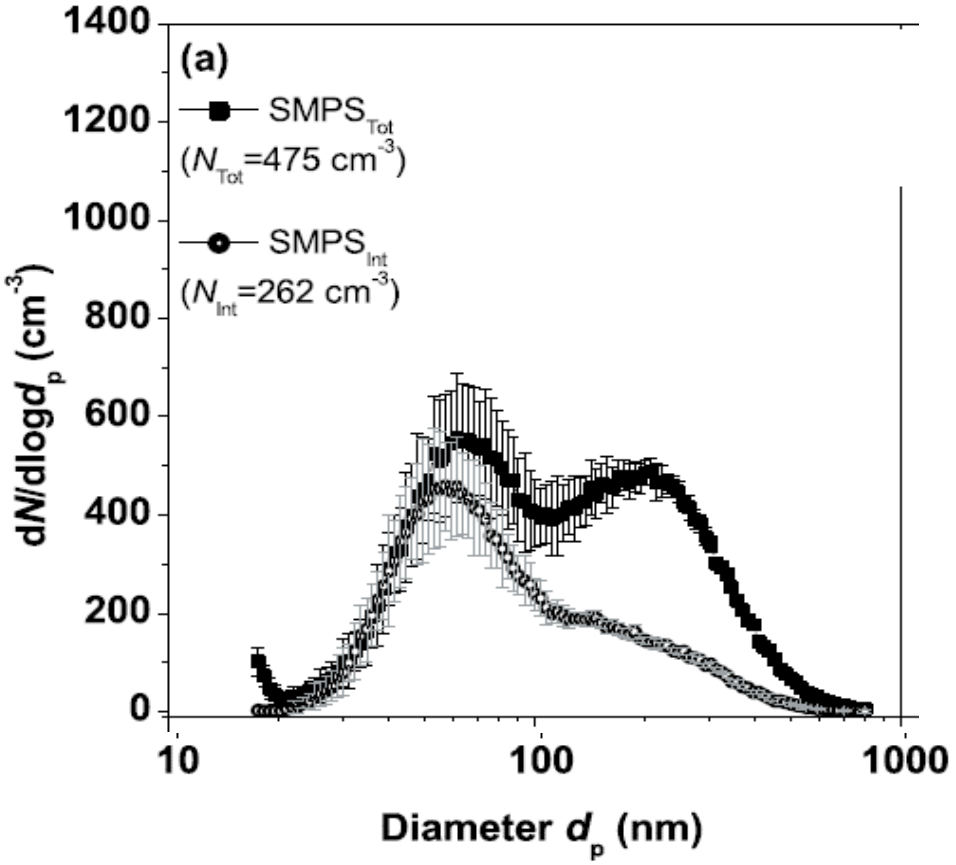
May 1998

December 1999

Väkevä, Kulmala, Stratmann, and Hämeri: Field measurements of hygroscopic properties and state of mixing of nucleation mode particles. Atmos. Chem. & Physics, 2002.

Aerosol Properties: Cloud (Activated) Phase

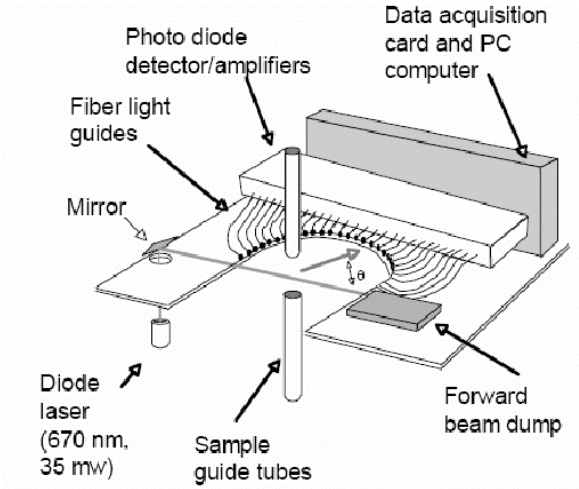
Current treatment	Target treatment	How MILAGRO can help
Prescribed fraction of total.	Predicted (Easter 2004; Koch 2005).	CVI+DMA measurements. Evaluation of integrated models.

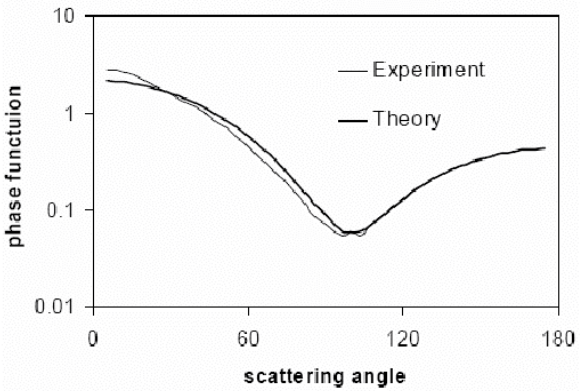


Henning, Bojinski, Diehl, Ghan, Nyeki, Weingartner, Wurzler,and Baltensperger: Aerosol partitioning in natural mixed-phase clouds. GRL 2004.

Aerosol Properties: Refractive Index

Current treatment	Target treatment	How MILAGRO can help
For external mixtures only. Prescribed.	Internal and external mixtures. Volume mixing rule for soluble. Maxwell-Garnett rule for inclusions.	DMA, AMS, BC, refractive index measurements. Phase function evaluation.

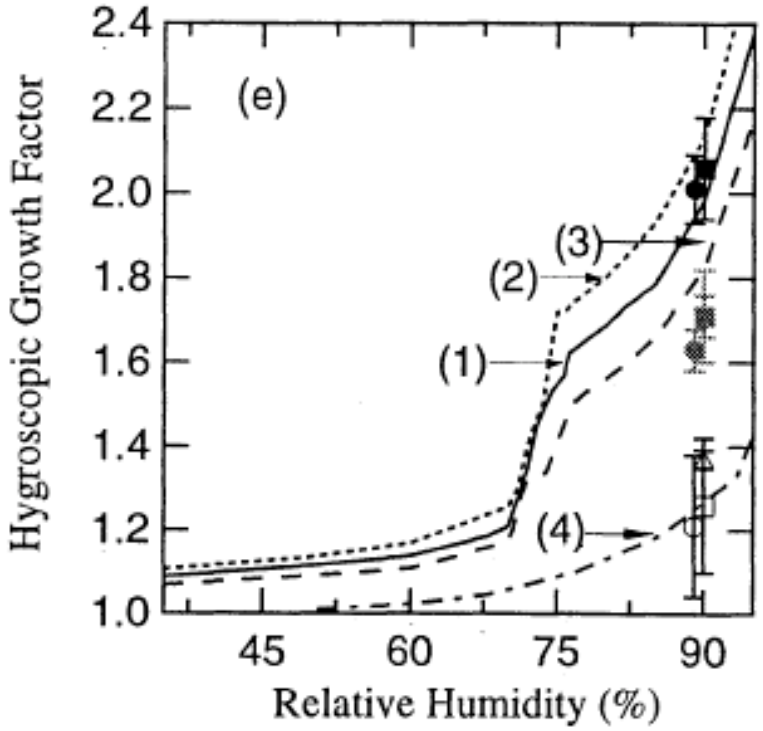




Barkey, Bailey, Liou, and Hallett, : Light scattering properties of plate and column ice crystals generated in a laboratory cold chamber. Appl. Opt. 2002.

Aerosol Properties: Hygroscopicity

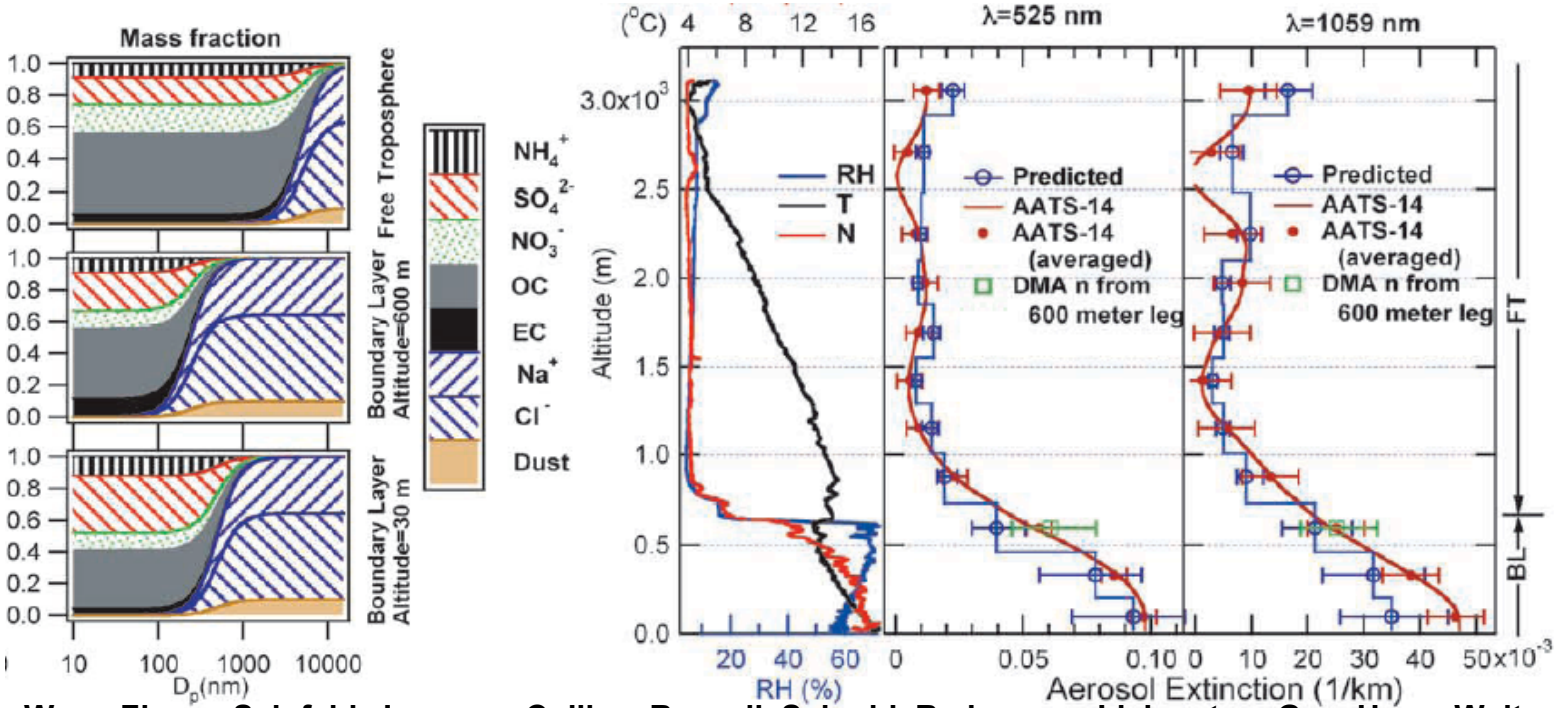
Current treatment	Target treatment	How MILAGRO can help
External mixtures only	Internal and external mixtures. Volume average.	H-TDMA, AMS, PILS measurements. Evaluation of diameter growth factor.



Ming and Russell: Predicted hygroscopic growth of sea salt & organic aerosol. JGR 2001

Aerosol Properties: Optical Properties

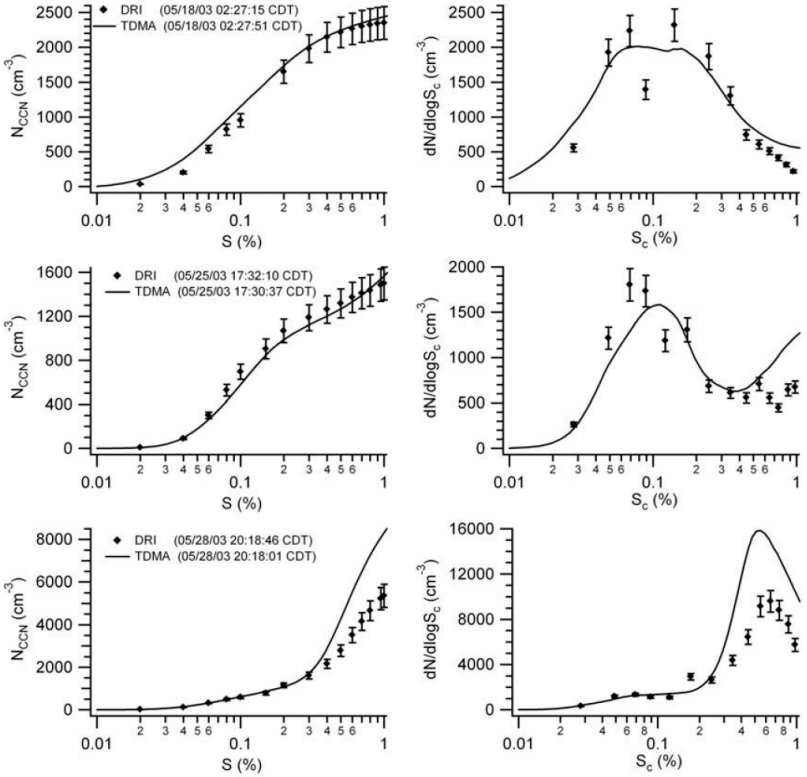
Current treatment	Target treatment	How MILAGRO can help
For external mixtures only. Prescribed properties as f(RH)	Internal and external mixtures. Parameterized in terms of r_{wet} and n_{wet} (Ghan 2001). *Dependence on particle shape.*	DMA, AMS, BC, refractive index, absorption, extinction, aerosol optical depth measurements. Local radiative closure. Aerosol optical depth validation.



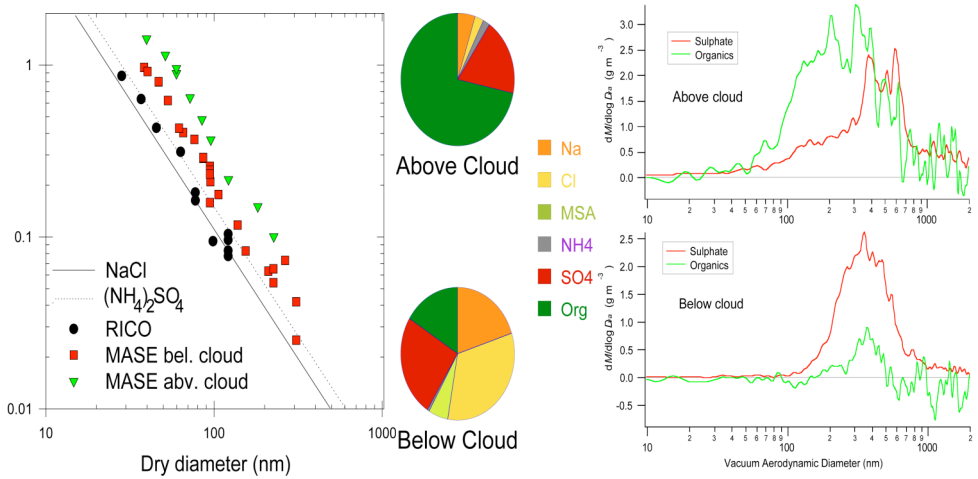
Wang,Flagan,Seinfeld, Jonsson, Collins, Russell, Schmid, Redemann, Livingston, Gao, Hegg, Welton, and Bates: Clear-column radiative closure during ACE-Asia: Comparison of multiwavelength extinction derived from particle size and composition with results from Sun photometry. JGR 2002.

Aerosol Properties: CCN Activity

Current treatment	Target treatment	How MILAGRO can help
Externally mixed inorganic salts.	Köhler theory for external mixtures of internally-mixed inorganic and soluble organic salts.	DMA, AMS, PILS, CCN field measurements. CCN closure.



Gasparini, Li, Collins, Ferrare, and Brackett: Application of aerosol hygroscopicity measured at the Atmospheric Radiation Measurement Program's Southern Great Plains site to examine composition and evolution. JGR 2006



J. Hudson (DRI) Y.-N. Lee (BNL) L. Alexander (PNNL)

The critical supersaturation for particle activation (left) measured above cloud at Point Reyes, CA during the ASP MASE experiment is higher than that below cloud and that expected for ammonium sulfate and sodium chloride because the composition above cloud (center) is dominated by organics rather than sulfate or sea salt, particularly for diameters less than 300 nm (right).

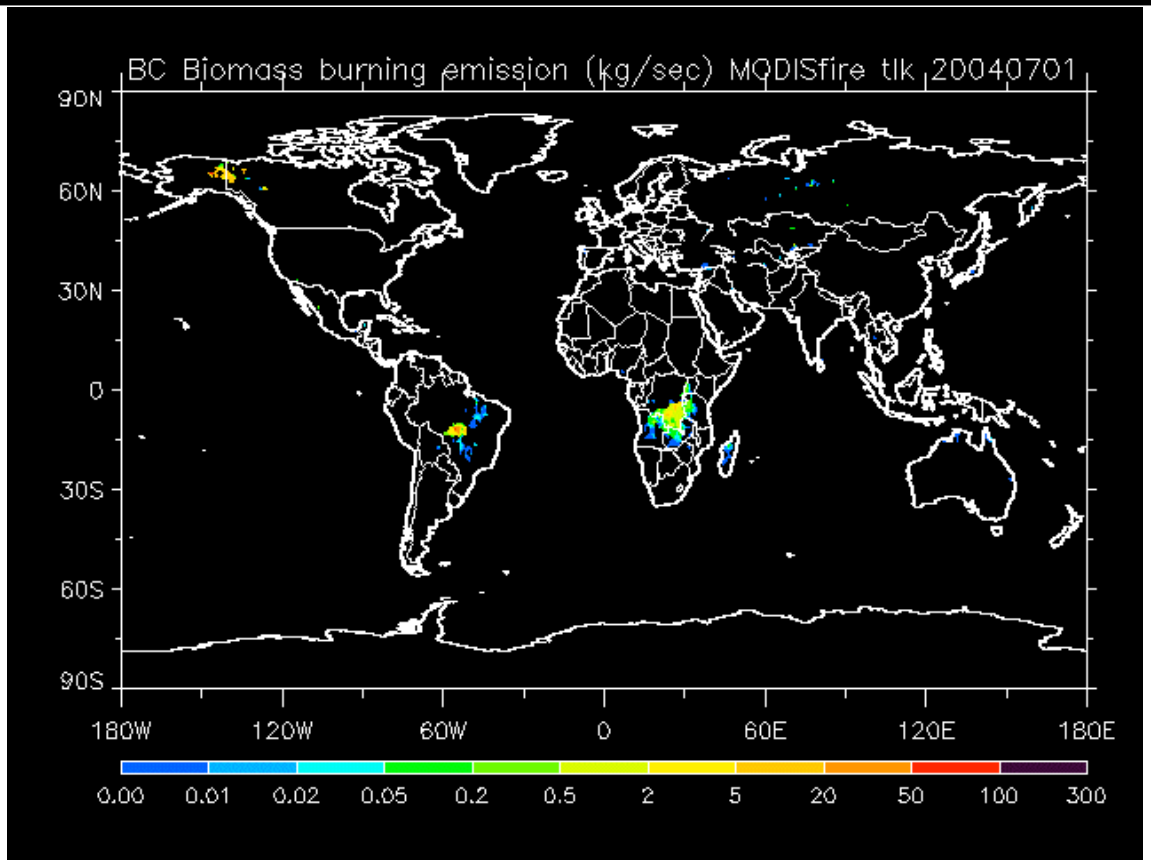
Chemistry and Aerosol Processes

Aerosol Processes

Process	Current status	Target treatment	How MILAGRO can help
New particle formation	Binary homogenous nucleation	Ternary nucleation •ammonia •organic	New parameterization Evaluation of parameterization
Formation of OC	Instantaneous conversion of precursors with prescribed uniform yield.	Reaction of precursors with oxidants. Multiple hydrocarbon groups. Dependence of yield on total organic aerosol.	Evaluation of process and integrated models
Aging of BC and OC	Prescribed hygrophobic-to-hydrophilic conversion time	*Parameterized treatment of coagulation, condensation and oxidation effects*	Lab experiments Evaluation of process models
Water uptake	For external mixtures only No hysteresis. Equilibrium	Internal and external. Hysteresis *Kinetic effects*	Evaluation of growth factor
Activation	Prescribed number activated	Diagnosed number activated	Evaluation of activation models
Aqueous chemistry	Bulk treatment (same for all cloud droplets). pH dependence	*Size dependent*	Detailed modeling
Convective transport and removal	Cumulus parameterization. Ambiguous cloud fraction	Cumulus parameterization with realistic cloud fraction, aqueous chemistry. *Statistics from embedded cloud models*	Evaluation of parameterization in integrated models.
In-cloud scavenging	Precipitation rate independent of aerosol	Precipitation rate depends on aerosol. Add influence of collision/coalescence on number. Subgrid variability in autoconversion.	Evaluation of collision / coalescence schemes (Daum). Evaluation of integrated models

Chemistry/Aerosol Processes: Primary Emissions

Current treatment	Target treatment	How MILAGRO can help
Prescribed for all species	Online for dust, sea salt. *Daily biomass burning estimates from area burned, biomass density (or fuel load), burning efficiency, & emission factor.*	Validation of satellite estimates of biomass burning emissions of particulates. Improved estimates of emissions factors.



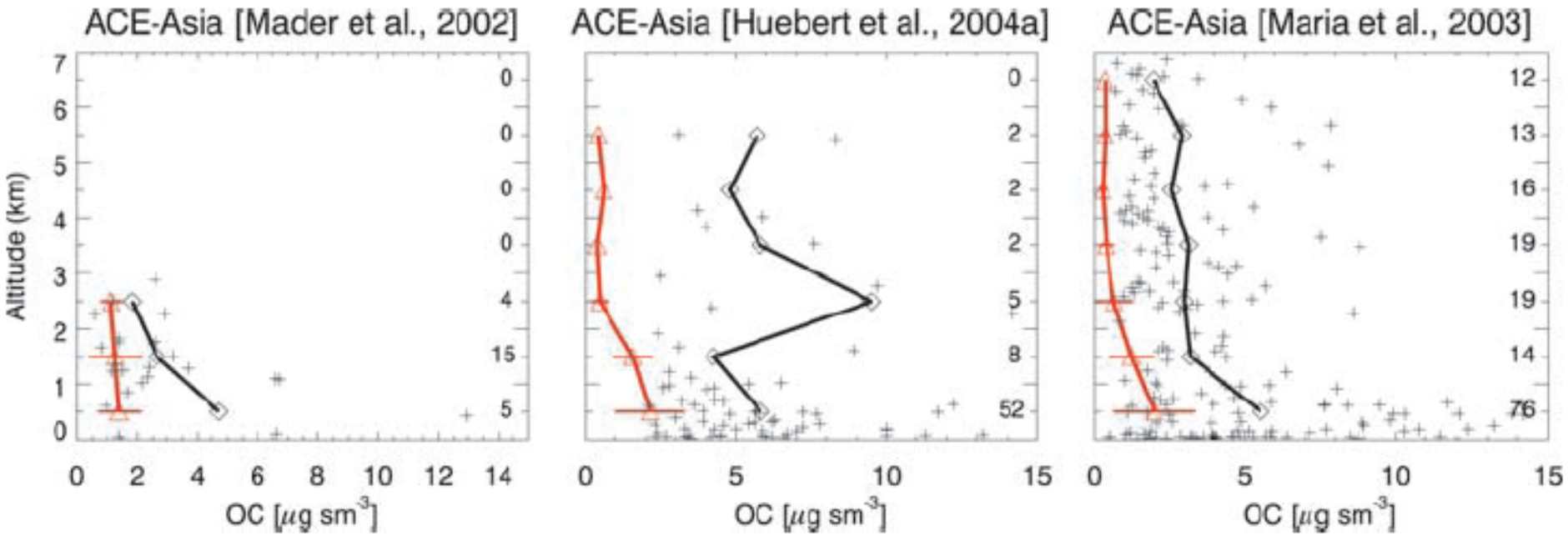
From "Using MODIS fire count data as an interim solution for estimating biomass burning emission of aerosols and trace gases", Mian Chen.

Chemistry/Aerosol Processes: Gas Emissions

Current treatment	Target treatment	How MILAGRO can help
Prescribed for all species. VOC not well speciated.	Speciated VOC emissions from different sources.	CO, speciated VOC emissions from biomass burning. Speciated VOC concentrations near urban sources.

Chemistry/Aerosol Processes: Oxidation of Precursors

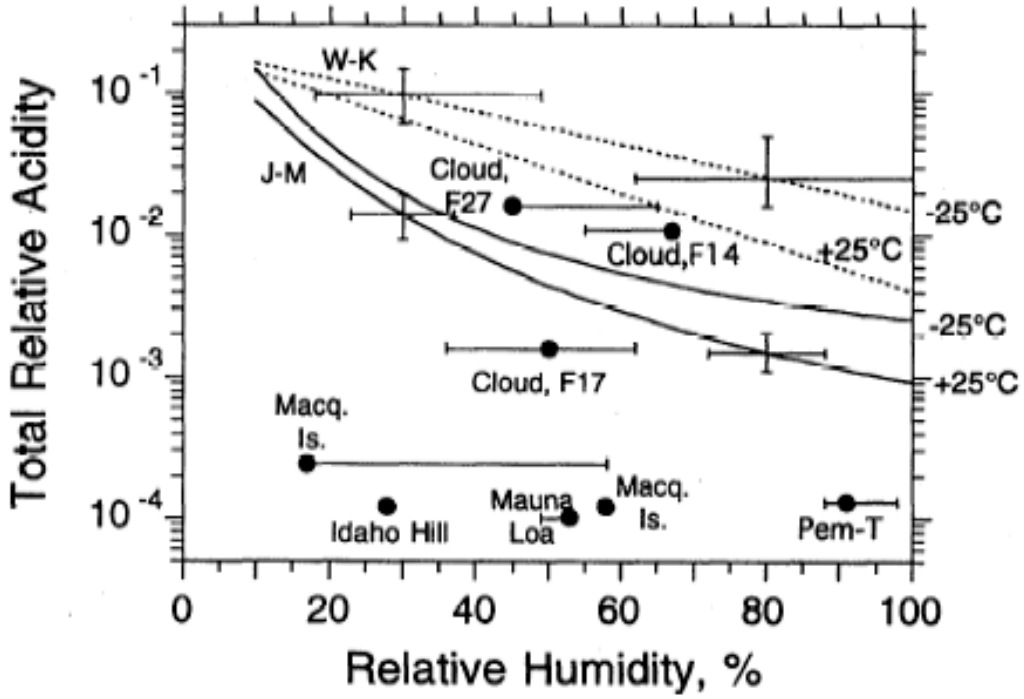
Current Treatment	Target Treatment	How MILAGRO can help.
Reaction of SO_2 and DMS with prescribed oxidant concentrations. Instantaneous oxidation of VOC with prescribed yield.	Reaction of all precursors with online oxidants. Multiple hydrocarbon groups, with dependence of yield on organic aerosol.	Validation of Master Mechanism. Development of improved condensed mechanism.



Heald, Jacob, Park, Russell, Huebert, Seinfeld, Liao, and Weber: A large organic aerosol source in the free troposphere missing from current models. GRL 2005.

Chemistry/Aerosol Processes: New particle formation

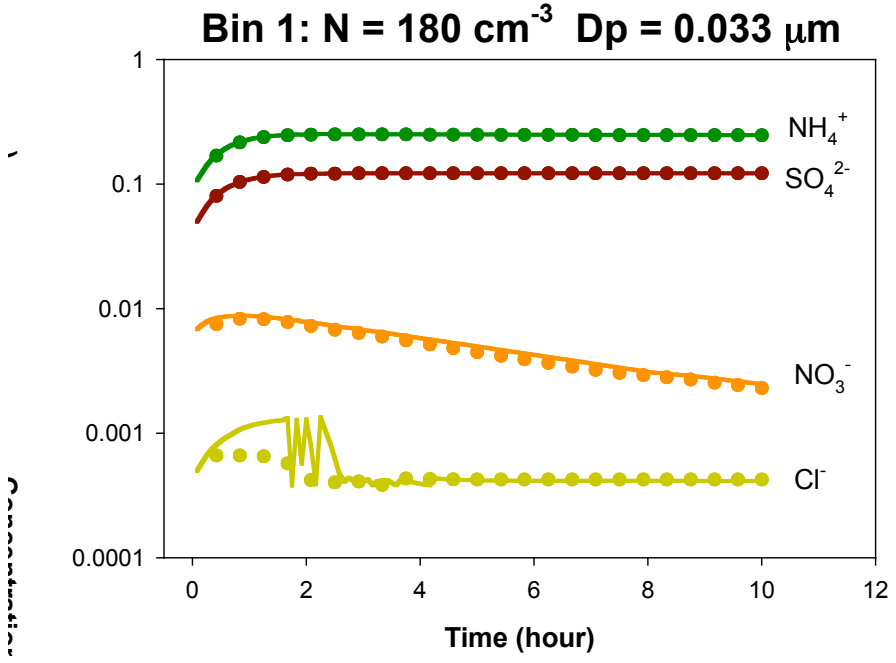
Current treatment	Target treatment	How MILAGRO can help
Neglected.	Ternary homogenous nucleation (water, sulfuric acid, and ammonia). *Organic nucleation.* *Ion-induced nucleation (water, sulfuric acid).*	CN, DMA, H_2SO_4, NH_3, organic measurements. New parameterizations. Evaluation of parameterizations.

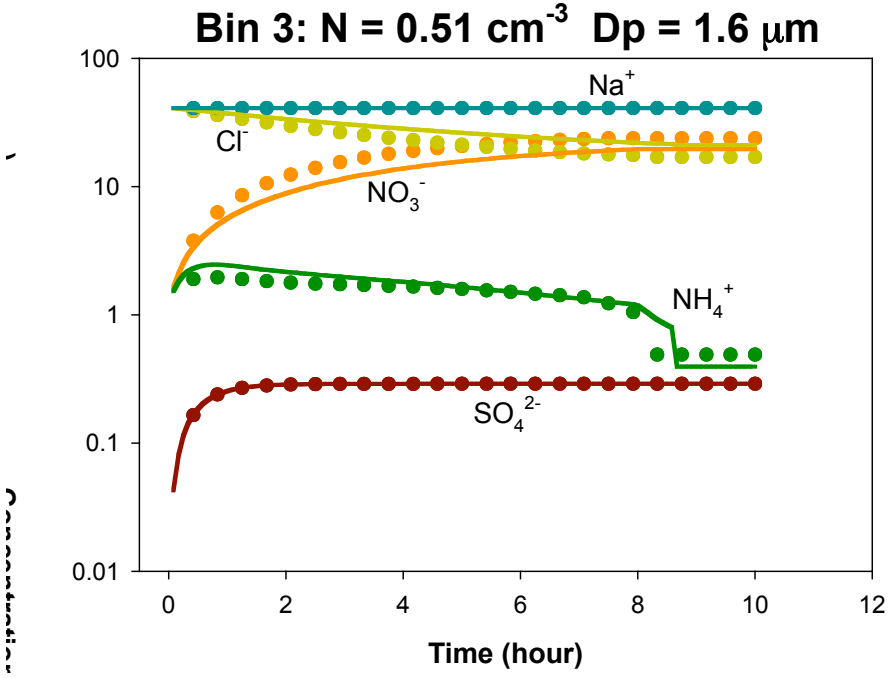


Weber, Tanner, Eisele, Clarke, Kapustin: New particle formation in the remote troposphere: a comparison of observations at various sites, Geophys. Res. Lett, 1999

Chemistry/Aerosol Processes: Condensation of Oxidized Precursor Gases

Current treatment	Target treatment	How MILAGRO can help
Instantaneous condensation	Size-dependent mass transfer treatment	Size-resolved composition measurements from AMS. Validation of parcel models.

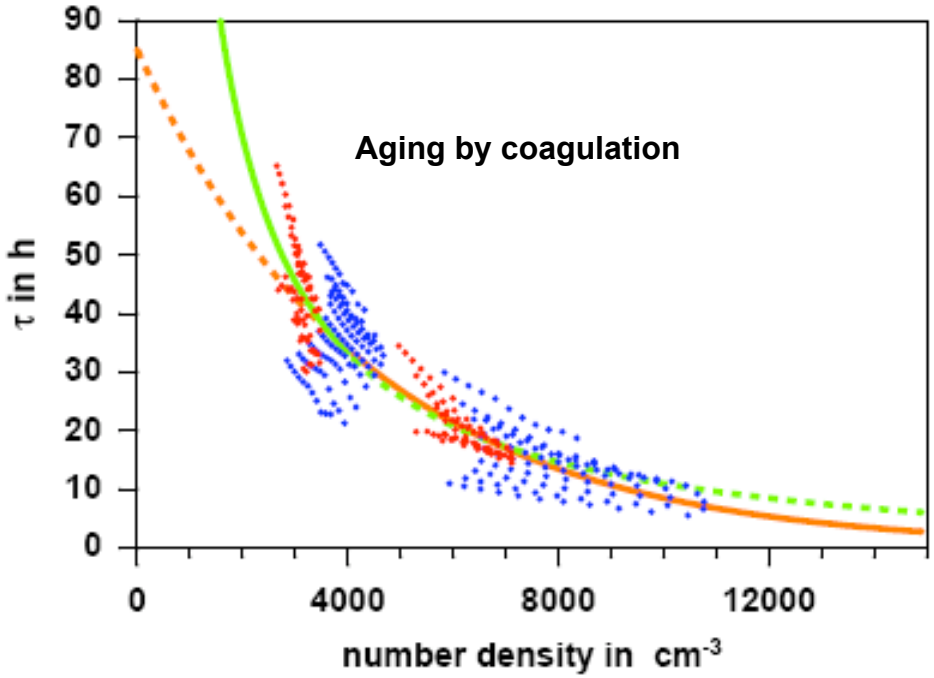




Zaveri, Easter, Fast, and Peters: A computationally efficient method for solving dynamic gas-particle mass transfer differential equations. American Association for Aerosol Research Conference, Austin TX, Oct. 2005.

Chemistry/Aerosol Processes: Evolution of Hygroscopicity

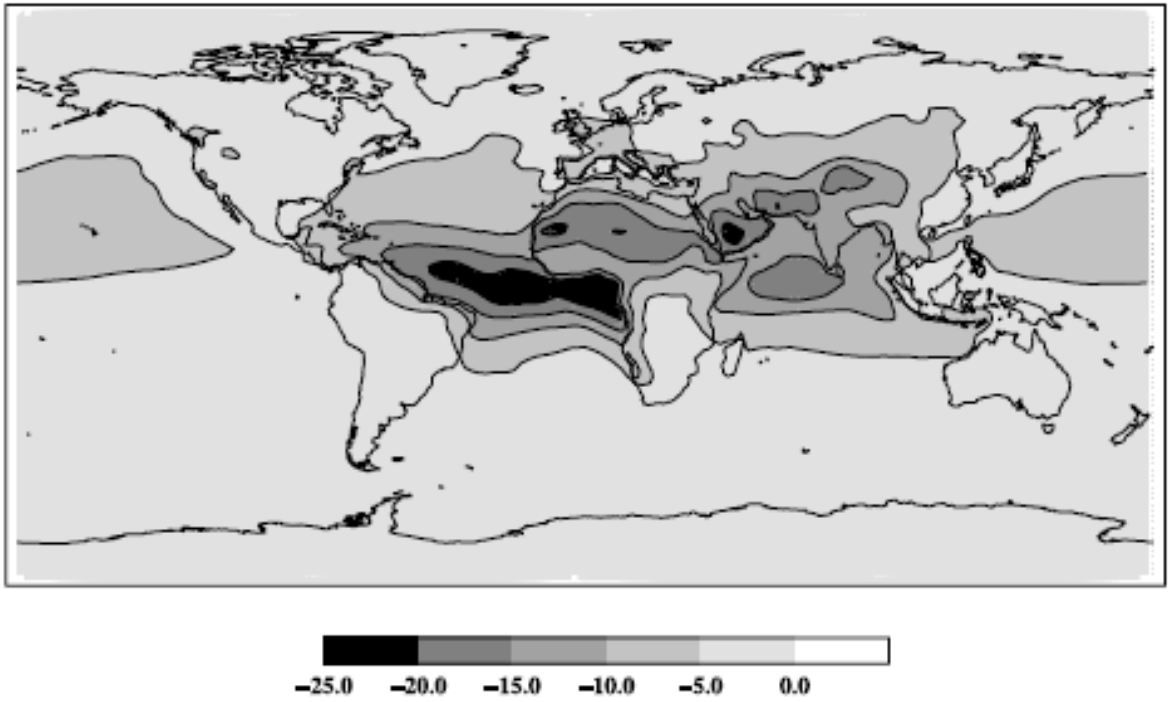
Current treatment	Target treatment	How MILAGRO can help
Prescribed hygrophobic-to-hydrophilic conversion time.	Separate treatment of coagulation and condensation effects for BC and OC, condensation effects for dust (Wilson 2001; Croft 2005).	Single particle analysis, HTDMA. Evaluation of box models



Riemer, Vogel, and Vogel, A parameterisation of the soot aging for global climate models. Atmos. Chem. Phys. Discuss., 2004.

Chemistry/Aerosol Processes: Heterogeneous Chemistry

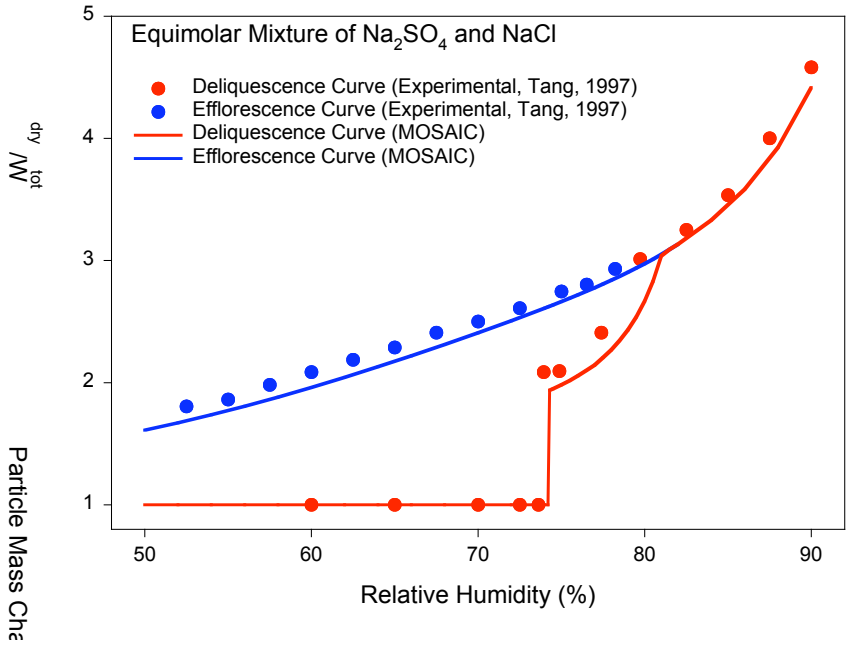
Current Treatment	Target Treatment	How MILAGRO can help.
Neglected.	*Uptake of HNO_3, NO_3, SO_2, O_3, HO_2 onto aerosol.*	Single particle analysis. Evaluation of simulations with and without heterogeneous chemistry.

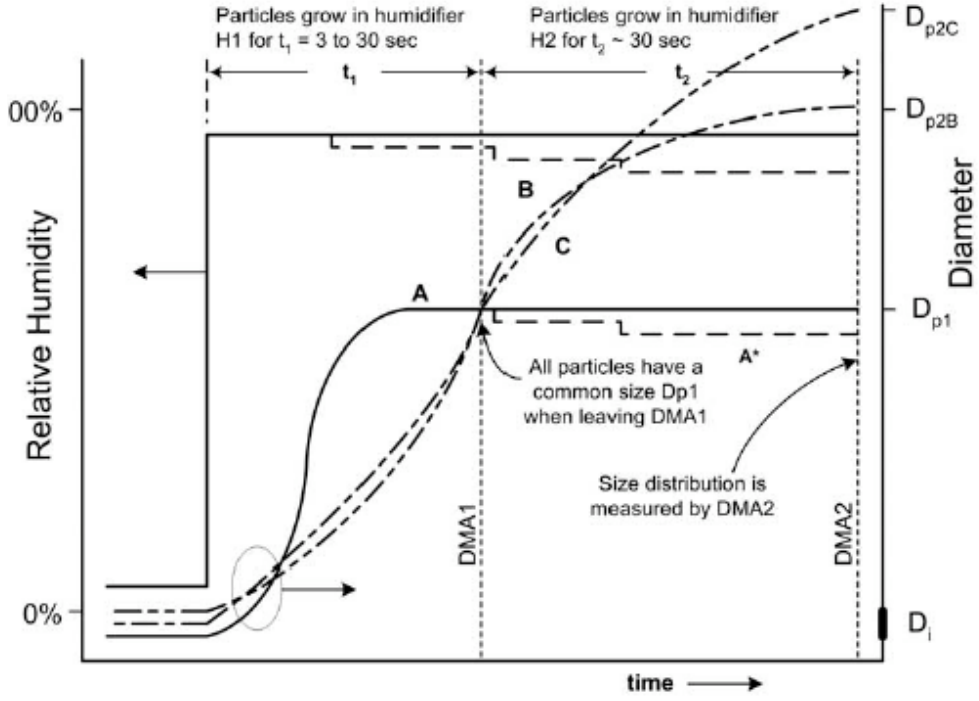


Percentage change in surface ozone due to heterogeneous chemistry on dust. Bauer, Balkansi, Schulz, and Hauglustaine: Globel modeling of heterogeneous chemistry on mineral aerosol surface: influence on tropospheric ozoen chemistry and comparison to observations. JGR 2004.

Chemistry/Aerosol Processes: Water uptake

Current treatment	Target treatment	How MILAGRO can help
External mixtures only. No hysteresis. Equilibrium Köhler theory.	Internal and external. Hysteresis (Easter 2004). *Thermodynamics. Kinetic effects (Zaveri 2005).*	H-TDMA, AMS measurements. Evaluation of growth factor.



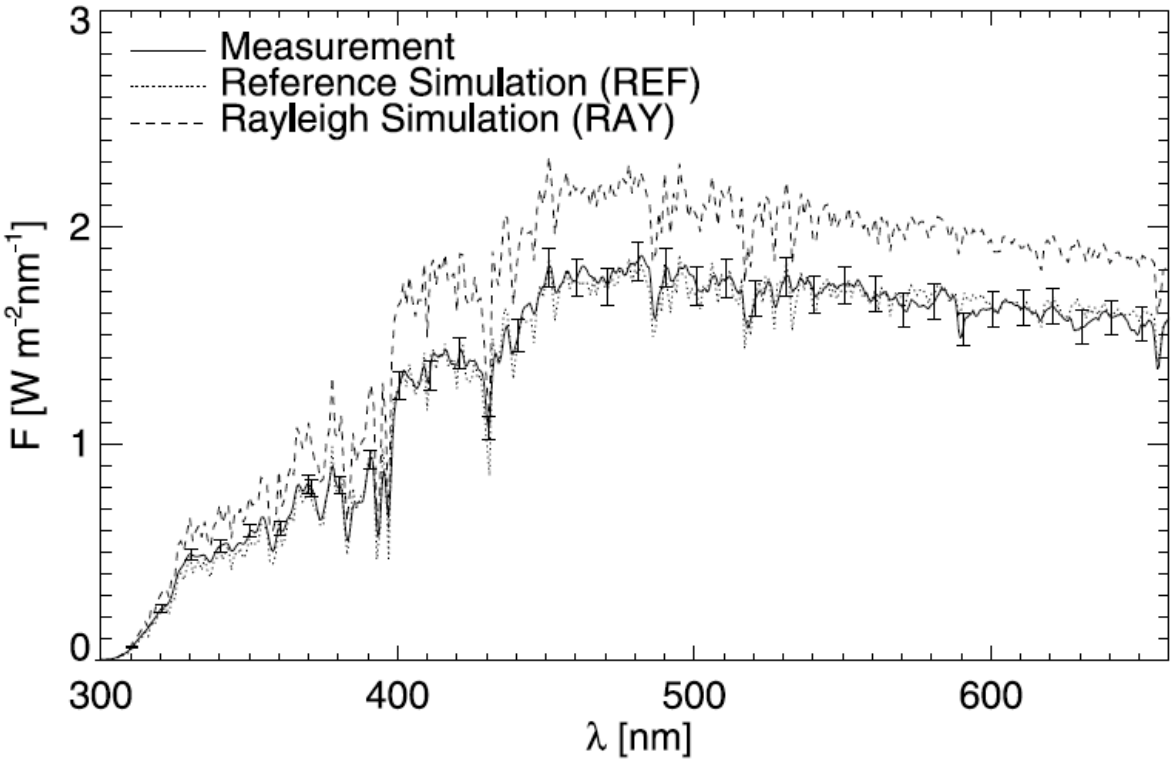


Zaveri, Easter, and Peters: A computationally efficient multicomponent equilibrium solver for aerosols (MESA), JGR 2005

Chuang, Measurement of the timescale of hygroscopic growth for atmospheric aerosols. JGR 2003

Chemistry/Aerosol Processes: Aerosol Impact on Actinic Flux and Chemistry

Current treatment	Target treatment	How MILAGRO can help
Neglected	Scaling of clean-sky flux by ratio of dirty-sky/clean-sky flux	Actinic flux, irradiance, extinction and oxidant measurements to evaluate simulations with and without feedback

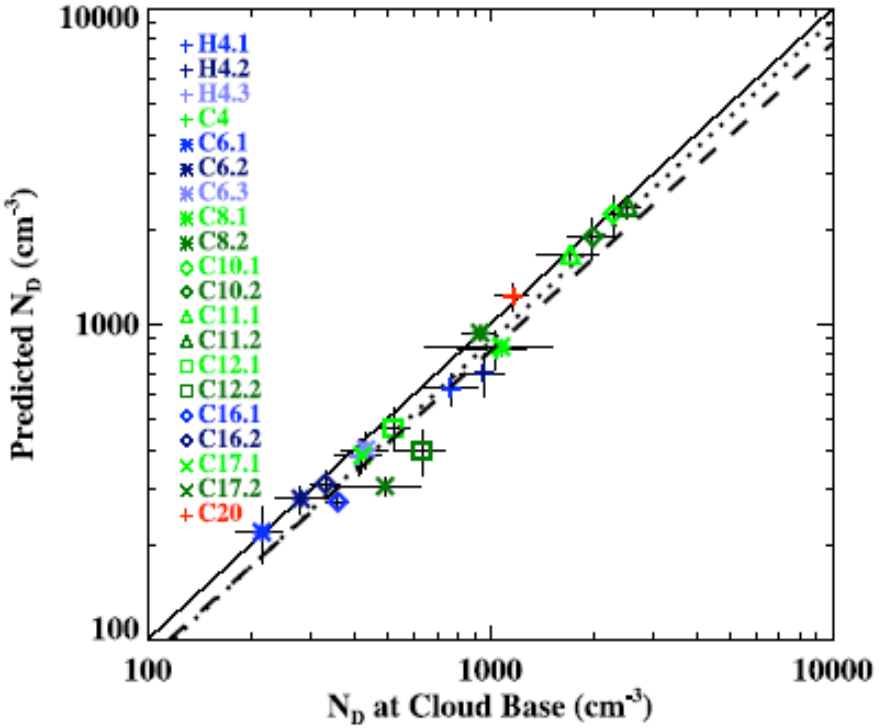


Actinic flux measured near Lindeberg Germany and calculated with and without aerosol effects. Fruh, Eckstein, Trautmann, Wendisch, Fiebig, and Feister: Ground-based measured and calculated spectra of actinic flux density and downward UV irradiance in cloudless conditions and their sensitivity to aerosol microphysical properties. JGR, 2003.

Aerosol Processes: Activation

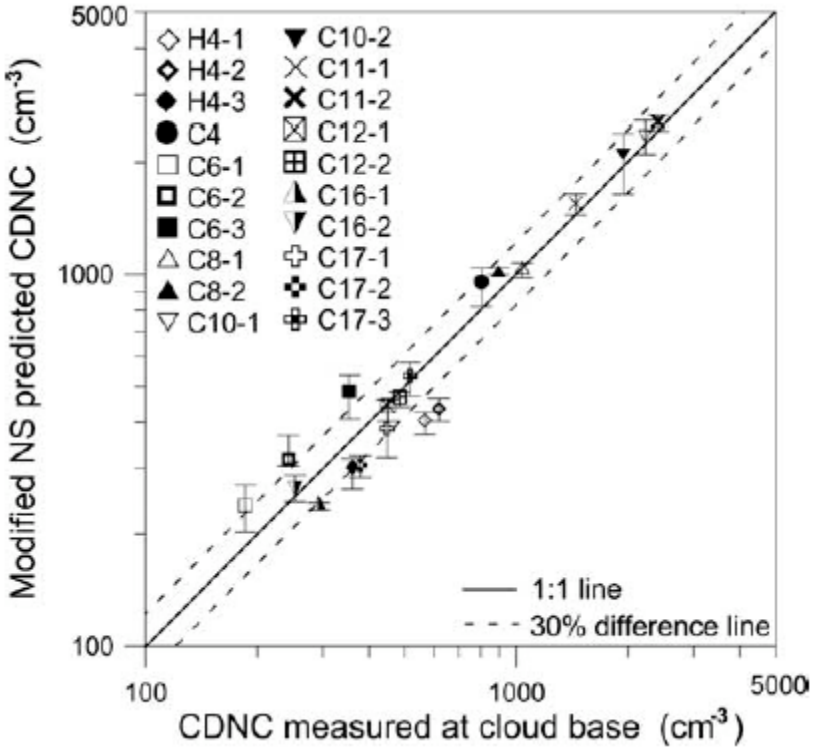
Current treatment	Target treatment	How MILAGRO can help
Prescribed number activated.	Diagnosed number activated (Nenes & Seinfeld 2003; Meskhidze et al 2005).	DMA, AMS, gustprobe, CAPS measurements. Evaluation of activation models.

Parcel model



Conant, VanReken, Rissman, Varutbangkul, Jonsson, Nenes, Jimenez, Delia, Bahreini, Roberts, Flagan, and Seinfeld: Aerosol--cloud drop concentration closure in warm cumulus. JGR 2004.

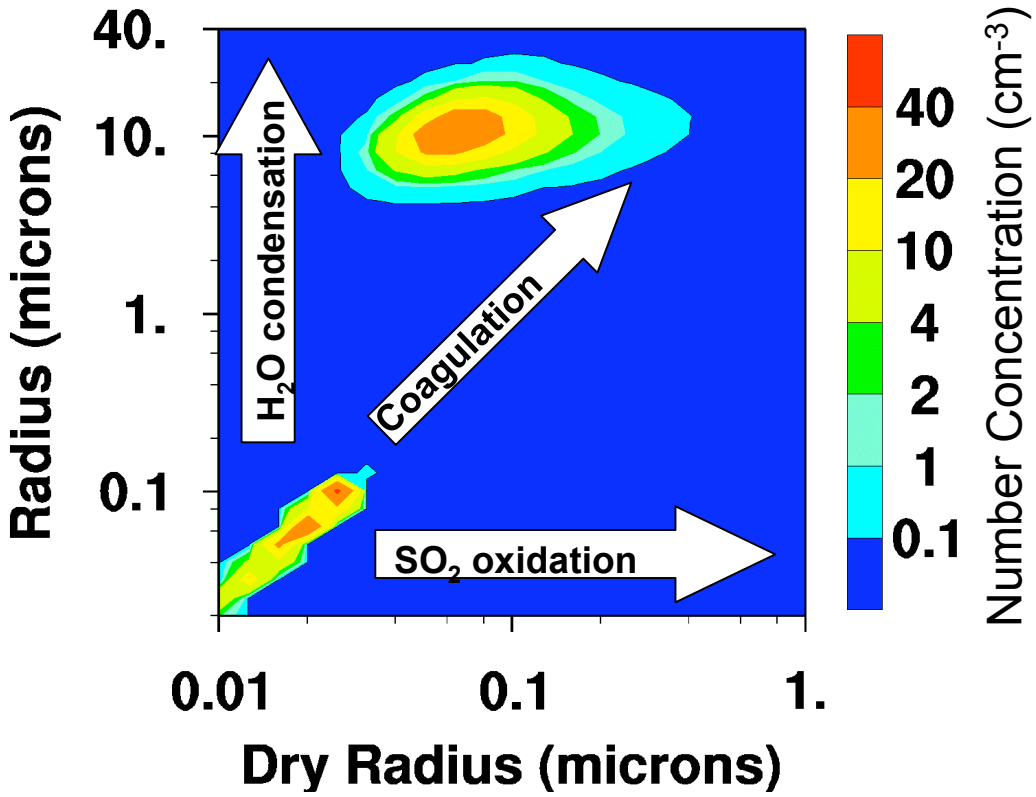
Parameterization



Meskhidze, Nenes, Conant, and John H. Seinfeld: Evaluation of a new cloud droplet activation parameterization with in situ data from CRYSTAL-FACE and CSTRIPE. JGR 2005.

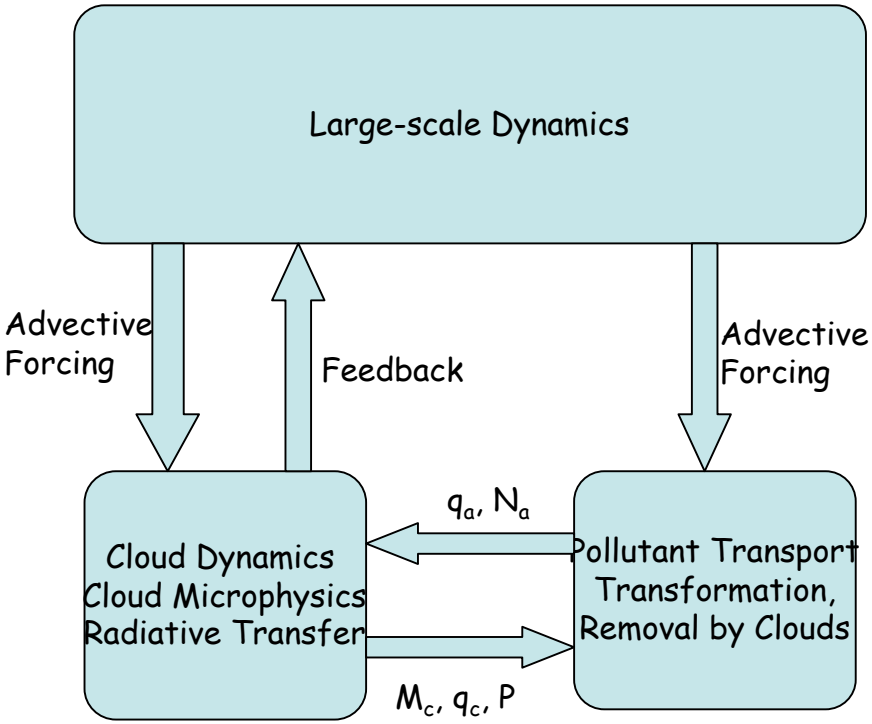
Aerosol Processes: Aqueous chemistry

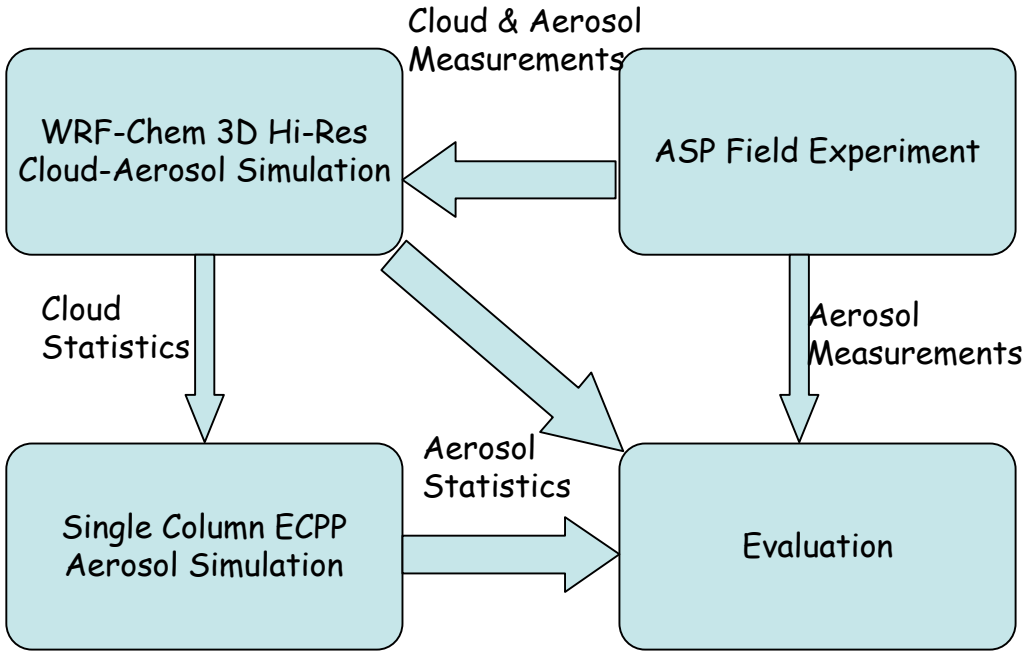
Current treatment	Target treatment	How MILAGRO can help
Bulk treatment (same for all cloud droplets). pH dependence.	*Size dependent*	Detailed modeling



Aerosol Processes:
Convective Cloud Transport, Transformation and Removal

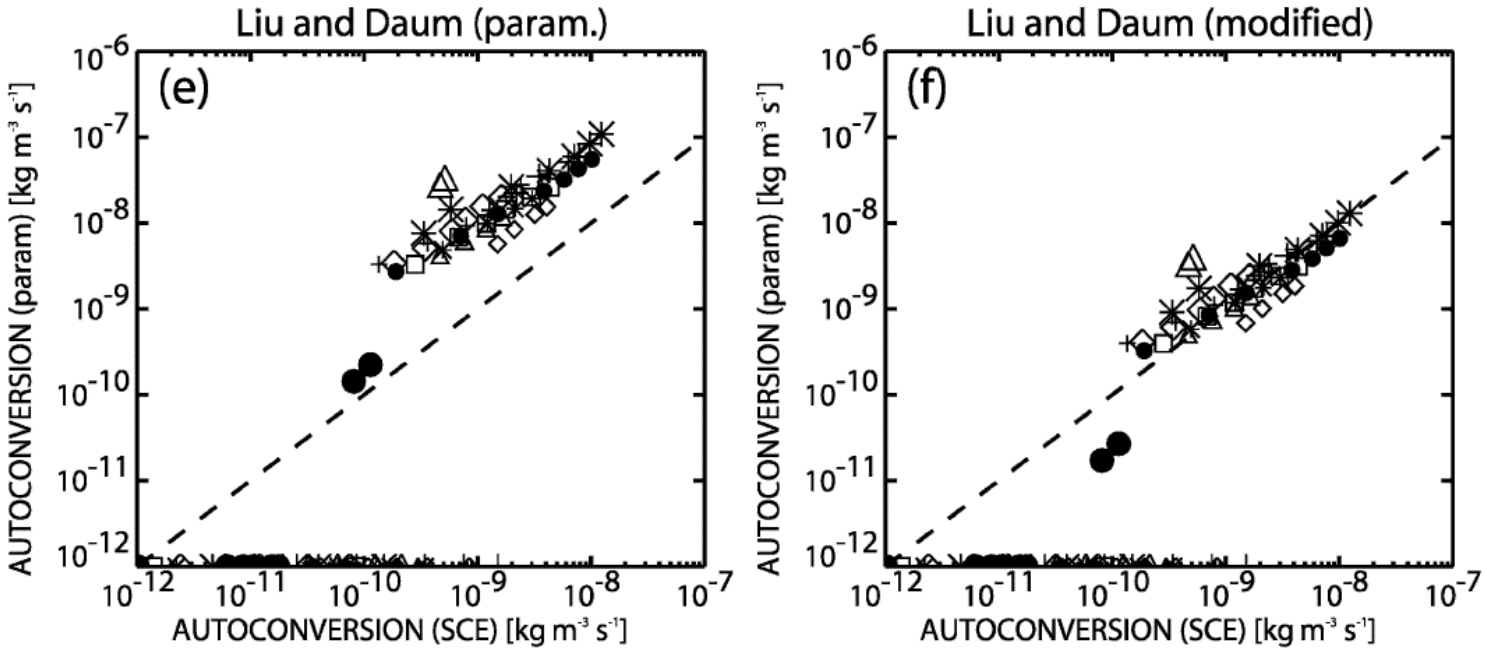
Current treatment	Target treatment	How MILAGRO can help
Cumulus parameterization. Ambiguous cloud fraction.	Cumulus parameterization with realistic cloud fraction, aqueous chemistry. *Statistics from embedded cloud models.*	Wind, gas, aerosol, cloud measurements. Evaluation of parameterizations in integrated models.





Aerosol Processes: In-cloud scavenging

Current treatment	Target treatment	How MILAGRO can help
Precipitation rate independent of aerosol	Precipitation rate depends on aerosol (Liu et al. 2005). Add influence of collision/coalescence on number. Subgrid variability in autoconversion.	CVI+DMA+CAPS measurements. Evaluation of collision / coalescence scheme. Evaluation of integrated models.



Wood: Drizzle in stratiform boundary layer clouds. Part II: Microphysical aspects. JAS 2005

Closure Experiments

- Focus on a single property or process
- Require precise measurements

Experiment	Input Data	Validation Data
Aerosol mass concentration	Size-resolved ions, BC, organic, dust	Aerosol size distribution
Refractive index	Size-resolved ions, BC, organic, dust, mixing state	Refractive index
Radiative absorption	Size-resolved ions, BC, organic, dust, mixing state, refractive indices	Absorption
Radiative scattering	Size-resolved ions, BC, organic, dust, mixing state , refractive indices	Scattering
Radiative extinction	Size-resolved ions, BC, organic, dust, mixing state , refractive indices	Extinction
New particle formation	CNC, ultrafine size distribution, RH, H_2SO_4, NH_3, organic vapor	CNC, ultrafine size distribution
Water uptake	Size-resolved ions, BC, organic, dust, RH	Hygroscopicity, humidification size factor, extinction factor

Closure Experiments

Experiment	Input Data	Validation Data	Reference
Aerosol mass concentration	Ions, BC, organic, dust concentration	Total mass concentration	Zhang et al. (2005)
Refractive index	Ions, BC, organic, dust, mixing state	Refractive index	
Radiative absorption	Ions, BC, organic, dust, size distribution, mixing state	absorption	
Radiative scattering	Ions, BC, organic, dust, size distribution, mixing state	scattering	Clarke et al. (2002)
Radiative extinction	Ions, BC, organic, dust, size distribution, mixing state	extinction	Wang et al. (2002)
New particle formation	CNC, ultrafine size distribution, RH, H_2SO_4, NH_3, organic vapor	CNC, ultrafine size distribution	McMurry et al. (2005)
CCN concentration	Ions, BC, organic, dust, aerosol size distribution	CCN concentration	Cantrell et al. (2001); Gasparini et al. (2006)
Water uptake	Ions, BC, organic, dust, aerosol size distribution, RH	Humidification size factor, extinction factor	Swietlicki et al. (1999); Dick et al. (2000)
Aerosol activation	CCN spectrum, ions, BC, organic, dust, aerosol size distribution, updraft velocity	Droplet number, activated aerosol size distribution	Conant et al. (2004); Meskhidze et al. (2005)
Droplet collision /coalescence	Vertical profile of droplet size distribution	Drizzle size distribution	Wood (2005)

Summary

- The representation of aerosol properties and processes in current global climate models is of necessity crude.

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- The climate modeling community seems to be receptive to devoting computational resources to improving the representation of aerosol properties and processes.

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- The climate modeling community seems to be receptive to devoting computational resources to improving the representation of aerosol properties and processes.
- The treatment could be improved significantly simply by applying treatments currently used in global aerosol models.

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- The climate modeling community seems to be receptive to devoting computational resources to improving the representation of aerosol properties and processes.
- The treatment could be improved significantly simply by applying treatments currently used in global aerosol models.
- MILAGRO can provide the measurements, process modeling, and integrated modeling needed to develop the next generation of parameterizations of aerosol properties and processes.

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- The representation of aerosol properties and processes in current global climate models is of necessity crude.
- The climate modeling community seems to be receptive to devoting computational resources to improving the representation of aerosol properties and processes.
- The treatment could be improved significantly simply by applying treatments currently used in global aerosol models.
- MILAGRO can provide the measurements, process modeling, and integrated modeling needed to develop the next generation of parameterizations of aerosol properties and processes.
- Issues of scale and uncertainty need to be addressed.

Summary

- The representation of aerosol properties and processes in current global climate models is of necessity crude.
- The climate modeling community seems to be receptive to devoting computational resources to improving the representation of aerosol properties and processes.
- The treatment could be improved significantly simply by applying treatments currently used in global aerosol models.
- MILAGRO can provide the measurements, process modeling, and integrated modeling needed to develop the next generation of parameterizations of aerosol properties and processes.
- Issues of scale and uncertainty need to be addressed.
- Lagrangian parcel models and regional chemical transport models will play an essential role in isolating transport effects from chemistry and aerosol physics.