Insights into the Kinetics of Secondary Organic Aerosols

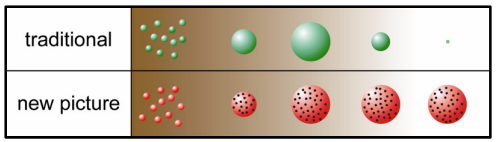
Alla Zelenyuk et al. / Pacific Northwest National Laboratory

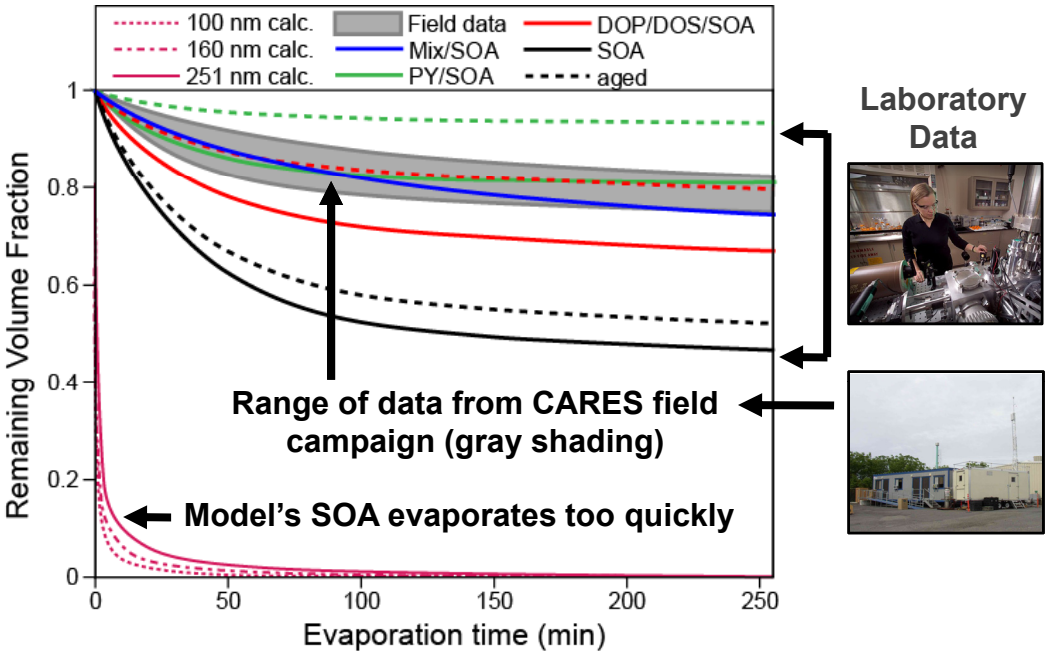
Science Question

- Do current assumptions regarding the role of evaporation on secondary organic aerosols (SOA) evolution explain why model predictions of organic aerosols are too low?

Approach

- Single particle mass spectrometer, SPLAT II, used to characterize the evaporation kinetics of populations of individual particles.
- Controlled laboratory experiments compared with samples collected during the June 2010 CARES campaign; both behave similarly and trap hydrophobic organics - slows evaporation with aging.





Findings

- Field and laboratory data indicate SOA evaporation is **10–100 times slower** than expected by models.
- SOA treatments in models all assume a liquid-like behavior in equilibrium with the gas phase not supported by this data; **that needs to be significantly revised**.

Vaden, T.D., D. Imre, J. Beranek, M. Shrivastava, and A. Zellenyuk. 2011. "Evaporation kinetics and phase of laboratory and ambient organic aerosol." *Proceedings of the National Academy of Science*, doi:10.1073/pnas.1013391108.

Vaden, T.D., C. Song, R.A. Zaveri, D. Imre, and A. Zelenyuk. 2010. "Morphology of mixed primary and secondary organic particles and the adsorption of spectator organic gases during aerosol formation." *Proceedings of the National Academy of Sciences*, 107(15), 6658–6663, doi:10.1073/pnas.0911206107.

How Important Is Organic Aerosol Hygroscopicity to Aerosol Indirect Forcing?

Xiaohong Liu (PNNL) and Jian Wang (BNL)

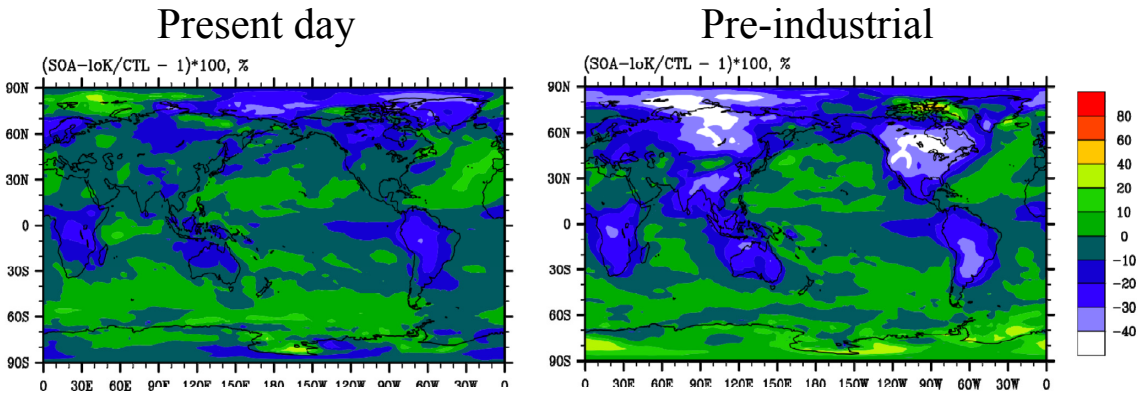
Science Question

What is the uncertainty in simulated global aerosol indirect forcing due to the uncertainties in our current understanding of organic hygroscopicity?

Approach

- A version of NCAR Community Atmospheric Model (CAM) is used to simulate CCN concentration, cloud droplet number concentration (CDNC), and Aerosol indirect forcing.
- The organic hygroscopicities are varied within typical ranges based on previous field and laboratory studies, and their impact on simulated CCN concentration, droplet number concentration and aerosol indirect forcing are examined.

Percentage change in CCN (*S*=0.1%) at 859 hPa when **k of secondary organics decreases from 0.14 to 0.07.**



Greater changes in CCN concentration for pre-industrial simulation.

Key Accomplishment

The uncertainty in our current understanding of organic hygroscopicity leads to a variation of simulated Global annual mean aerosol indirect forcing by 30%. This large variation is caused by the disproportional changes in cloud droplet number concentration between the present-day and pre-industrial conditions when relative loadings of sulfate versus organic are significantly different.

Publication

Liu, X., and J. Wang (2010), How Important Is Organic Aerosol Hygroscopicity to Aerosol Indirect Forcing? *Environmental Research Letters*, *5*, 044010, doi:10.1088/1748-9326/5/4/044010.

Aerosol Modeling Testbed: A Community Tool

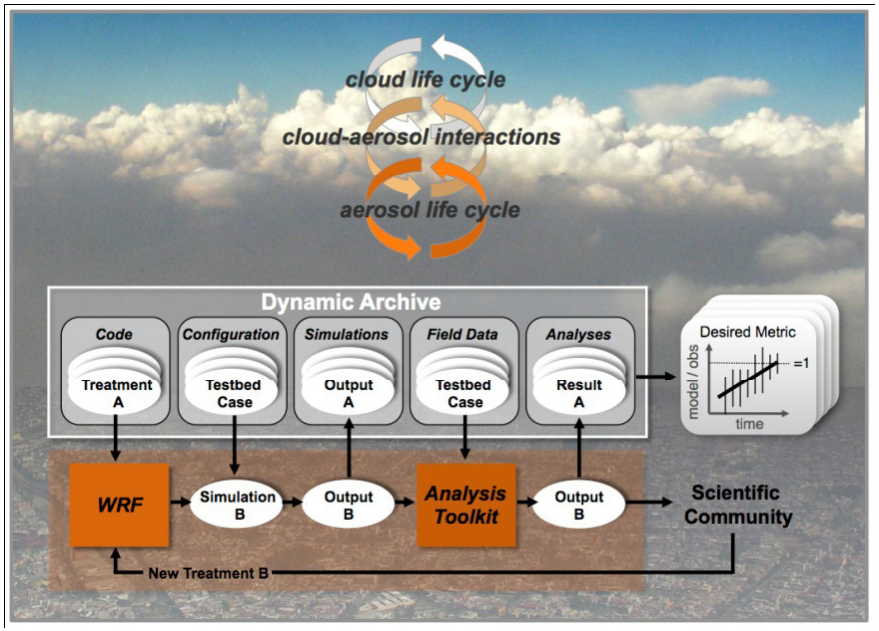
Jerome Fast et al. / Pacific Northwest National Laboratory

Science Question

- Can we reduce the large uncertainties in predicted aerosols and aerosol radiative forcing by changing the haphazard approach currently employed by the scientific community?

Approach

- Streamline the process of testing and evaluating aerosol and clouds process modules over a **range of spatial scales**.
- Target specific processes to **objectively and systematically** evaluate aerosol process modules and better quantify model uncertainties.
- Provide tools that **facilitate science** by minimizing redundant tasks.
- **Document** computational expense and performance, share all research activities with scientific community.



Findings

- The Aerosol Modeling Testbed, a **new aerosol modeling paradigm**, provides researchers a comprehensive tool to better understand new factors and parameters and link their results to the research of others.
- A **shared, systematic approach** across the community will generate better understanding of model uncertainties and allow scientists to better advise policy makers.

Fast, J.D., W.I. Gustafson Jr., E.G. Chapman, R.C. Easter, J.P. Rishel, R.A. Zaveri, G.A. Grell, and M.C. Barth. 2011. "The Aerosol Modeling Testbed: A Community Tool to Objectively Evaluate Aerosol Process Modules," Bull. Amer. Meteor. Soc., 92, 343-360. DOI: 10.1175/2010BAMS2868.1.

Laboratory Studies of Secondary Organic Aerosol CCN Activity as a Function of Oxygen-to-Carbon Ratio for a Range of Organic Precursors

A.T. Lambe[1,2], D.R. Croasdale[1], J.P. Wright[1], P. Davidovits[1], T.B. Onasch[1,2], P. Massoli[2], L. R. Williams[2], D.R. Worsnop[2], J.T. Jayne[2], C.E. Kolb[2], and W.H. Brune[3] / [1]Boston College , [2]Aerodyne Research Inc. [3]Penn State

Science Question

Can CCN activity and chemical composition of secondary organic aerosol (SOA) be related using a single-parameter representation for use in climate models?

Approach

• Generate SOA particles using a laboratory aerosol flow reactor.

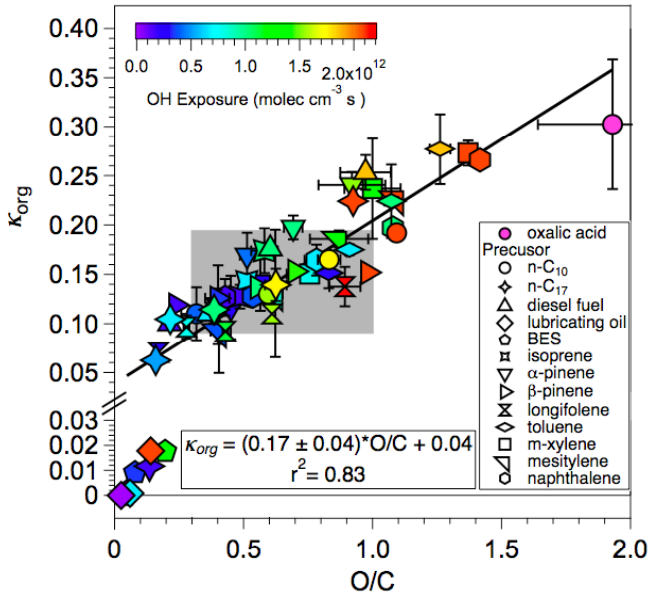
• Measure hygroscopicity of SOA (κ_{org}) over 1-20 days of equivalent atmospheric oxidation by OH radicals.

• Represent κ_{org} as a function of oxygen-to-carbon (O/C) ratio of the SOA.

Key Accomplishment

SOA hygroscopicity measurements indicate a linear relationship between κ_{org} - O/C over range of typical ambient values (grey region). The slope of a linear fit is 0.17, with ±1σ value of 0.04. Measurements for O/C < 0.3 fall off the trend line, indicating a different κ_{org} - O/C relationship in this range that will be explored in future work.

κ_{org} as a function of O/C ratio



Publication

Lambe, A. T., et al., 2011: Laboratory studies of the chemical composition and cloud condensation nuclei (CCN) activity of secondary organic aerosol (SOA) and oxidized primary organic aerosol (OPOA), *Atmos. Chem. & Phys. Discuss.*, 11, 13617-13653, 10.5194/acpd-11-13617-2011.

Comparing Simple and Complex Treatments of Organics

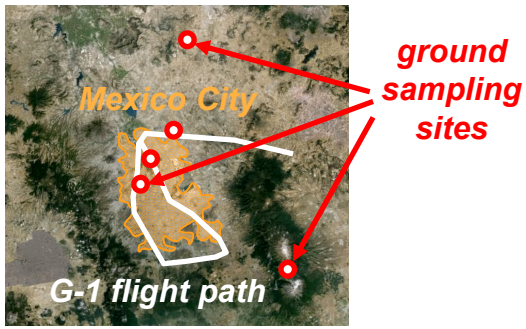
Manish Shrivastava et al. / Pacific Northwest National Laboratory

Science Question

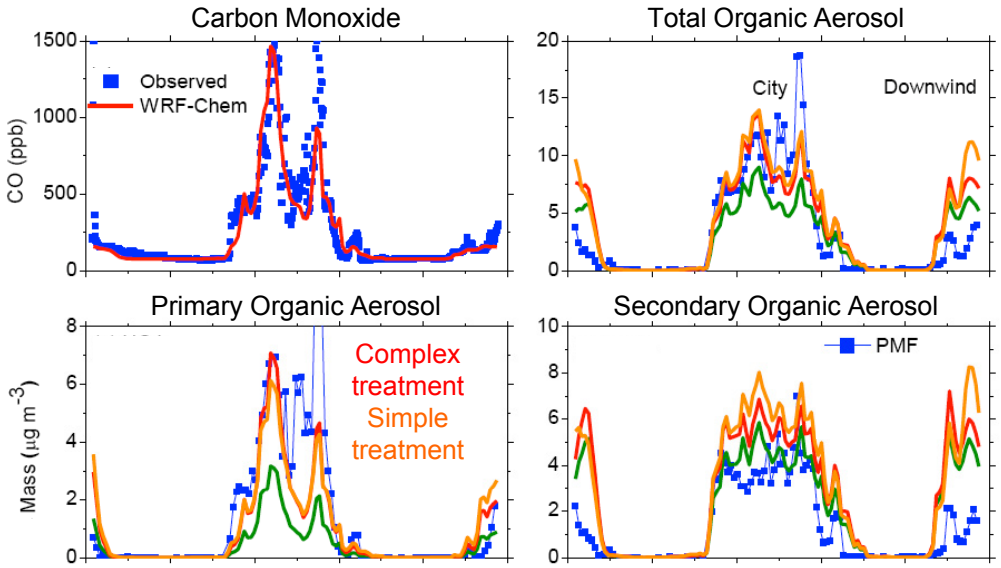
- Is it possible to reduce the complexity of representing secondary organic aerosol (SOA) in models?

Approach

- Implement a simple and computationally efficient treatment of SOA coupled with the MOSAIC aerosol model based on the Volatility Basis Set approach and compare it to a more complex benchmark treatment.
- Utilize the Aerosol Modeling Testbed tools to compare predictions using the extensive organic aerosol data collected during the MILAGRO campaign near Mexico City.



Comparison of Two Approaches for the G-1 Flight on March 15, 2006



Findings

- Developed for the first time **a simplified way to represent SOA** that is more suitable for 3D applications than more complex representations
- Both methods produced too little SOA in the city, SOA that was similar to observed aloft over areas surrounding the city, and too much SOA further downwind.

Shrivastava, M, J. Fast, RC Easter, WI Gustafson Jr., RA Zaveri, A Hodzic, and J Jimenez. 2011. "Modeling organic aerosols in a megacity: comparison of simple and complex representations of the volatility basis set approach." *Atmos. Chem. Phys.*, 11, 6639–6662.

The Importance of Aerosol Mixing State on CCN Spectrum

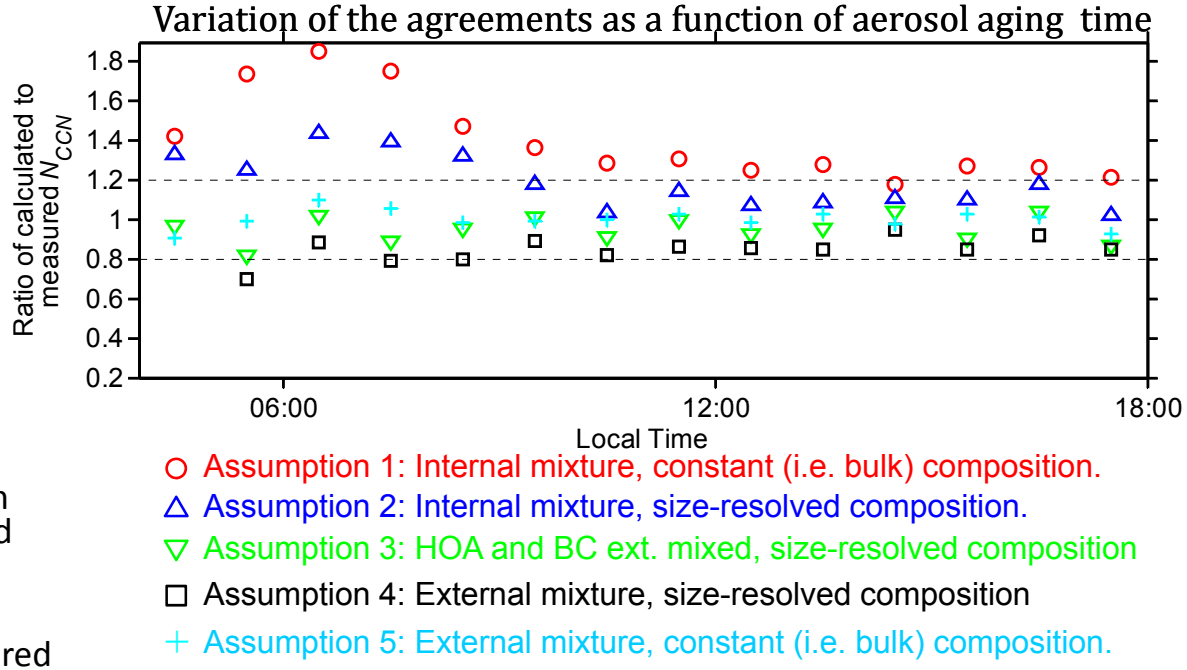
J. Wang (BNL), M. J. Cubison, A. C. Aiken, J. L. Jimenez, and D. R. Collins

Science Question

How important is the assumed mixing state on simulated CCN spectrum, and how does this importance vary with atmospheric aging of aerosols?

Approach

- Aerosol microphysics and chemical composition were characterized at an urban site during the Megacity Initiative: Local and Global Research Observations (MILAGRO) study.
- CCN spectrum is calculated from the measured aerosol size distributions and various simplified scenarios of aerosol mixing state and chemical composition.
- The agreement between measurement and calculation based on various simplifications and the variation of the agreement with the aging of the aerosol are examined.
- The influence of assumed mixing state on predicted CCN spectrum is studied using both aerosols observed during MILAGRO and representative aerosol types.



Key Accomplishment

The rapid mixing of primary non-hygroscopic aerosol particles with photochemically produced hygroscopic species suggests that during daytime, a few tens of kilometers away from aerosol sources, CCN spectrum may be derived with sufficient accuracy by assuming an internal mixture and using bulk chemical composition.

Publication

Wang, J. et al., (2010), The importance of aerosol mixing state and size-resolved composition on CCN concentration and the variation of the importance with atmospheric aging of aerosols, *Atmos. Chem. Phys.*, 10, 7267–7283.

Understanding Unresolved Aerosol in Global Climate Models

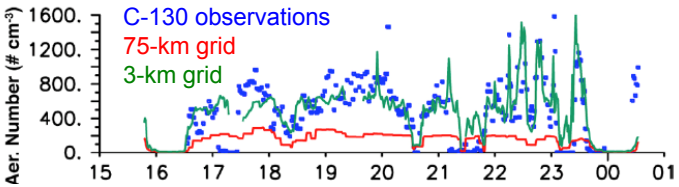
William Gustafson et al. / Pacific Northwest National Laboratory

Science Question

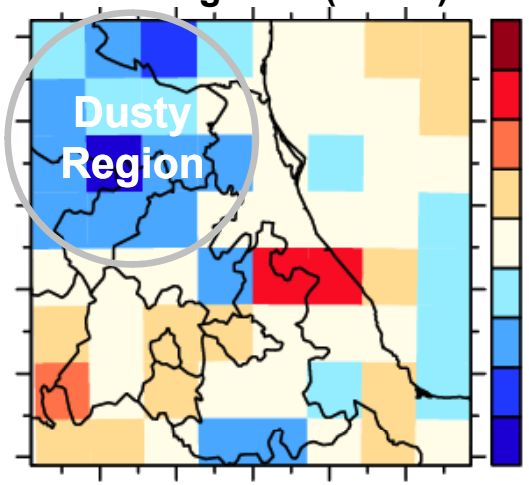
- Is neglecting unresolved aerosol processing significant in terms of coarse climate model predictions?

Approach

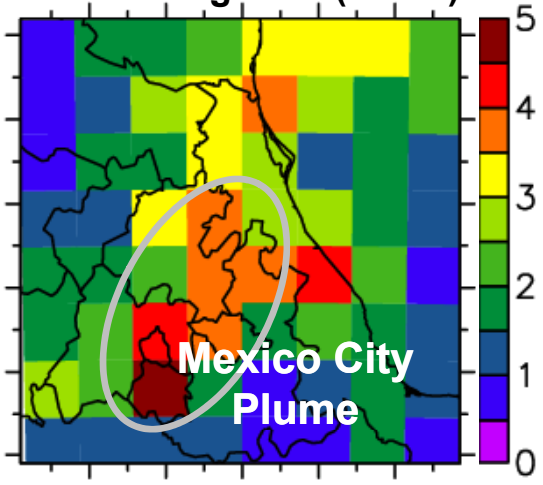
- Understand and quantify subgrid aerosol processes.
- Use the WRF-Chem model and low and high resolutions to identify missing aerosol information at low resolution.
- Use the Aerosol Modeling Testbed with observational data from the MILAGRO field campaign to test and evaluate the model.



Mean Bias
Aerosol Radiative Forcing, Including Dust (W m^{-2})



RMSE
Aerosol Radiative Forcing, Excluding Dust (W m^{-2})



Findings

- Neglecting subgrid aerosol processes can lead to a **30% error in direct radiative forcing** around Mexico City.
- Coarse global models need to account for subgrid scale meteorological impacts on online emission sources such as dust and sea-salt.

Gustafson Jr. WI, Y Qian, and JD Fast. "Downscaling Aerosols and the Impact of Neglected Subgrid Processes on Direct Aerosol Radiative Forcing for a Representative GCM Grid Spacing." Journal of Geophysical Research-Atmospheres, 116, D13303, 28 PP., 2011. DOI:10.1029/2010JD015480. http://www.agu.org/pubs/crossref/2011/2010JD015480.shtml

Kinetics versus Thermodynamics in the Growth of SOA: Implication from studies of α-Pinene Oxidation

B. J. Finlayson-Pitts / University of California, Irvine

Science Question

What is the mechanism of SOA growth from simultaneous oxidation of a-pinene by O_3 and NO_3 radicals ?

Approach

•Formation of SOA was studied in a unique large volume slow-flow aerosol flow tube system from reaction of a-pinene with a mixture of NO_2 at variable concentrations and a fixed O_3 concentration.

•Organic nitrates were measured in the gas phase and in the SOA using aerosol mass spectrometers (AMS and SPLAT-II), FTIR, and LC-UV.

•Partition coefficients (K_p) of organic nitrates between the gas phase and SOA particles were determined from the experimental data:

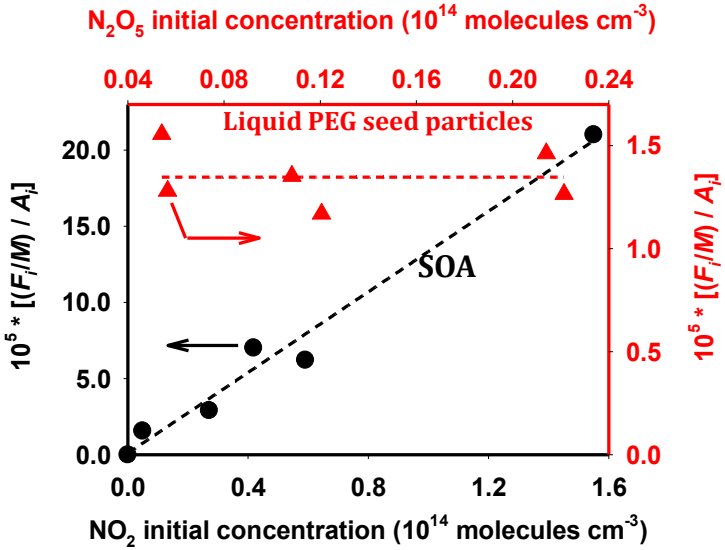
$$K_p = \frac{F_i / M}{A_i}$$

M = Total particle mass concentration (μg m^{-3})

A_i = Concentration of organic nitrates in the gas phase (μg m^{-3})

F_i = Concentration of organic nitrates in the SOA (μg m^{-3})

Dependence on NO_2 of K_p for organic nitrates partitioning into SOA or liquid polyethylene glycol particles



Key Accomplishment

First demonstration that uptake of organic nitrates between the gas phase and particles is not consistent with thermodynamic equilibrium partitioning, but rather with a kinetically determined growth mechanism. Atmospheric models that assume equilibrium partitioning will have to be reformulated.

Publication

V. Perraud , E. A. Bruns, M. J. Ezell, S. N. Johnson, Y. Yu, L. M. Alexander, A. Zelenyuk, D. Imre, W. L. Chang, D. Dabdub, J. F. Pankow, B. J. Finlayson-Pitts, 2011: Importance of Kinetic Effects in Atmospheric Secondary Organic Aerosol Formation, submitted.

Nighttime Aerosol Chemistry from Urban / Industrial Sources

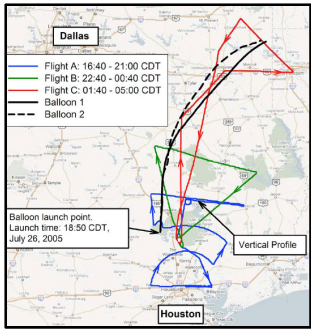
Rahul Zaveri et al. / Pacific Northwest National Laboratory

Science Question

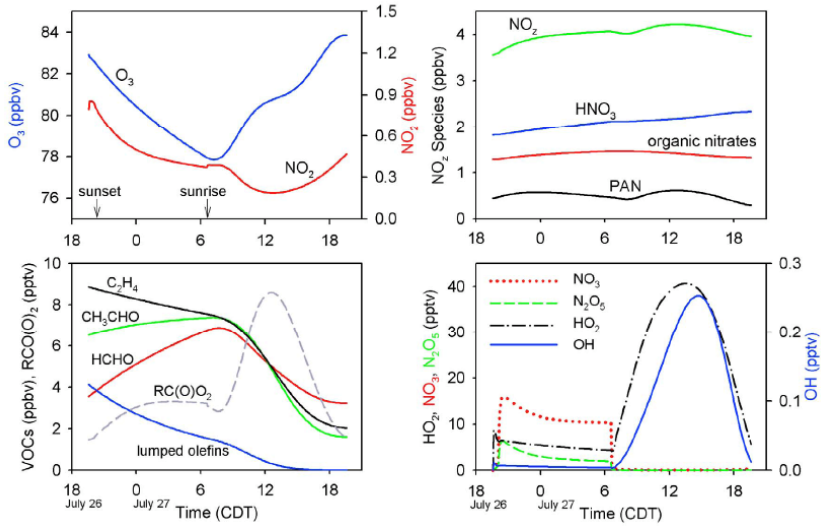
- How does chemistry at night affect aerosol aging the next day?

Approach

- There have been relatively few measurements of trace gases and aerosols at night; therefore, Twin Otter aircraft flights measured in situ chemistry downwind of Houston.
- Tetroons used to define Lagrangian trajectories of air masses.
- Constrained box modeling using the MOSAIC aerosol model performed to simulate detailed trace gas and aerosol chemistry associated with aerosols from urban and industrial sources that are transported far downwind of Houston.



Evolution of Various Species as a Function of Processing Time



Findings

- High concentrations of volatile organic compounds were found at the altitude of the low-level jet, demonstrating that pollutants are transported over long distances at night.
- The model showed that **organic nitrates formed at night** via NO_3 reaction with olefins, potentially contributing to secondary organic aerosol formation several hundred kilometers downwind the next day.

Zaveri RA, PB Voss, CM Berkowitz, E Fortner, J Zheng, R Zhang, RJ Valente, RL Tanner, D Holcomb, TP Hartley, and L Baran. 2010. "Overnight atmospheric transport and chemical processing of photochemically aged Houston urban and petrochemical industrial plume." *Journal of Geophysical Research-Atmospheres 115: D23303.* DOI:10.1029/2009JD013495.

Understanding Emission Sources of Organic Carbon

Alma Hodzic et al. / National Center for Atmospheric Research

Science Question

- Do models adequately represent aerosol emissions and processes consistent with the observed relative contribution of fossil and modern carbon the atmosphere?

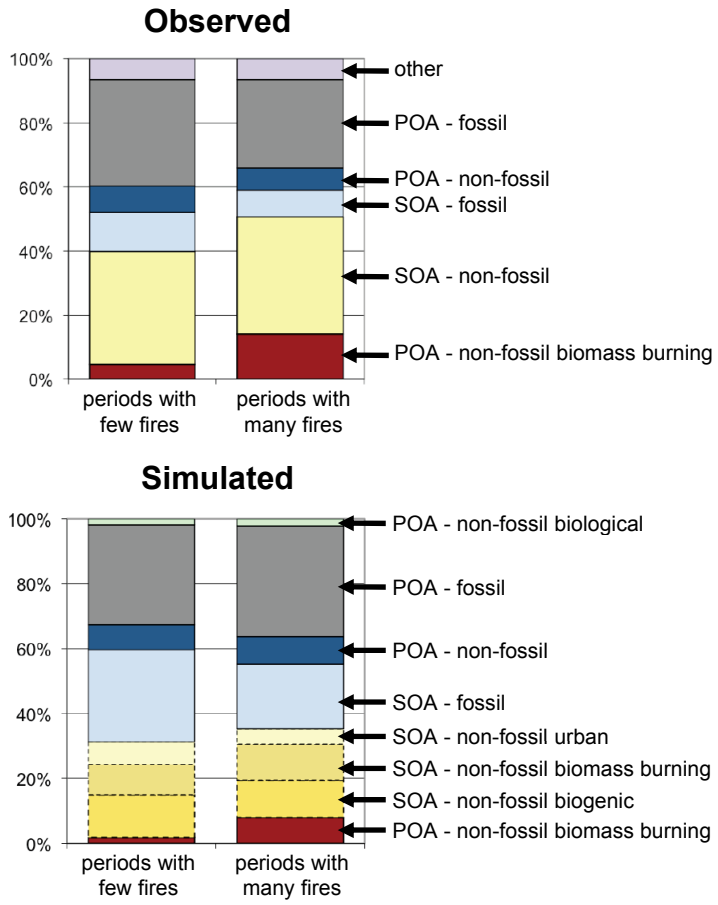
Approach

- A regional model with a state-of-the-science treatment of secondary organic aerosols used in conjunction with various data, including filter samples.
- Radioactive carbon data used to evaluate the performance of a regional atmospheric model when simulating the contributions of fossil fuels and modern carbon.

Modern carbon sources include biomass burning, forest fires, and cooking.

Findings

- C-14 data confirmed that organic matter surrounding Mexico City is comprised of surprisingly large amounts of modern carbon.
- Demonstrated **first-ever model simulations on modern carbon**.



Hodzic A, JL Jimenez, ASH Prévôt, S Szidat, JD Fast, and S Madronich. 2010. "Can 3-D models explain the observed fractions of fossil and non-fossil carbon in and near Mexico City?" *Atmospheric Chemistry and Physics* 10(22):10997–11016. DOI: 10.5194/acp-10-10997-2010.

Cloud Impact on Solar Radiation at Surface: Spectral Changes

Kassianov E., Barnard J., Berg L.K., Long C.N., and C. Flynn *Pacific Northwest National Laboratory*

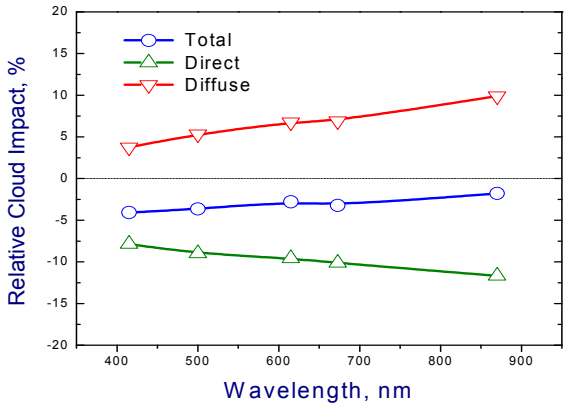
Science Question

- Typically, radiative transfer parameterizations applied in climate models have been evaluated using observed broadband radiative properties.
- Application of the broadband values alone could mask parameterization errors and therefore render them undetectable.
- *Can we provide spectrally resolved observational constraints for improved model evaluations?*

Approach

- We introduce a framework for estimating the *spectral* values of normalized cloud radiative forcing. This framework combines spectrally resolved all-sky solar fluxes measured by ground-based radiometers and the corresponding clear-sky fluxes estimated by a new physically-based approach.
- Application of this framework to summertime days with cumuli at the SGP site reveals that the magnitude and sign of the cloud impact on the amount of direct and scattered solar radiation reaching the surface can change substantially with wavelength.

The total cloud impact on the amount of solar energy reaching the Earth's surface is governed by its direct and diffuse components. These components have comparable magnitude and opposite signs. Aerosol is likely responsible for their spectral changes.



Key Accomplishment

The observed spectral changes of the total cloud impact and its direct and diffuse components should be considered as essential constraints for improved evaluations of cloud-aerosol-radiation interactions described by numerical models of the atmosphere.

Publication

Kassianov E., Barnard J., Berg L.K., Long C.N., and C. Flynn, 2011: Shortwave Spectral Radiative Forcing of Cumulus Clouds from Surface Observations, *Geophys. Res. Lett.*, L07801.

New Measurements of Carbonaceous Aerosol Properties

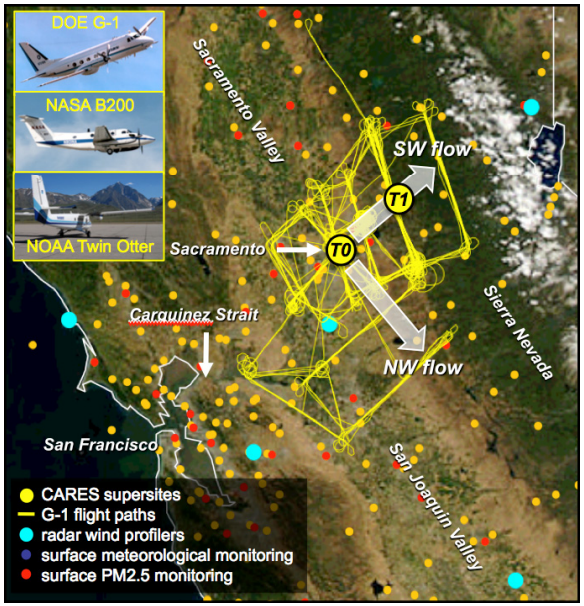
Rahul Zaveri et al. / Pacific Northwest National Laboratory

Objective

- Obtain measurements of black carbon, organic carbon, aerosol mixing state, and aerosol optical properties as part of the July 2010 Carbonaceous Aerosol and Radiative Effects Study (CARES) needed to evaluate and improve models.

Approach

- Two supersites and the G-1 research aircraft obtained extensive in-situ aerosol property measurements; an airborne lidar obtained vertical profiles of aerosol optical properties.
- 70 scientists from 20 institutions participated in CARES.
- Data available in ARM Data Archive for analysis.
- Collaboration and coordination with NOAA's CalNex campaign.



Measurements during CARES in central California

Findings

- **Interactions between biogenic and anthropogenic** emission sources likely important for secondary organic aerosol formation in the region.
- Found different **mixing states** of organic compounds mixed with black carbon, sulfates and nitrates.
- Observed very high levels of **ultrafine particles**, likely associated with new particle formation events.

Zaveri, R, et al. 2011. "Overview of the 2010 Carbonaceous Aerosols and Radiative Effects Study (CARES)." To be submitted to *Atmospheric Chemistry and Physics*.

Fast JD, et al.. 2011. "Transport and mixing patterns over central California during the Carbonaceous Aerosol and Radiative Effects Study (CARES)." To be submitted to *Atmospheric Chemistry and Physics*.

Improving Aerosol Optical Depth Estimates Near Clouds

Evgueni Kassianov et al. / Pacific Northwest National Laboratory

Science Question

- Can aerosol optical depth (AOD) near clouds be better quantified from observations?

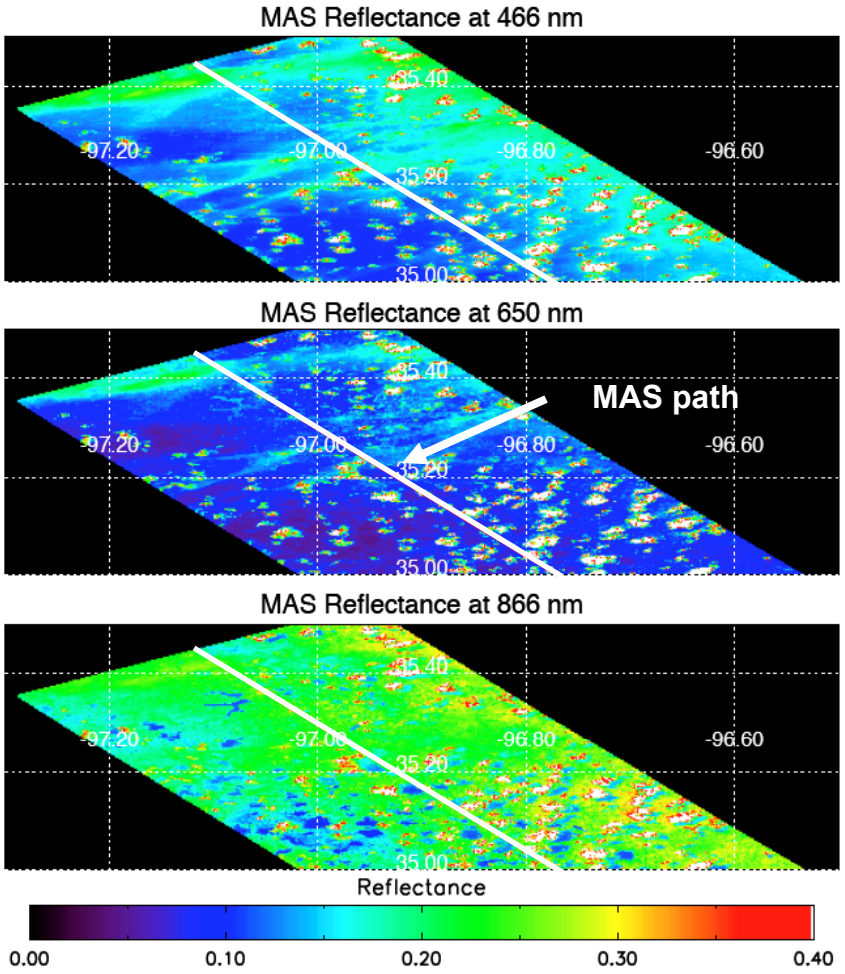
Approach

- Employ recently developed reflectance ratio method to generalized aerosol retrievals for partly cloudy conditions.
- Apply reflectance ratio against extensive measurements collected during the Cumulus Humulis Aerosol Processing Study (CHAPS) over the ARM Southern Great Plains site.

Findings

- The reflectance ratio-based AOD estimates were in good agreement (within 30%) with those from independent airborne lidar and surface radiometric observations.
- New methodology available to evaluate model predictions of AOD during partly cloudy conditions.

MODIS Airborne Simulation (MAS) Measurements over Oklahoma



Kassianov, E, M Ovchinnikov, LK Berg, SA McFarlane, C Flynn, C Ferrare, C Hostetler, and M Alexandrov. 2010. "Retrieval of aerosol optical depth in the vicinity of broken clouds from reflectance ratios: Case study." *Atmospheric Measurement Techniques,* 3, 1333–1349.

An Investigation Into the Aerosol Semi-Direct Effect at Barrow, AK

Zafer Boybeyi & John Lindeman
George Mason University, Fairfax, VA

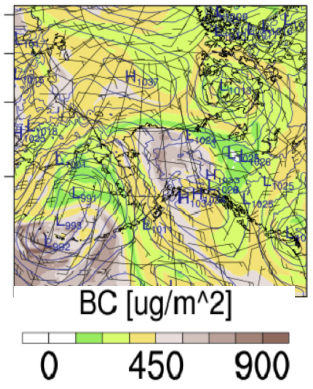
Science Question

How does the aerosol semi-direct effect influence atmospheric stability and cloud cover at Barrow, AK during an episode of Arctic haze? How sensitive is the atmosphere to variations in black carbon?

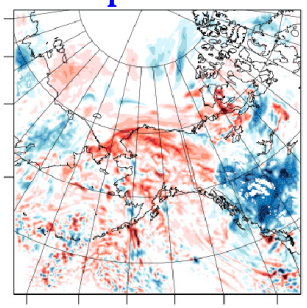
Approach

- WRF/Chem simulations of polluted conditions at Barrow during the ISDAC field experiment.
- Compare polluted case with identical baseline WRF/Chem run that does not include aerosol direct or semi-direct effects **(BASE)**.
- Sensitivity studies with WRF/Chem simulations containing twice the black carbon concentration **(BC)** and no black carbon **(no BC)**.

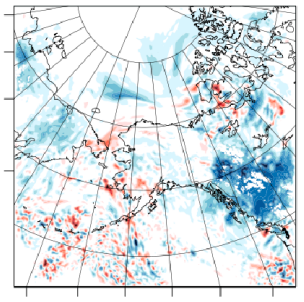
Black carbon concentration



850 mB potential temperature difference

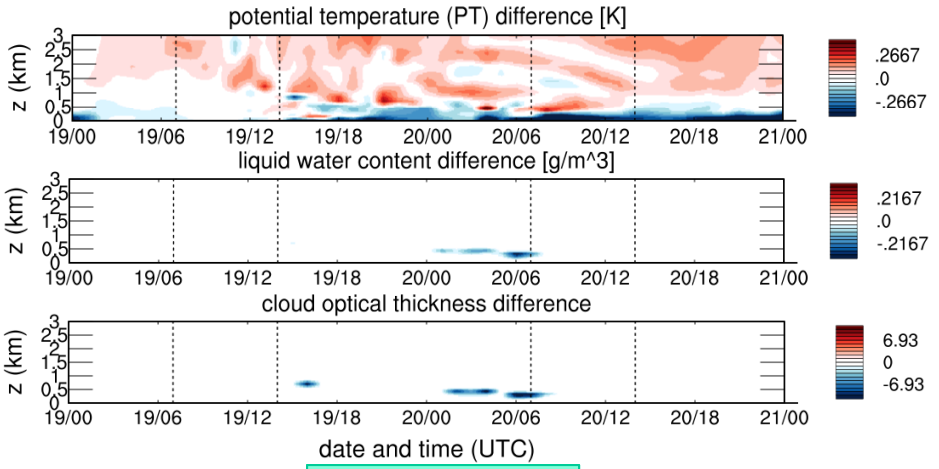


BC - BASE



no BC - BASE

Time series at Barrow, AK (April 19-21, 2008)



BC - BASE

Findings and conclusions

The aerosol semi-direct effect increases stability and dries the lower troposphere at Barrow, AK. The atmosphere is particularly sensitive to the black carbon concentration, which considerable enhances tropospheric warming.

Publication

Lindeman, J. & Z. Boybeyi., 2011: An examination of the aerosol semi-direct effect for a polluted case of the ISDAC field campaign, *Journ. Geo. Res.*, under review.

Fast Time-Resolved Aerosol Collector

Xiao-Ying Yu/ Pacific Northwest National Laboratory

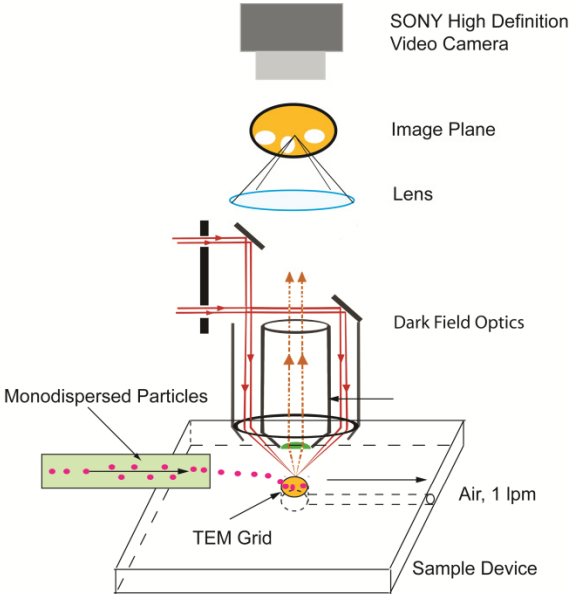
Science Question

How do we probe dust within microstructures of clouds from a traversing aircraft on the scale of 1 meter or less?

Approach

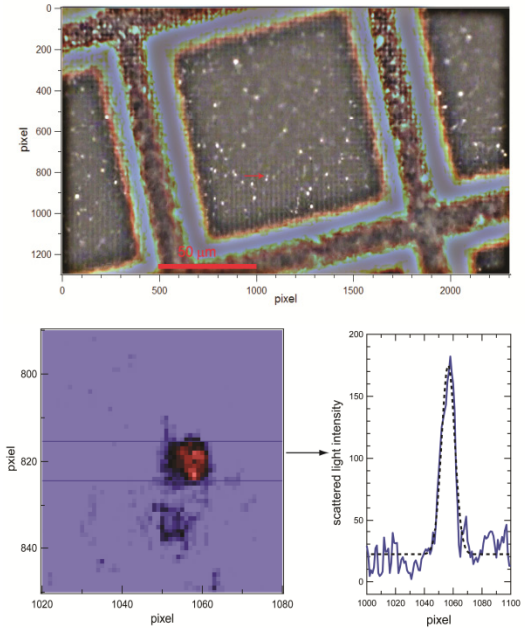
We choose:

- A non-scanning approach, i.e. no moving parts.
- The use of a commercial high definition digital video camera for imaging, low cost.
- Particle collection by microsieve: low flow impedance and good for post-field analysis.



A schematic of the experimental setup

A single 100 nm particle detected within 4 ms.



Key Accomplishment

In situ particle size and number concentration are resolved to 4–17 millisecond by optical tagging, making probing of cloud microstructure by airborne platform possible.

Publication

Yu, X.-Y. et al., 2010: Fast time-resolved aerosol collector: proof of concept, Atmos. Meas. Tech., 3, 1377–1384, doi: 10.5194/amt-3-1377-2010

Aerosol Aging on a Substrate without Wall-Effects by a Cross-Flow

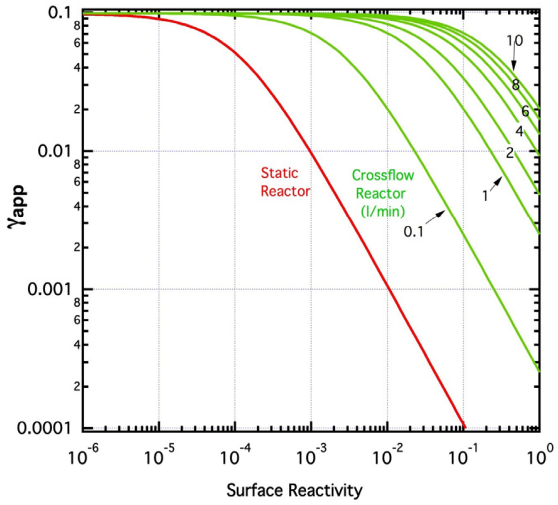
Xiao-Ying Yu/ Pacific Northwest National Laboratory

Science Question

How do we study aerosol aging and eliminate transport-limited trace gas depletion near the particles and substrates?

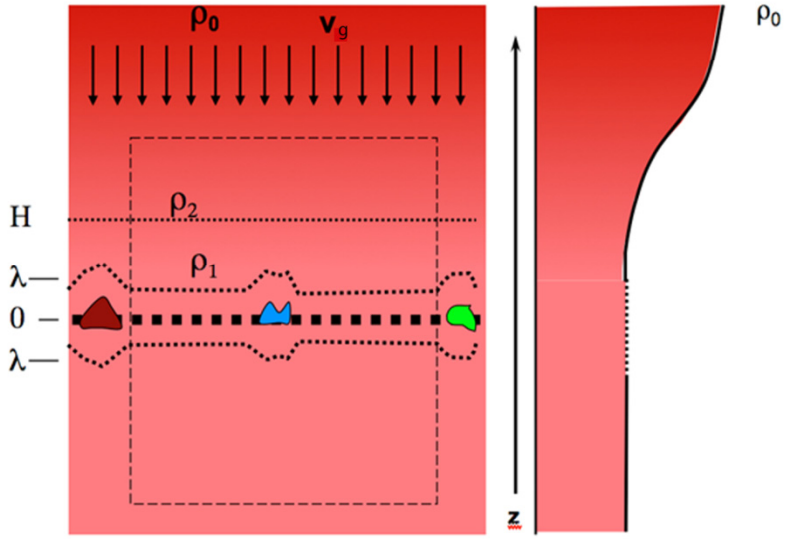
Approach

- Conducted theoretical analyses of
  1) an isolated single particle in a static gas
  2) multiple particles on a substrate in a static gas
  3) multiple particles on a substrate with a cross-flow.
- Showed that a cross-flow can be used to remove wall effects and establish a "zone of isolation."



The apparent reactive uptake coefficient γ_{app} versus surface reactivity, for M=1000 1 mm diameter particles with γ_p=0.1.

Gas transport in the cross-flow over a substrate. The dashed square represents the "zone of isolation" surrounding each particle (red, blue, and green).



Key Accomplishment

We have shown theoretically that pulling air through pre-deposited particles on microgrid substrates in cross-flow conditions is well-suited for long-term atmospheric chemistry studies. It is a convenient replacement for large aerosol chambers.

Publication

Cowin, J. P. et al., 2011: Theoretical analyses of aerosol aging on a substrate without wall-effects by a cross-flow, The Open Atmos. Sci. J., submitted.

Measuring the Increase in Organic Carbon within Individual Particles During CARES

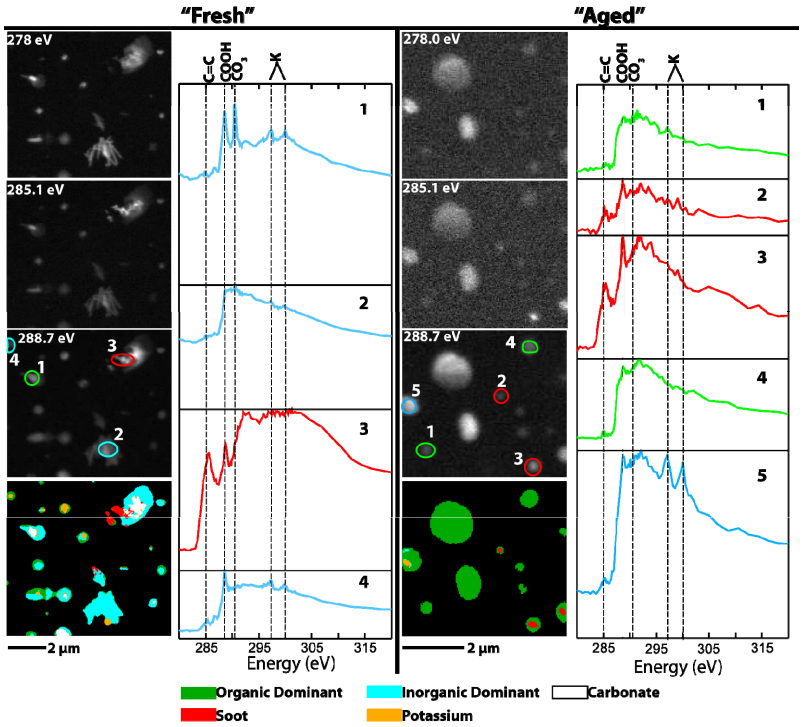
Ryan C. Moffet, Alexander Laskin, and Mary K. Gilles
Lawrence Berkeley National Laboratory/Pacific Northwest National Laboratory

Science Question

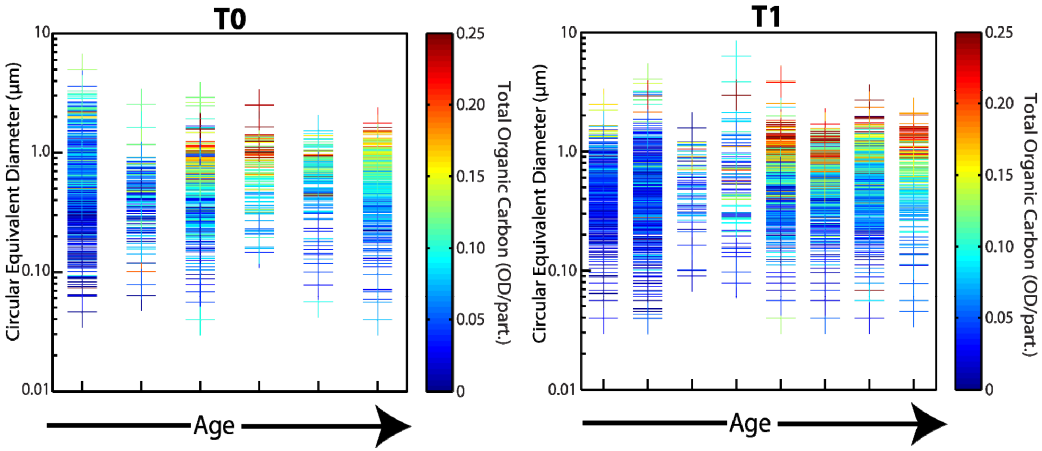
How does the mixing state of carbonaceous aerosols change as the urban aerosol plume ages?

Approach

- Chose period of photochemical aerosol formation during the CARES campaign.
- Use scanning transmission X-ray microscopy to quantitatively identify carbonaceous mixing states during CARES
- Image at different energies to obtain spatially resolved chemical mixing state for fresh and aged cases.



Organic Carbon Content for Individual Particles



Key Accomplishment

Identified a dramatic change in mixing state during a period of photochemical smog buildup. The carbon content of individual particles in Sacramento is at least a factor of two lower than in Mexico City.

Publications

Moffet, R. C., et al., Microscopic Characterization of Carbonaceous Aerosol Particle Aging in the Outflow from Mexico City, *Atmos. Chem. & Phys.*, 2010, 10, 961–976.

Moffet, R.C., et al., 2011: Spectro-Microscopic Studies of Carbonaceous Aerosol Aging in Central California, *Atmos. Chem. & Phys.*, In preparation.

Airborne HSRL Measurements and WRF-Flexpart Simulations of Aerosol Plume Transport and Transformation during MILAGRO

Benjamin de Foy/ Saint Louis U. – Sharon Burton, Rich Ferrare, Chris Hostetler, John Hair/ NASA - LaRC

Science Question

Can WRF-Flexpart simulations be used to qualitatively classify aerosol origin and estimate the age of aerosol plumes?

Approach

- Infer aerosol type using airborne HSRL measurements of aerosol optical properties.
- Simulate particle trajectories for different aerosol types using WRF-Flexpart.

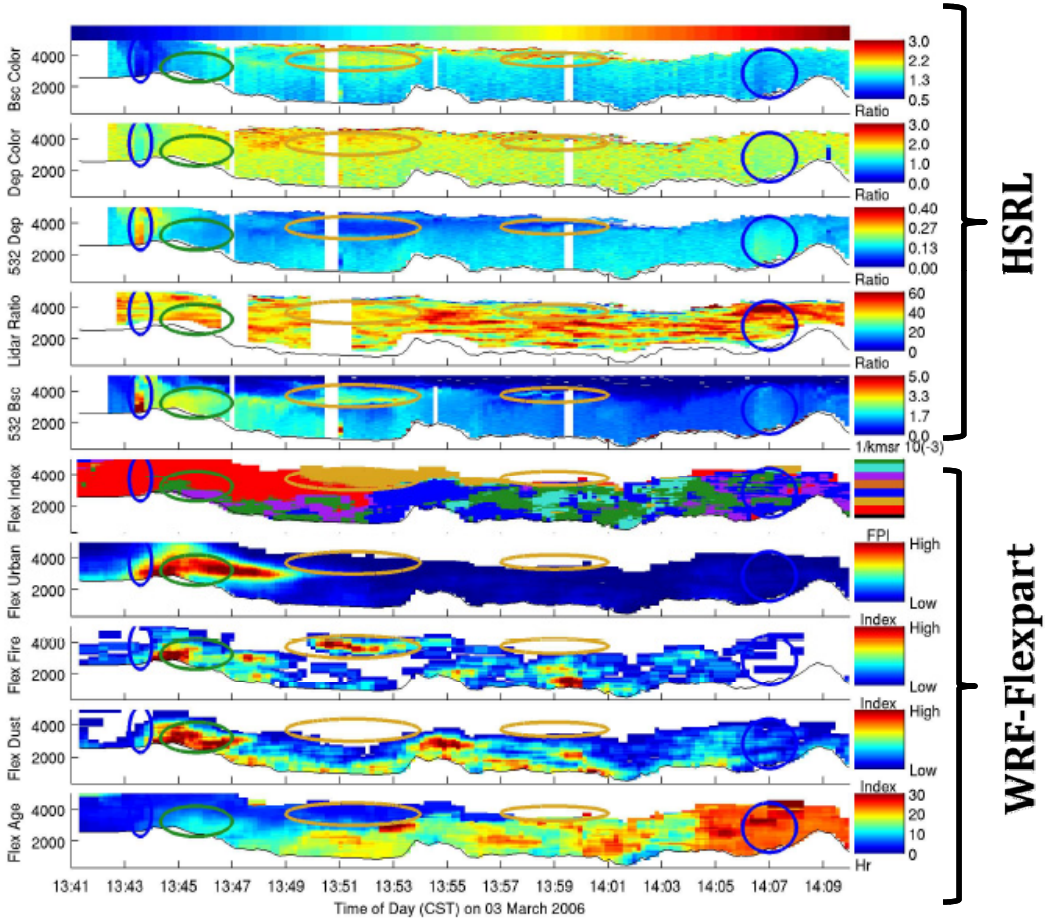
Key Accomplishment

• Used airborne HSRL measurements to infer aerosol type and evaluate WRF-Chem simulations of aerosol type and transport over Mexico City region during MILAGRO.
• Identified atmospheric tar balls in the hot plumes from MODIS-detected forest fires.

Publication

de Foy, B., et al. , Aerosol Plume Transport and Transformation in High Spectral Resolution Lidar measurements and WRF-Flexpart simulations during the MILAGRO Field Campaign, Atmos. Chem. Phys., 11, 3543–3563, 2011.

HSRL measurements and Flexpart Simulations
March 3, 2006



Curtain plots of HSRL backscatter coefficients and intensive properties for March 3, 2006 along with Flexpart average age and particle count index for dust, fire, and urban emissions. The yellow ellipses show biomass burning plumes, the blue ellipses show dust plumes, and the green ellipse shows a mixed plume.

Raman Lidar and HSRL Measurements of Aerosol and Water Vapor Variability

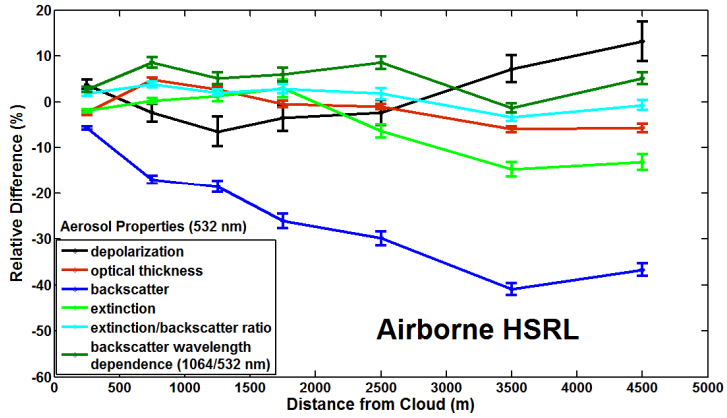
Rich Ferrare[1], Marian Clayton[2], Dave Turner[3], Chris Hostetler[3], John Hair[1], Mike Obland,[1] Ray Rogers[1]
[1]NASA/LaRC, [2]SSAI, [3]NOAA/NSSL

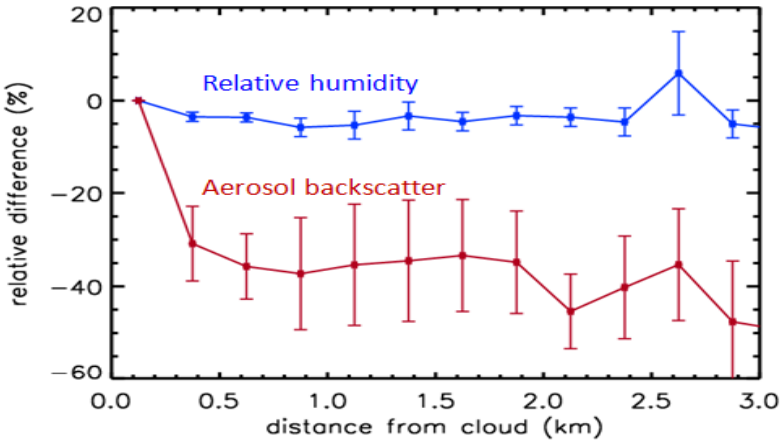
Science Questions

- How do aerosol optical and physical properties vary near clouds?
- How are these variations related to changes in relative humidity?
- How well can we use lidar to measure or infer these variations?

Approach

SGP Raman lidar aerosol and water vapor measurements and NASA Langley Research Center airborne High Spectral Resolution Lidar (HSRL) measurements acquired during the CHAPS and RACORO campaigns are used to investigate aerosol hygroscopicity and variations in aerosol properties near clouds in the daytime boundary layer.





Key Accomplishments

- SGP Raman lidar measured increased relative humidity (5–10%) near clouds.
- Raman lidar and HSRL measured increased aerosol backscatter (20–40%) and aerosol optical depth (5–10%) near clouds.
- HSRL measured decreased aerosol depolarization near clouds (10–20%), indicating that aerosols become more spherical with higher RH near clouds.
- Variations in aerosol properties and RH are largest at or within about 200 m below cloud base.

Towards a Particle-Resolved Aerosol Representation for the Simulation of the Aerosol Impact on Regional Scales

Matthew West and Nicole Riemer, UIUC

Science Questions

Couple the new stochastic, particle-resolved aerosol model, PartMC-MOSAIC (Riemer et al., 2009), with the community model system WRF-Chem to investigate the evolution of aerosol particles and their optical and hygroscopic properties during atmospheric transport, while accurately taking account of the full aerosol mixing state.

Approach

Improve the efficiency of PartMC-MOSAIC to enable its extension from a Lagrangian box model to a model that is coupled to a 1D (and eventually 3D) transport code by:
(1) a new algorithm for parallel simulation,
(2) "superparticles" as a coarse-graining method, with each superparticle representing a collection of physical particles.

Publication

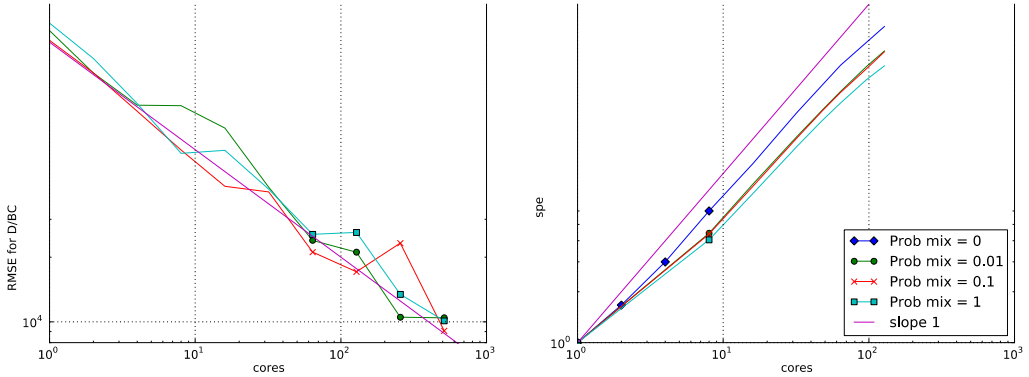
Riemer, N., M. West, R.A. Zaveri, R.C. Easter, Simulating the evolution of soot mixing state with a particle-resolved aerosol model, *J. Geophys. Res.* 114(D09202), 2009.

Key Accomplishments

Developed two algorithmic advances that lead to a 2-3 order of magnitude speedup:

1. Parallel Simulation via Mixing: The particle population is distributed over several processors. At each timestep processors simulate independently, then exchange particles with a given probability. We showed that this algorithm converges and scales near-linearly up to hundreds of cores.

2. Superparticle Method: This allows coarse-graining where each "superparticle" in our new scheme represents a family of identical physical particles. This method is useful for applications where different types of particles need to be tracked that have very disparate concentrations.



Global Modeling of Aerosol Absorption and Climate Impact

Yan Feng and Rao Kotamarthi/ Argonne National Laboratory

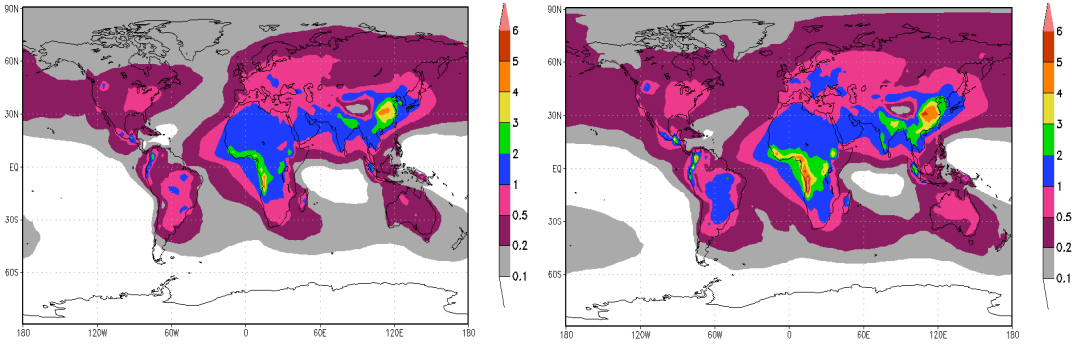
Science Question

What are the major uncertainties in aerosol absorption by global models, compared with surface and satellite observations?

Approach

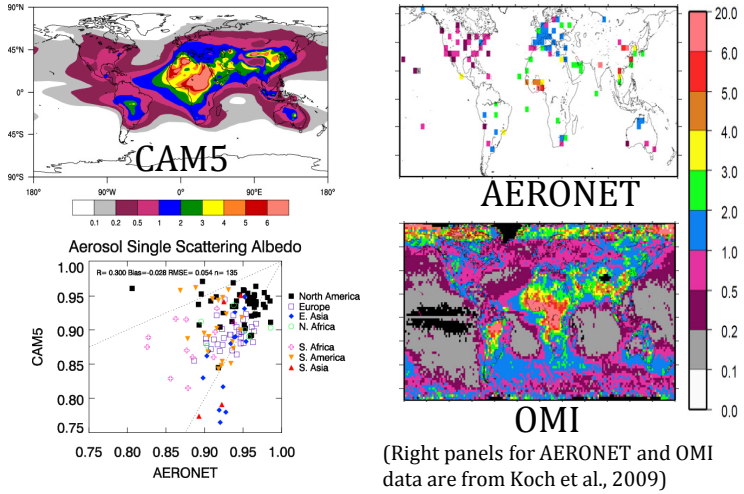
- Develop an optical treatment to account for absorbing organic carbon (brown carbon, BrC) in a global aerosol transport model.
- Evaluate simulated aerosol optical properties and vertical profiles using in situ and remote sensing data.
- Estimate uncertainty related to emissions.

Enhancement in absorbing aerosol optical depth



Standard BrC Strong absorbing BrC

Absorbing aerosol optical depth (x100)



(Right panels for AERONET and OMI data are from Koch et al., 2009)

Key Accomplishment

- **The inclusion of brown carbon could increase aerosol absorption by 6% to 38%.**
- **Preliminary evaluation shows that CAM5 underestimates aerosol absorption optical depth for its low-biased column loadings than surface concentrations or single-particle absorption.**

Publication

Feng, Y., et al., 2011: Effects of black carbon and brown carbon on surface solar radiation, in preparation.

Modeling Water and Solute Content in Atmospheric Aerosols

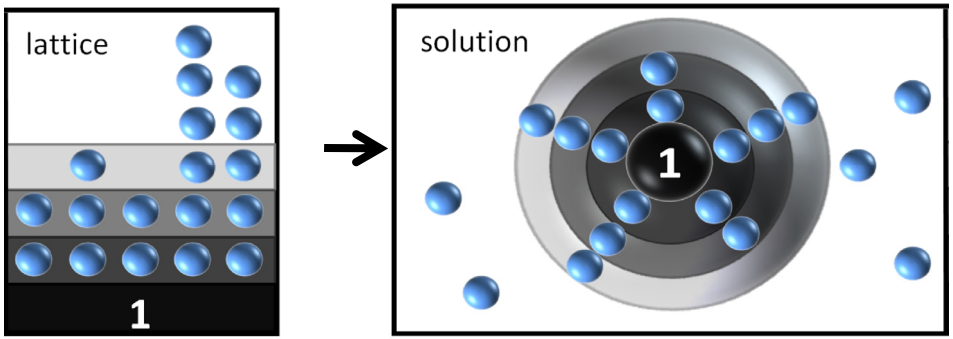
Cari Dutcher, Xinlei Ge, Simon Clegg and Anthony Wexler
University of California, Davis, and University of East Anglia

Importance

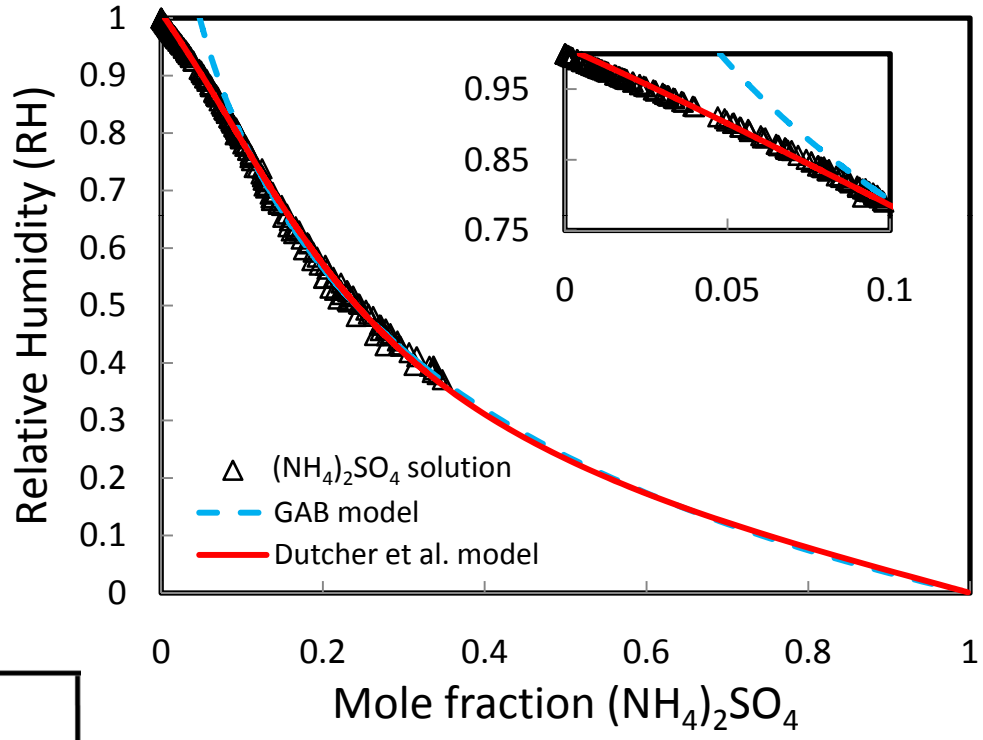
Accurate predictions of water and solute activities in atmospheric aerosols to very low relative humidities (*RH*) and over wide ranges of temperature are central to predictions of aerosol size, optical properties, and cloud formation.

Approach

- Apply the statistical mechanics of lattice adsorption on a solid substrate to the water / salt relationship in solution *(figure below, left to right).* This is a particularly powerful method for low relative humidity systems.
- Improve the model predictions by accounting for the energetic costs of hydration of salt ions in solution *(figure below, gray rings).*
- Expand the model to multi-salt solutions using standard mixing relationships.



Key accomplishment: Ability to represent *RH* / concentration relationships in soluble aerosols from 95% *RH* to dryness with a very simple model.



Publication

Dutcher, C.S., et al., Statistical mechanics of multilayer sorption: Extension of the Brunauer-Emmett-Teller (BET) and Guggenheim-Anderson-deBoer (GAB) adsorption isotherms. *J. Phys. Chem.* [in press], 2011.

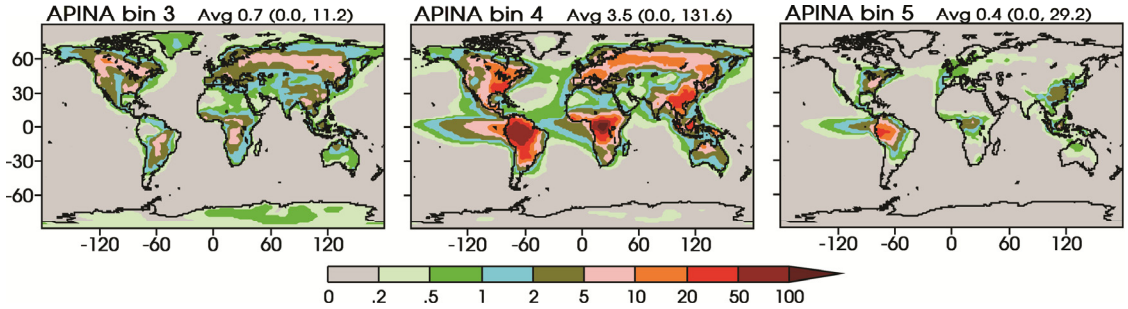
Aerosol Processes Including Their Interactions with Clouds

Cathy Chuang/ Lawrence Livermore National Laboratory

Science Question

- How does SOA formation affect the regional and global aerosol characteristics?
- How significant is aerosol microphysics in the effects of aerosols on cloud and precipitation?

Preliminary CAM5 simulation of surface SOA (in 10^{-2} mg/kg) from a-pinene for size bin 3 (0.10–0.22 mm), bin 4 (0.22–0.46 mm), and bin 5 (0.46–1.0 mm).



Approach

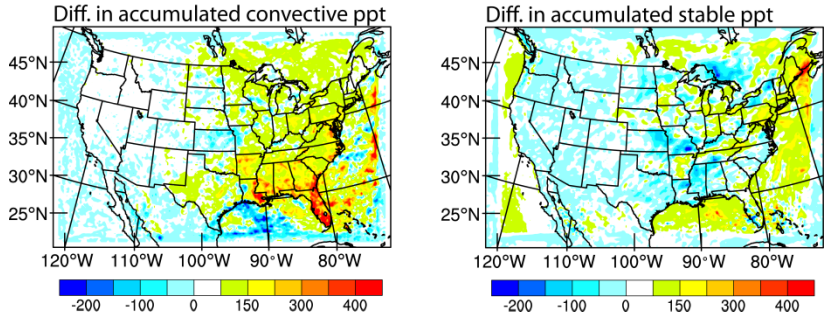
●Implement a sectional aerosol package (MADRID) into CAM5 and integrate with the MOZART gas chemistry.

●Add 10 reactions for SOA chemistry.

●Modify the land model to enable 15 interactive biogenic emissions of volatile organic carbon compounds.

●Explore aerosol/cloud interactions in regional scale with WRF.

Differences in yearly accumulated convective and stable precipitation (mm) between WRF simulations with prognostic and prescribed N_{drop}.



Key Accomplishment

The first attempt to introduce a size-resolved capability with the detailed treatment of SOAs in CAM.