FINAL REPORT

Cloud Condensation Nuclei measurements during the 2010 CalNex campaign: observations, analysis and impacts.

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1. Objectives of this proposal

While a major regulatory goal is the reduction of particulate matter and its precursors (e.g., SO_2, NO_x, VOCs, NH_4) to improve air quality, this also reduces the climatic cooling associated with these particles scattering sunlight back into space. Measurements of Cloud Condensation Nuclei (CCN) are essential in quantifying the extent to which PM regulations would influence clouds and climate on the local to regional scale. In May through June of 2010, the NOAA WP-3D and CIRPAS Twin Otter aircrafts were deployed in southern California as part of CalNex with comprehensive aerosol, gas-phase and cloud instrumentation to characterize atmospheric trace constituents. The Georgia Tech team deployed a cloud condensation nuclei (CCN) spectrometer aboard each platform for quantifying the aerosol-cloud link.

Using residual funds left from the proposal (and through a no-cost extension) we carried out measurements of cloud condensation nuclei (CCN) data collected aboard the NOAA WP-3D and a ground site operated at Centerville, AL during the June-July 2013 Southeast Atmosphere Study (SENEX 2013), to address the following scientific questions:

1. What are the hygroscopicity, CCN activity and activation kinetics of aerosols in the Southeast United States (SE US)? How do these parameters relate to aerosol chemical composition and size distribution? What is the spatiotemporal evolution of these parameters?
2. How does the aerosol mixing state in the SE US change over time and distance from their sources? How does this evolution affect CCN activity, hygroscopicity, and droplet activation kinetics of aerosol?
3. What are the similarities and differences of SE US aerosol against other locations of the globe (urban, rural, continental, remote, anthropogenic, biogenic, biomass burning)? How does the mixing of anthropogenic and biogenic aerosol precursors affect observed hygroscopicity and activation kinetics?
4. What is the indirect forcing associated with SE US aerosol? How complex should be the size distribution-composition-CCN-activity relationship, to adequately represent the aerosol-CCN link?
5. What are the impacts of point sources (such as power plants) on the aerosol and CCN number as a function of distance from the source?

2. Work done between Last and Final Reports.

Since the last (Year 4) report was submitted, a number of tasks in progress have been completed (and detailed in section 3, Milestone 1), while others are ongoing. The latter will be completed over the next few months, and will acknowledge support from this grant; the expected publications from work in progress beyond this report are also listed in Section 5, under manuscripts in preparation.

3. Summary of primary results and milestones for the whole proposal.

Milestone 1: Results from SOAS.

During the SOAS campaign, at the Centerville, AL site, a suite of instruments including a CCN counter, a thermodenuder (TD) and a high resolution time-of-flight aerosol mass spectrometer (AMS) were used to measure CCN activity, aerosol volatility, composition and oxidation state (Figure 1). Particles were either sampled directly from ambient or through a Particle Into Liquid Sampler (PILS) allowing the investigation of the water-soluble aerosol component.

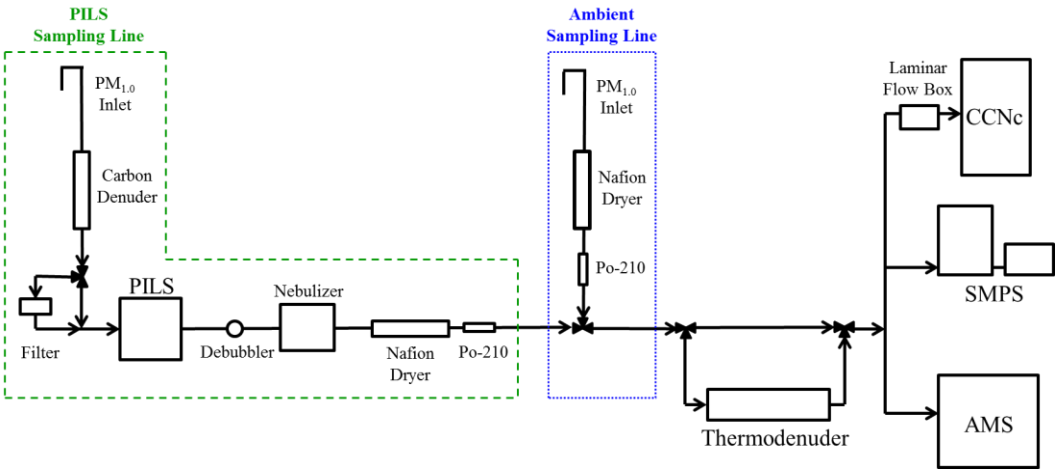

Figure 1. Instrument setup used during the SOAS Centerville, AL site, combining a PILS, thermodenuder, CCNc, AMS, and SMPS to measure the water-soluble fraction of ambient aerosol (green dashed line) and ambient aerosol (blue dotted line).

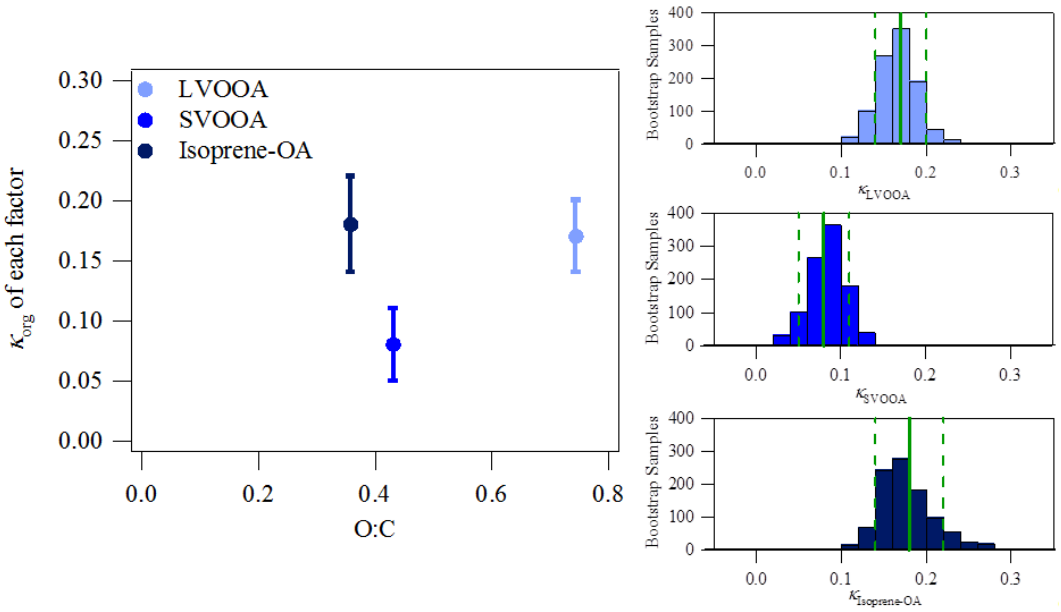

Figure 2. κ_{org} found for each organic PMF factor through linear regression versus O:C for non-denuded PILS aerosol at s=0.40% (left panel), where error bars represent the standard deviation. Examples of binned κ_{org} solutions from bootstrap analysis results of the linear regression are shown on the right, where the solid and dashed green lines represent the average and one standard deviation in each factor κ_{org}.

Analysis of the data sampled showed that ambient aerosol exhibited size-dependent composition with larger particles being more hygroscopic. The hygroscopicity of thermally denuded aerosol was similar between ambient and PILS-generated aerosol and showed limited dependence on volatilization (not shown). PMF analysis for the aerosol showed that the most hygroscopic components are both the most and the least volatile features of the aerosol, with the least volatile component appearing to be the most oxidized, as expected (Figure 2). No clear relationship, though, was found between organic hygroscopicity and organic to carbon ratio, most probably because of the small range of observed O:C during this study (Figure 3). This work is currently under review in Cerully et al., (2014)

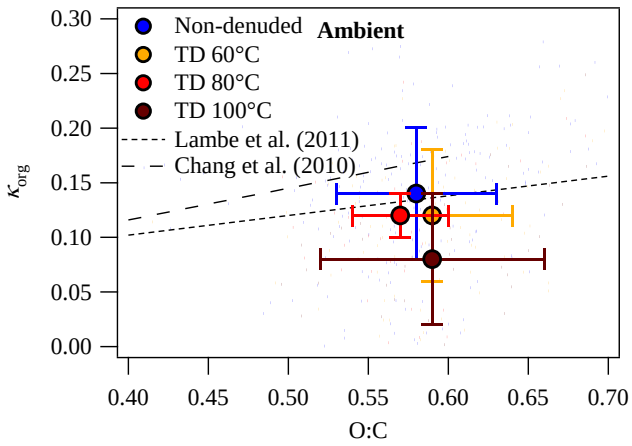

Figure 2. Variation in κ_{org} with O:C for ambient aerosol at s=0.20% and PILS aerosol at s=0.40% for data collected at the Centerville SOAS site. Small colored dots indicate all measured points while larger circles and squares indicate the averages for while errors bars indicate one standard deviation in measured values for ambient and PILS aerosol, respectively. Also shown are dashed lines indicating the parameterizations of κ_{org} with O:C from published studies.

Milestone 2: Improvements to Scanning Flow CCN Analysis.

We have developed a new numerical model for the non-steady-state operation of the Droplet Measurement Technologies (DMT) Cloud Condensation Nuclei (CCN) counter. The model simulates the Scanning Flow CCN Analysis (SFCA) instrument mode (Moore and Nenes, 2009), where a wide supersaturation range is continuously scanned by cycling the flow rate over 20-120 seconds. Model accuracy is verified using a broad set of data which include ammonium sulfate calibration data (under conditions of low CCN concentration) and airborne measurements where either the instrument pressure was not controlled or where exceptionally high CCN loadings were observed. It is shown here for the first time that small pressure and flow fluctuations can have a disproportionately large effect on the instrument supersaturation due to localized compressive/expansive heating and cooling. The model shows that, for fast scan times, these effects can explain the observed shape of the SFCA supersaturation-flow calibration curve and transients in the outlet droplet sizes. The extent of supersaturation depletion from the presence of CCN during SFCA operation is also examined; we found that depletion effects can be neglected below 4000 cm^{-3} for CCN number.

In addition to the numerical model improvements, we have considerably improved the flow control hardware during the SFCA operation of the CCN instrument. For this, we have developed a new control system, that uses an Arduino microprocessor together with an MKS mass flow controller; the whole system aircraft-grade and mounted upon the DMT CCN instrument as an independent hardware "module" that runs in parallel with the other hardware controlling the instrument. The module is attached in a way that does not disconnect or deactivate any existing hardware, so that switching into the conventional mode of operation, if needed, is a matter of turning a valve. The new module however controls the flow considerably better than before, and, with substantial improvement in instrument performance and data quality. In addition, the Arduino and mass flow controller are responsive enough to allow scan flow profiles that are sinusoidal (instead of linear), which reduce the pressure gradients and supersaturation swings during instrument operation. The new flow profile also allow considerable data quality even during flow downscans, which historically have been discarded from analysis (Moore and Nenes, 2009).

Milestone 3: Developing a comprehensive approach for studying droplet activation kinetics from measurements of CCN activity.

This study presents an approach to study droplet activation kinetics from measurements of CCN activity by the Continuous Flow Streamwise Thermal Gradient CCN Chamber (CFSTGC) and a comprehensive model of the instrument and droplet growth. The model, which can be downloaded from http://nenes.eas.gatech.edu/Experiments/CFSTGC.html, is evaluated against a series of experiments with ammonium sulfate calibration aerosol. Observed and modeled droplet sizes are in excellent agreement for a water vapor uptake coefficient ~0.2, which is consistent with theoretical expectations. The model calculations can be considerably accelerated without significant loss of accuracy by assuming simplified instrument geometry and constant parabolic flow velocity profiles. With these assumptions, the model can be applied to large experimental data sets to infer kinetic growth parameters while fully accounting for water vapor depletion effects and changes in instrument operation parameters such as the column temperature, flow rates, sheath and sample flow relative humidities, and pressure. When the effects of instrument operation parameters, water vapor depletion and equilibrium dry particle properties on droplet size are accounted for, the remaining variations in droplet size are most likely due to non-equilibrium processes such as those caused by organic surface films, slow solute dissociation and glassy or highly viscous particle states. As an example of model application, data collected during a research flight in the ARCTAS 2008 campaign are analyzed. The model shows that water vapor depletion effects can explain changes in the observed average droplet size. This work was published in Raatikainen et al., 2012 and is the basis for all subsequent studies studying droplet activation kinetics, including the Moore et al. (2012) study on aerosol sampled over the Deep Water Horizon spill, and, the analysis of global aerosol data published in the *Proceedings of the National Academy of Sciences* (Raatikainen et al., 2013).

Milestone 4: Analysis of CCN activity of California Aerosol

This study presents an overview and analysis of cloud condensation nuclei (CCN) sampled in California by the WP-3D aircraft during CalNex. Four distinct geographical regions are characterized, including the Los Angeles basin, the San Joaquin and Sacramento Valleys, and the eastern Pacific Ocean west of southern California. Median size distributions in the Central Valley were unimodal (Dg ~ 25 nm) with a larger fraction of organic species and smaller fraction of nitrate species in the Sacramento Valley aerosol than in the San Joaquin Valley aerosol. Size distributions in the Los Angeles basin and marine outflow were bimodal (geometric mean diameter, Dg= 30, 90-100 nm) with similar organic fractions and some replacement of nitrate with sulfate in the marine outflow. Both fine particle and CCN concentrations were found to decrease rapidly above the planetary boundary layer (~2 km altitude), with CCN concentrations in the boundary layer ranging from 10^2 –10^4 cm^{-3} STP, while fine particle concentrations (0.004–1μm diameters) ranged from 10^3 – 10^5 cm^{-3} STP. The CCN-active number fraction varied between 0 – 100% in the Los Angeles Basin and Marine Outflow, but was substantially lower (0 – 40%) in the San Joaquin and Sacramento Valleys. Values of the hygroscopicity parameter, κ, inferred from the CCN measurements varied from 0.1 – 0.25, with the highest values in the marine outflow and the lowest values in the Sacramento Valley. The κ values agreed well with the predictions based on size-resolved aerosol composition, but were overpredicted by almost twofold when size-averaged composition was used. CCN closure was assessed for simplified compositional and mixing state assumptions, and it was found that assuming the aerosol to be internally mixed overpredicted CCN concentrations by 30 – 75% for all air mass types except within the Sacramento Valley, where good closure (overprediction < 10%) was achieved by assuming insoluble organics. Assuming an externally-mixed aerosol fraction or incorporating size-resolved composition data improved closure in the other three regions, consistent with the bimodal nature of the aerosol size distribution.

A study focusing in the Los Angeles basin was also carried out using data collected with support from this proposal and onboard the CIRPAS Twin Otter during CalNex (Hersey et al., 2013). Aerosol composition was found to evolve from source-rich areas in the western Basin to downwind sites in the eastern Basin, evidenced by transition from an external to internal mixture, as well as enhancements in organic O:C ratio, the amount of organics and nitrate internally mixed on almost all particle types, and coating thickness on refractory black carbon (rBC). Transport into hot, dilute out flow regions leads to signifycant volatilization of semivolatile material, resulting in a unimodal aerosol comprising primarily oxygenated, low-volatility, water-soluble organics and sulfate. The fraction of particles with rBC or soot cores is between 27 and 51% based on data from a Single Particle Soot Photometer (SP2) and Aerosol Time of Flight Mass Spectrometer (ATOFMS). Secondary organics appear to inhibit subsaturated water uptake in aged particles, while CCN activity is enhanced with photochemical age. A biomass-burning event resulted in suppression of subsaturated hygroscopicity but enhancement in CCN activity, suggesting that BB particles may be nonhygroscopic at subsaturated RH but are important sources of CCN. Aerosol aging and biomass burning can lead to discrepancies between subsaturated and supersaturated hygroscopicity that may be related to mixing state. In the cases of biomass burning aerosol and aged particles coated with secondary material, more than a single parameter representation of subsaturated hygroscopicity and CCN activity is needed.

Milestone 5: Analysis of CCN activity and droplet activation kinetics of aerosol formed in the vicinity of the Deep Water Horizon Oil Spill

Secondary organic aerosol (SOA) resulting from the oxidation of organic species emitted by the Deepwater Horizon oil spill were sampled during two survey flights conducted by the WP-3D aircraft in June 2010. A new technique for fast measurements of cloud condensation nuclei (CCN) supersaturation spectra called Scanning Flow CCN Analysis was deployed for the first time on an airborne platform. Retrieved CCN spectra show that most particles act as CCN above (0.3±0.05)% supersaturation, which increased to (0.4± 0.1)% supersaturation for the most organic-rich aerosol sampled. The aerosol hygroscopicity parameter, κ, was inferred from both measurements of CCN activity and from humidified-particle light extinction, and varied from 0.05 to 0.10 within the emissions plumes. However, κ values were lower than expected from chemical composition measurements, indicating a degree of external mixing or size-dependent chemistry, which was reconciled assuming bimodal, size-dependent composition. The CCN droplet effective water uptake coefficient ,γ_{cond}, was inferred from the data using the comprehensive instrument model described in Milestone 3, and no significant delay in droplet activation kinetics from the presence of organics was observed, despite a large fraction of hydrocarbon-like SOA present in the aerosol. This work was published in Moore et al., (2012)

Milestone 6: Worldwide data sets constrain the water vapor uptake coefficient in cloud formation

This study is a synthesis of globally-relevant CCN data from ten field campaigns, including the NOAA-funded CalNex (2010), DeepWater Horizon (2010), ARCPAC (2008), GoMACCs (2006) and ITCT2k4 (2004) to study the variability of the water uptake coefficient, α_c, a parameter which has a profound impact on the water uptake kinetics and droplet number. Estimates of α_c for droplet growth from activation of ambient particles vary considerably and represent a critical source of uncertainty in estimates of global cloud droplet distributions and the aerosol indirect forcing of climate. We find that rapid activation kinetics ($\alpha_c > 0.1$) is uniformly prevalent. Important implications arise from this result. First, α_c is effectively constant for all the data considered, even for particles composed largely of organics with very low oxygen content. Anthropogenic (compositional) impacts on α_c are therefore limited to the 0.1–1.0 range, considerably reducing the uncertainty for cloud droplet number prediction in climate models.

This study resolves a decades-long uncertainty in cloud physics on the value of α_c, because it appears that α_c for ambient aerosol can be represented in models with a constant value in the 0.1–1.0 range. This study was published in the Proceedings of the National Academy of Sciences (Raatikainen et al., *PNAS*, 2013).

Milestone 7: Usage of the CalNex data in global model frameworks to better understand aerosol-cloud interactions

The data collected in this project will be used by the community for years to come in global model studies. We have already used it the study of Moore et al., (2013); where a global aerosol model (the NASA Global Modelling Initiative chemical transport model) coupled with a cloud droplet parameterization adjoint was used to quantify the sensitivity of cloud droplet number concentration to uncertainties in predicting CCN concentrations. Published CCN closure uncertainties for six different sets of simplifying compositional and mixing state assumptions are used as proxies for modelled CCN uncertainty arising from application of those scenarios. We found that cloud droplet number concentrations (N_d) are fairly insensitive to the number concentration (Na) of aerosol which act as CCN over the continents ($\partial \ln Nd/\partial \ln Na$ ~10–30%), but the sensitivities exceed 70% in pristine regions such as the Alaskan Arctic and remote oceans. This means that CCN concentration uncertainties of 4–71% translate into only 1–23% uncertainty in cloud droplet number, on average. The study does highlight the sensitivity of some remote areas to pollution brought into the region via long-range transport (e.g., biomass burning) or from seasonal biogenic sources (e.g., phytoplankton as a source of dimethylsulfide in the southern oceans).

4. Personnel supported by this grant

Jack Lin, PhD student (current), Earth and Atmospheric Sciences
Richard Moore, PhD student (graduated), Chemical and Biomolecular Engineering
Kate Cerully, PhD student (graduated), Chemical and Biomolecular Engineering
Dr. Aikaterini Bougiatioti, Postodoctoral scholar, Earth and Atmospheric Sciences (travel support only)

5. Manuscripts acknowledging support from this grant

Published/In press

Lathem, T.L and Nenes, A. (2011) Water vapor depletion in the DMT Continuous Flow CCN Chamber: effects on supersaturation and droplet growth, *Aeros.Sci.Tech.*, 45, 604–615, doi:10.1080/02786826.2010.551146

Lack, D.A., Cappa, C.D., Langridge, J., Bahreni, R., Buffaloe, G., Brock, C., Cerully, K., Coffman, D., Fahey, D.W., Hayden, K., Holloway, J., Lerner, B., Massoli, P., Li, S-M., McLaren, R., Middlebrook, A., Moore, R., Nenes, A., Nuaanan, I., Onasch, T., Peischl, J., Perring, A., Quinn, P., Ryerson, T., Schwartz, J.P., Spackman, R., Wofsy, S.C., Worsnop, D., Xiang, B. and E. Williams (2011) Observed Changes in Climate and Air Quality – Relevant Shipping Emissions Due to Vessel Fuel Quality and Speed Regulation, *Env.Sci.Tech.*, doi: 10.1021/es2013424*s*

Moore, R.H., Raatikainen, T., Langridge, J.M., Bahreini, R., Brock, C.A., Holloway, J.S., Lack, D.A., Middlebrook, A.M., Perring, A.E., Schwarz, J.P., Spackman J.R., and Nenes, A. (2012) CCN Spectra, Hygroscopicity, and Droplet Activation Kinetics of Secondary Organic Aerosol Resulting from the 2010 Deepwater Horizon Oil Spill, *Env.Sci.Tech.*, doi: 10.1021/es203362w

Moore, R.H., Cerully, K., Bahreini, R., Brock, C.A., Middlebrook, A.M., and Nenes, A. (2012) Hygroscopicity and Composition of California CCN During Summer 2010, *J.Geoph.Res.*, 117, D00V12, doi:10.1029/2011JD017352

Raatikainen, T., Moore, R. H., Lathem, T. L. and A. Nenes (2012) A coupled observation–modeling approach for studying activation kinetics from measurements of CCN activity, *Atmos.Chem.Phys.*, **12**, 4227-4243

Karydis, V.A., Capps, S.L., Moore, R.H., Russell, A., Henze, D.K. and A. Nenes (2012) Using a global aerosol model adjoint to unravel the footprint of spatially-distributed emissions on cloud droplet number and cloud albedo, *Geoph.Res.Let.*, **39**, L24804, doi:10.1029/2012GL053346

Raatikainen,T., Nenes,A., Seinfeld, J. H., Morales,R., Moore, R. H., Lathem, T. L., Lance, S., Padro, L. T., Lin, J. J., Cerully, K., Bougiatioti, A., Cozic, J., Ruehl, C., Chuang, P. Y., Anderson, B., Flagan, R.C., Jonsson, H., Mihalopoulos, N., and J. N. Smith (2013) Worldwide data sets constrain the water vapor uptake coefficient in cloud formation, *Proc.Nat.Acad.Sci.*, doi: 10.1073/pnas.1219591110

Ryerson, T.B., Andrews, A.E., Angevine, W.M., Bates, T.S., Brock, C.A., Cairns, B., Cohen, R.C., Cooper, O.R., de Gouw, J.A., Fehsenfeld, F.C., Ferrare, R.A., Fischer, M.L., Flagan, R.C., Goldstein, A.H., Hair, J.W., Hardesty, R.M., Hostetler, C.A., Jimenez, J.L., Langford, A.O., McCauley, E., McKeen, S.A., Molina, L.T., Nenes, A., Oltmans, S.J., Parrish, D.D., Pederson, J.R., Pierce, R.B., Prather, K., Quinn, P.K., Seinfeld, J.H., Senff, C.J., Sorooshian, A., Stutz, J., Surratt, J.D., Trainer, M., Volkamer, R., Williams, E.J., and Wofsy, S.C. (2013) The 2010 California Research at the Nexus of Air Quality and Climate Change (CalNex) field study, J.Geoph.Res., 118, doi:10.1002/jgrd.50331

Moore, R.H., Karydis, V.L., Capps, S.L., Lathem, T.L. and Nenes, A. (2013) Droplet Number Prediction Uncertainties From CCN: An Integrated Assessment Using Observations and a Global Model Adjoint, Atmos.Chem.Phys., 13, 4235–4251

Cerully, K., Hite, J., McLaughlin, M., and Nenes., A. (2014) Toward the Determination of Joint Volatility-Hygroscopicity Distributions: Development and Response Characterization for Single-Component Aerosol, Aer.Sci.Tech., 48, doi:10.1080/02786826.2013.870326

Raatikainen, T., Lin, J.J, Cerully, K., Lathem, T.L., Moore, R.H. and Nenes, A. (2014) CCN data interpretation under dynamic operation conditions, Aer.Sci.Tech., 48, doi:10.1080/02786826.2014.899429

In review

Lathem, T. L., Nenes, A., Deng, Y., Zhang, H., Anderson, B. E., Crumeyrolle, S., Beyersdorf, A. J., Ziemba, L. D., Thornhill, K. L., Winstead, E. L., Black, R. A., Murillo, S. T., Blowing salt into the eye infuriates hurricanes, *Proc.Nat.Acad.Sci.*

Cerully, K., Bougiatioti, A., Guo, H., Xu, L., Hite, J.R., Ng, N.L., Weber, R., Nenes, A., On The Link Between Hygroscopicity, Volatility, And Oxidation State Of Ambient and Water-Soluble Aerosol In The Southeastern United States, *Atmos.Chem.Phys. Disc.*

In preparation

Xu , L., Middlebrook , A.N., Liao ,J., Welti, A., Guo, H., Lin, J.J., Bougiatioti, A., Weber, R.J., Nenes, A., Holloway, J.S., Gilman, J. , Lerner, B.M., Graus, M. , Warneke, C., Trainer, M., De Gouw, J., and Ng, N.L., Aircraft Measurement of Isoprene-derived Organic Aerosol during the Southeast Nexus (SENEX) Campaign Using an Aerosol Mass Spectrometer

Lin, J.J., Bougiatioti, A., Nenes, A., Anderson, B.E., Beyersdorf, A.J., Brock, C.A. , Gordon, T.D. , Lack, D. , Law, D.C., Liao, J., Middlebrook, A.M. , Richardson, M. , Thornhill, K.L. , Winstead, E., Wagner, N.L., Welti, A., and Ziemba, L.D.,Vertical profiles of cloud condensation nuclei, aerosol hygroscopicity, water uptake, and scattering across the United States

References

Cerully, K., Bougiatioti, A., Guo, H., Xu, L., Hite, J.R., Ng, N.L., Weber, R., Nenes, A. (2014) On The Link Between Hygroscopicity, Volatility, And Oxidation State Of Ambient and Water-Soluble Aerosol In The Southeastern United States, *Atmos.Chem.Phys. Disc.*

Hersey, S., Craven, J., Metcalf, A., Lin, J., Lathem, T., Suski, K., Cahill, J., Duong, H., Sorooshian, A., Jonsson, H., Nenes, A., Prather, K., Flagan, R., Seinfeld, J. (2013) Composition and Hygroscopicity of the Los Angeles Aerosol: CalNex, *J.Geoph.Res.*, **117**, doi:10.1002/jgrd.50307

Moore, R.H. and Nenes, A. (2009) Scanning Flow CCN Analysis - A Method for Fast Measurements of CCN Spectra, Aer.Sci.Tech., 43, 1192-1207

Moore, R.H., Cerully, K., Bahreini, R., Brock, C.A., Middlebrook, A.M., and Nenes, A. (2012) Hygroscopicity and Composition of California CCN During Summer 2010, *J.Geoph.Res.*, **117**, D00V12, doi:10.1029/2011JD017352

Moore, R.H., Raatikainen, T., Langridge, J.M., Bahreini, R., Brock, C.A., Holloway, J.S., Lack, D.A., Middlebrook, A.M., Perring, A.E., Schwarz, J.P., Spackman J.R., and Nenes, A. (2012) CCN Spectra, Hygroscopicity, and Droplet Activation Kinetics of Secondary Organic Aerosol Resulting from the 2010 Deepwater Horizon Oil Spill, *Env.Sci.Tech.*, doi: 10.1021/es203362w

Moore, R.H., Karydis, V.L., Capps, S.L., Lathem, T.L. and Nenes, A. (2013) Droplet Number Prediction Uncertainties From CCN: An Integrated Assessment Using Observations and a Global Model Adjoint, *Atmos.Chem.Phys.*, **13**, 4235–4251

Raatikainen, T., Lin, J.J, Cerully, K., Lathem, T.L., Moore, R.H. and Nenes, A. (2014) CCN data interpretation under dynamic operation conditions, Aer.Sci.Tech., 48, doi:10.1080/02786826.2014.899429

Raatikainen,T., Nenes,A., Seinfeld, J. H., Morales,R., Moore, R. H., Lathem, T. L., Lance, S., Padro, L. T., Lin, J. J., Cerully, K., Bougiatioti, A., Cozic, J., Ruehl, C., Chuang, P. Y., Anderson, B., Flagan, R.C., Jonsson, H., Mihalopoulos, N., and J. N. Smith (2013) Worldwide data sets constrain the water vapor uptake coefficient in cloud formation, Proc.Nat.Acad.Sci., doi: 10.1073/pnas.1219591110