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Cloud Condensation Nuclei Activity of Aerosols during GoAmazon 2014/15 Field Campaign Report

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Executive Summary

Aerosol indirect effects, which represent the impact of aerosols on climate through influencing the properties of clouds, remain one of the main uncertainties in climate predictions (Stocker et al. 2013). Reducing this large uncertainty requires both improved understanding and representation of aerosol properties and processes in climate models, including the cloud activation properties of aerosols. The Atmospheric System Research (ASR) science program plan of January 2010 states that: "A key requirement for simulating aerosol-cloud interactions is the ability to calculate *cloud condensation nuclei and ice nuclei* (CCN and IN, respectively) concentrations as a function of supersaturation from the chemical and microphysical properties of the aerosol." The Observations and Modeling of the Green Ocean Amazon (GoAmazon 2014/15) study seeks to understand how aerosol and cloud life cycles are influenced by pollutant outflow from a tropical megacity (Manaus)—in particular, the differences in cloud-aerosol-precipitation interactions between polluted and pristine conditions. One key question of GoAmazon2014/5 is: "What is the influence of the Manaus pollution plume on the cloud condensation nuclei (CCN) activities of the aerosol particles and the secondary organic material in the particles?" To address this question, we measured size-resolved CCN spectra, a critical measurement for GoAmazon2014/5.

The pollution plume from Manaus meanders north and south on two- to three-day cycles so that the main research site were in and out of the Manaus plume and pristine conditions every few days. The contrasts between pristine air and the pollution plume provided excellent opportunities to look into how and to what extent different aerosol size and compositions impact the CCN activity of aerosol. The CCN spectra and activation fraction of size selected particles allow a clear separation of the impact of aerosol composition from size. The diurnal and seasonal variations of CCN activity, and the influences from both aerosol size and composition, were examined under a variety of conditions ranging from pristine to heavily polluted. The influence of aerosol composition on CCN activity is manifested in particle hygroscopicity, which was derived from measured size-resolved CCN spectra. When combined with aerosol composition measurements, the derived particle hygroscopicity allowed us to characterize the average value and range of hygroscopicity for major aerosol components, including secondary organic aerosols (SOA). The long-term deployment (one-year) and the opportunities to sample both pristine air and Manaus plumes provided a sufficient data set for examining the hygroscopicities of both primary organic aerosol (POA) and SOA from the urban region, as well as SOA formed by natural biogenic precursors (including modification by the anthropogenic influence). These data sets and results can be incorporated into advanced process models for improved representation of CCN concentrations and cloud droplet formation to address the influence of tropical megacities on climate.

Acronyms and Abbreviations

ACSM	Aerosol Chemical Speciation Monitor
AMS	Aerosol Mass Spectrometer
ARM	Atmospheric Radiation Measurement Climate Research Facility
ASR	Atmospheric System Research
BBOA	biomass burning organic aerosol
BVOC	biogenic volatile organic carbon
CCN	cloud condensation nuclei
CCNc	CCN counter
CPC	Condensation Particle Counter
DMA	Differential Mobility Analyzer
Dp	particle diameter, equivalent to mobility diameter for spherical particles
f44	fraction of organic mass measured by the AMS found at m/z = 44
IN	ice nuclei
IPCC	Intergovernmental Panel on Climate Change
LBA	Large-Scale Biosphere-Atmosphere Experiment in Amazonia
MAOS	Mobile Aerosol Observing System
MPS	Scanning Mobility Particle Sizer
O:C	oxygen-to-carbon elemental ratio measured by the AMS
PMF	Positive Matrix Factorization
POA	primary organic aerosol
P-ToF	Particle Time of Flight
S NOx	nitrogen oxides
SOA	secondary organic aerosol
SP2	Single Particle Soot Photometer
VOC	volatile organic carbon
κ / κ_{CCN}	hygroscopicity parameter/hygroscopicity parameter measure by supersaturation
κ_{org}	hygroscopicity parameter of the organic fraction of an aerosol

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1.0 Background

Observations and Modeling of the Green Ocean Amazon (GoAmazon2014/5), a field campaign by the Atmospheric Radiation Measurement (ARM) Climate Research Facility, a U.S. Department of Energy Office of Science user facility sponsored by the Office of Biological and Environmental Research, focused on the influences of pollutant outflow from a tropical megacity on aerosol and cloud life cycles—in particular, the differences in cloud-aerosol-precipitation interactions between polluted and pristine conditions. One key question of GoAmazon 2014 is: "What is the Influence of the Manaus pollution plume on the cloud condensation nuclei (CCN) activities of the aerosol particles and the secondary organic material in the particles?" To answer this question, we measured size-resolved CCN spectra, a critical measurement for GoAmazon2014/5. A system consisting of a scanning mobility particle sizer and a cloud condensation nuclei counter operated in series was deployed at the main research site (T3 site, ARM site MAO, 3°12'47.82"S, 60°35'55.32"W) near Manacapuru during the GoAmazon2014/5 campaign (February 2014 to March 2015). Aerosol particles ranging from 50 to 250 nm in diameter, a size range over which aerosol CCN activity is strongly influenced by chemical composition, were size selected and the CCN spectra of the size-selected particles were characterized under a number of atmospherically representative supersaturations. The measurement was conducted continuously, and results are now available on the ARM Data Archive.

The measurement of CCN spectra and activation fraction of size selected particles allows a clear separation of the impact of aerosol composition from size. Combined with trace gas and other aerosol measurements co-located at the T3 site, the size-solved CCN spectra provided key information to address the following scientific objectives:

1. Variability (diurnal and seasonal) and controlling processes of aerosol CCN activity under a variety of conditions ranging from pristine to heavily polluted.
2. The hygroscopicity of organic components, and its variation as aerosols age (i.e., the increase in organics oxidation level and atomic O:C ratio).
3. Average value and range of hygroscopicity for major aerosol organic components, such as primary organics aerosol (POA) and secondary organic aerosol (SOA) due to urban emissions, and SOA formed by natural biogenic volatile organic carbon (BVOCs) (including possible modification by the anthropogenic influence).

1.1 Collaborating Agencies

- U.S. Department of Energy
- Large Scale Biosphere Atmosphere Experiment in Amazonia (LBA) (Brazilian-funded program)
- Principal Investigator: Jian Wang, *DOE Brookhaven National Laboratory*
- Co-Principle Investigators: Scot Martin, *Harvard University*; Lawrence Kleinman, *DOE Brookhaven National Laboratory*

We acknowledge the support from the Central Office of the Large Scale Biosphere Atmosphere Experiment in Amazonia (LBA), the Instituto Nacional de Pesquisas da Amazonia (INPA), and the

Instituto Nacional de Pesquisas Espaciais (INPE). The work was conducted under 001262/2012-2 of the Brazilian National Council for Scientific and Technological Development (CNPq).

1.2 Collaborators/Team Members

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2.0 Notable Events or Highlights

- February 1, 2014 – Start data of Campaign, begin installation of instrumentation.
- March 12, 2014 – Start data of data acquisition. Though data acquisition had begun earlier (Feb 10) instrument leaks and flooding of the working fluid of the CPC delayed collection of quality data.
- March 24, 2014 – Co-located sampling with the Max Plank Institute size-resolved CCN instrument that was deployed at the ATTO site (T0a).
- March 31, 2014 – End of Intensive Operational Period 1.
- May 20 – 26, 2014 – Computer crash took instrument offline.
- June 27 – 30, 2014 – Program glitch caused data not to be saved.
- July 28 – 30, 2014 – Water in CCN ran dry and needed to be refilled.
- August 6 – 8, 2014 – USB malfunctioned while scientist was en route to the site.
- August 15, 2015 – Start of Intensive Operational Period 2.
- August 20 – 23, 2014 – Intensive biomass burning plume impacting site.

- October 5 – 15, 2014 – CCN operated behind oxidation flow reactor half of the time and ambient sampling the rest to investigate the changes in particle hygroscopicity upon further aging of ambient particles.
- October 15, 2014 – End of Intensive Operational Period 2.
- November 4, 2014 – Solenoid water supply pump in CCN replaced by technicians on site.
- March 5, 2015 – Co-located sampling and calibration with the University of Sao Paulo size-resolved CCN instrument that was located at the TIWA (T2) site.
- March 5, 2015 – End of instrument operation; calibration and instrument de-installation.

3.0 Lessons Learned

We discovered that the temperature fluctuation inside the IOP instrument container at the T3 site was too large (as much as 10°C change night to day) and that this significantly impacted the calibration and stability of the CCN. We corrected for this change by calibrating the instrument over a range of room temperatures (this variability was related to the change in the temperature at the top of the CCN column). This greatly increased the time needed for calibration (by a factor of 3).

4.0 Results

4.1 Seasonal Trend and Size Dependence of Hygroscopicity

The monthly average particle hygroscopicity (κ_{CCN}) at the T3 site ranged from 0.1 to 0.2 over all six particle sizes investigated (75, 94, 112, 142, 171, and 222 nm), and is substantially lower than 0.3 suggested for continental sites (Andreae and Rosenfeld 2008). With the transition from the wet season to the dry season, the κ_{CCN} decreases by 0.03 – 0.05 with the absolute minimum of 0.115 for all sizes occurring in September and October (Figure 1).The low κ_{CCN} during the dry season can be attributed to the combination of high particle organic volume fraction and low organic hygroscopicity (e.g., for 94 nm $\Delta\kappa_{CCN}$ = 0.038). The 75 nm aerosol exhibits the same seasonal trend as the other sizes but has the lowest κ_{CCN} over the course of the year.

κ_{CCN} increased with increasing size for all the seasons (Figure 2). Data for 222 nm particles were not considered for this comparison due to a systematic under-sampling of κ_{CCN} > 0.21 from limitations in the achievable instrument supersaturation, a similar but opposite result is seen for 51 nm data, where particles did not activate under the range of supersaturations investigated (> 50% of the data). Figure 2 also shows data from a previous study in the Amazon basin during the AMAZE-08 campaign by Gunthe et al. (2009). The size dependence during GoAmazon2014/5 generally agrees with the AMAZE-08 result for the κ_{CCN}, with the current data set having similar hygroscopicity for the background air relative to the AMAZE-08 data. A weaker trend in κ_{CCN} relative to the AMAZE-08 data might be expected because the aerosol investigated in this study spanned a much lower contribution of sulfate.

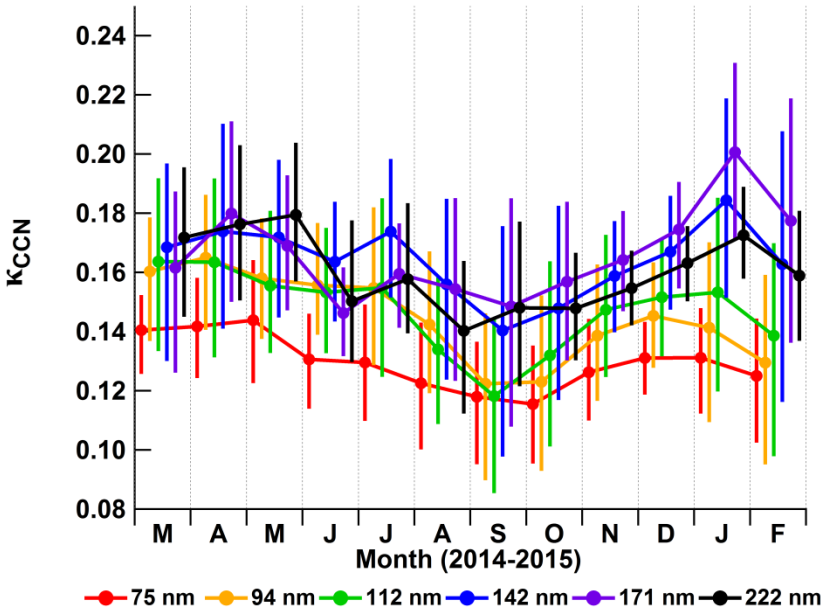

Figure 1. Annual variations of κ_{CCN}. Error bars here represent the 25th and 75th percentiles.

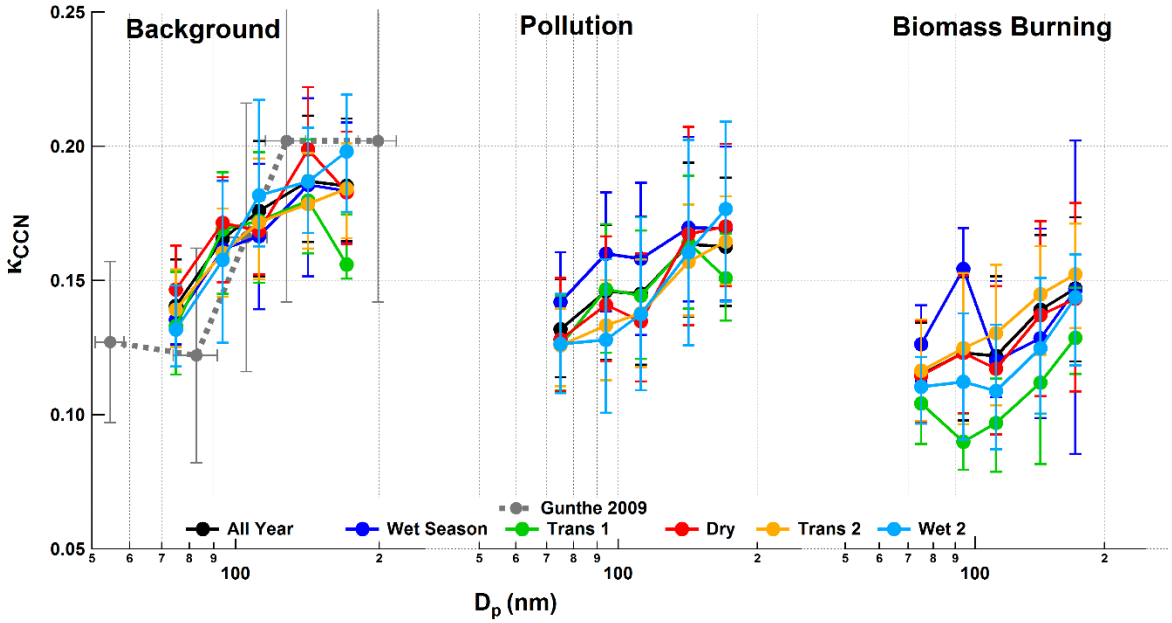

Figure 2. Dependence of κ_{CCN} on particle size compared to a previous study in the Amazon for all available data shown according to plume type and divided by season. The error bars represent the 25th and 75th percentiles.

4.2 Diel Trends of Particle Hygroscopicity

The results of the three largest sizes (D_p = 112, 142 and 171 nm) were binned together to increase the data available to create diel profiles (Figure 3). Figure 3 shows the diel variations for the wet season under background and polluted conditions. There were only a handful of cases classified as biomass burning in the wet season and not enough to evaluate diel trends. Background air appears to have a relatively high hygroscopicity (0.2) that displays no significant diel cycle, indicating nearly constant chemical composition. The lack of diel trend agrees with the observations at the T0a site by the Max Plank Institute using a separate SCCN instrument operated by stepping the size at a given CCNc super saturation (Rose et al. 2008). Figure 4 shows the diel trend at T0a during April and May and the month of August averaged over similar critical particle sizes as the T3 data (D_{pc} = 100 – 175 nm) compared to background aerosol at T3 over similar periods of time.

During the wet season the diel trend of the polluted air (Figure 3) shows an increase in κ_{CCN} towards solar noon (LT = UTC – 4 hours). The observed daily cycle of κ_{CCN} at T3 is influenced by the modification of the background by more local sources at night as well as daytime photo-oxidation.

Figure 5 shows the diel variability for background, polluted, and biomass burning conditions during the dry season. The background aerosol appears very similar to that of the wet season, with no diel cycle (Figure 3). The biomass burning air masses during the dry season (Figure 5) exhibit the same high daytime hygroscopicity but with slightly lower hygroscopicity at night compared to the polluted-type aerosol. The biomass burning classification during the dry season represents intense biomass burning beyond that already found in the background.

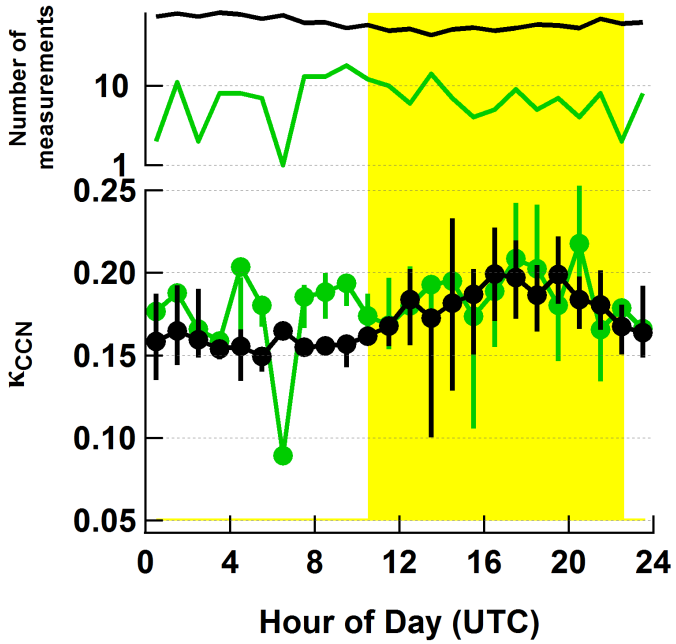

Figure 3. Diel trend in κ_{CCN} for the wet season (March-May) under background (green) and polluted (black) conditions. The top axis shows the number of points included in the average for each hour. The aerosol in polluted air masses approaches the same level of hygroscopicity as the background aerosol at local noon (yellow-shaded region represents the local daytime). The error bars represent the 25th and 75th percentiles of the averaged data.

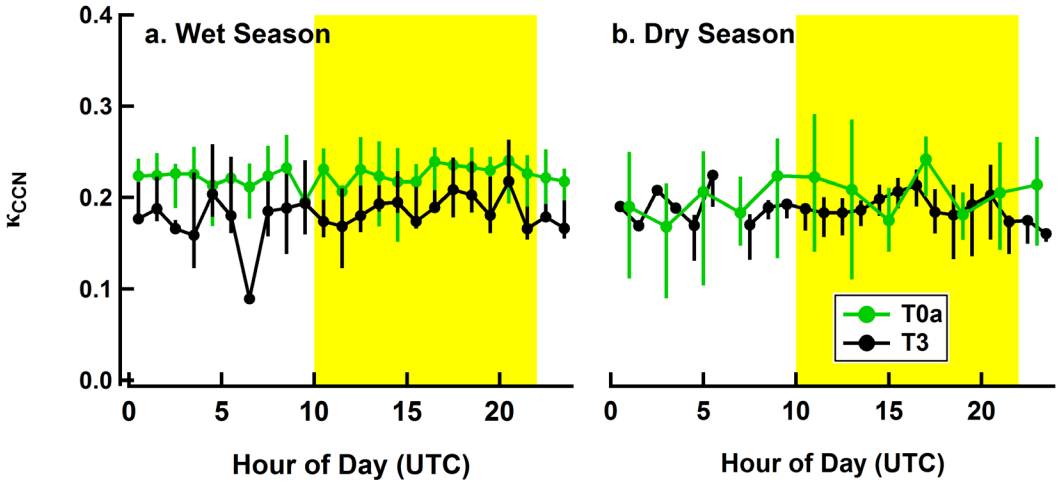

Figure 4. Comparison of measurements of κ_{CCN} at T0a and T3 during the months of April and May (panel a) and August (panel b) 2014 and T3 (background conditions) during the wet and dry season. The Error bars represent the 25th and 75th percentiles of the data.

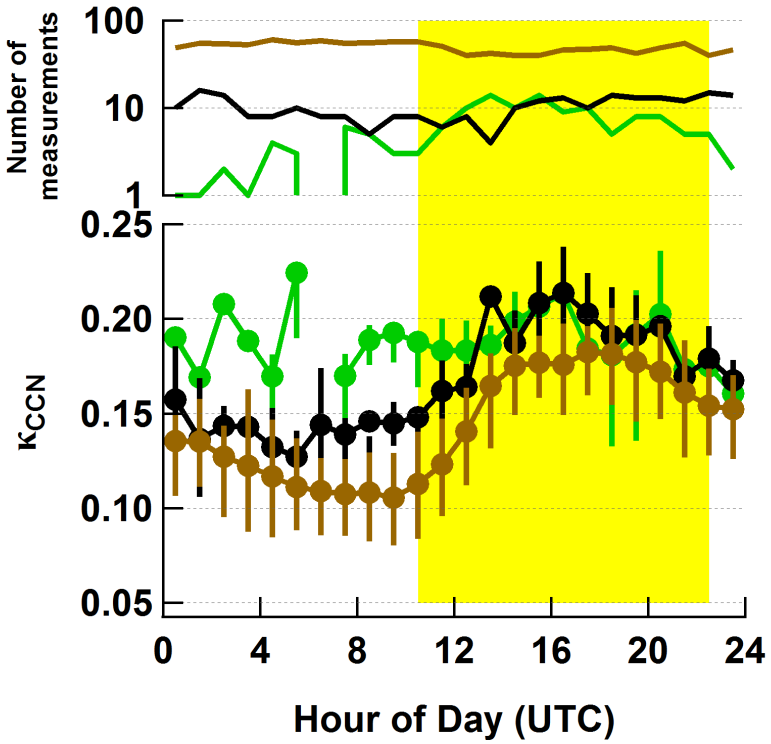

Figure 5. Dry season variation of the κ_{CCN} of the three air types: background (green), polluted (black), and biomass burning (brown). The number of data points in each hour bin of the diel average is shown on the top panel. The pollution and biomass burning aerosols approach the same level of hygroscopicity as the background aerosol at local noon (yellow-shaded region represents the local daytime). The error bars represent the 25th and 75th percentiles of the averaged data.

5.0 Public Outreach

GoAmazon 2014/15 web links:

- http://campaign.arm.gov/goamazon2014/ Campaign web page
- http://www.arm.gov/sites/amf/mao/ ARM mobile deployment web page

News Releases:

- February 19, 2014: Brookhaven National Laboratory news release: BNL's Jian Wang Will Help Understand Rainforest Atmosphere Dynamics, https://www.bnl.gov/newsroom/news.php?a=24654 (accessed 15 July 2015).

6.0 CCN Activity of Aerosols during GoAmazon 2014 Publications

6.1 Journal Articles/Manuscripts

In preparation: Thalman, R, S de Sa, ML Alexander, HMJ Barbosa, P Compuzano-Jost, DA Day, W Hu, JL Jimenez, M Krüger, C Kuang, A Manzi, BD Palm, U Pöschl, G Senum, A Sedlacek, S Springston, R Souza, T Watson, P Artaxo, S Martin, and J Wang. "Measurements of size-resolved sub-micron aerosol hygroscopicity in the Amazon River Basin at a downwind site during Green Ocean Amazon (GoAmazon) 2014/5." Planned for submission to *Atmospheric Chemistry and Physics*. (GoAmazon2014/5 Special Issue)

6.2 Meeting Abstracts/Presentations/Posters

2014 American Geophysical Union – Fall Meeting (Poster), San Francisco, CA December 2014

Measurements of size-resolved cloud condensation nuclei (CCN) spectra were performed at the T3 site of the Green Ocean Amazon (GoAmazon) field project located near Manacapuru, Brazil during 2014. The T3 site is a receptor site for both polluted urban downwind (Manaus is a city of several million 70 km upwind) and background (Amazon rainforest) air-masses and can provide a contrast between clean and polluted conditions. Particle hygroscopicity (kappa) and mixing state were calculated from the particle activation spectrum measured by size-selecting aerosols and exposing them to a wide range of supersaturation in the CCN counter (Droplet Measurement Technologies Continuous-Flow Streamwise Thermal Gradient CCN Chamber). The supersaturation was varied between 0.07 and 1.1% by changing a combination of both total flow rate and temperature gradient in the CCN counter. Measured spectra were examined for air masses with different levels of influence from the Manaus plume. Particle hygroscopicity generally peaked near noon local time, which was broadly consistent with the trend in aerosol sulfate. The average kappa values during the first intensive operational period were 0.14±0.05, 0.14±0.04 and 0.16±0.06 for 75, 112 and 171 nm particles respectively. Evaluation of particle hygroscopicity, dispersion (mixing state), and organic hygroscopicity will be presented with respect to size and level of pollution.

GoAmazon2014/5 Science Conference, Harvard University, Cambridge, MA, May 18-20 2015

During the Green Ocean Amazon (GoAmazon) 2014/15 campaign, measurements of size-resolved cloud condensation nuclei (CCN) spectra were made at the T3 site. These measurements ran from March 12, 2014 to March 3, 2015. Particle hygroscopicity (kappa) and mixing state were calculated from the particle activation spectrum measured by size-selecting aerosols and exposing them to a wide range of supersaturation in the CCN counter (Droplet Measurement Technologies Continuous-Flow Streamwise Thermal Gradient CCN Chamber). The supersaturation was varied between 0.07 and 1.1% by changing a combination of both total flow rate and temperature gradient in the CCN counter. During the second intensive operational period (IOP2), measurements were made in combination with the Oxidation Flow Reactor (OFR) system used by the Aerosol Mass Spectrometer (AMS). Here we evaluate the range of measured organic particle hygroscopicity with respect to indicators of oxidation as measured by the AMS (f44 and O:C) and the Aerosol Chemical Speciation Monitor (ACSM) found in the Department of Energy Atmospheric Radiation Measurement (ARM) suite. We observe overall diurnal trends, in particle hygroscopicity (κ_{CCN}), organic hygroscopicity (κ_{org}), f44 and particle hygroscopicity dispersion ($\sigma_\kappa/\kappa_{CCN}$) for all of the seasons and air mass sources.

AAAR Annual Meeting 2015

During the Green Ocean Amazon (GoAmazon) 2014/15 campaign, measurements of size-resolved cloud condensation nuclei (CCN) spectra were made at a measurement site (T3) 5 hours downwind of the city of Manaus (pop. 2 million). These measurements ran from March 12, 2014 to March 3, 2015. Particle hygroscopicity (κ) and mixing state were calculated from the particle activation spectrum measured by size-selecting aerosols and exposing them to a wide range of supersaturation in the CCN counter (Droplet Measurement Technologies Continuous-Flow Streamwise Thermal Gradient CCN Chamber). The supersaturation was varied between 0.07 and 1.1% by changing a combination of both total flow rate and temperature gradient in the CCN counter. During the second intensive operational period (IOP2), measurements were made in combination with the Oxidation Flow Reactor (OFR) system coupled to an Aerosol Mass Spectrometer (AMS). Here we evaluate the range of measured organic particle hygroscopicity with respect to indicators of oxidation as measured by the AMS (f_{44} and O:C) and the Aerosol Chemical Speciation Monitor (ACSM) found in the Department of Energy Atmospheric Radiation Measurements (ARM) suite. We observe overall diel trends, in particle hygroscopicity (κ_{CCN}), organic hygroscopicity (κ_{org}), f_{44} and particle dispersion ($\sigma_\kappa/\kappa_{CCN}$) for all of the seasons and air mass sources. We discuss the factors contributing to the changing particle hygroscopicity throughout the day with respect to air mass sources and relative to the regional background aerosol. The annual κ_{CCN} trend ($\kappa_{CCN} = 0.14 \pm 0.04$ (wet season), 0.12 ± 0.04 (dry season)) can mostly be explained by the changing aerosol composition and contributions of the various air mass sources at the T3 site, as the regional background changes during the transition from the wet to dry season from clean to heavily influenced by biomass burning. Background conditions are sampled in ~19% of the available data while polluted and biomass-burning conditions each make up 40% of the data.

7.0 References

Andreae, MO, and D Rosenfeld. 2008. “Aerosol–cloud–precipitation interactions. Part 1. The nature and sources of cloud-active aerosols.” *Earth-Science Reviews* 89: 13-41, doi: 10.1016/j.earscirev.2008.03.001.

Boyd, CM, J Sanchez, L Xu, AJ Eugene, T Nah, WY Tuet, MI Guzman, and NL Ng. 2015. "Secondary organic aerosol formation from the β-pinene+NO3 system: effect of humidity and peroxy radical fate." *Atmospheric Chemistry and Physics* 15: 7497-7522, doi:10.5194/acp-15-7497-2015.

Canagaratna, MR, JL Jimenez, JH Kroll, Q Chen, SH Kessler, P Massoli, L Hildebrandt Ruiz, E Fortner, LR Williams, KR Wilson, JD Surratt, NM Donahue, JT Jayne, and DR Worsnop. 2015. "Elemental ratio measurements of organic compounds using aerosol mass spectrometry: characterization, improved calibration, and implications." *Atmospheric Chemistry and Physic*s 15: 253-272, doi:10.5194/acp-15-253-2015.

Cerully, KM, A Bougiatioti, JR Hite Jr, H Guo, L Xu, NL Ng, R Weber, and A Nenes. 2015. "On the link between hygroscopicity, volatility, and oxidation state of ambient and water-soluble aerosol in the Southeastern United States." *Atmospheric Chemistry and Physic*s 15(15): 8679-8694, doi:10.5194/acp-15-8679-2015.

Chang, RYW, JG Slowik, NC Shantz, A Vlasenko, J Liggio, SJ Sjostedt, WR Leaitch, and JPD Abbatt. 2010. "The hygroscopicity parameter (κ) of ambient organic aerosol at a field site subject to biogenic and anthropogenic influences: relationship to degree of aerosol oxidation." *Atmospheric Chemistry and Physics* 10: 5047-5064, doi:10.5194/acp-10-5047-2010.

de Gouw, JA, AM Middlebrook, C Warneke, PD Goldan, WC Kuster, JM Roberts, FC Fehsenfeld, DR Worsnop, MR Canagaratna, AAP Pszenny, WC Keene, M Marchewka, SB Bertman, and TS Bates. 2005. "Budget of organic carbon in a polluted atmosphere: Results from the New England Air Quality Study in 2002." *Journal of Geophysical Research: Atmospheres* 110: D16305, doi:10.1029/2004jd005623.

Duplissy, J, PF DeCarlo, J Dommen, MR Alfarra, A Metzger, I Barmpadimos, ASH Prevot, E Weingartner, T Tritscher, M Gysel, AC Aiken, JL Jimenez, MR Canagaratna, DR Worsnop, DR Collins, J Tomlinson, and U Baltensperger. 2011. "Relating hygroscopicity and composition of organic aerosol particulate matter." *Atmospheric Chemistry and Physics* 11: 1155-1165, doi:10.5194/acp-11-1155-2011.

Fisch, G, J Tota, LAT Machado, MAF Silva Dias, RF da F. Lyra, CA Nobre, AJ Dolman, and JHC Gash. 2004. "The convective boundary layer over pasture and forest in Amazonia." *Theoretical and Applied Climatolology* 78: 47-59, doi:10.1007/s00704-004-0043-x.

Gunthe, SS, SM King, D Rose, Q Chen, P Roldin, DK Farmer, JL Jimenez, P Artaxo, MO Andreae, ST Martin, and U Pöschl. 2009. "Cloud condensation nuclei in pristine tropical rainforest air of Amazonia: size-resolved measurements and modeling of atmospheric aerosol composition and CCN activity." *Atmospheric Chemistry and Physics* 9: 7551-7575, doi:10.5194/acp-9-7551-2009.

Huisman, A, J Hottle, M Galloway, J DiGangi, K Coens, W Choi, I Faloona, J Gilman, W Kuster, J de Gouw, N Bouvier-Brown, A Goldstein, B LaFranchi, R Cohen, G Wolfe, J Thornton, K Docherty, D Farmer, M Cubison, J Jimenez, J Mao, W Brune, and F Keutsch. 2011. "Photochemical modeling of glyoxal at a rural site: observations and analysis from BEARPEX 2007." *Atmospheric Chemistry and Physics* 11: 8883-8897, doi:10.5194/acp-11-8883-2011.

Jimenez, JL, MR Canagaratna, NM Donahue, ASH Prevot, Q Zhang, JH Kroll, PF DeCarlo, JD Allan, H Coe, NL Ng, AC Aiken, KS Docherty, IM Ulbrich, AP Grieshop, AL Robinson, J Duplissy, JD Smith,

KR Wilson, VA Lanz, C Hueglin, YL Sun, J Tian, A Laaksonen, T Raatikainen, J Rautiainen, P Vaattovaara, M Ehn, M Kulmala, JM Tomlinson, DR Collins, MJE Cubison, J Dunlea, JA Huffman, TB Onasch, MR Alfarra, PI Williams, K Bower, Y Kondo, J Schneider, F Drewnick, S Borrmann, S Weimer, K Demerjian, D Salcedo, L Cottrell, R Griffin, A Takami, T Miyoshi, S Hatakeyama, A Shimono, JY Sun, YM Zhang, K Dzepina, JR Kimmel, D Sueper, JT Jayne, SC Herndon, AM Trimborn, LR Williams, EC Wood, AM Middlebrook, CE Kolb, U Baltensperger, and DR Worsnop. 2009. "Evolution of Organic Aerosols in the Atmosphere." *Science* 326(5959): 1525-1529, doi:10.1126/science.1180353.

Knote, C, A Hodzic, JL Jimenez, R Volkamer, JJ Orlando, S Baidar, J Brioude, J Fast, DR Gentner, AH Goldstein, PL Hayes, WB Knighton, H Oetjen, A Setyan, H Stark, R Thalman, G Tyndall, R Washenfelder, E Waxman, and Q Zhang. Q. 2014. "Simulation of semi-explicit mechanisms of SOA formation from glyoxal in a 3-D model." *Atmospheric Chemistry and Physics* 14: 6213-6239, doi:10.5194/acp-14-6213-2014.

Lambe, AT, AT Ahern, LR Williams, JG Slowik, JPS Wong, JPD Abbatt, WH Brune, NL Ng, JP Wright, DR Croasdale, DR Worsnop, P Davidovits, and TB Onasch. 2011. "Characterization of aerosol photooxidation flow reactors: heterogeneous oxidation, secondary organic aerosol formation and cloud condensation nuclei activity measurements." *Atmospheric Measurement Techniques* 4: 445-461, doi:10.5194/amt-4-445-2011.

Mei, F, PL Hayes, A Ortega, JW Taylor, JD Allan, J Gilman, W Kuster, J de Gouw, JL Jimenez, and J Wang. 2013a. "Droplet activation properties of organic aerosols observed at an urban site during CalNex-LA." *Journal of Geophysical Research: Atmospheres* 118(7): 2903-2917, doi:10.1002/jgrd.50285.

Mei, F, A Setyan, Q Zhang, and J Wang. 2013b. "CCN activity of organic aerosols observed downwind of urban emissions during CARES." *Atmospheric Chemistry and Physics* 13: 12155-12169, doi:10.5194/acp-13-12155-2013.

Padró, LT, D Tkacik, T Lathem, CJ Hennigan, AP Sullivan, RJ Weber, LG Huey, and A Nenes. 2010. "Investigation of cloud condensation nuclei properties and droplet growth kinetics of the water-soluble aerosol fraction in Mexico City." *Journal of Geophysical Research: Atmospheres* 115(D9), doi:10.1029/2009jd013195.

Rissler, J, A Vestin, E Swietlicki, G Fisch, J Zhou, P Artaxo, and MO Andreae. 2006. "Size distribution and hygroscopic properties of aerosol particles from dry-season biomass burning in Amazonia." *Atmospheric Chemistry and Physics* 6: 471-491, doi:10.5194/acp-6-471-2006.

Rose, D, SS Gunthe, E Mikhailov, GP Frank, U Dusek, MO Andreae, and U Pöschl. 2008. "Calibration and measurement uncertainties of a continuous-flow cloud condensation nuclei counter (DMT-CCNC): CCN activation of ammonium sulfate and sodium chloride aerosol particles in theory and experiment." *Atmospheric Chemistry and Physics* 8: 1153-1179, doi:10.5194/acp-8-1153-2008.

Stocker, TF, Q Dahe, and G-K Plattner. Climate Change 2013: The Physical Science Basis, Working Group I Contribution to the Fifth Assessment Report of the Intergovernmental Panel on Climate Change. Summary for Policymakers (IPCC, 2013). https://www.ipcc.ch/pdf/assessment-report/ar5/wg1/WGIAR5_SPM_brochure_en.pdf

Vestin, A, J Rissler, E Swietlicki, GP Frank, and MO Andreae. 2007. "Cloud-nucleating properties of the Amazonian biomass burning aerosol: Cloud condensation nuclei measurements and modeling." *Journal of Geophysical Research: Atmospheres* 112(D14), doi:10.1029/2006jd008104.

Volkamer, R, JL Jimenez, F San Martini, K Dzepina, Q Zhang, D Salcedo, LT Molina, DR Worsnop, and MJ Molina. 2006. "Secondary organic aerosol formation from anthropogenic air pollution: Rapid and higher than expected." *Geophysical Research Letters* 33: L17811, doi:10.1029/2006GL026899.

Volkamer, R, F San Martini, LT Molina, D Salcedo, JL Jimenez, and MJ Molina. 2007. "A missing sink for gas-phase glyoxal in Mexico City: Formation of secondary organic aerosol." *Geophysical Research Letters* 34(19): L19807, doi:10.1029/2007GL030752.

Washenfelder, RA, CJ Young, SS Brown, WM Angevine, EL Atlas, DR Blake, DM Bon, MJ Cubison, JA de Gouw, S Dusanter, J Flynn, JB Gilman, M Graus, S Griffith, N Grossberg, PL Hayes, JL Jimenez, WC Kuster, BL Lefer, IB Pollack, TB Ryerson, H Stark, PS Stevens, and MK Trainer. 2011. "The glyoxal budget and its contribution to organic aerosol for Los Angeles, California, during CalNex 2010." *Journal of Geophysical Research: Atmospheres* 116: D00V02, doi:10.1029/2011JD016314.

Whitehead, JD, MW Gallagher, JR Dorsey, N Robinson, AM Gabey, H Coe, G McFiggans, MJ Flynn, J Ryder, E Nemitz, and F Davies. 2010. "Aerosol fluxes and dynamics within and above a tropical rainforest in South-East Asia." *Atmospheric Chemistry and Physics* 10: 9369-9382, doi:10.5194/acp-10-9369-2010.

Xu, L, H Guo, CM Boyd, M Klein, A Bougiatioti, KM Cerully, JR Hite, G Isaacman-VanWertz, NM Kreisberg, C Knote, K Olson, A Koss, AH Goldstein, SV Hering, J de Gouw, K Baumann, S-H Lee, A Nenes, RJ Weber, and NL Ng. 2015. "Effects of anthropogenic emissions on aerosol formation from isoprene and monoterpenes in the southeastern United States." *Proceedings of the National Academy of Sciences* 112(1): 37-42, doi:10.1073/pnas.1417609112.

Zaveri, RA, CM Berkowitz, LI Kleinman, SR Springston, PV Doskey, WA Lonneman, and CW Spicer. 2003. "Ozone production efficiency and NOx depletion in an urban plume: Interpretation of field observations and implications for evaluating O3-NOx-VOC sensitivity." *Journal of Geophysical Research: Atmospheres* 108(D14), doi:10.1029/2002jd003144.

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