Insight into Particle Growth Rates and Controls from Two Years of SGP SMPS and HTDMA Data

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I. Introduction

Frequent aerosol formation and subsequent growth are observed over the DOE SGP ARM site. Aerosol growth appears to occur primarily due to condensation of low volatility gases, but other mechanisms (e.g., coagulation) can sometimes contribute significantly. Number size and size resolved hygroscopicity distributions of identified growth episodes were fitted with lognormals. Time series profiles of the growing mode median particle size and hygroscopic growth factors (HGF) were fitted using appropriate mathematical forms. Condensational growth for all episodes were estimated by eliminating coagulation loss. Time dependent slopes of particle size and HGF were used to quantify the net addition of aerosol mass and its characteristics and subsequently were interpreted into hygroscopicity parameter, kappa, to link the addition of generic aerosol components (i.e., more hygroscopic / "inorganic-like" and less hygroscopic / "organic-like") to the observed growth. Those additions were further related to requisite gas phase concentrations to study temporal variations.

II. Identifying & fitting growth modes

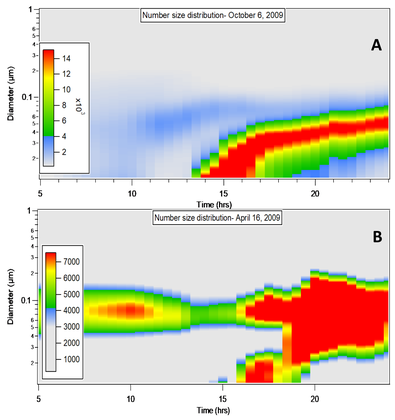

Figure 1. Aerosol number size distribution within A. clean background and B. polluted background

- **Size distribution range:** 12 nm - 15µm
- **Size resolved hygroscopicity:** 13, 25, 50, 100, 200, 400, and 600 nm
- **Identification:** High number concentration in nucleation mode and consistent growth for hours
- **Fit:** Polynomial, Logarithmic

III. Seasonality of growth events

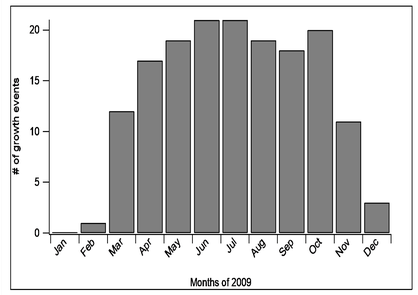

Figure 2. Variation in growth event frequency during 2009

- **Result:** Clear seasonality in 2009
- **Frequent in summer:** Intense solar irradiation, high biogenic emissions, and frequent precipitation
- **Rare in winter**

IV. Effect of coagulation

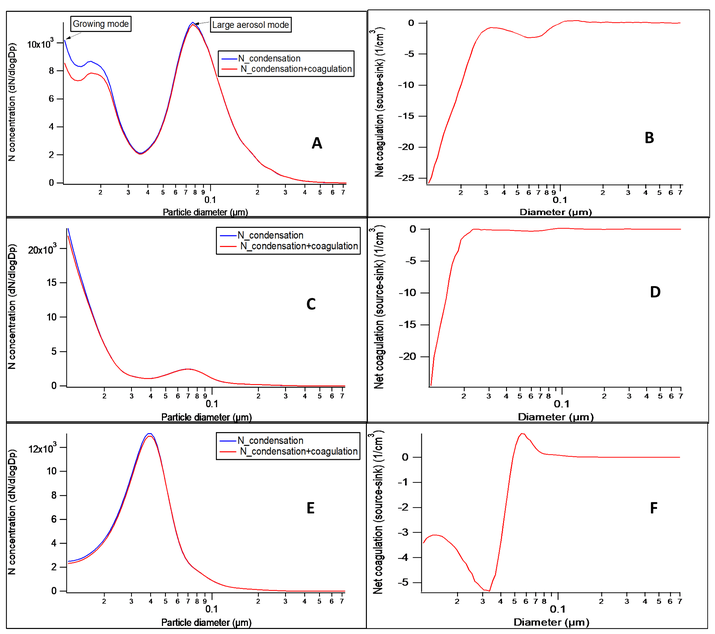

Figure 3. Effect of coagulation based on 3 scenarios

A: N concentration on April 16, 2009 following nucleation: *Nucleation & accumulation mode N conc. very high*
B: Net coagulation on April 16, 2009 following nucleation: *coagulation loss* ***maximum of 16%***
C: N concentration on October 6, 2009 following nucleation: *Nucleation mode N conc. high*
D: Net coagulation on October 6, 2009 following nucleation: *Coagulation loss 7%*
E: N concentration on October 6, 2009 later that day: *Accumulation mode conc. high*
F: Net coagulation on October 6, 2009 later that day: *Coagulation loss 6%*

V. Aerosol growth profile

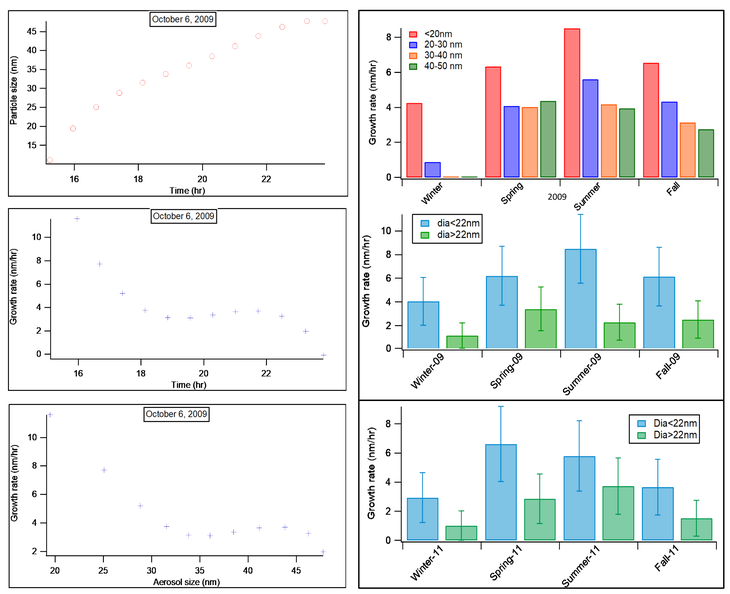

Figure 4: Diurnal growth profile
1A: Aerosol size VS. time
1B: Aerosol growth rate VS. time
1C: Aerosol growth rate VS. time

Figure 5: Seasonal growth profile
2A: Growth rate VS. season in 2009 for 4 size classes
2B: Growth rate VS. season in 2009 for 2 size classes
2C: Growth rate VS. season in 2011 for 2 size classes

VI. Hygroscopicity analysis

- **Growth mode HGF:** Interpolated from 7 aerosol size HGF matrix
- **HGF Diurnal pattern:** Often high during day (more inorganic or more oxidized organic), low at night (less inorganic or less oxidized organic?)
- **Kappa Diurnal pattern:** 0.2 - 0.3 during day, 0.1 - 0.2 at night

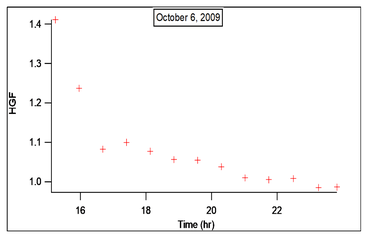

Figure 6. Diurnal variation of growing mode hygroscopicity on October 6, 2009

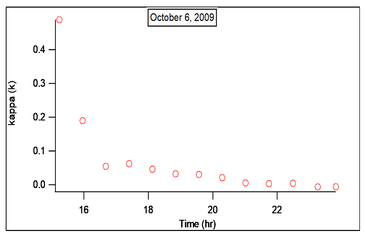

Figure 7. Diurnal variation of growing mode kappa on October 6, 2009

- **Organic kappa, k_{org}:** Using ACSM, SMPS, HTDMA data & mixing rule
- **Assumptions:** Bulk and size dependent composition
- **Result:** Some deviation, but, similar pattern

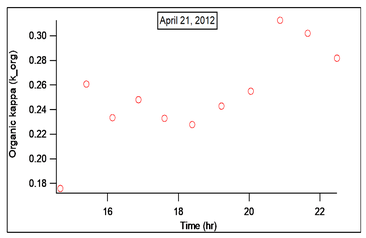

Figure 8. Diurnal variation of organic content kappa on April 21, 2012 assuming size independent volume fraction of particle phase chemical species

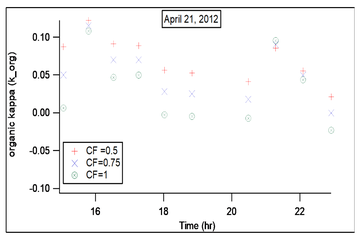

Figure 9. Diurnal variation of organic content kappa on April 21, 2012 assuming size dependent volume fraction of particle phase chemical species

VII. Precursor concentration profile

- **Assumptions:** Aerosols are internal mixtures of inorganic and organic species . Gas phase component ratio is similar to added particle phase (r). Organic kappa, K_{org} is (i) constant at 0.1 or (ii) temporally variable

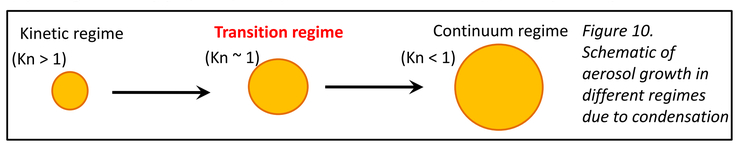

Figure 10. Schematic of aerosol growth in different regimes due to condensation

- **Total precursor concentration (TC) :** From mass balance in appropriate regime
- **Inorganic-like / organic-like precursor concentration**: Using TC and r
- **Result:** Similar profiles for both *Korg* assumptions, nighttime growth due to organic-like & daytime growth due to both types

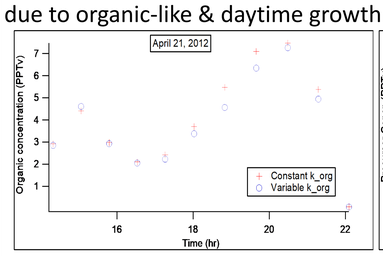

Figure 11. Comparison between "organic" precursor concentrations assuming Korg as constant and as variable

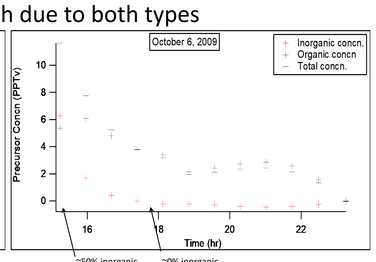

Figure 12. "Inorganic", "organic" and total precursor concentration assuming Korg as 0.1

VIII. Precursor concentration profile (contd.)

- **Result:** Seasonal profile of average precursor concentration
- **Inorganic-like concentration:** No definite pattern
- **Organic-like concentration:** Higher concentration during summer
- **VOC emission from vegetation:** *fn*(light intensity, leaf temperature)

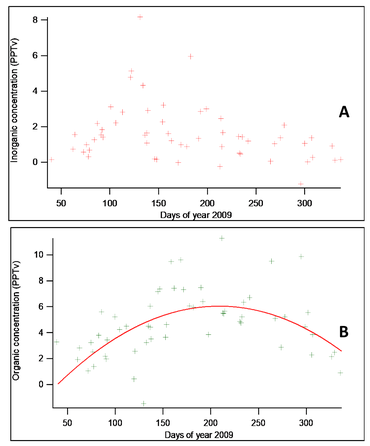

Figure 13. A. Variation in inorganic-like and B. Variation in organic-like precursor concentrations during 2009

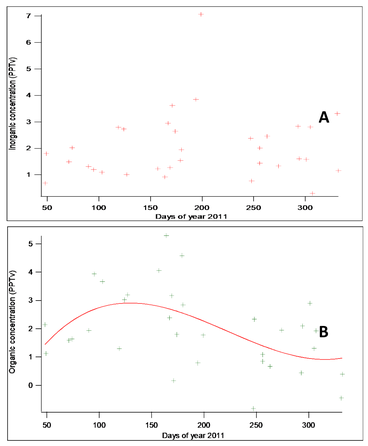

Figure 14. A. Variation in inorganic-like and B. Variation in organic-like precursor concentrations during 2011

IX. Summary

- Growth frequency was highest in summer 2009, presumably due to intense sunlight, abundant VOCs and frequent accumulation-mode scrubbing precipitation.
- Condensation was found to contribute more to particle growth for all growth episodes compared to coagulation.
- Diurnal profile of growing aerosol mode (~12 – 50 nm) indicates a decreasing growth rate with increasing size (e.g., 8.5 mm/hr for 20 nm, 5.6 nm/hr for 20-30 nm, 4.1 nm/hr for 30-40 nm in 2009 summer).
- Seasonal variation of the growth rate showed an increasing trend towards summer (e.g., winter: 4.2 nm/hr, spring: 6.3 nm/hr, summer: 8.5 nm/hr, and fall: 6.5 nm/hr during 2009)
- The HGF profile of the growing mode aerosol was typically higher during the day, suggesting condensation of more hygroscopic (e.g., inorganic-like) species and lower at night, suggesting condensation of less hygroscopic (e.g., organic-like) species. kappa (k) of these small particles was also higher during the day (0.2-0.3) and lower at night (0.1-0.2). The inferred kappa for the organic content computed for 6 days in 2012 showed a diurnal pattern likely reflecting chemical evolution of the organic components.
- Estimated gas phase precursor concentrations assuming k_{org} = 0.1 varied seasonally in the organic-like precursor concentration with a maximum during summer, which likely reflects increased organic precursor emissions from the surrounding agricultural fields and increased oxidant concentrations and photo-chemical reaction rates.
- Diurnal profiles showed higher inorganic-like precursor concentration during day. Introducing k_{org} as variable caused some change, but did not influence the overall trend.