Modeling Evolution of Aerosol Size Distribution in the Manaus Urban Plume During the GoAmazon 2014 Campaign

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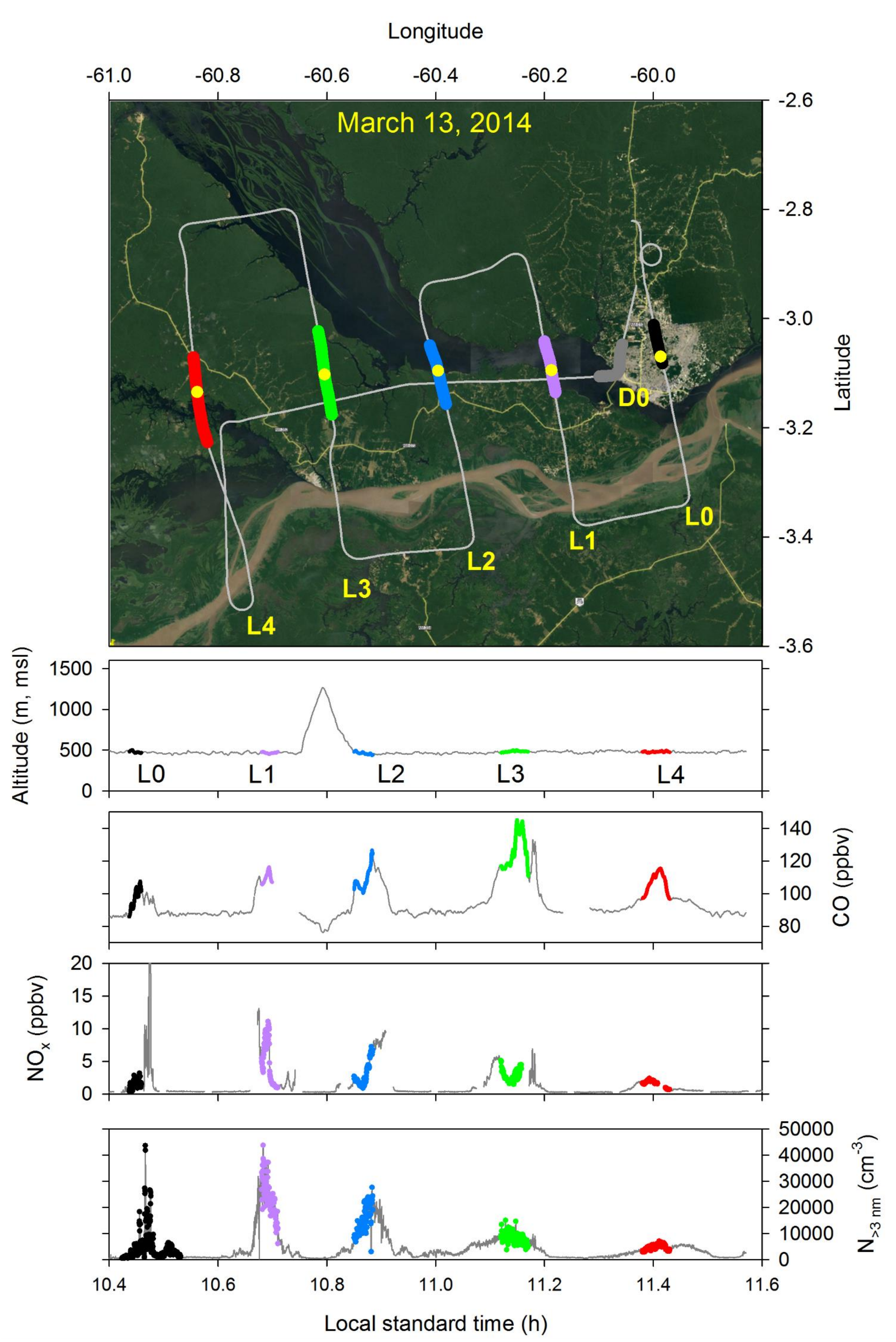
1. Introduction

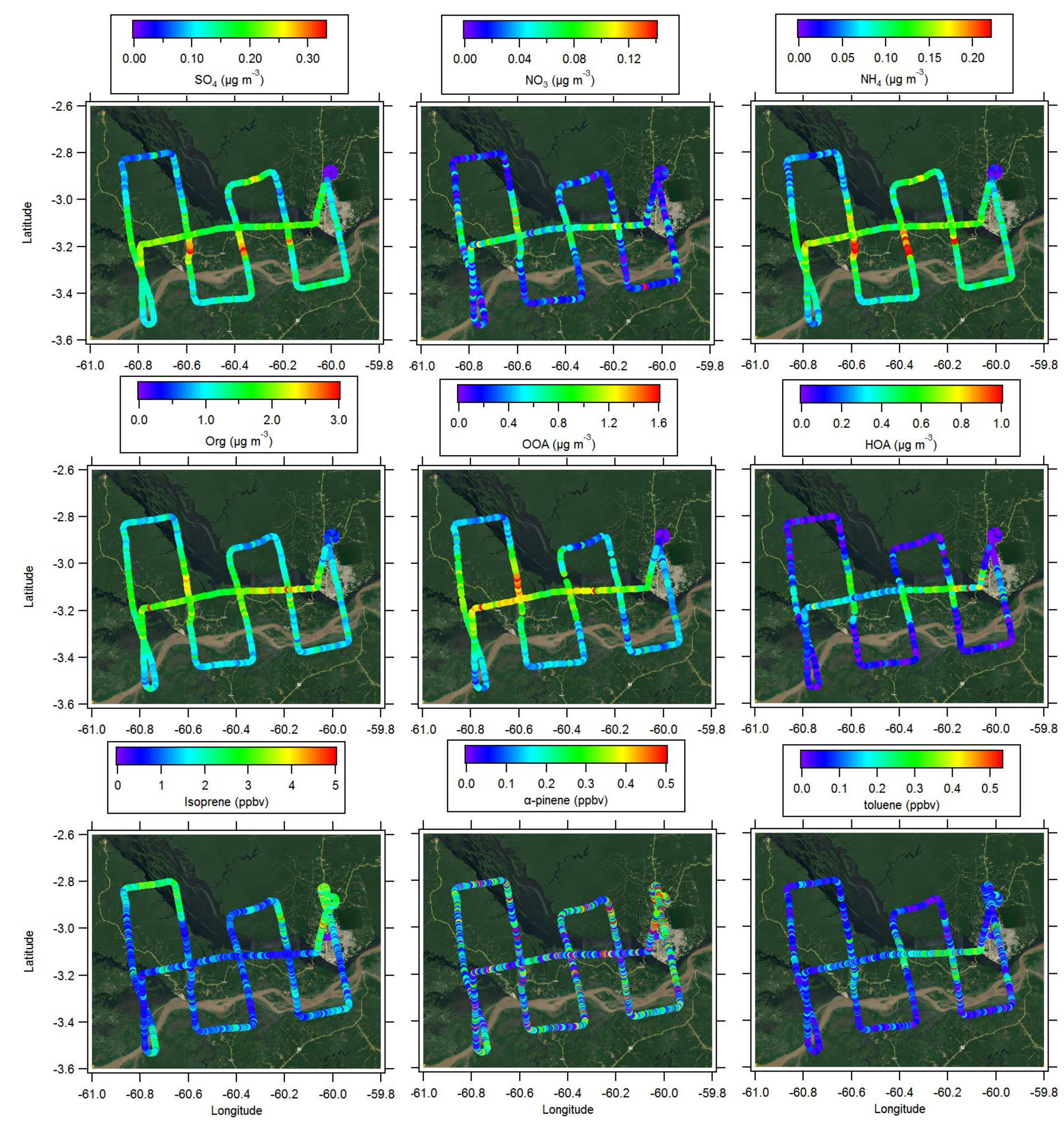
One of the objectives of the GoAmazon field campaign is to understand how the urban aerosol from Manaus interacts with natural emissions from the surrounding relatively pristine rainforest. Here we analyze the semi-Lagrangian aircraft measurements made downwind of Manaus on March 13, 2014, with a focus on the growth of the ultrafine urban aerosol to cloud condensation nuclei (CCN) sizes. The comprehensive sectional aerosol box model MOSAIC is applied to interpret the observed evolution of aerosol size distribution and gain insights into the mechanisms of secondary organic aerosol (SOA) formation and particle growth in the Manaus plume. The roles of coagulation and condensation are evaluated in shaping the aerosol size distribution with time.

2. Manaus Urban Plume

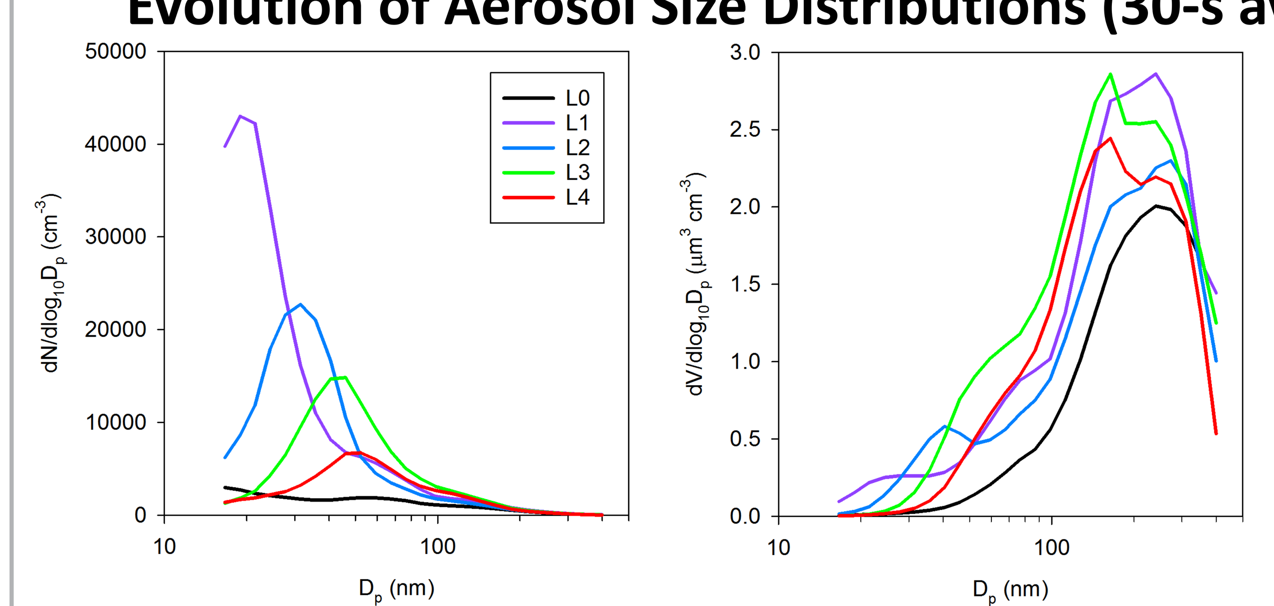


The Manaus urban plume is identified at increasing downwind distances, characterized by enhanced CO, NOx, and particle number concentrations. The colored legs are selected here for further analysis. Yellow dots in each leg represents the selected plume peak.

Aerosol (HR-AMS) and Trace Gas (PTRMS) Composition

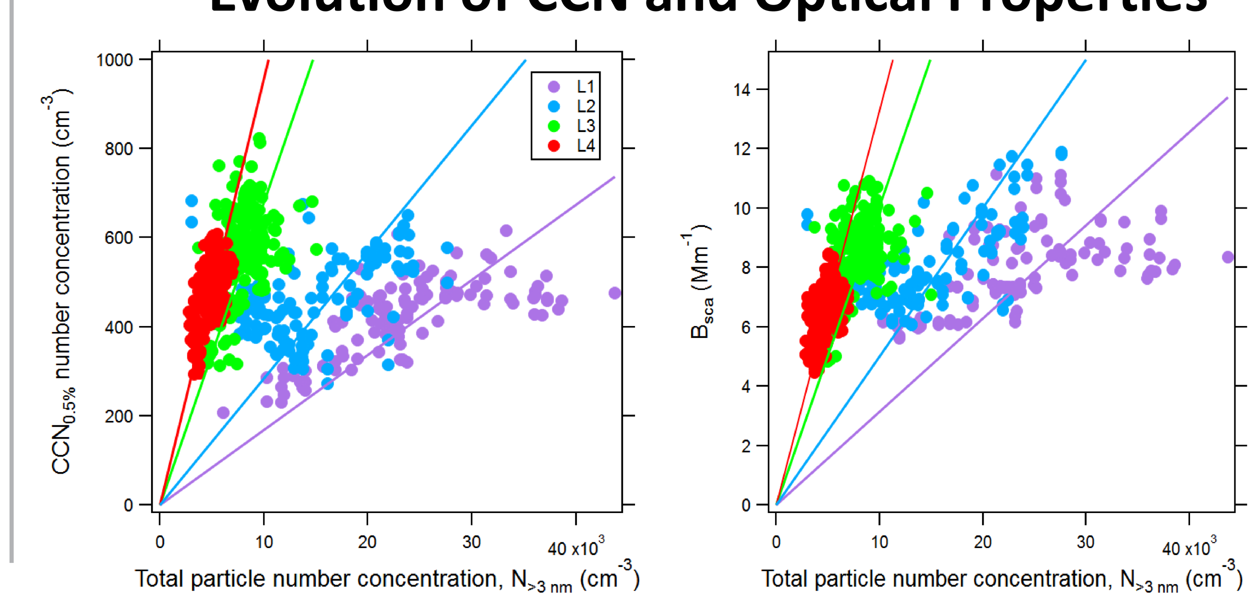


Evolution of Aerosol Size Distributions (30-s avg FIMS data)



Ultrafine aerosol is observed to grow with plume aging, but the accumulation mode aerosol size appears to remain constant (as seen in the volume size distribution).

Evolution of CCN and Optical Properties

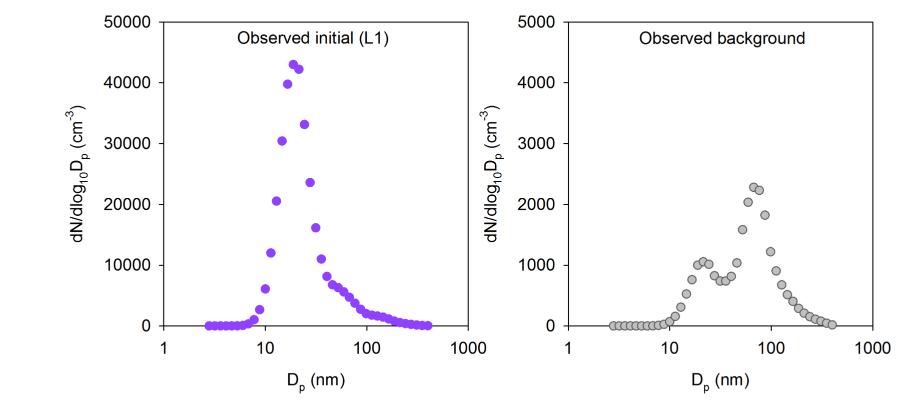


CCN concentrations and scattering coefficients are found to significantly increase with plume aging.

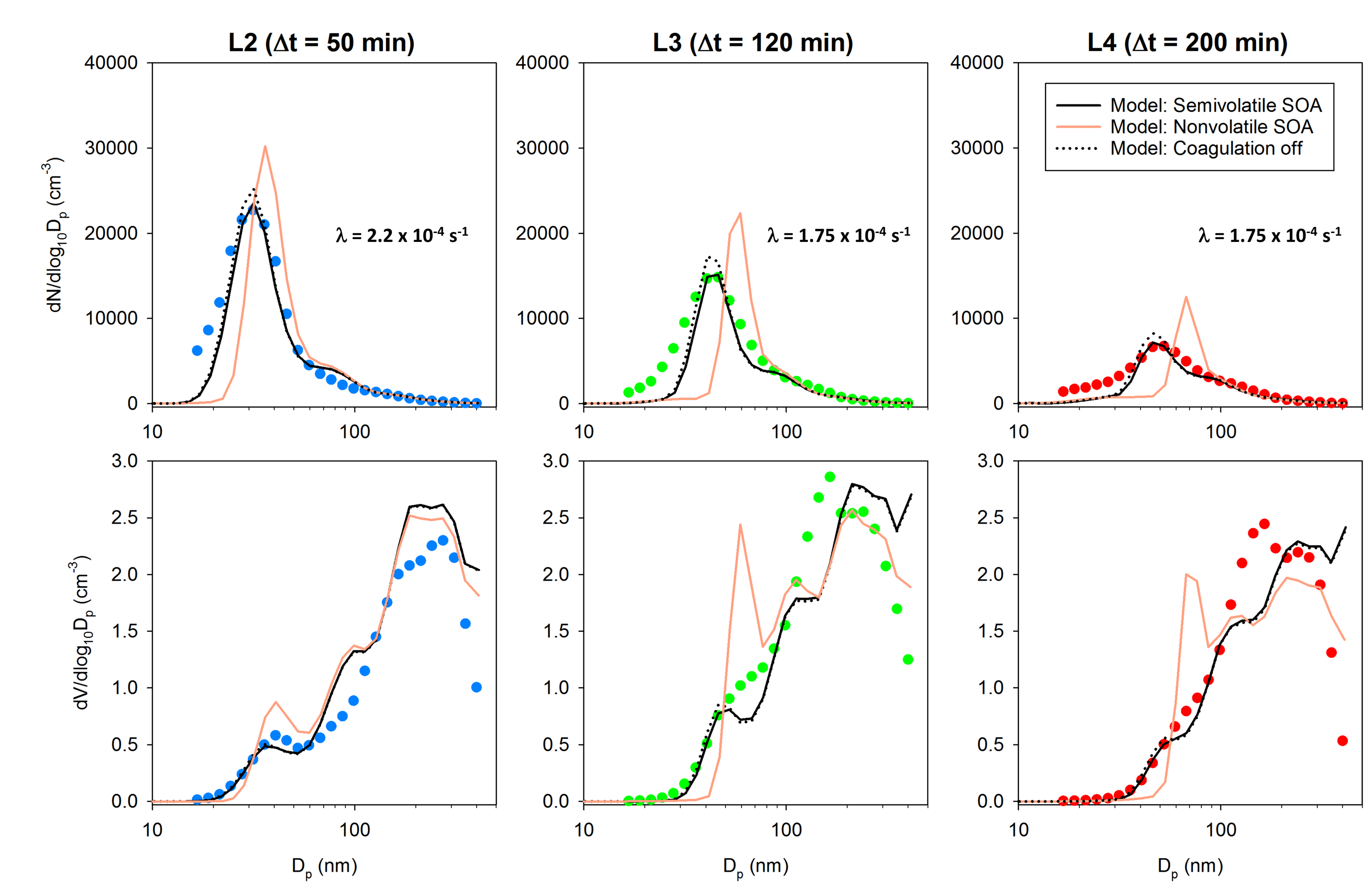
3. Model Analysis of Aerosol Growth

MOSAIC is applied in a Lagrangian box modeling framework to examine the impact of the volatility of condensing SOA species on the evolution of aerosol size distribution.

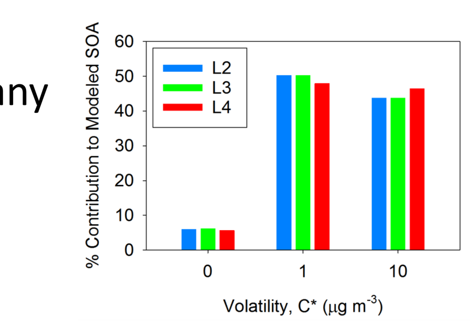
The model is initialized using observations at L1 and allowed to mix with background aerosol due to dilution at a prescribed first-order rate, λ (s^{-1}). Performance is evaluated using observations at L2, L3, and L4.



Model Evaluation Results



A semivolatile SOA model is able to reproduce the observed size distribution evolution without invoking any bulk diffusion limitation. The nonvolatile condensation model overpredicts the growth of ultrafine aerosol. Coagulation appears to play a minor role due to rapid dilution with the background.



	Observed		Model Semivolatile SOA		Model Nonvolatile SOA		Model Coagulation Off	
Plume Leg	N_{tot} (cm^{-3})	V_{tot} (μm^3 cm^{-3})	N_{tot} (cm^{-3})	V_{tot} (μm^3 cm^{-3})	N_{tot} (cm^{-3})	V_{tot} (μm^3 cm^{-3})	N_{tot} (cm^{-3})	V_{tot} (μm^3 cm^{-3})
L1	14803.6	4.71						
L2	9106.5	1.44	8054.7	1.72	8074.6	1.74	8475.1	1.72
L3	6526.0	1.87	4838.4	1.86	4871.3	1.92	5241.5	1.86
L4	3594.1	1.50	2782.3	1.52	2801.9	1.49	3006.0	1.52

4. Conclusions & Future Directions

- Rapid growth of Manaus ultrafine aerosol occurred as the urban plume was advected downwind, causing a sharp increase in CCN concentrations and light scattering.
- Condensation of semivolatile organic species (as opposed to mostly nonvolatile) appears to be necessary to reproduce the observed size distribution evolution.
- The present case can be used to test detailed SOA mechanisms and further investigate anthropogenic-biogenic interactions that affect climate-relevant aerosol properties..

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