Assessing the Impact of Measured New Particle Formation on Aerosol Size Distributions Downwind of Sources

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Introduction

The University of Denver Nucleation-Mode Aerosol Size Spectrometer (NMASS) measured the aerosol size distribution ranging from 4.0 nm to 60.0 nm over a total of 12 flights on board the Spec Learjet during the SEAC^{4}RS campaign. On-going new particle formation events can create particles with diameters of one to two nanometers. In-situ measurements of particles with the NMASS provide evidence for recent new particle formation since nanometer sized particles are removed rapidly by coagulation. In this investigation, we combine in-situ measurements with air parcel trajectory and coagulation calculations to indicate how far these particles may be transported before they are removed. Calculations of condensational to sufficient size to serve as cloud condensation nuclei will be made later permitting the impact of NPF to be assessed.

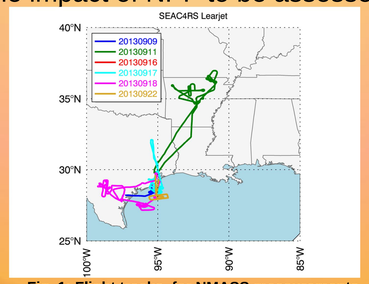

Fig. 1: Flight tracks for NMASS measurements

Instrumentation

- The NMASS consists of 5 continuous flow CPCs operating in parallel. The d_{p50} for each of the CPCs are 5.3 nm, 8.4 nm, 15 nm, 30 nm and 53 nm. The lowest detection limit is 4.0 nm. Aerosol particles are counted at 10Hz and these data are reported at 1Hz for analysis.
- The University of Denver designed inlet for the NMASS allows for near-isokinetic sampling, reducing errors in measured aerosol concentrations.

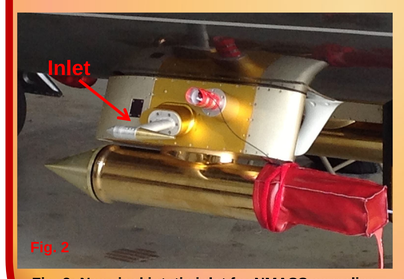

Fig. 2: Near-isokintetic inlet for NMASS sampling

Fig. 3 : The University of Denver Nucleation-Mode Aerosol Size Spectrometer (NMASS) as deployed during SEAC^{4}RS.

Detecting New Particle Formation

The number of particles in the size range 5.3-8.4 nm was determined by subtracting the mixing ratio of particles in the second channel (M2) from that in the first channel (M1). The "relative difference" is defined as the ratio of the reported difference (M_1 - M_2) to the standard deviation in this value expected to result from the counts ($\sigma \approx \sqrt{\text{counts}}$) (1).

$$\text{Relative difference} = \frac{M_1 - M_2}{(b_1^2 C_1 - b_2^2 C_2)^{0.5}} \quad (1)$$

where $M_i = b_i C_i$, M_i are mixing ratios calculated from CPC channel i, C_i are counts (particles/s) from CPC channel i and b_i is dependent on sample flow, instrument pressure and temperature. An NPF event is identified when relative difference is greater than 3. Using this criterion, the chance of misclassifying NPF events as a result of statistical fluctuations is less than 0.2%. In order to reduce false NPF detections from ice shattering on the inlet, a composite cloud mask was generated using data from SPEC's Hawk 10 and Hawk 50 2D-S (Stereo) Probes to filter out measurements inside clouds.

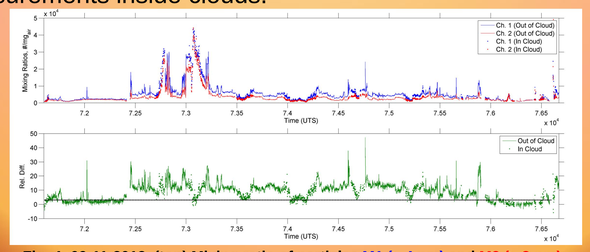

Fig. 4: 09-11-2013 (top) Mixing ratio of particles M1 (> 4 nm) and M2 (> 8 nm); (bottom) Relative difference

NPF Events During SEAC^{4}RS

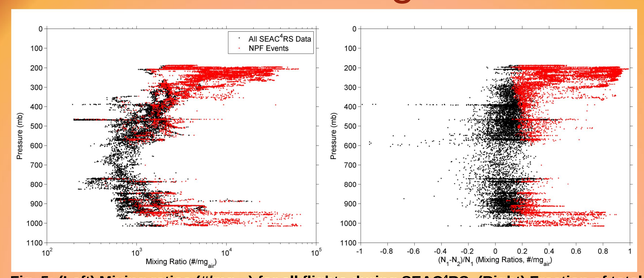

Fig. 5: (Left) Mixing ratios (#/mg$_{Air}$) for all flights during SEAC^{4}RS. (Right) Fraction of total number measured particles that were in the smallest size range (5.3-8.4 nm) . Points in red are identified NPF events based on criteria (1) above.

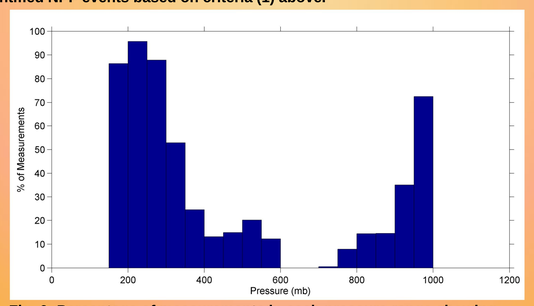

Fig. 6: Percentage of measurements in each pressure range showing evidence of new particle formation.

Impact of NPF Events

We did not measure full size distributions from the Learjet in SEAC4RS. We will estimate pre-existing aerosol surfaces based on overlaps with other platforms measuring aerosol in SEAC4RS and calculate evolution of the SEACR4S aerosol. Similar NPF events (multiple 100+ second duration events, altitudes near 10 km) were seen during the MACPEX campaign which also used the NMASS and covered a similar region.

In order to estimate the impact of NPF events downwind of the source, a four day forward trajectory calculation (see Fig. 7) of the air parcel containing a 484 sec NPF event from MACPEX (04-26-2011) were determined using NOAA's Hybrid Single Particle Lagrangian Integrated Trajectory Model (HYSPLIT).The evolution of one size distribution from this event was determined using the MAEROS coagulation model (see Fig. 8).

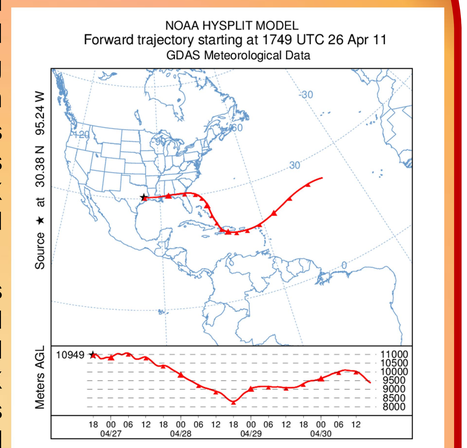

Fig. 7: (Top) Calculated horizontal trajectory of air parcel sampled during MACPEX campaign for 4 day HYSPLIT model run. (Bottom) Vertical trajectory.

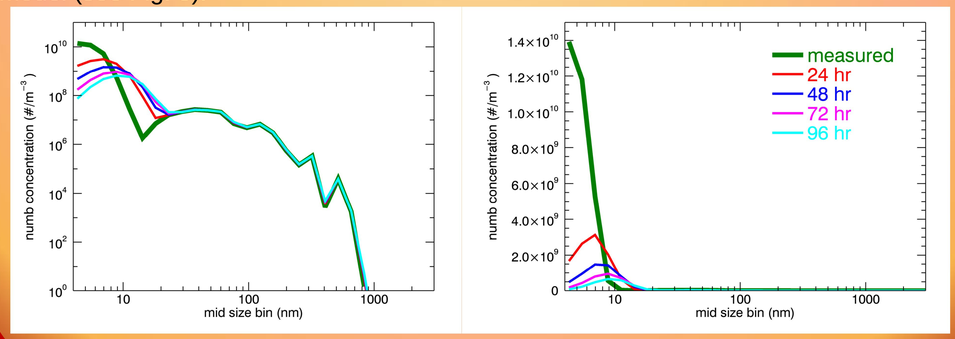

Fig. 8: Number size distribution evolution over 4 day coagulation calculation. Shown on logarithmic (Left) and linear (Right) axis.

Summary

Many instances of tropospheric new particle formation were seen during the SEAC4RS campaign. The highest percentage of measurements which meet NPF criteria were located at altitudes in the ranges 0-0.5 km and 8-12 with NPF events accounting for up to 95.7% of all measurements at 12 km. At mid altitudes (2-7 km), much fewer NPF events were seen correlating strongly with a decrease in total measured particle concentrations.

Current and future work will focus on the impact of these new particles on total aerosol loading and potential cloud interactions by serving as CCN. Recent work on size distributions obtained during the MACPEX campaign determined the evolution of the size distribution due to coagulation. The smallest particles (< 8 nm diameter) were removed from the size distribution over a period of 4 days, but growth in the larger bins was minimal. Future calculations will include condensation as this is expected to play a large role in the growth to larger sizes. Similar calculations will be performed on the SEAC4RS size distributions.

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