Chemical and Meteorological Controls on New Particle Formation in the Southern Great Plains

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Intro/Background

New particle formation (NPF) and growth is influenced by both chemical precursors and physical, meteorological conditions. NPF accounts for ~ 50% of cloud condensation nuclei globally, and a more complete understanding of this process will help constrain the global energy budget.[1]

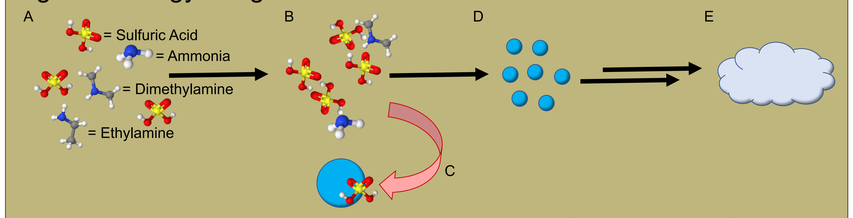

Figure 1. NPF schematic. A) NPF over land is typically driven by sulfuric acid.[2] Gas phase bases such as ammonia and amines increase nucleation rates of new particles.[3] B) Nucleation occurs when sulfuric acid forms clusters. If bases are present, they may enhance nucleation rates. C) Newly formed nanoparticles may be lost to coagulation with existing aerosol. D) If particles are not lost, they may grow and become CCN-active. E) CCN-active particles may form clouds.

Methods

Figure 2. View from guest instrumentation facility at SGP, and a map showing the location of the site.

Instrumentation deployed to the DOE Atmospheric Radiation Measurement Southern Great Plains site (SGP) in October 2021 and **April/May 2022**

- Chemical Ionization Time-of-Flight Mass Spectrometer with Ethanol Reagent Ion (EtOH-CIMS)
- Atmospheric Pressure Interface Time-of-Flight Mass Spectrometer (APi-ToF)

In addition, long-term measurements of particle size distribution are made at the site using a Scanning Mobility Particle Sizer (SMPS) and a nanoSMPS.

EtOH-CIMS includes an ammonia and amine calibration system to measure gas-phase bases quantitatively.

APi-ToF measures ambient ions, including charged clusters, which may correspond with the beginning of a NPF event.

NPF events were identified using the nanoSMPS and SMPS during the campaign, and they occurred on about 1/3 of days during both measuring periods.[4] This poster will focus on three consecutive days in April (below) that each had an NPF event.

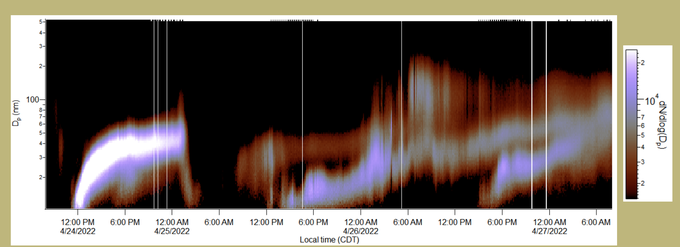

Figure 3. Particle size distribution from the SMPS at SGP showing three growth events on consecutive days.[5]

Results

APi-ToF Bisulfate and Sulfuric Acid Clusters

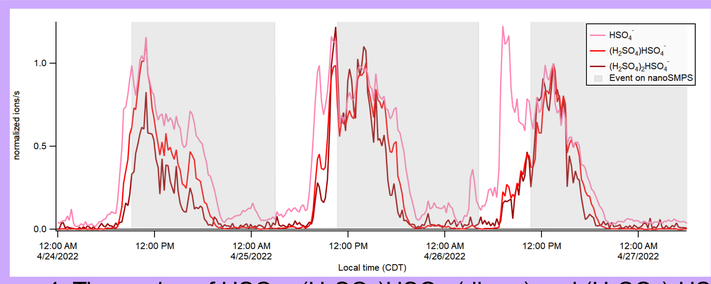

Figure 4. Timeseries of HSO_4^-, $(H_2SO_4)HSO_4^-$ (dimer) and $(H_2SO_4)_2HSO_4^-$ (trimer). Before the NPF events, we observe an increase in bisulfate, the dimer, and the trimer, corresponding with the growth of molecular clusters that occurs at the beginning of NPF.

Amines and Ammonia Participate in NPF

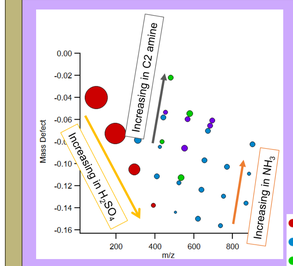

Figure 5. Mass defect plots of selected APi-Tof signals help to visualize composition of clusters. Markers are sized by signal intensity and colored by composition. Larger signals are more abundant. Signals related by the same slope are related by loss or gain of the same molecule.

- HSO_4^- and H_2SO_4 only clusters
- HSO_4^-, H_2SO_4 and NH_3 clusters
- HSO_4^-, H_2SO_4 and C2 amine clusters
- HSO_4^-, H_2SO_4, NH_3 and C2 amine clusters

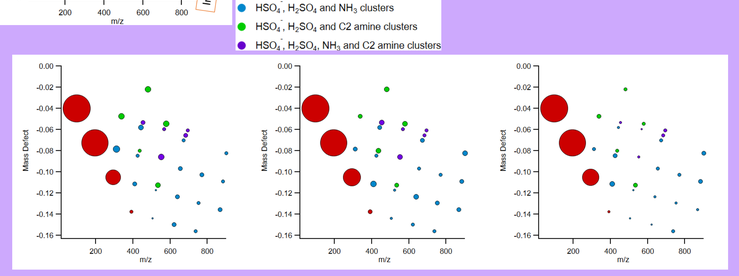

Figure 6. Mass defect plots of APi-ToF signals averaged over 8 hours during a NPF event and normalized to the dimer signal intensity. Markers are sized by signal intensity and colored by composition. Larger signals are more abundant.

EtOH-CIMS NH_3

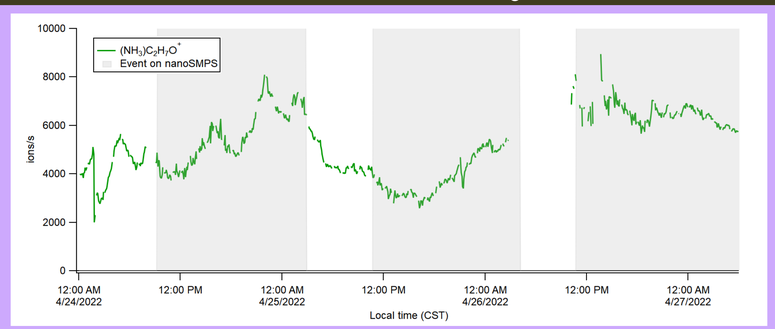

Figure 7. Timeseries of NH_3 during the three consecutive NPF events. NH_3 was always present during our measurements at the site.

HYSPLIT Back Trajectories during Events

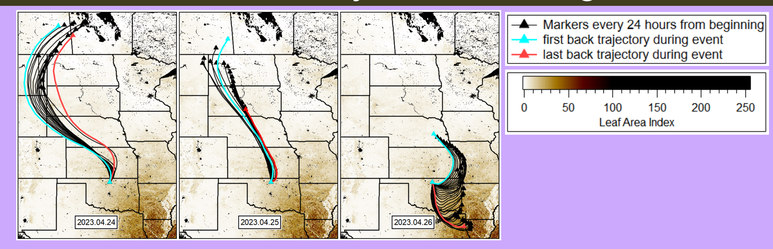

Figure 8. 24-hour HYSPLIT back trajectories calculated each hour during the NPF events on April 24th, 25th, and 26th 2022 overlaid on the Leaf Area Index from the MODIS satellite.[6] The first two days show transport from the north, while the third shows more transport from the east. The east also has more vegetation than the north, and oxidation products from biogenic emissions may participate in particle growth.

Meteorological Data during NPF Events

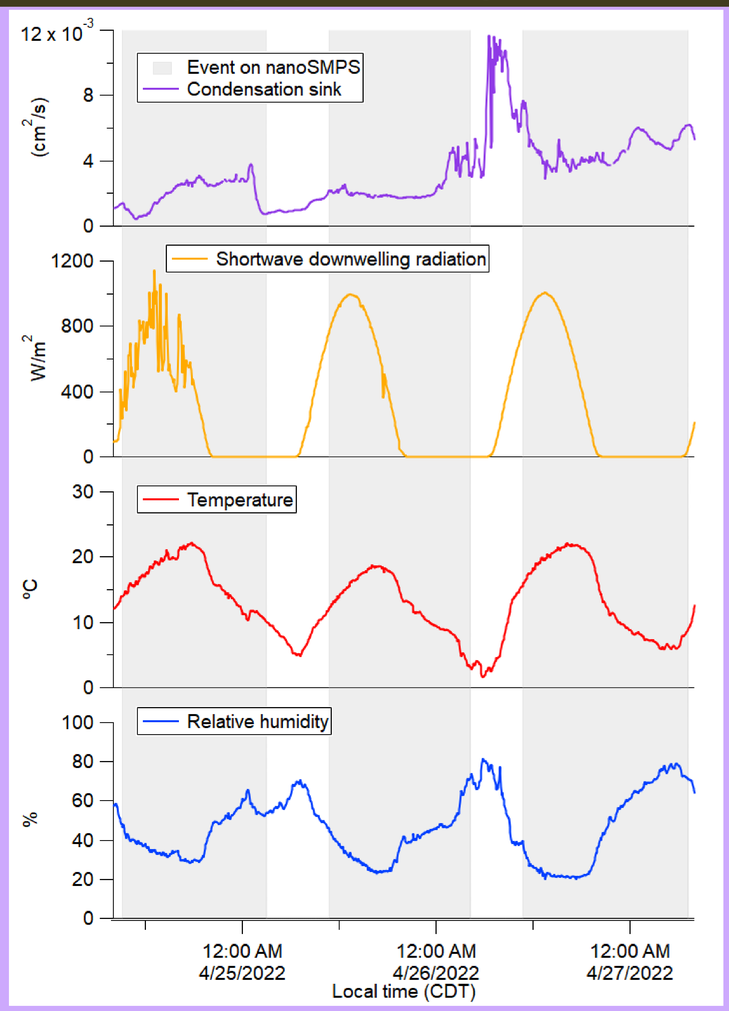

Figure 9. Temperature, relative humidity, and shortwave downwelling radiation are all similar during the three events. The condensation sink is relatively constant on the first two days, but much larger before the third event likely due to the presence of previously formed particles.

Conclusions

- Meteorological conditions were similar on the three NPF event days.
- There are differences in the chemical precursors and composition of ionized clusters measured on the APi-ToF.
  - More amines and ammonia are measured on the days when HYSPLIT back trajectories show transport from the north.
- Particles grew largest on the last of the three days when HYSPLIT back trajectories show transport from more vegetated areas in the east.

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(1) Gordon et al. *J. Geophys. Res. Atmos.* **2017.** (2) Kulmala et al. *Science*. **2013.** (3) Almeida et al. *Nature*. **2013.** (4) Kulmala et al. *Nat. Proc.* **2012.** (5) Atmospheric Radiation Measurement (ARM) user facility. 2016. Scanning mobility particle sizer (AOSSMPS). 2021-09-29 to 2022-05-07, Southern Great Plains (SGP) Lamont, OK (Extended and Co-located with C1) (E13). Compiled by C. Kuang, A. Singh and J. Howie. ARM Data Center. Data set accessed 2022-06-23. (6) Myneni, R., Knyazikhin, Y., Park, T. (2021). MODIS/Terra Leaf Area Index/FPAR 8-Day L4 Global 500m SIN Grid V061. NASA EOSDIS Land Processes DAAC. Accessed 2023-03-29.

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