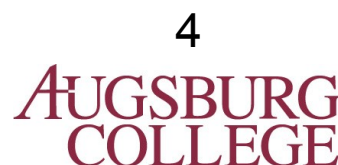


# New-particle formation and growth at the DOE Southern Great Plains field site in Oklahoma

Jeff Pierce<sup>1</sup>, Jim Smith<sup>2</sup>, Anna Hodshire<sup>1</sup>, Peter McMurry<sup>3</sup>, Jun Zhao<sup>3,a</sup>, Michael Lawler<sup>2</sup>, John Ortega<sup>2</sup>, David Hanson<sup>4</sup>, Kelley Barsanti<sup>5</sup>

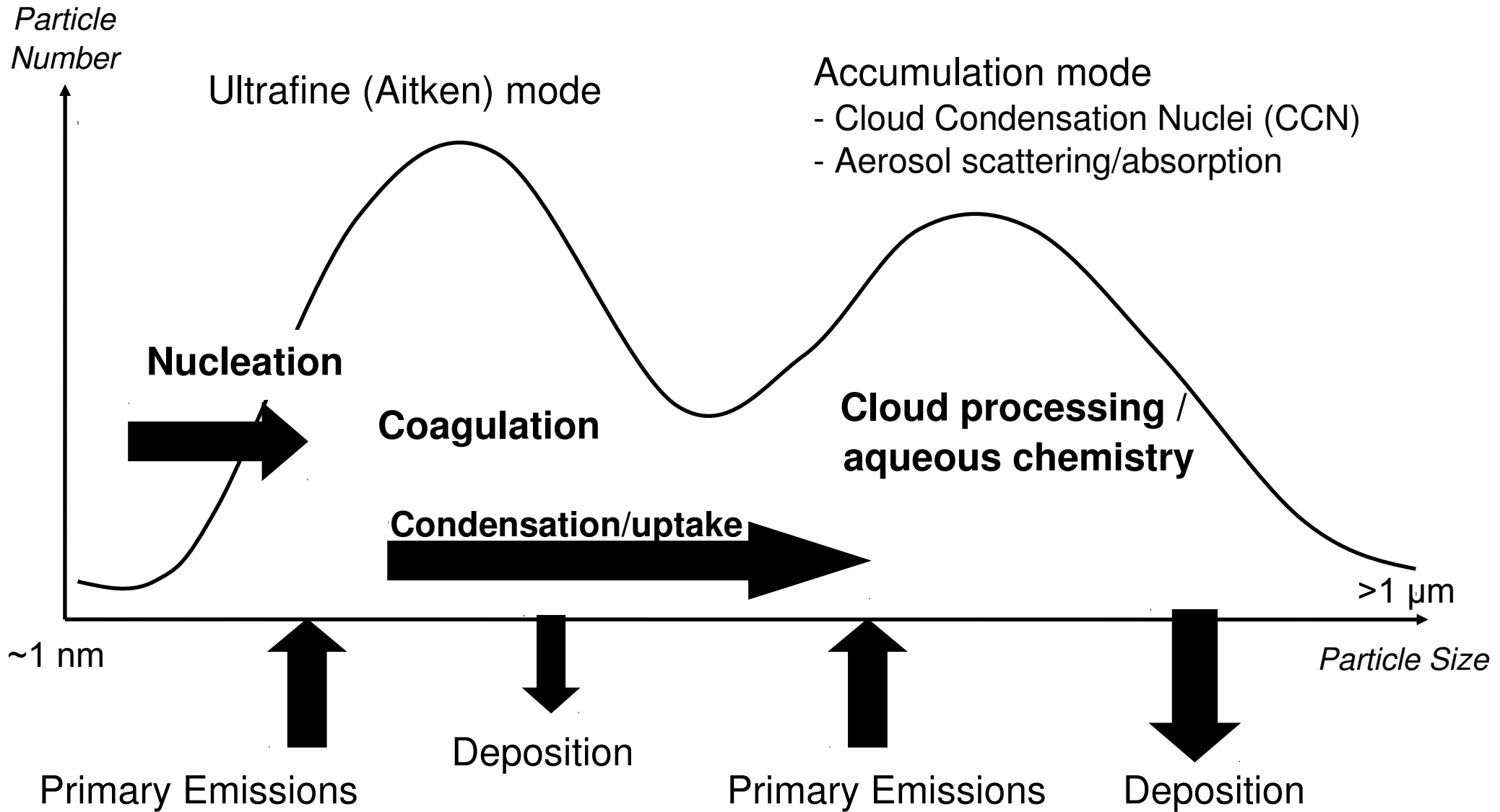


<sup>a</sup>Now at Sun Yat-Sen University

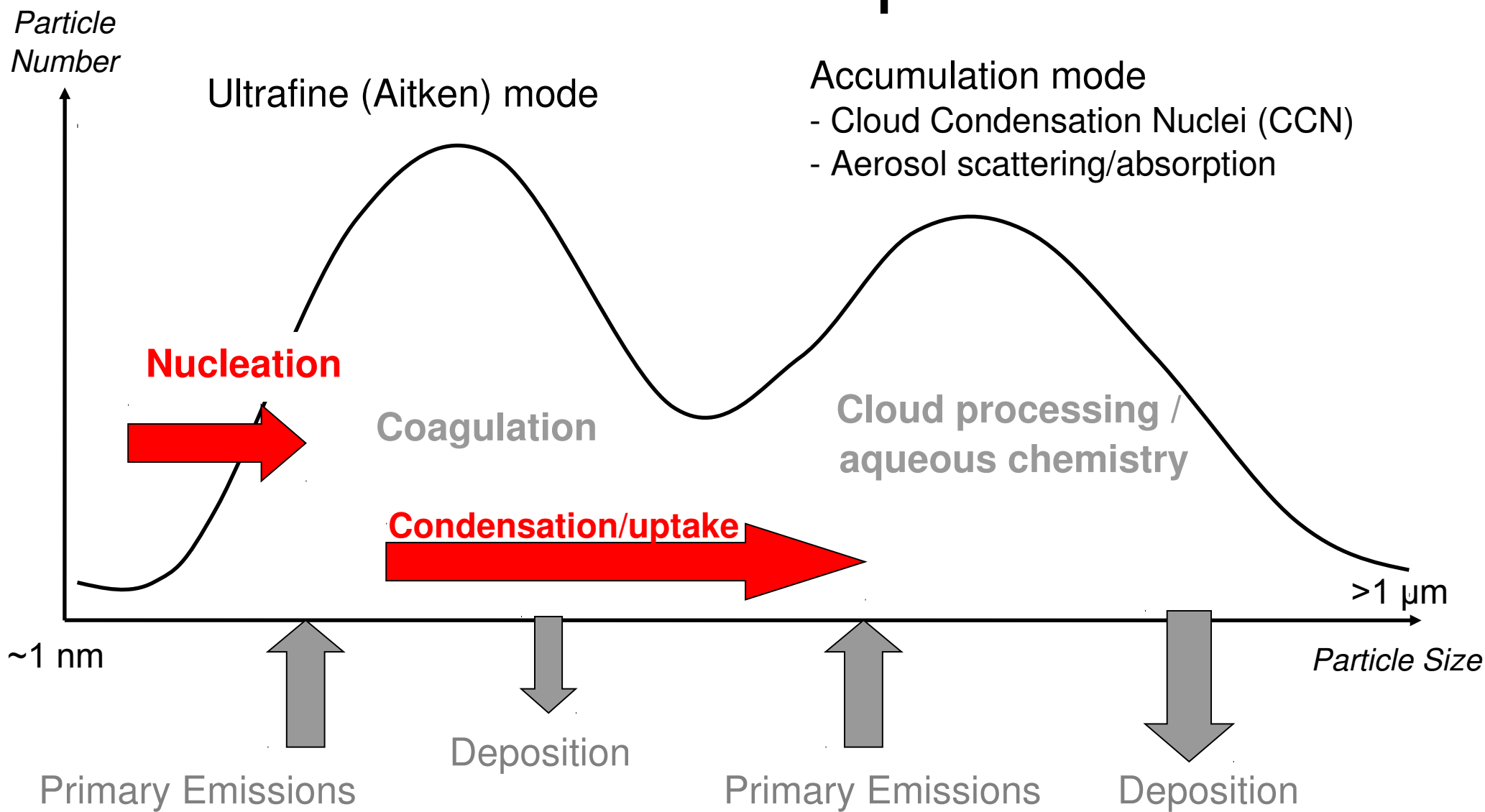


This research was supported in part by the U.S. Department of Energy's Atmospheric System Research, an Office of Science, Office of Biological and Environmental Research program, under Grant No. DE-SC0011780.

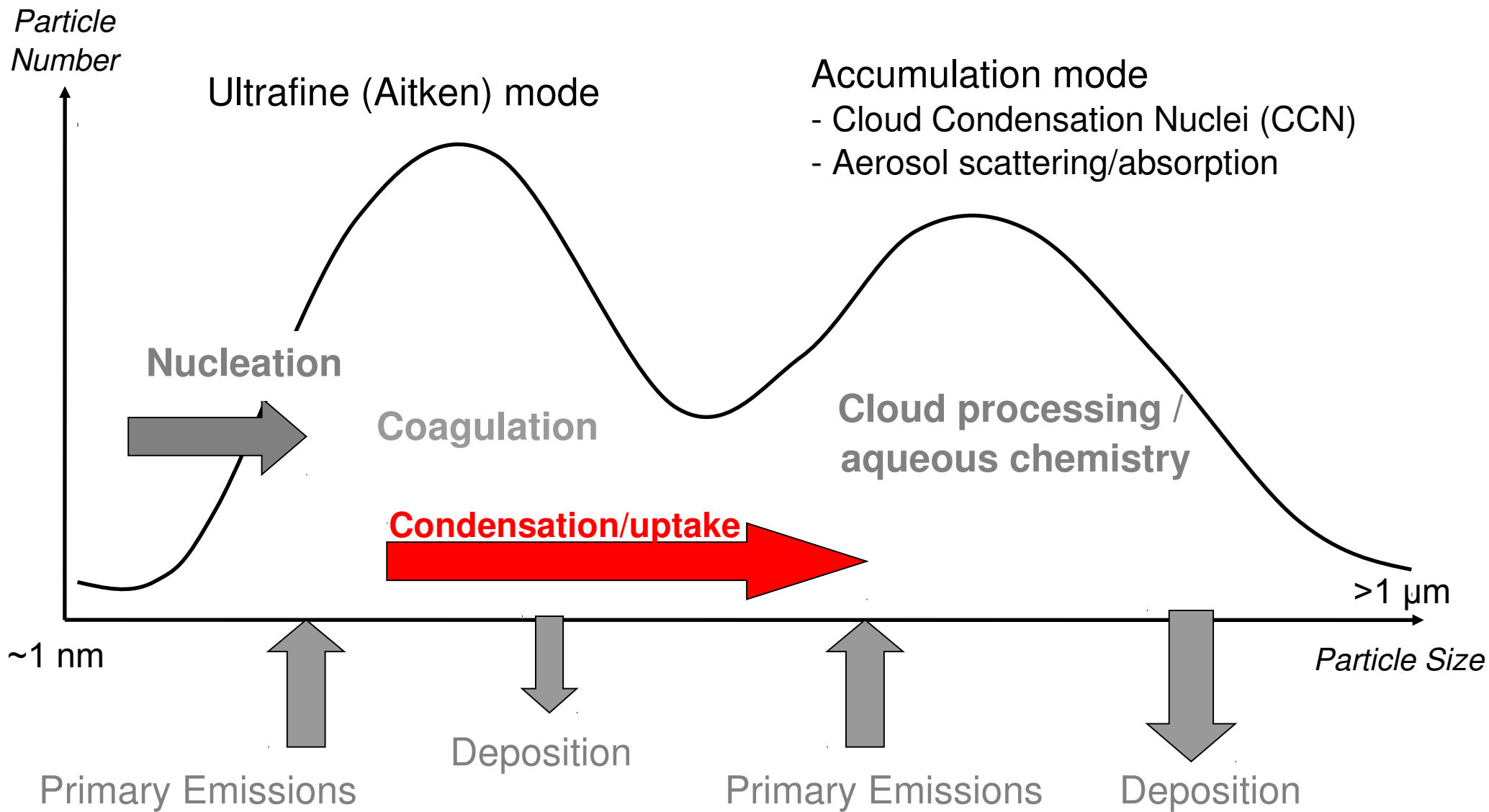
# Processes shaping aerosol size distribution



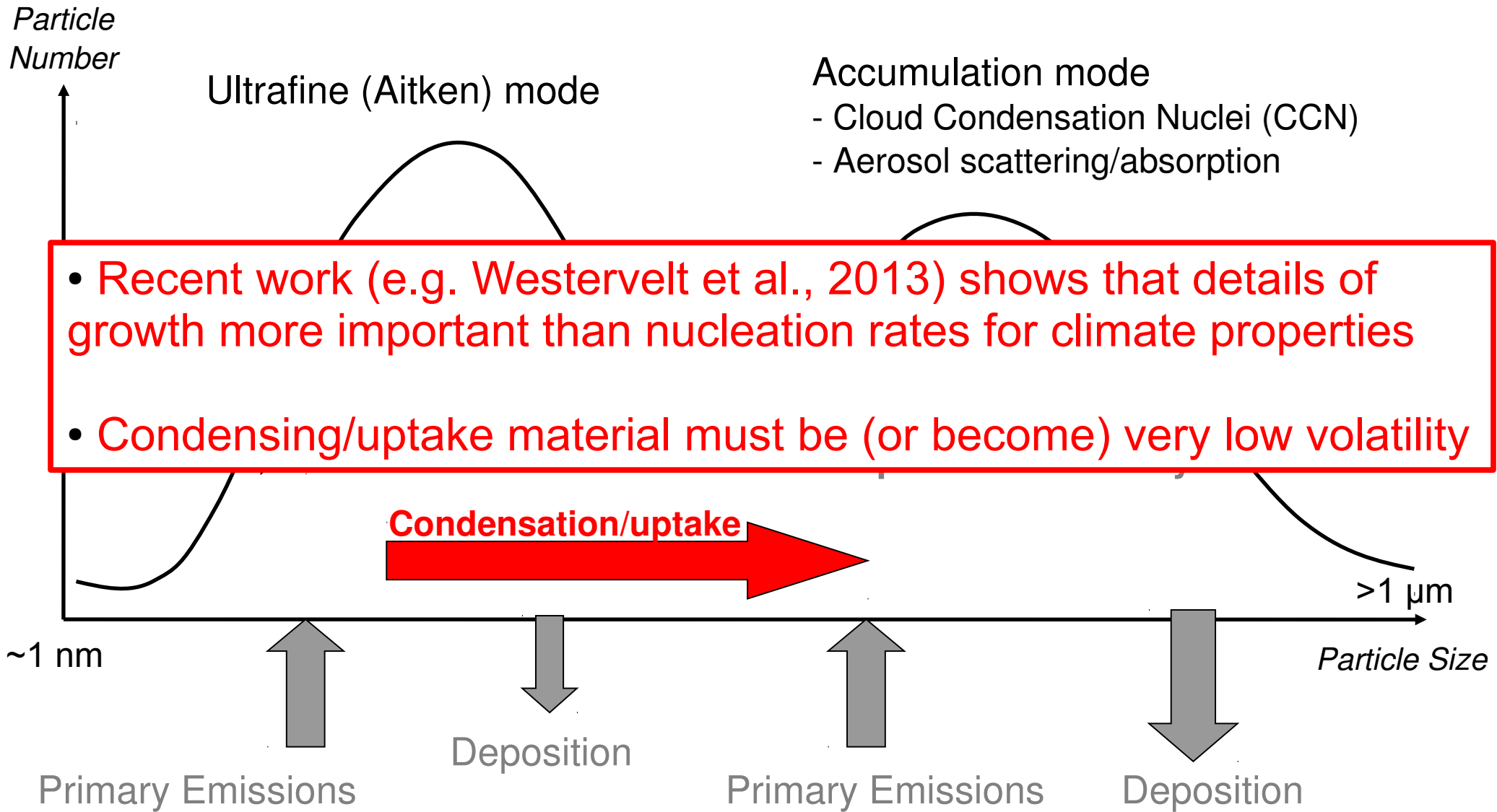
# Nucleation/growth contribute climate relevant particles



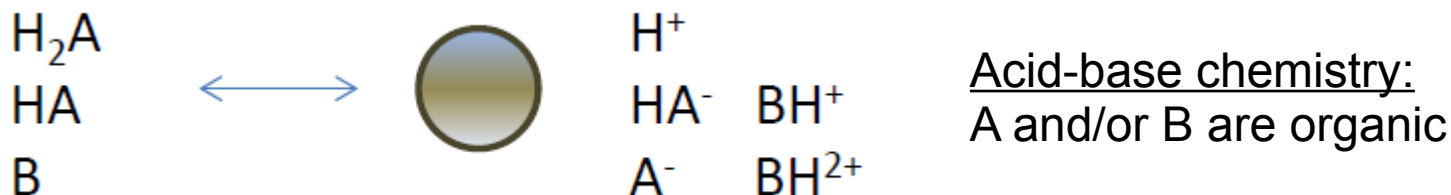
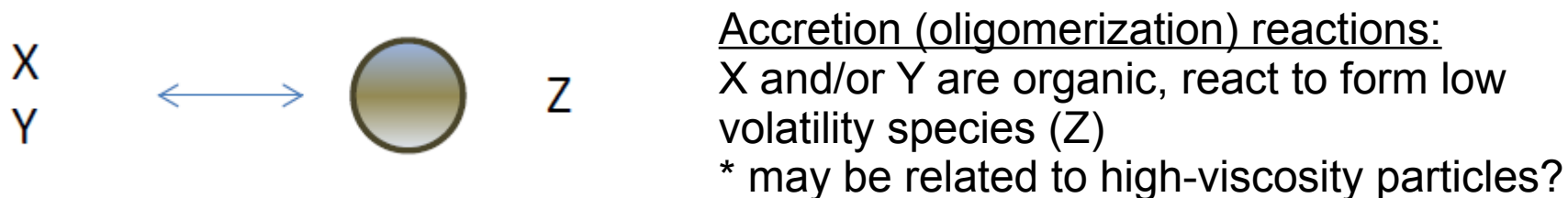
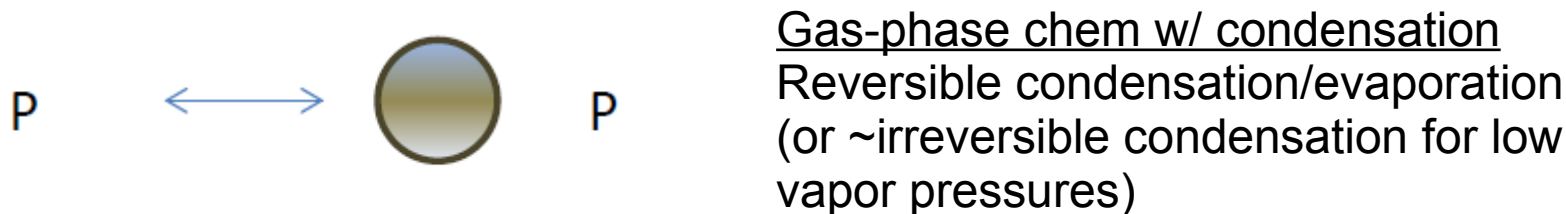
# Early growth processes less studied than nucleation



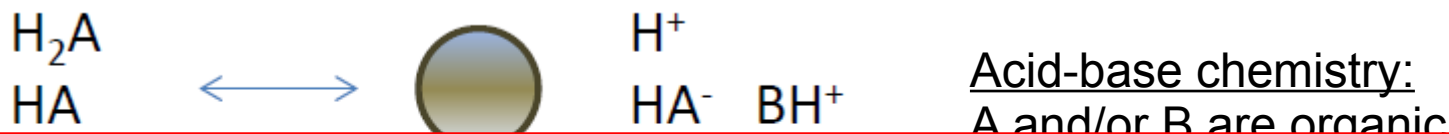
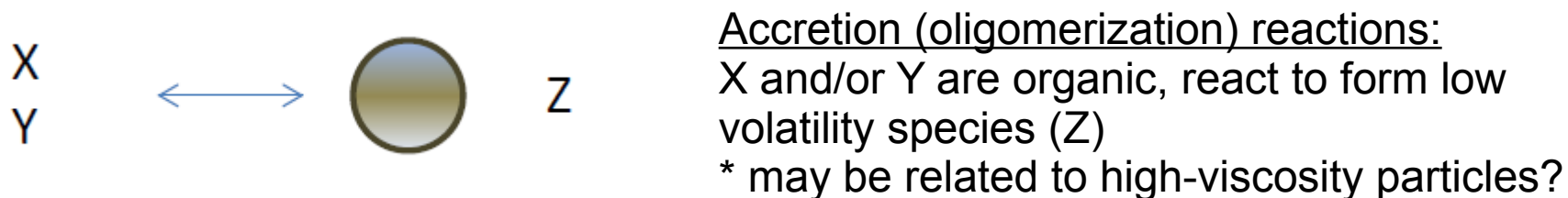
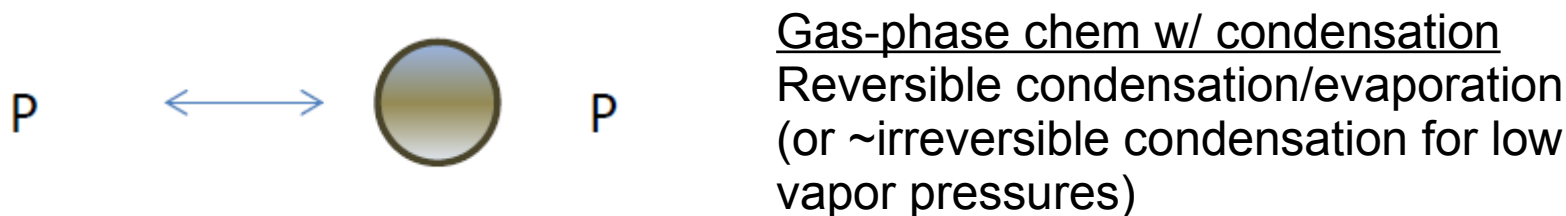
# Early growth processes less studied than nucleation



# Potential pathways of low-volatility material for growth



# Potential pathways of low-volatility material for growth



- Only “Gas-phase chem w/ condensation” explicitly included in 3D aerosol models.
- Models may fudge condensation to “correct” for lack of other processes to get growth rates right on average.
  - Will lack day-to-day variations chemical details.

# Potential pathways of low-volatility material for growth



Gas-phase chem w/ condensation  
Reversible condensation/evaporation  
(or ~irreversible condensation for low vapor pressures)



Accretion (oligomerization) reactions:  
X and/or Y are organic, react to form low volatility species (Z)  
\* may be related to high-viscosity particles?



- Only “Gas-phase chem w/ condensation” explicitly included in 3D aerosol models.
- Models may fudge condensation to “correct” for lack of other processes to get growth rates right on average.
  - Will lack day-to-day variations chemical details.
- **NEED PROCESS-BASED UNDERSTANDING FOR MODELS**



## DOE DE-SC0011780:

# Contributions of organic compounds to the growth of freshly nucleated atmospheric nanoparticles

- Goals

- Analysis of formation/growth during DOE ASR: “Southern Great Plains – New Particle Formation Study (NPFS)”
- Controlled laboratory studies of growth mechanisms
- Parameterize growth for aerosol models
- Test in regional/global aerosol models

# DOE DE-SC0011780:

## Contributions of organic compounds to the growth of freshly nucleated atmospheric nanoparticles

- Goals

Current focus (in first year of work)

- Analysis of formation/growth during DOE ASR: “Southern Great Plains – New Particle Formation Study (NPFS)”
- Controlled laboratory studies of growth mechanisms
- Parameterize growth for aerosol models
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# DOE DE-SC0011780:

## Contributions of organic compounds to the growth of freshly nucleated atmospheric nanoparticles

- Goals

This talk

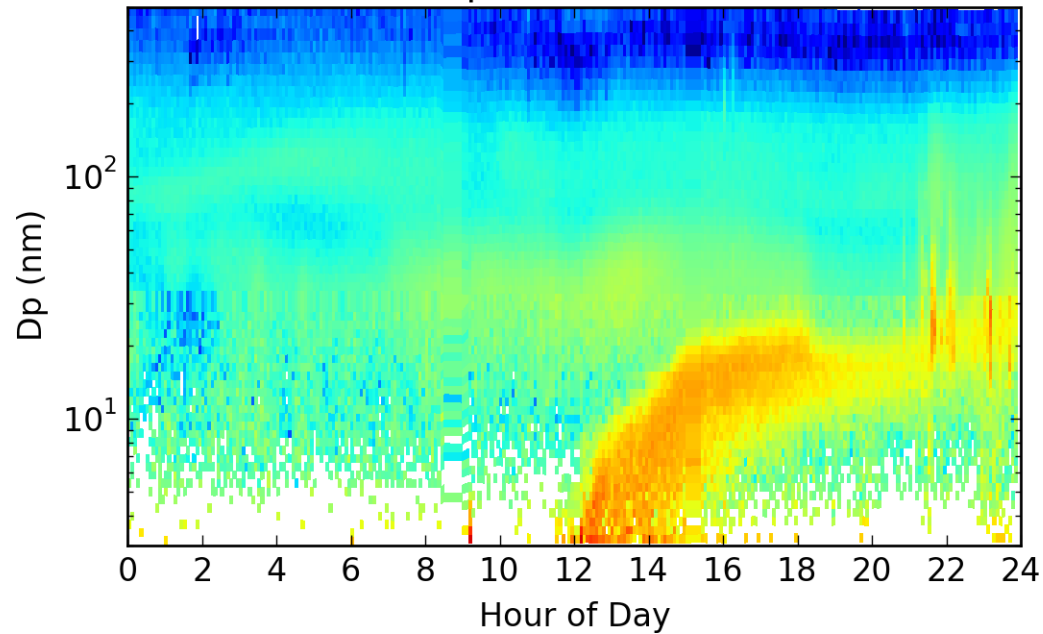
- Analysis of formation/growth during DOE ASR: “Southern Great Plains – New Particle Formation Study (NPFS)”
- Controlled laboratory studies of growth mechanisms
- Parameterize growth for aerosol models
- Test in regional/global aerosol models

# SGP New Particle Formation Study (NPFS)

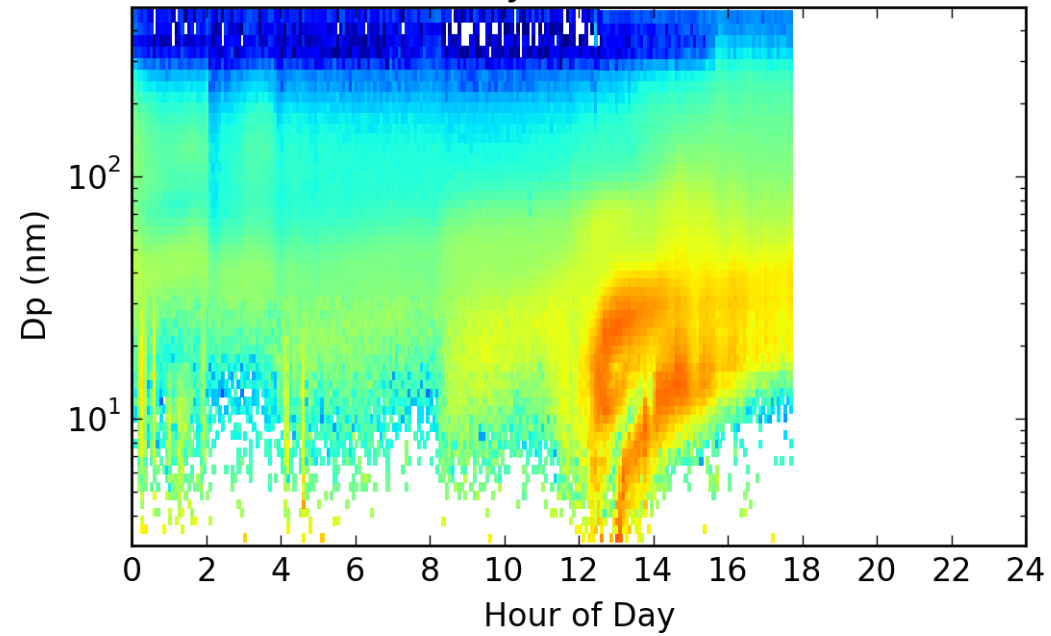
- April - May 2013: 18 observed NPF events
- Measurements in addition to core ARM observations (UMN, Augsburg, NCAR, UDEL)
  - Nanoparticle composition (TDCIMS and NAMS)
  - Particle size distribution: 1.5 nm to 1  $\mu\text{m}$  (SMPS + DEG-CPC)
  - Size-resolved nanoparticle volatility and hygroscopicity (UV/UHTDMA)
  - Aerosol gas-phase precursors (ammonia, amines, sulfuric acid; CIMS)
  - Vertically resolved aerosol number concentrations (dual-CPCs on tethered balloon)

# A tale of 3 growth events

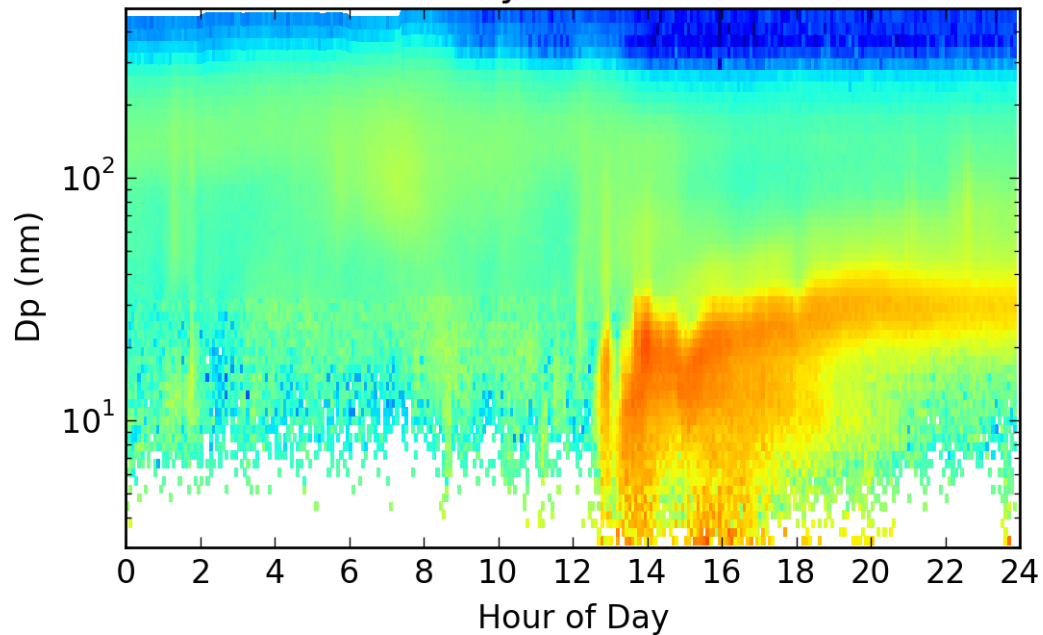
April 19, 2013



May 09, 2013

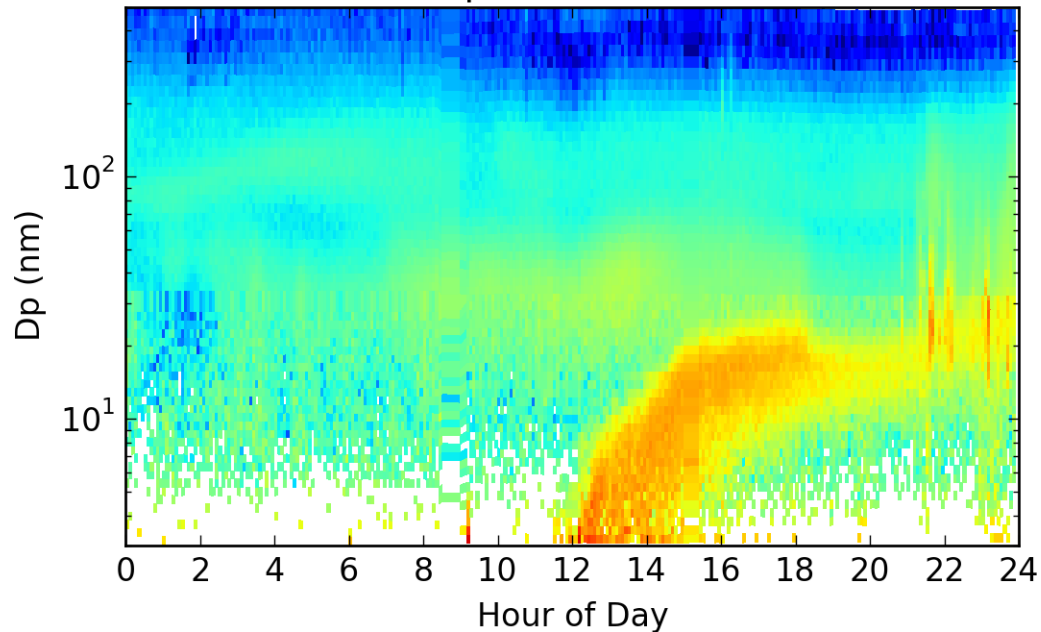


May 11, 2013

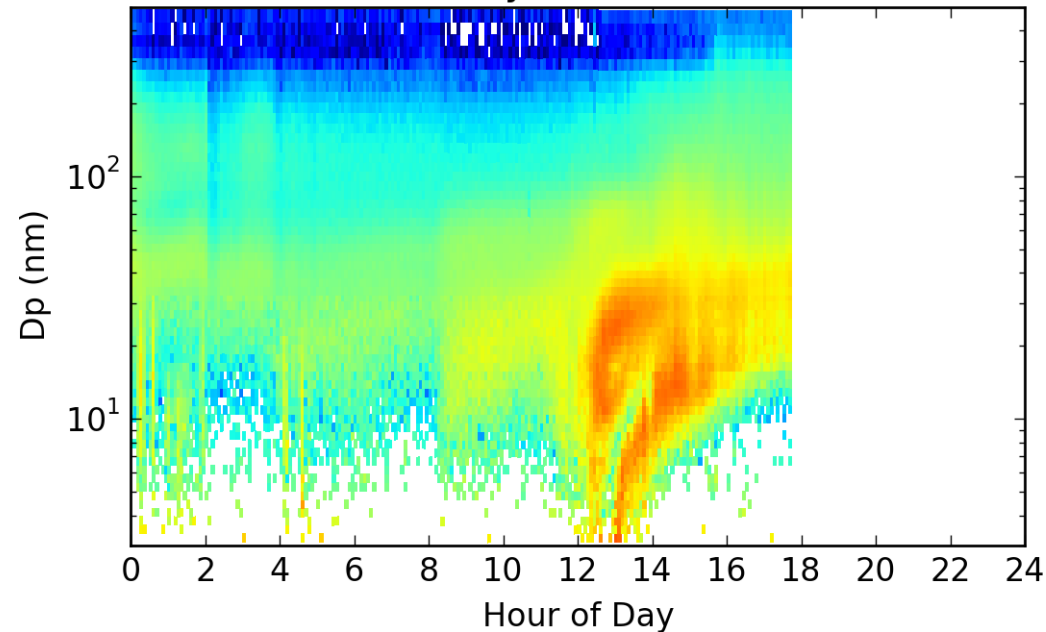


# A tale of 3 growth events

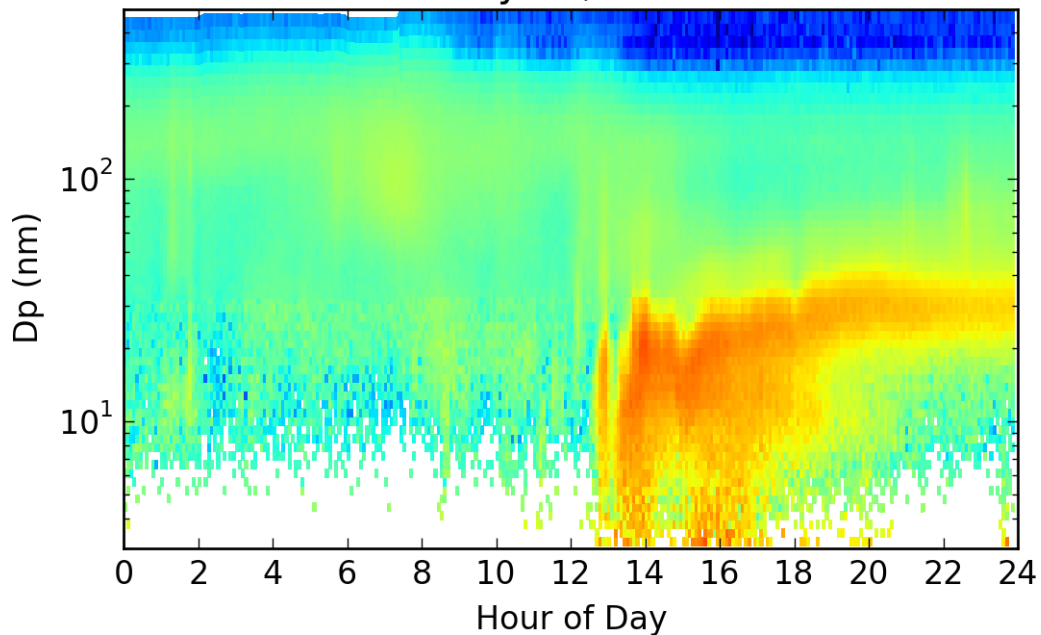
April 19, 2013



May 09, 2013



May 11, 2013

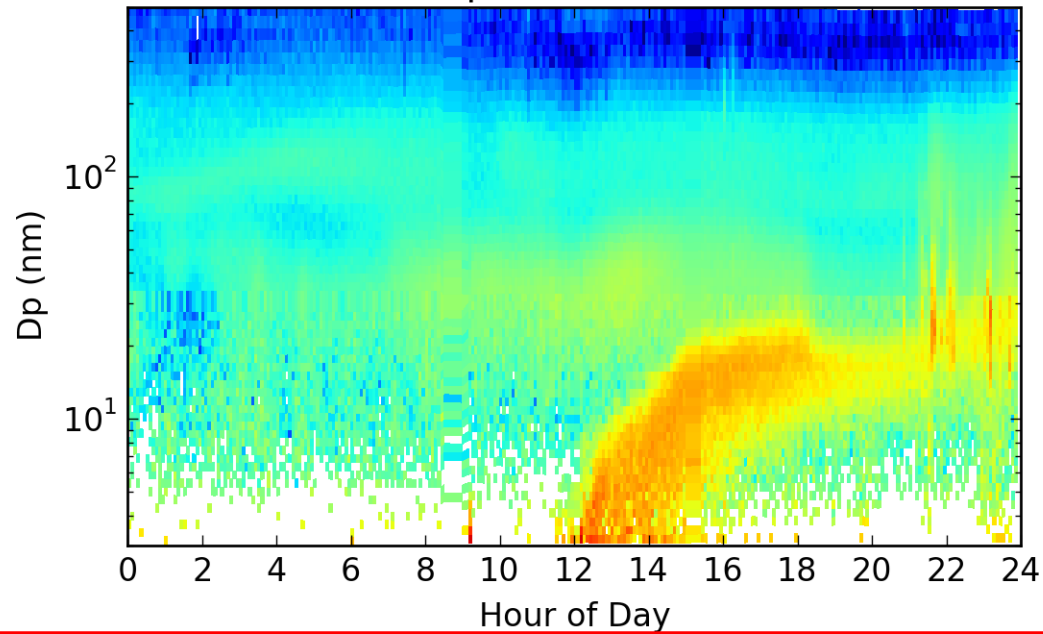


From size distributions alone, events look qualitatively similar in many ways.

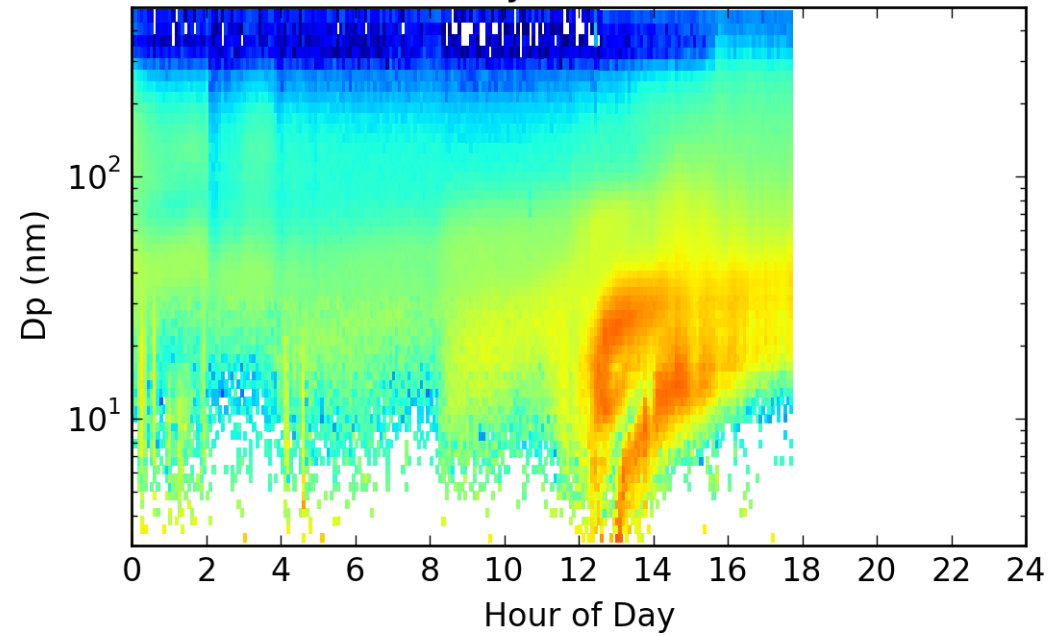
Only with the combinations of instruments available during SGP-NPFS we learn that growth on each day was through a different pathway.

# May 11

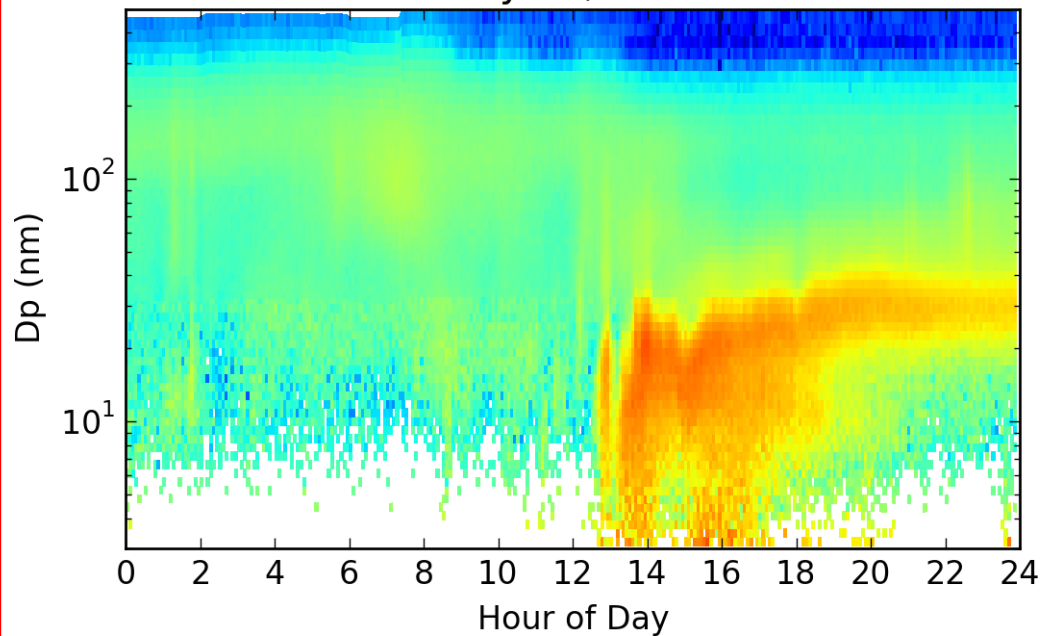
April 19, 2013



May 09, 2013



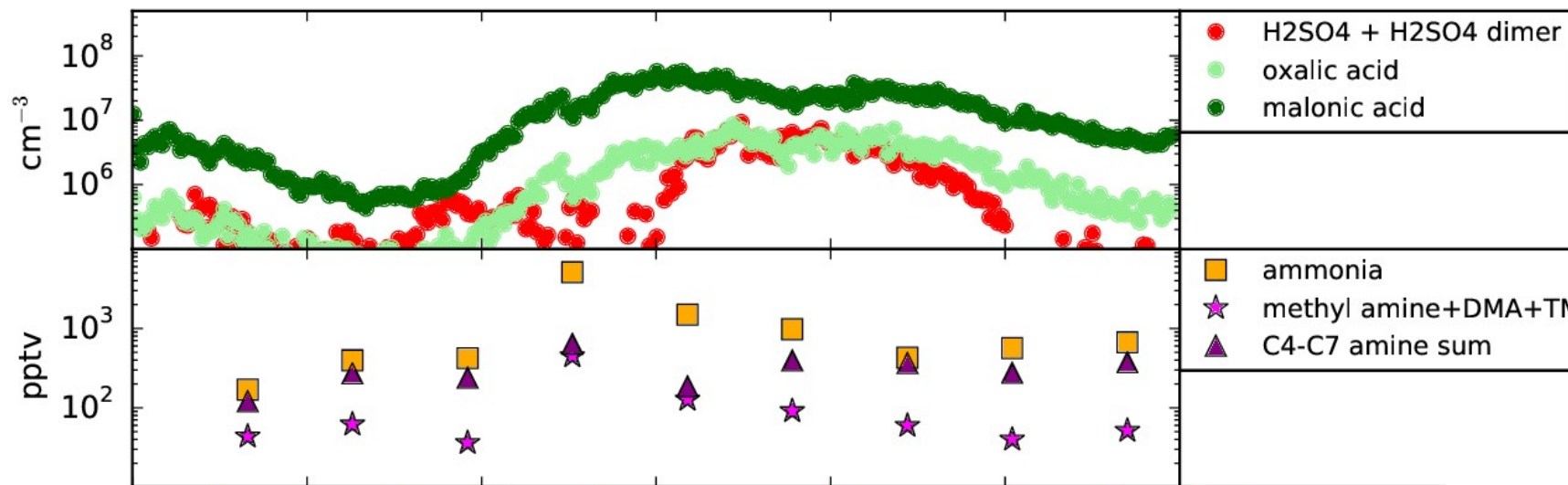
May 11, 2013



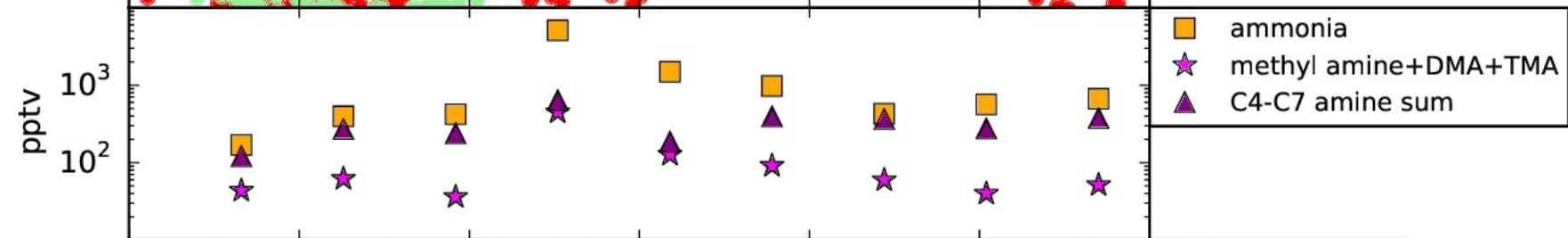
# May 11, 2013: Growth by sulfuric-acid/amines/organics

5.11.13

Gas-phase acids



Gas-phase bases

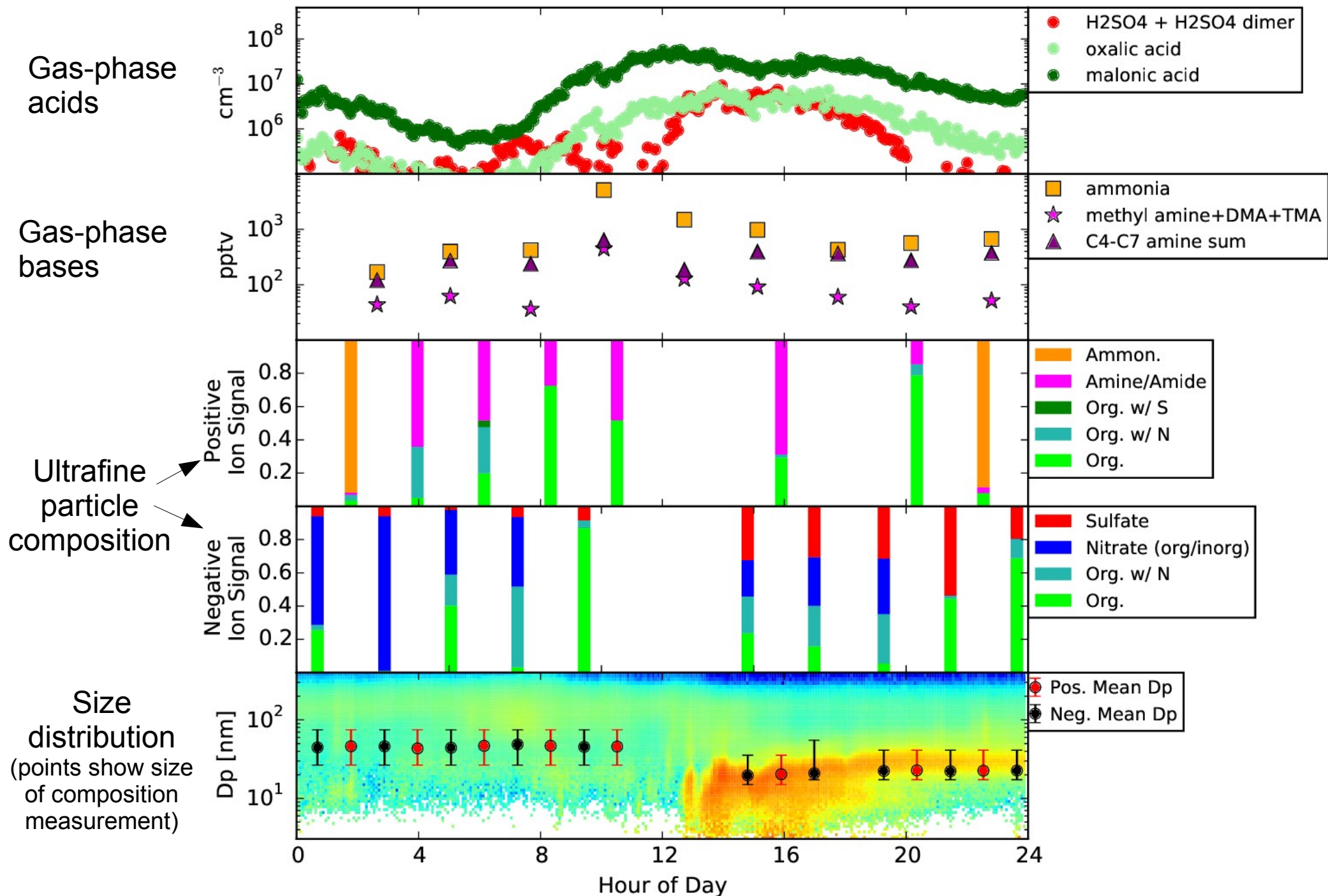


0 4 8 12 16 20 24  
Hour of Day



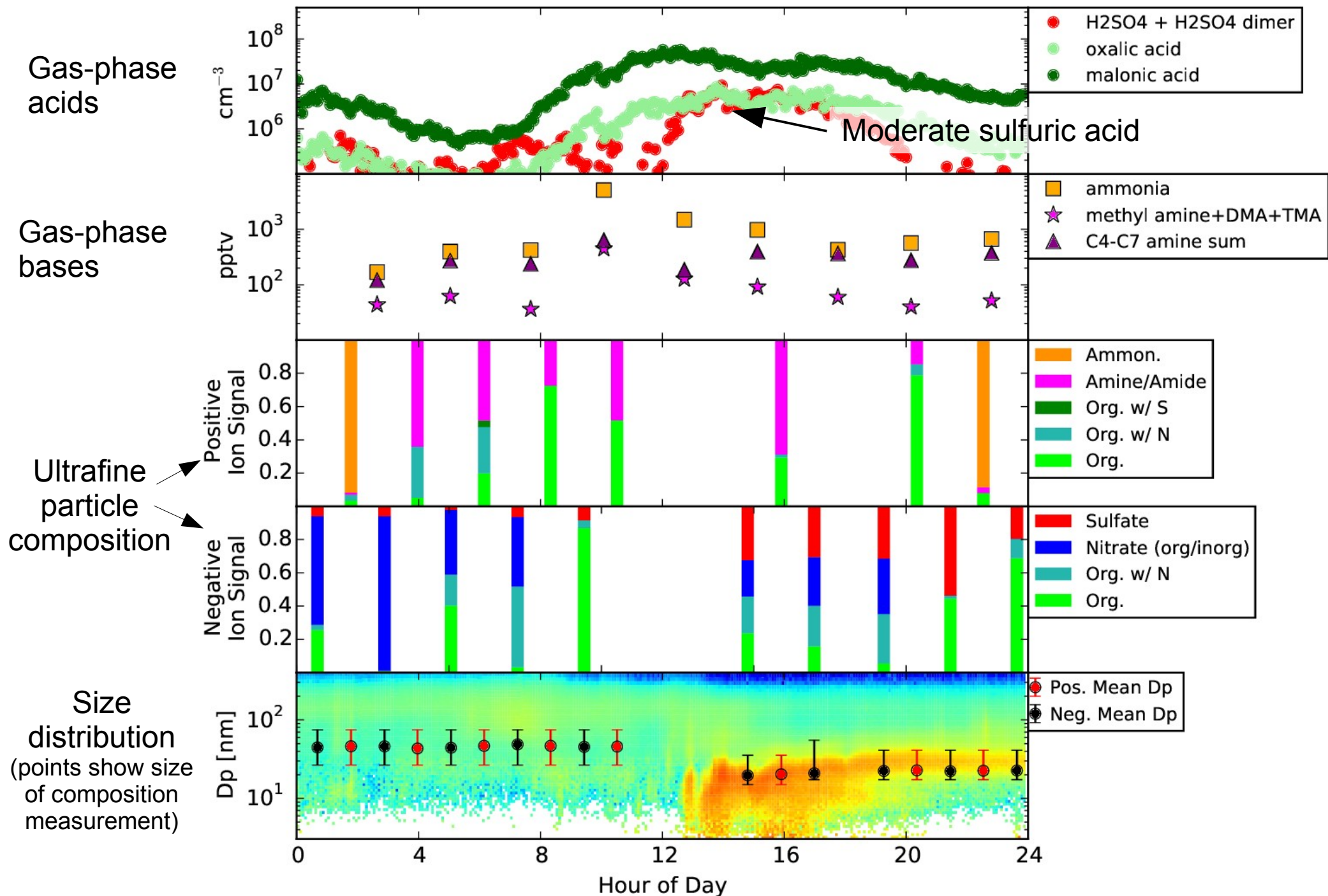
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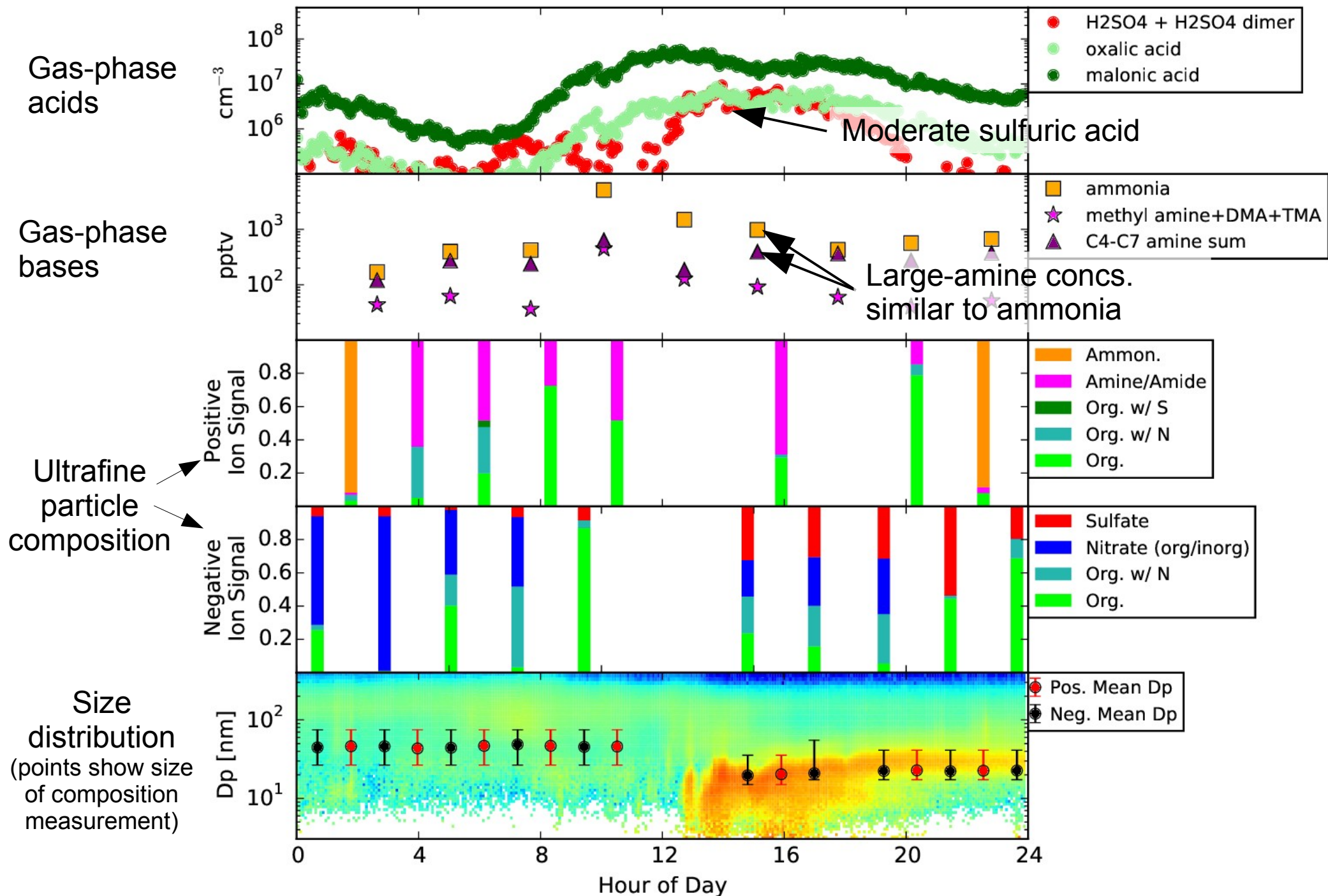
# May 11, 2015: Growth by sulfuric-acid/amines/organics

5.11.13



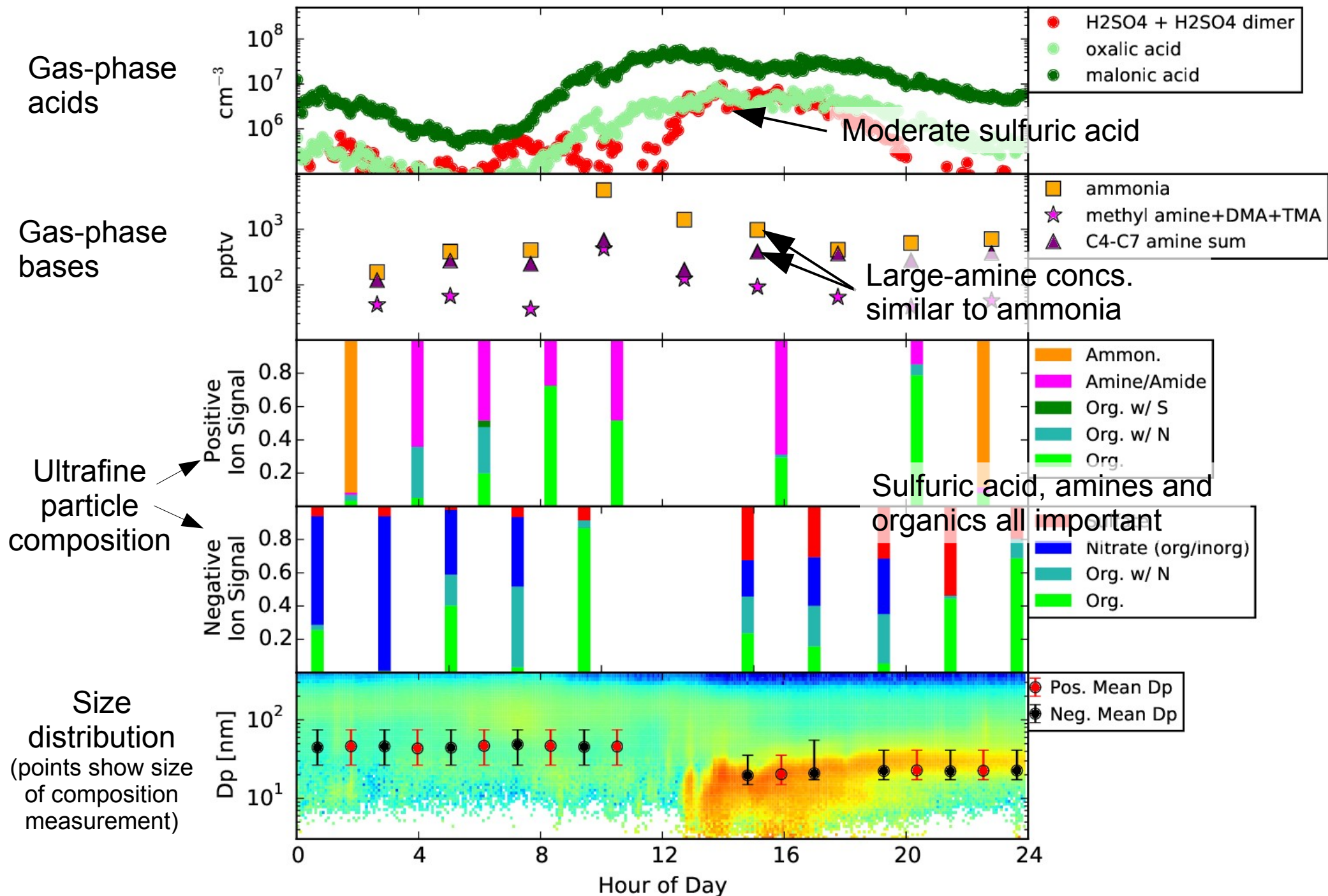
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5.11.13



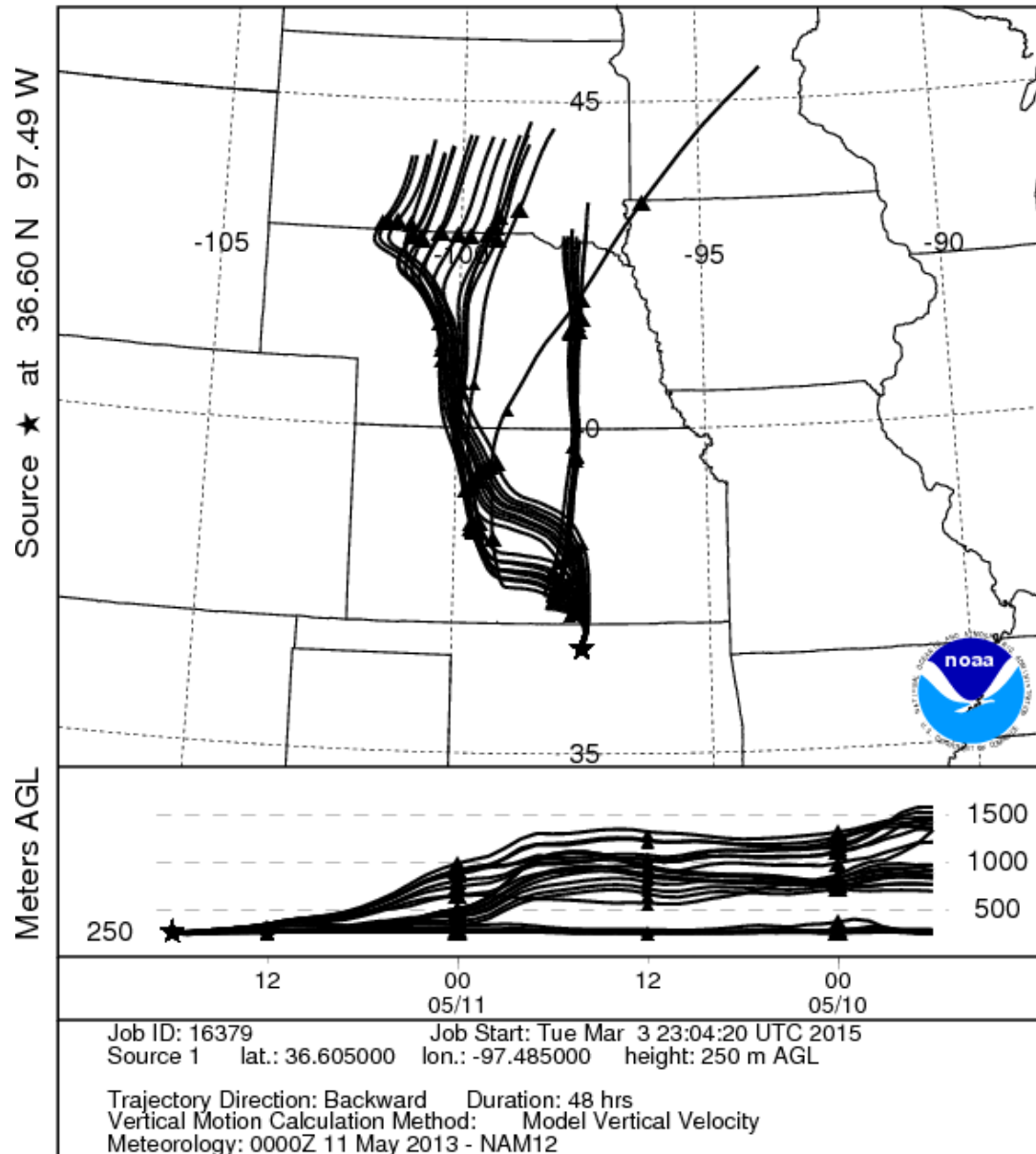
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5.11.13



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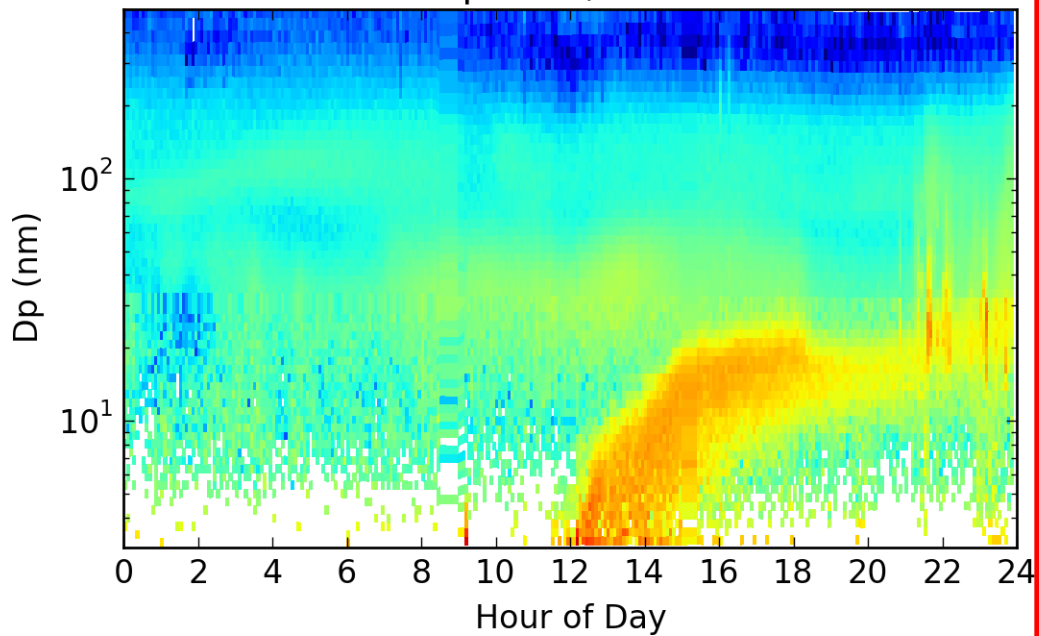
NOAA HYSPLIT MODEL  
Backward trajectories ending at 1800 UTC 11 May 13  
NAM Meteorological Data



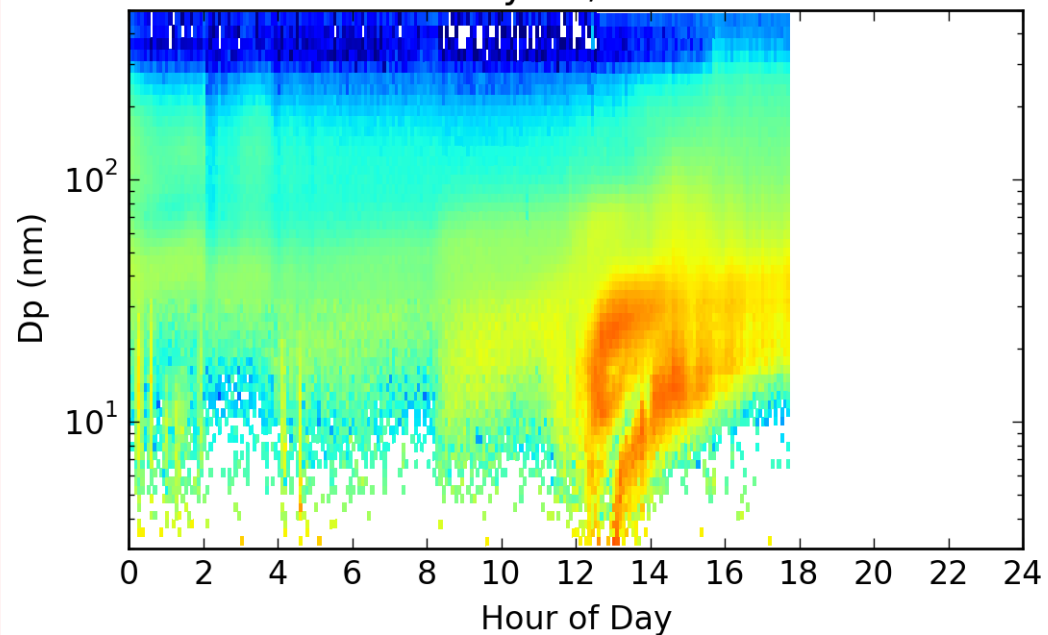
Flow from North:  
No major urban areas  
Agriculture

# April 19

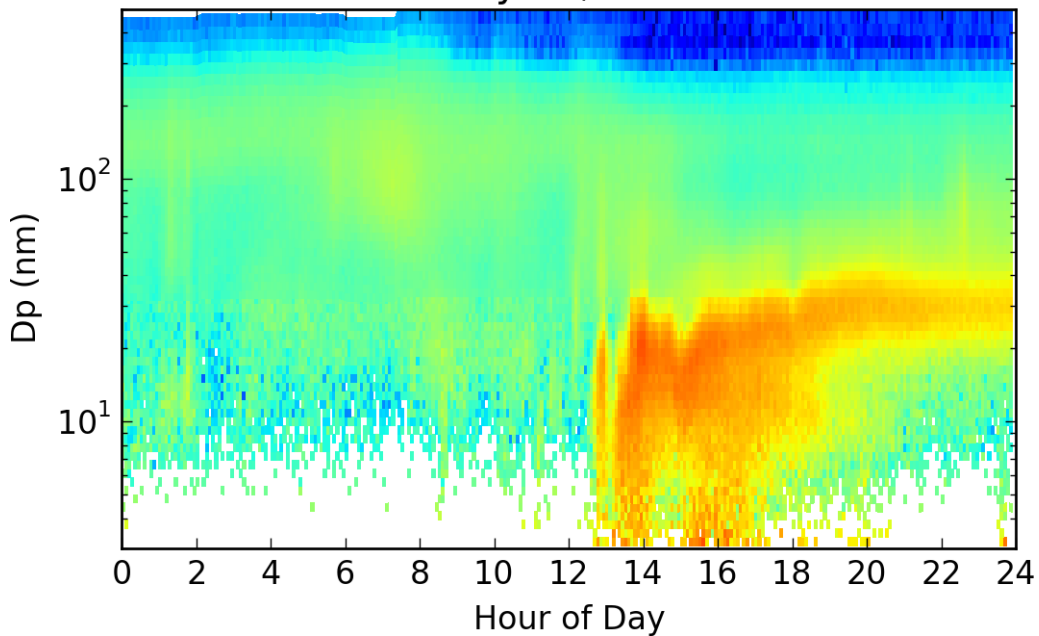
April 19, 2013



May 09, 2013

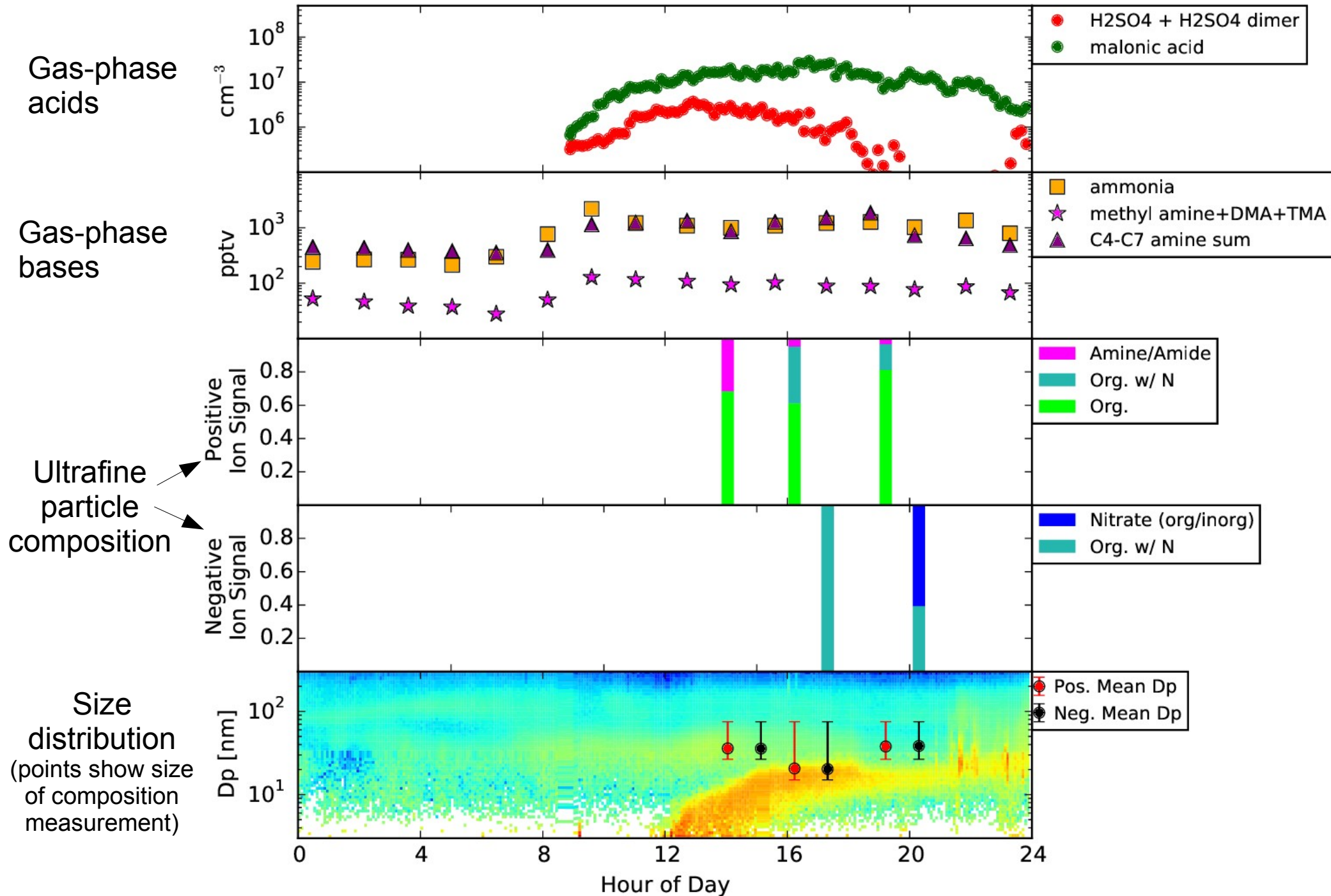


May 11, 2013



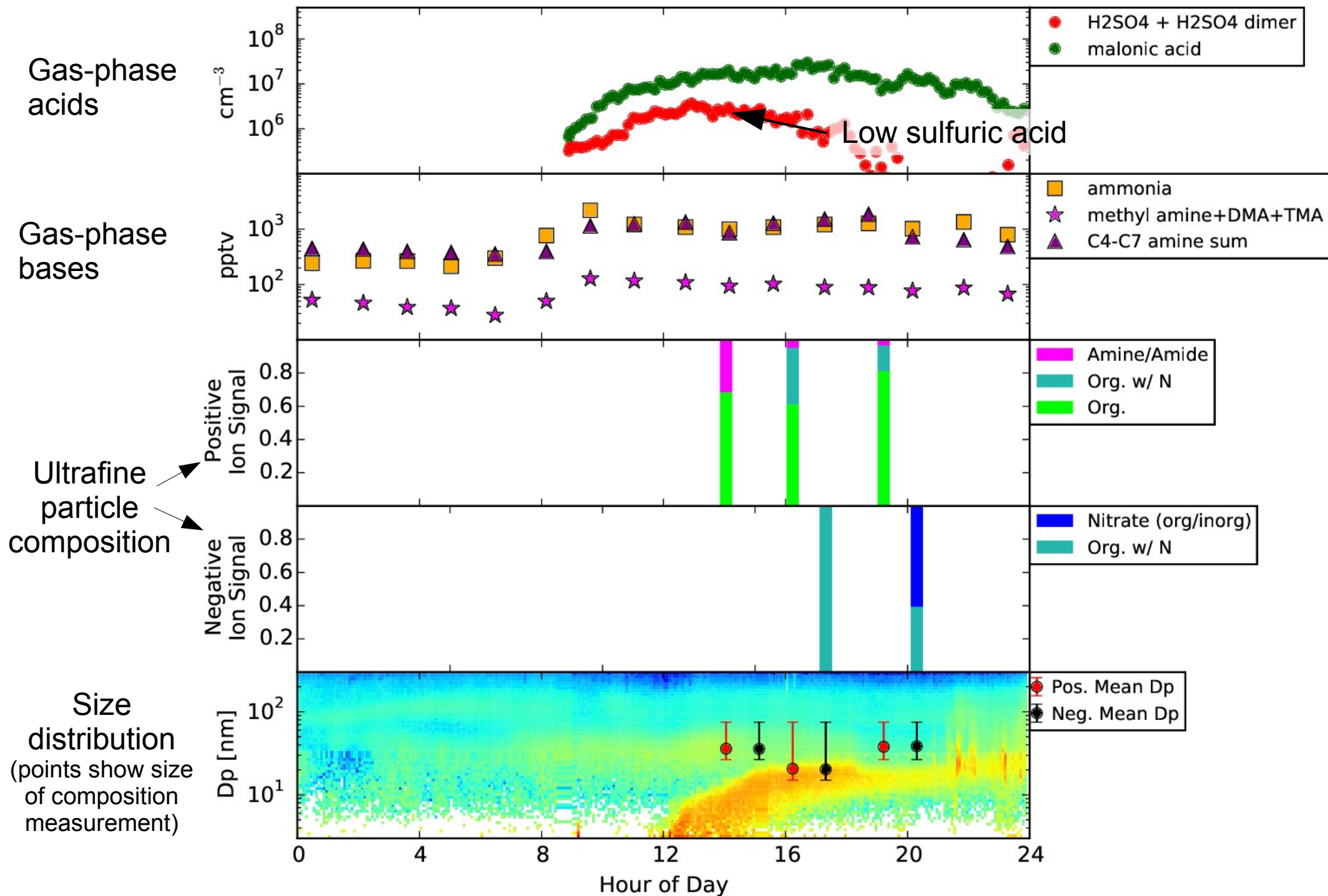
# April 19, 2013: Growth by organics

4.19.13



# April 19, 2013: Growth by organics

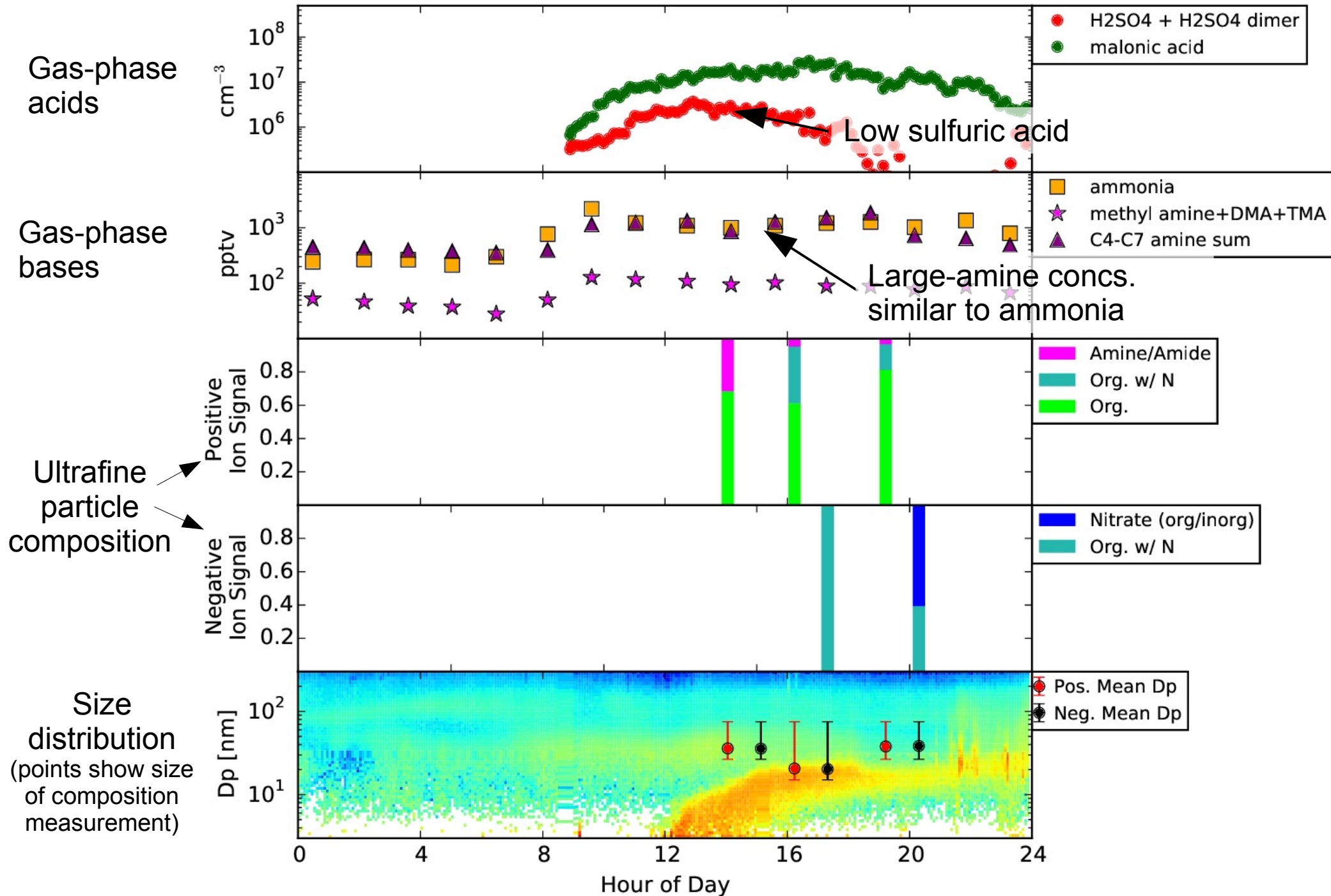
4.19.13





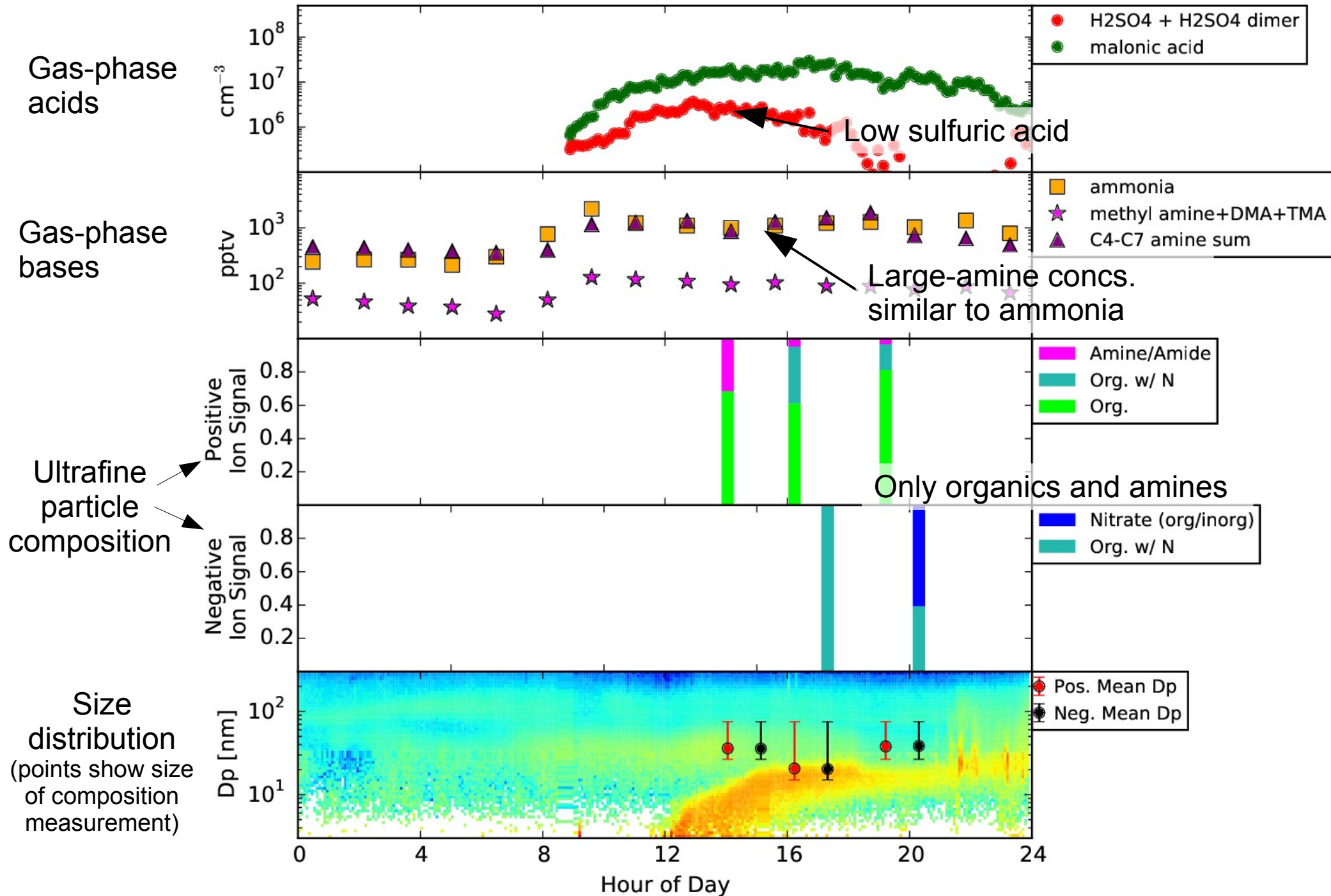
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4.19.13



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4.19.13

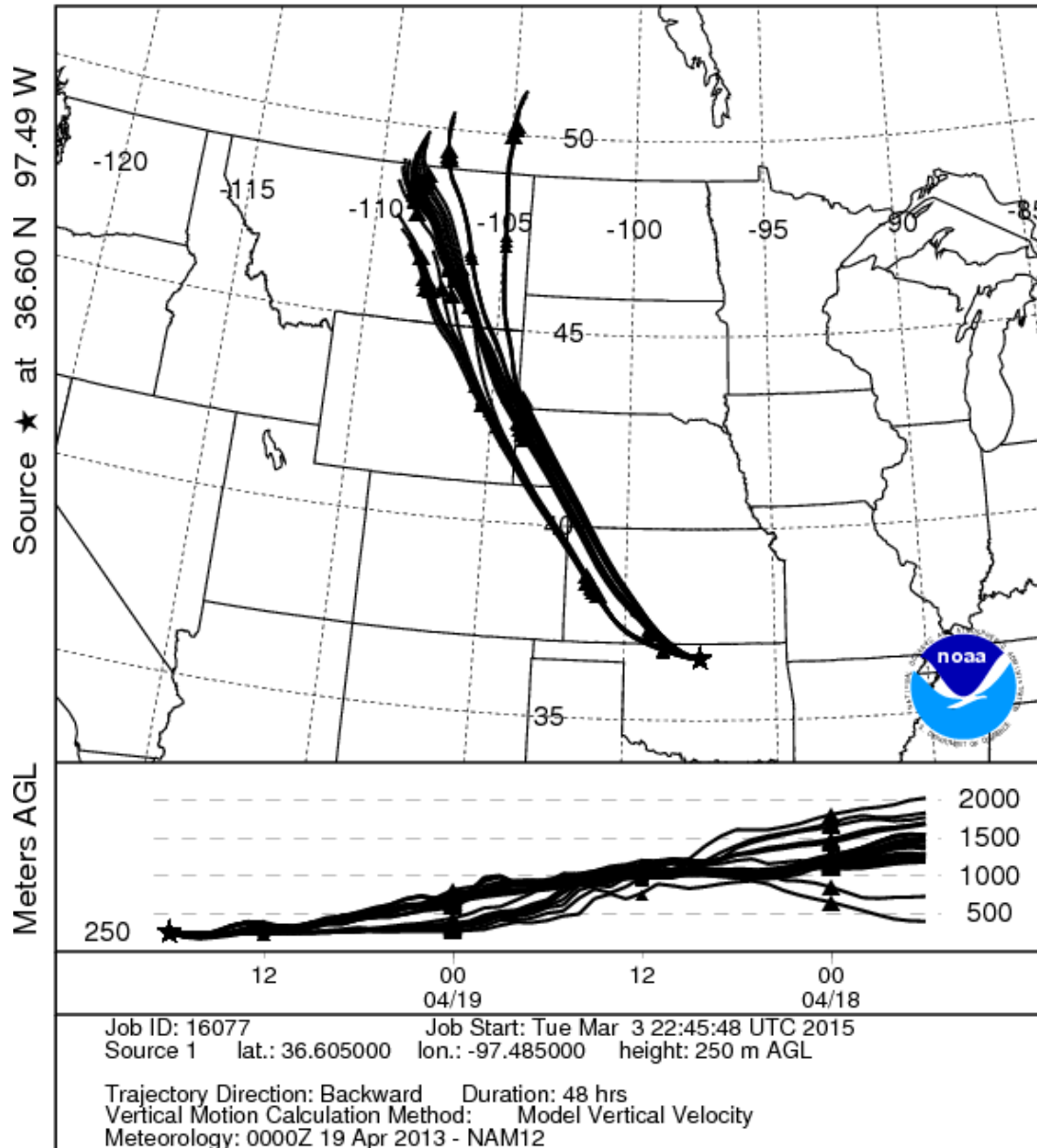


# April 19, 2013: Growth by organics

NOAA HYSPLIT MODEL

Backward trajectories ending at 1800 UTC 19 Apr 13

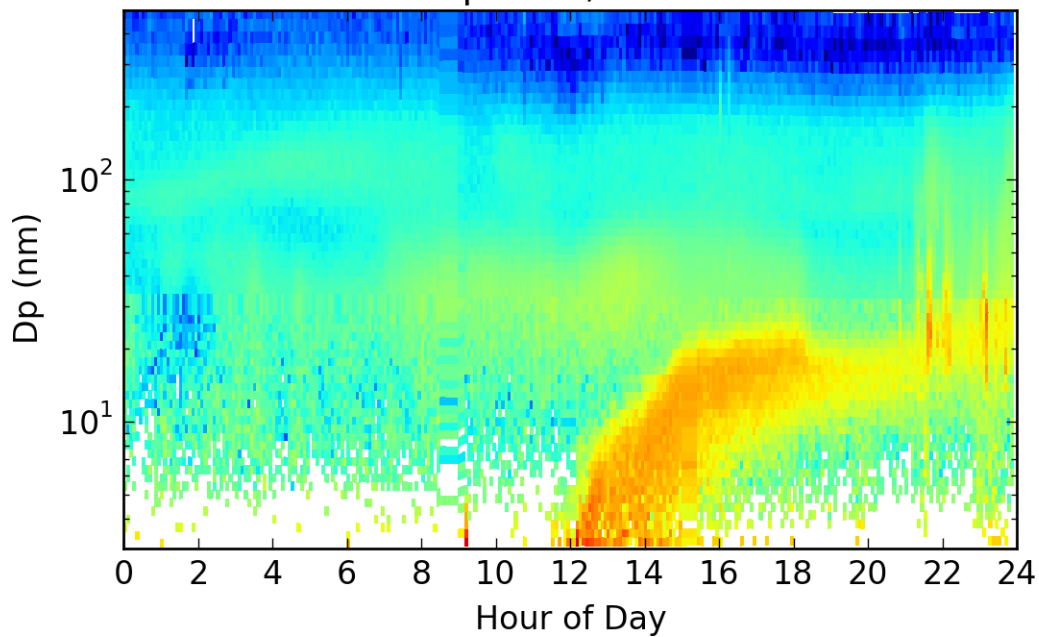
NAM Meteorological Data



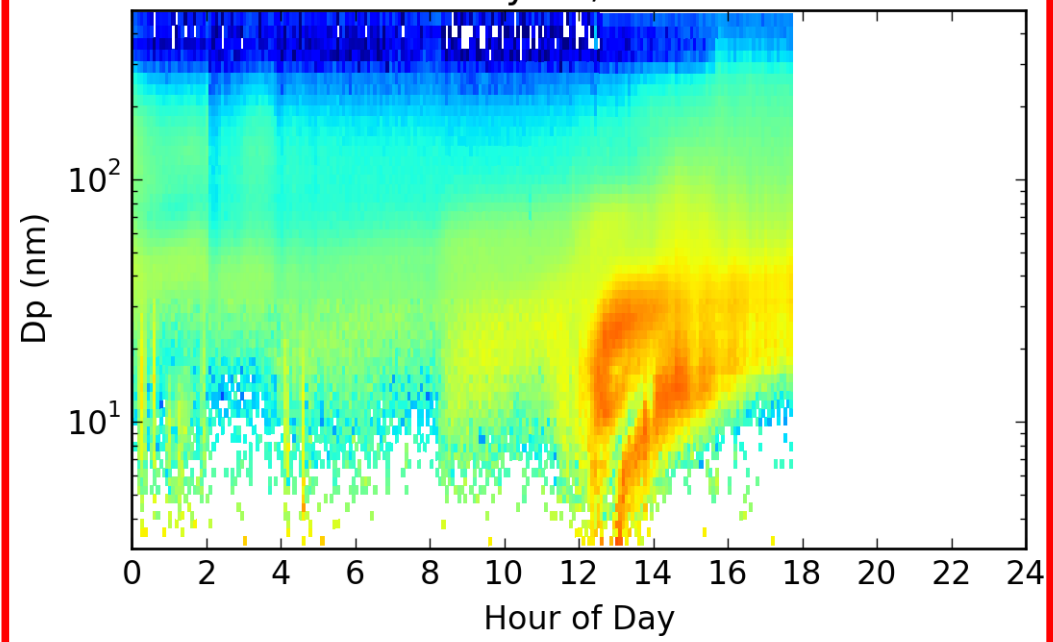
Flow from North:  
No major urban areas  
Agriculture

# May 9

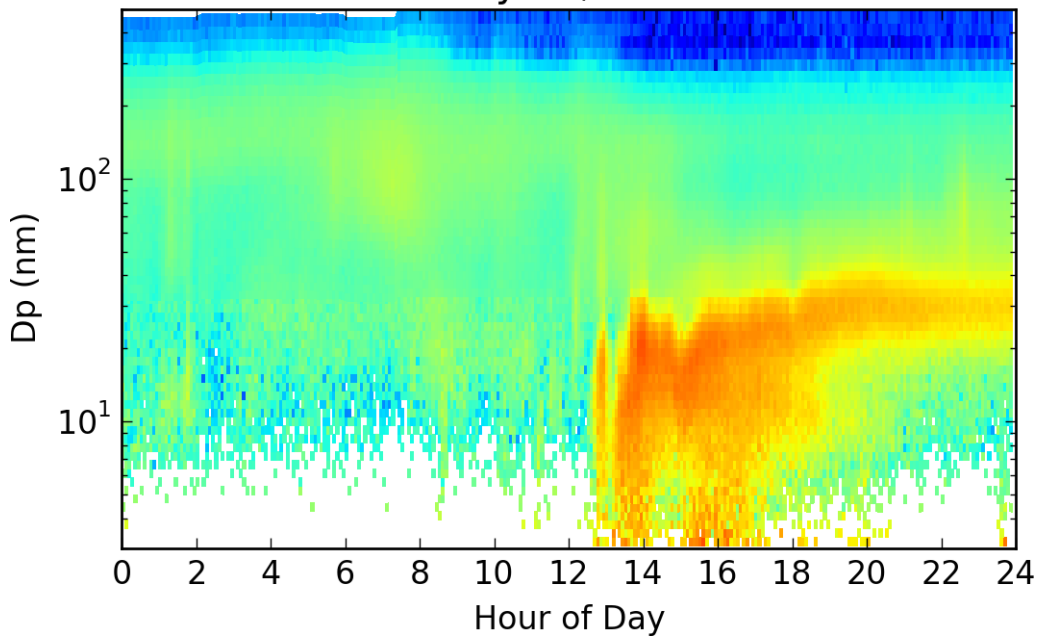
April 19, 2013



May 09, 2013

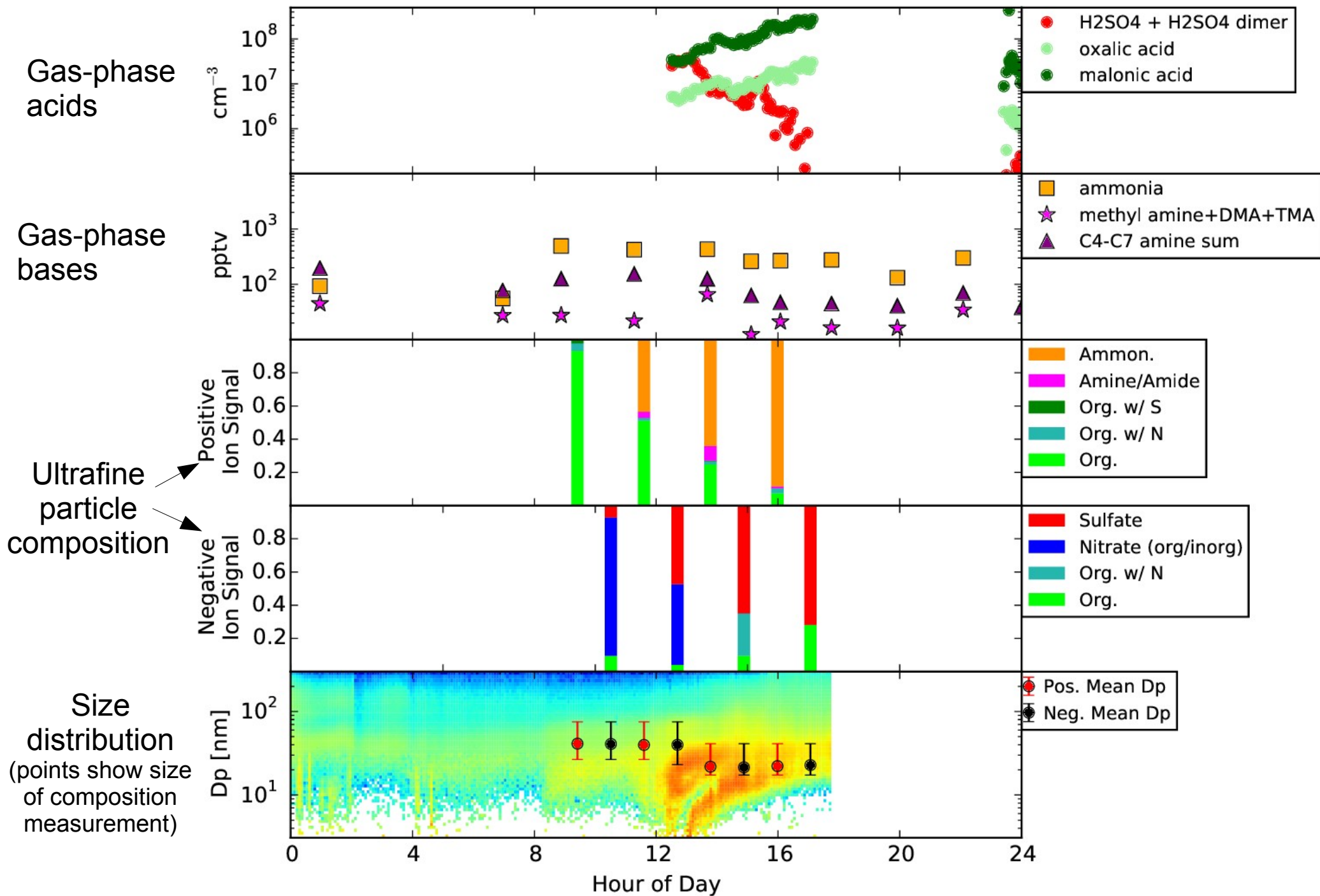


May 11, 2013



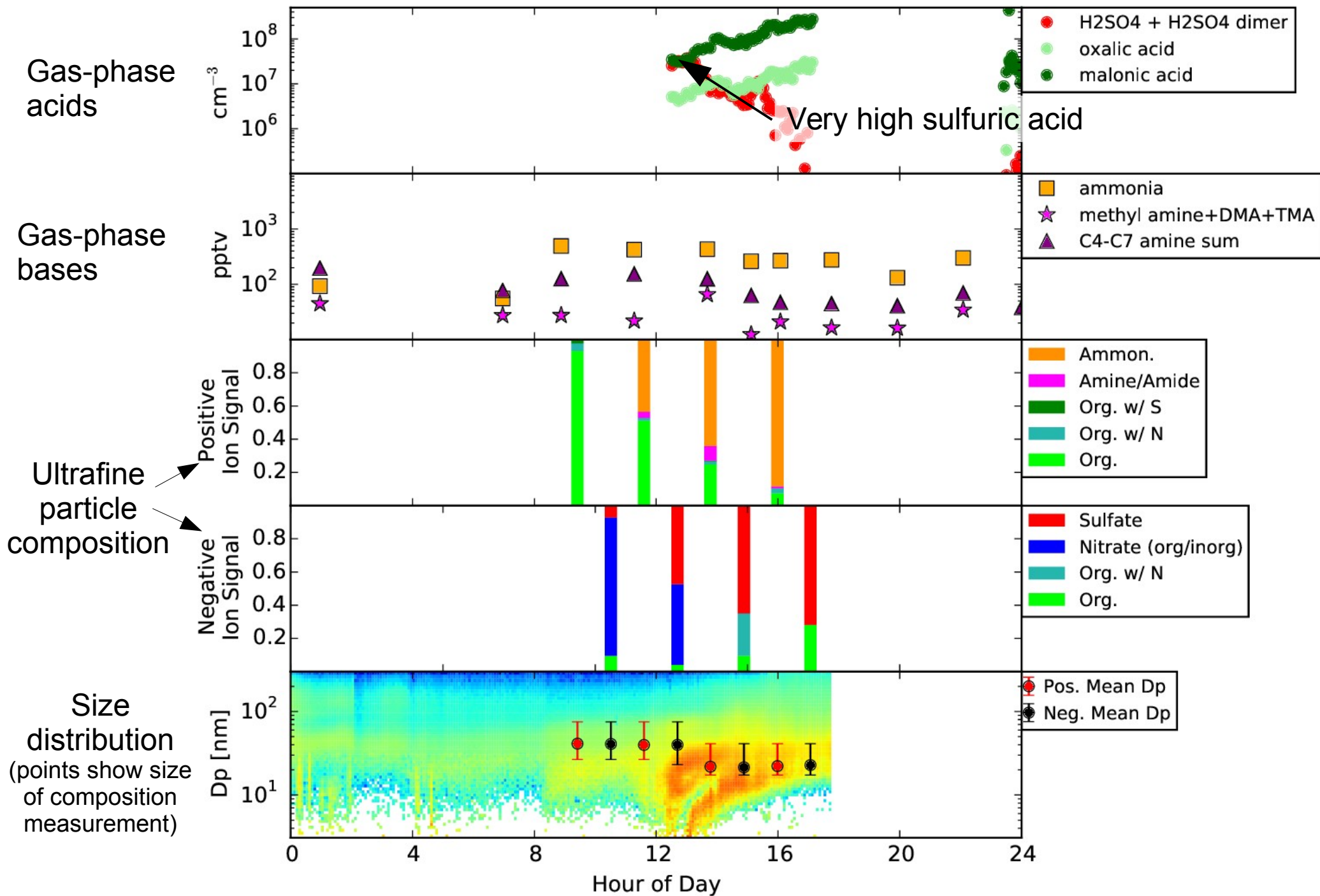
# May 9, 2013: Growth by sulfuric-acid/ammonia

5.9.13



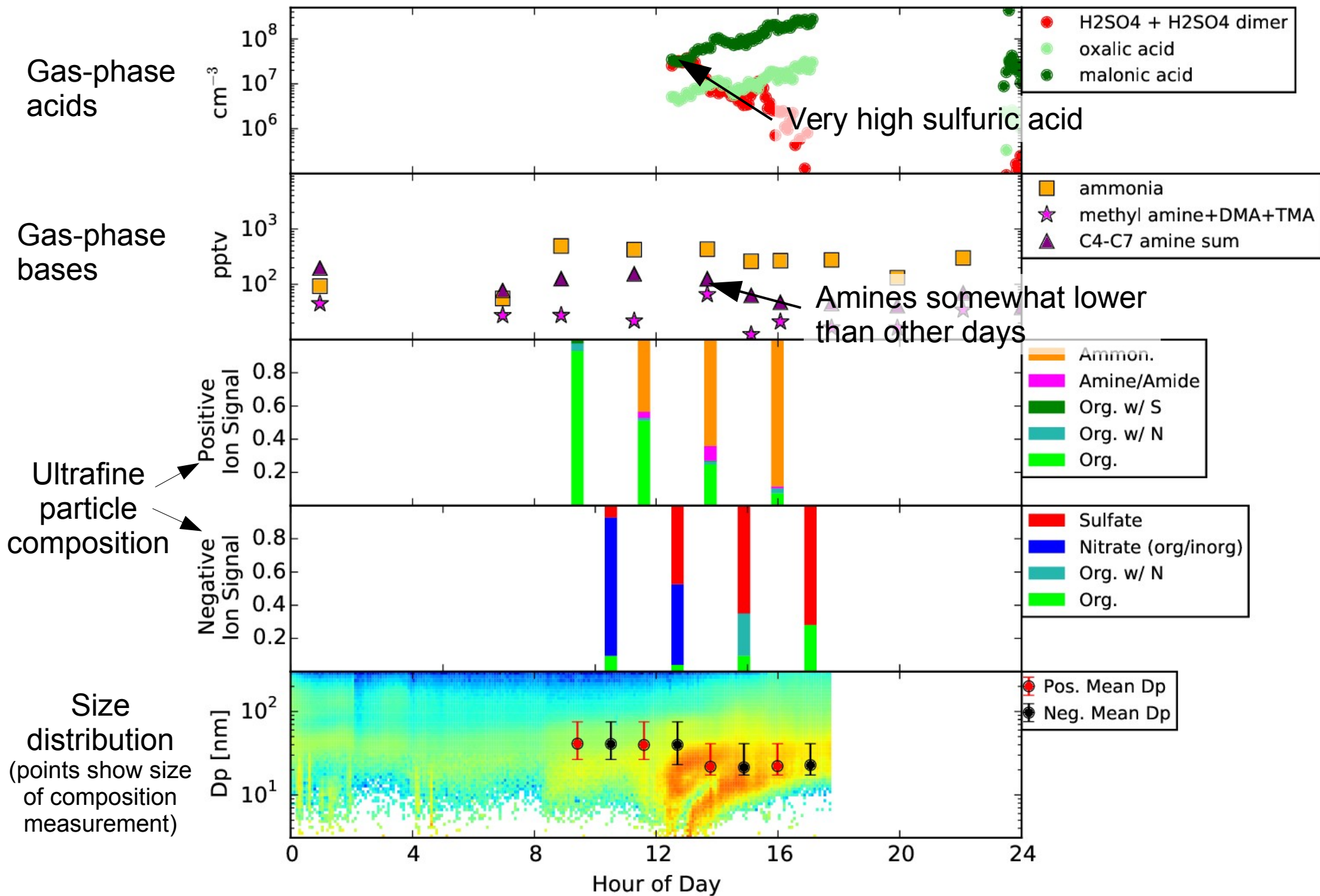
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5.9.13



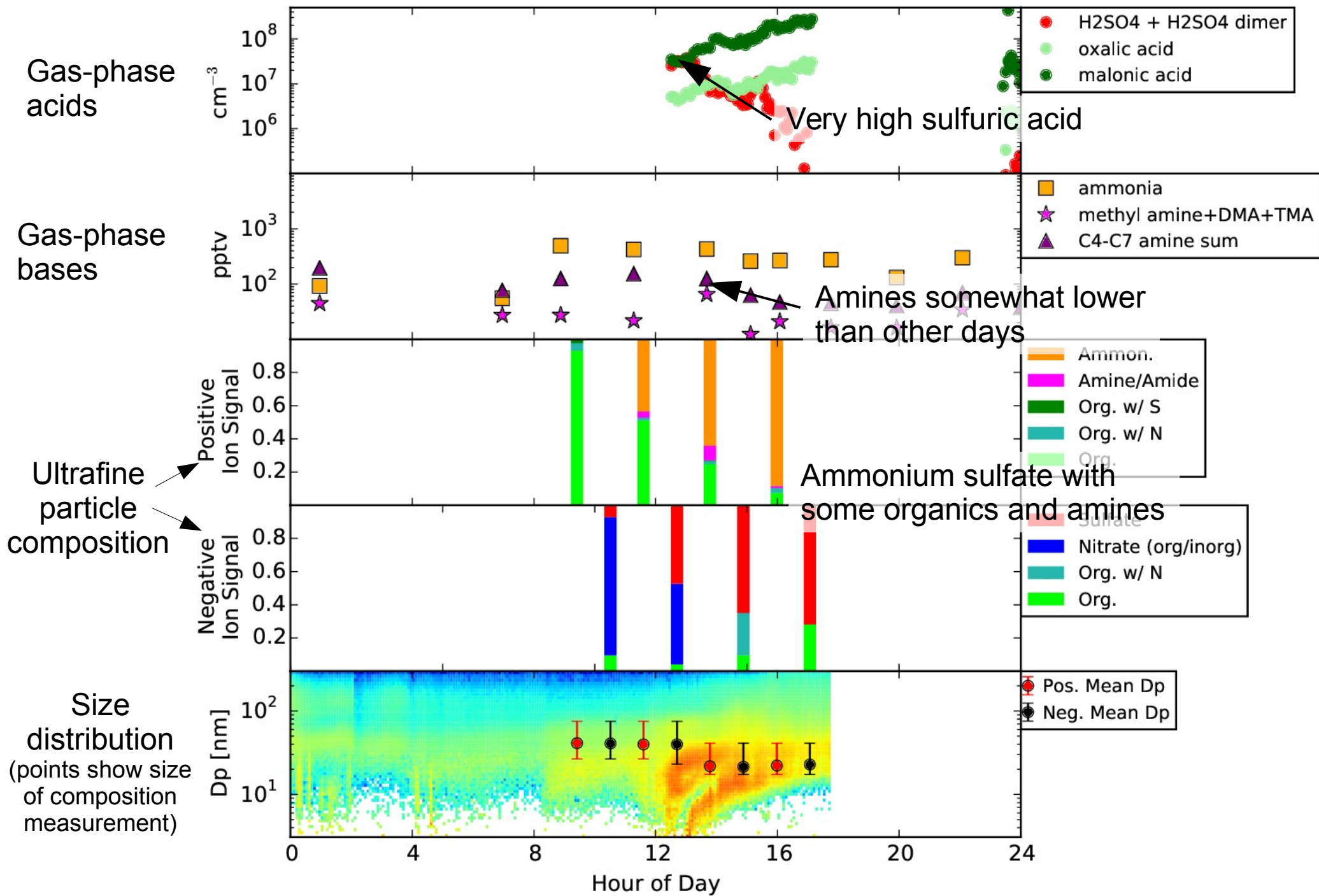
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5.9.13



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5.9.13



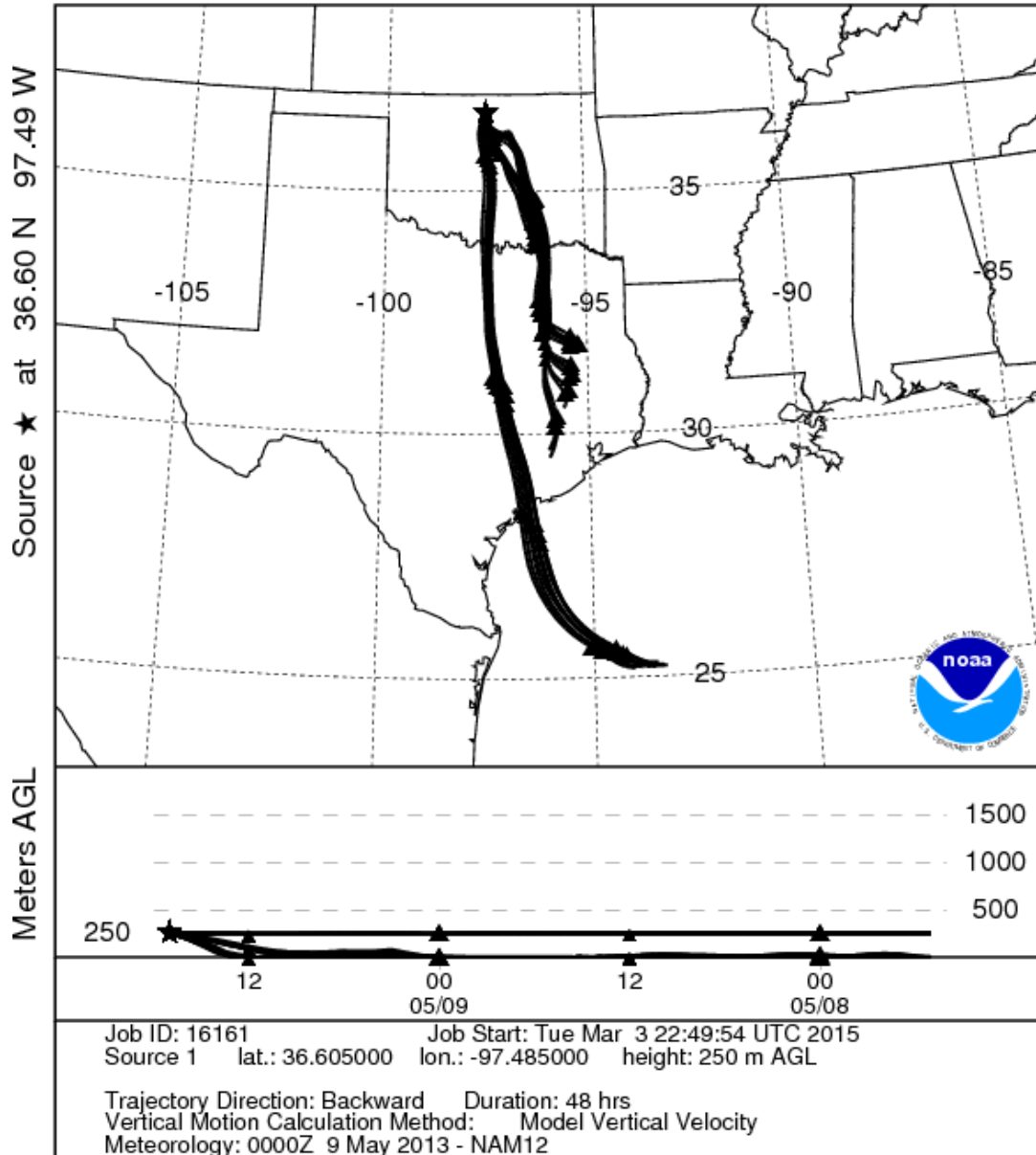


# May 9, 2013: Growth by sulfuric-acid/ammonia

NOAA HYSPLIT MODEL

Backward trajectories ending at 1700 UTC 09 May 13

NAM Meteorological Data

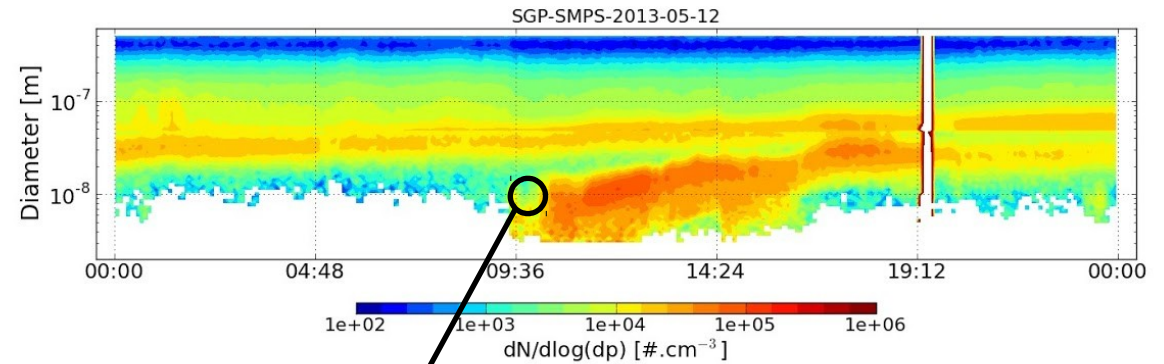


Flow from South:  
Major urban areas (e.g. Dallas)  
Industrial gulf coast

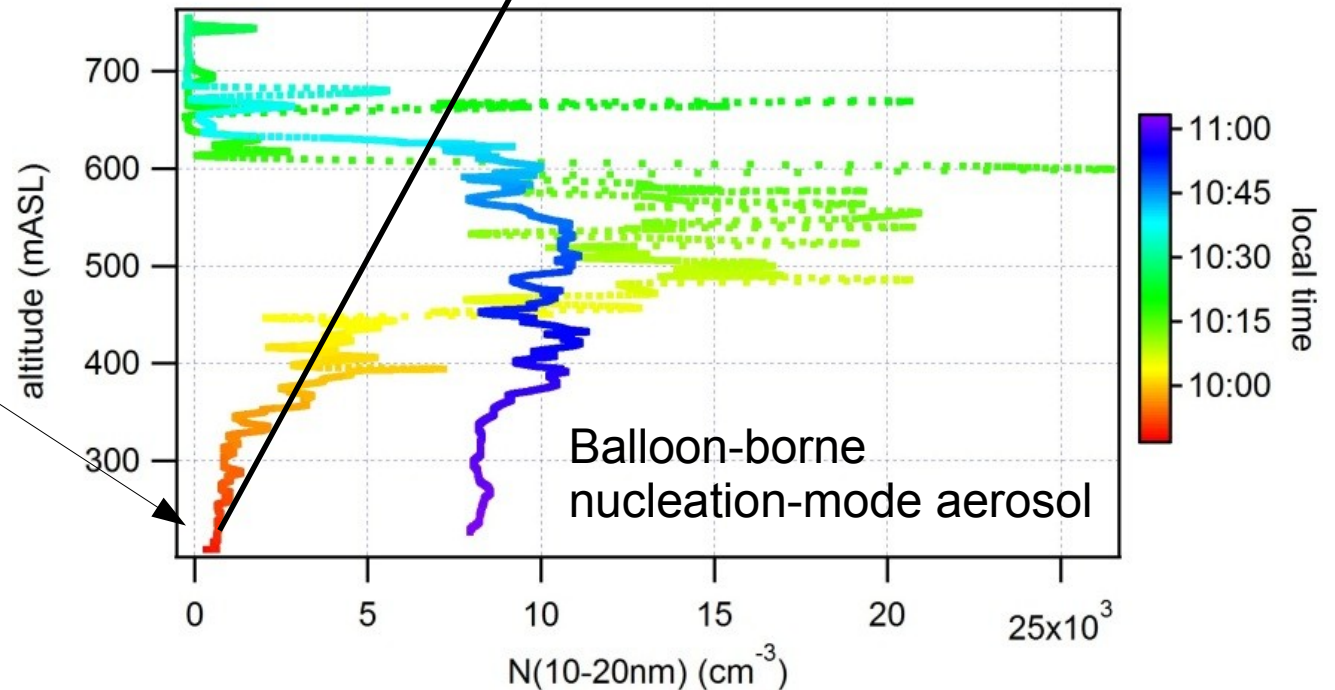
# NPFS-2013: vertical profiles of 10-20 nm diameter aerosol



Surface size distribution



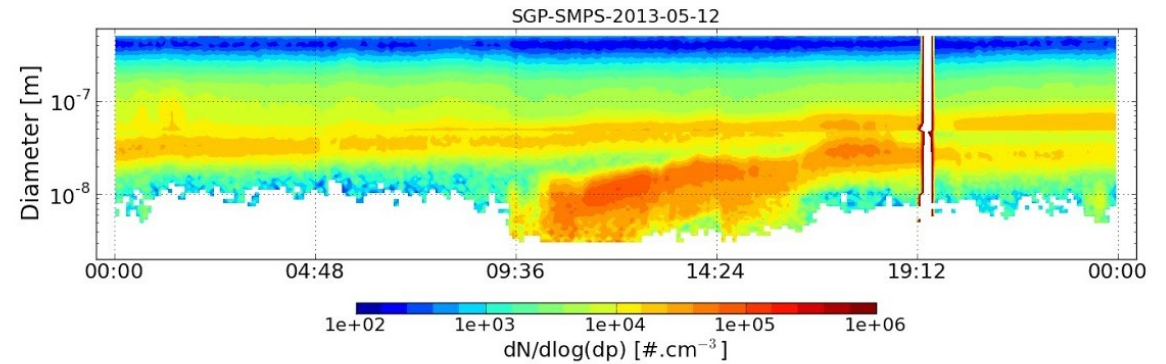
No nucleation at surface at start of flight



# NPFS-2013: vertical profiles of 10-20 nm diameter aerosol

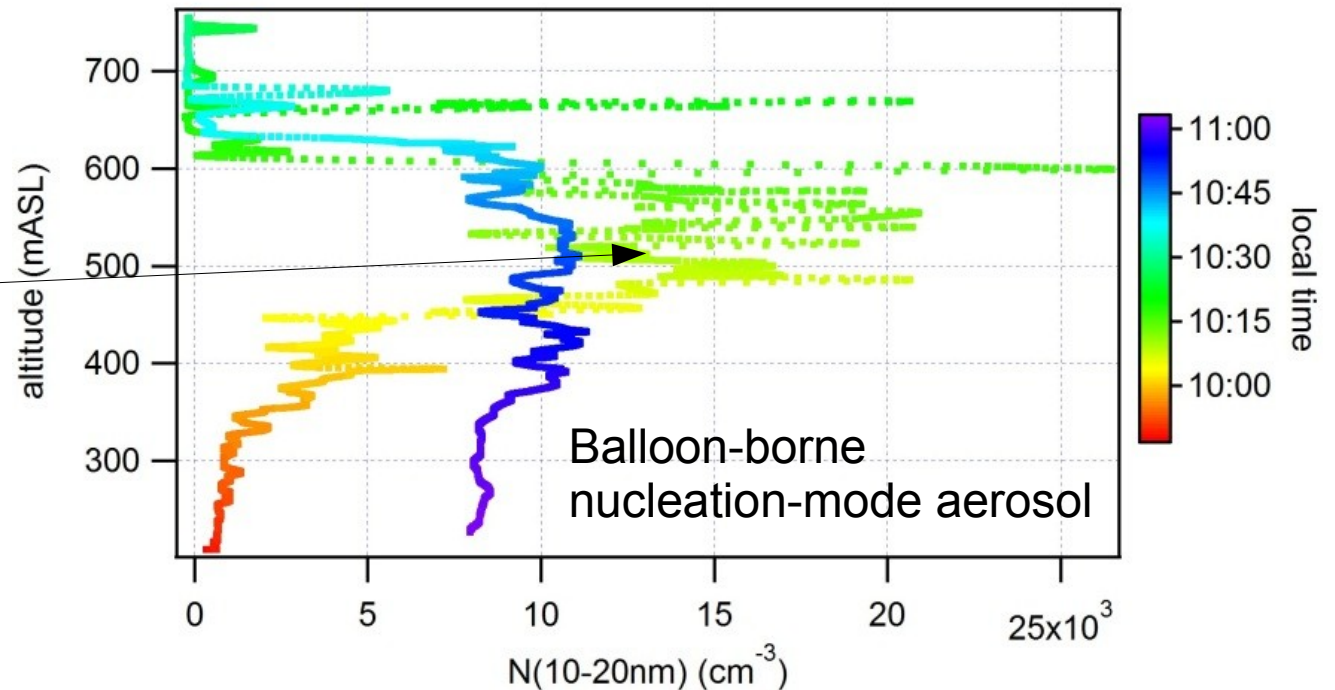


Surface size distribution



No nucleation at surface at start of flight

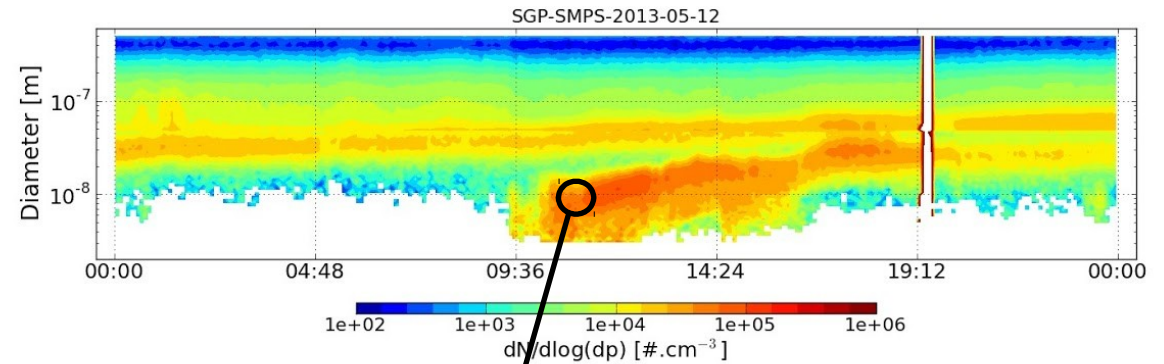
Nucleation aloft during ascent



# NPFS-2013: vertical profiles of 10-20 nm diameter aerosol



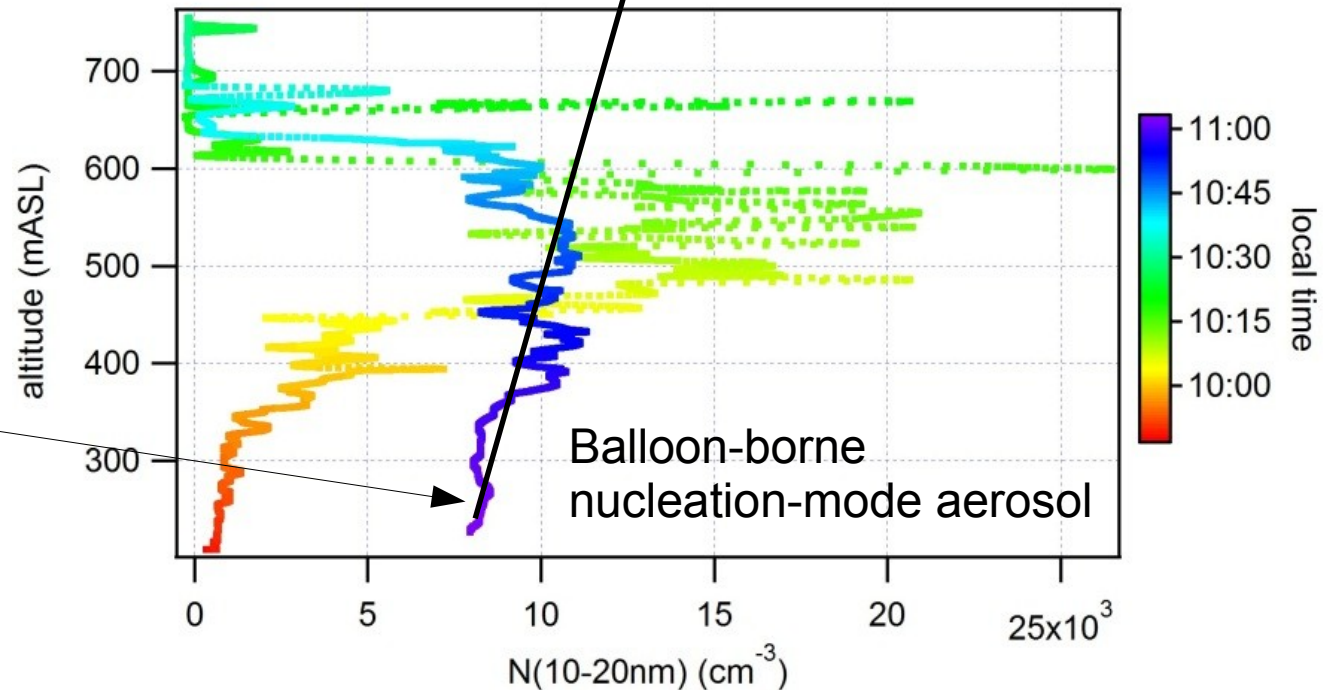
Surface size distribution



No nucleation at surface at start of flight

Nucleation aloft during ascent

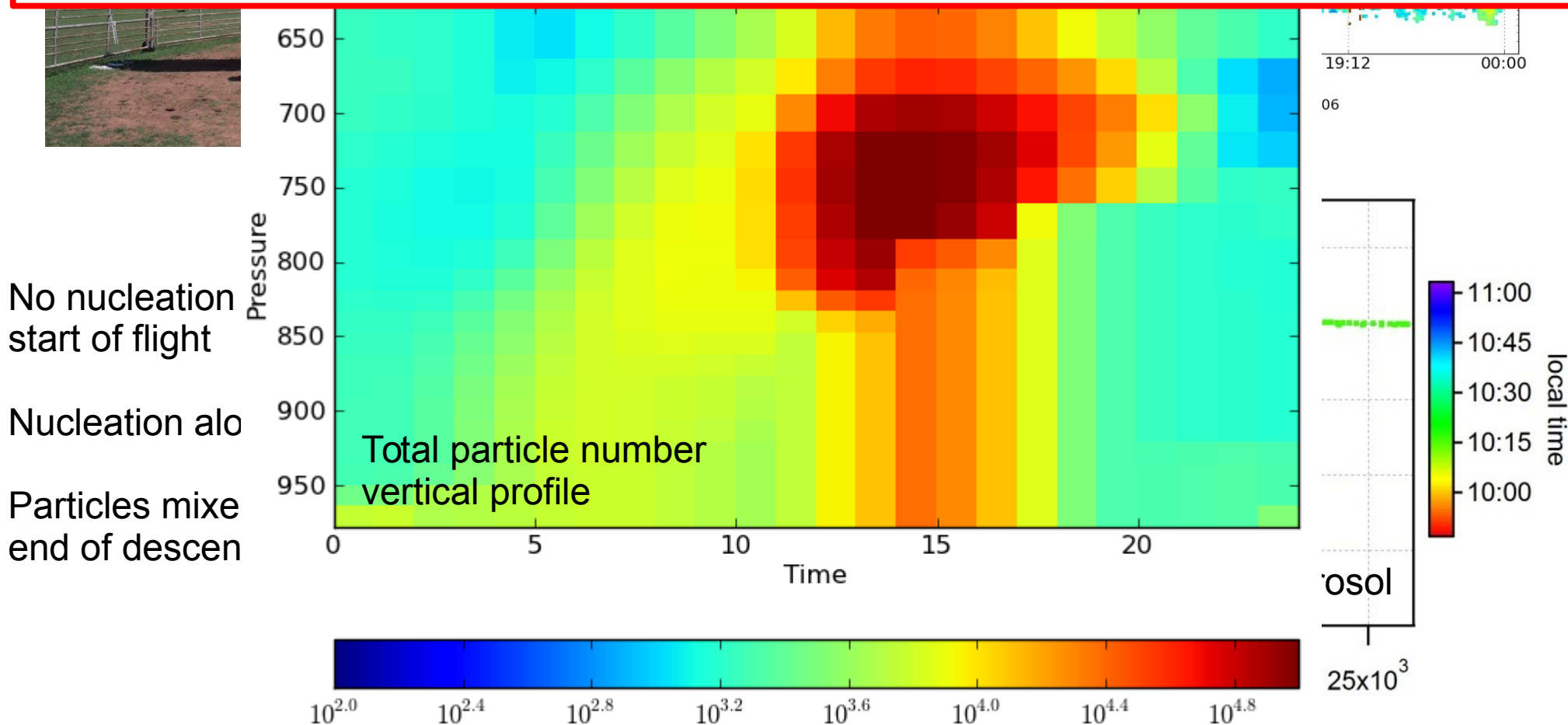
Particles mixed to surface by end of descent



# Nucleation aloft!

Implication: yesterday's residual layer important  
Does dry deposition in surface layer overnight remove precursors?

Captured in GEOS-Chem-TOMAS regional modeling results (below)



# Take home points

- DOE-funded New Particle Formation Study at ARM SGP site
  - Unprecedented collection instruments for NPF/growth/precursor measurements
  - At least 3 distinct pathways for new-particle growth observed
  - Various pathways not resolved in models
  - Nucleation observed to start aloft

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  - At least 3 distinct pathways for new-particle growth observed
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  - Nucleation observed to start aloft
- Next steps with field study
  - Do gas-phase species and particle growth fit our understanding of thermodynamics and kinetics?
  - Explore all field-study days... patterns?

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- DOE-funded New Particle Formation Study at ARM SGP site
  - Unprecedented collection instruments for NPF/growth/precursor measurements
  - At least 3 distinct pathways for new-particle growth observed

## Future goals

- Controlled lab studies of growth pathways
- Growth parameterization building
- Test against field observations

- Do gas-phase species and particle growth fit our understanding of thermodynamics and kinetics?
- Explore all field-study days... patterns?



# May 11, 2013: Can track through night... similar event on May 12

5.11.13-5.12.13

