New Particle Formation in the UT/LS:

Project Overview and Preliminary Results

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Outline

- Project Overview
  - Background
  - Objectives
  - Study design
  - Progress
- Preliminary Results
  - Sunset and sunrise experiments
  - Tropopause folding episodes

Background

- Environmental and Human Health Concerns
  - Particle growth → cloud condensation nuclei
  - Human inhalation exposure to ultrafine particles

- New particle formation (NPF, or nucleation) is one of the chain of events that leads to cloud formation, yet its precise mechanism is not well understood.

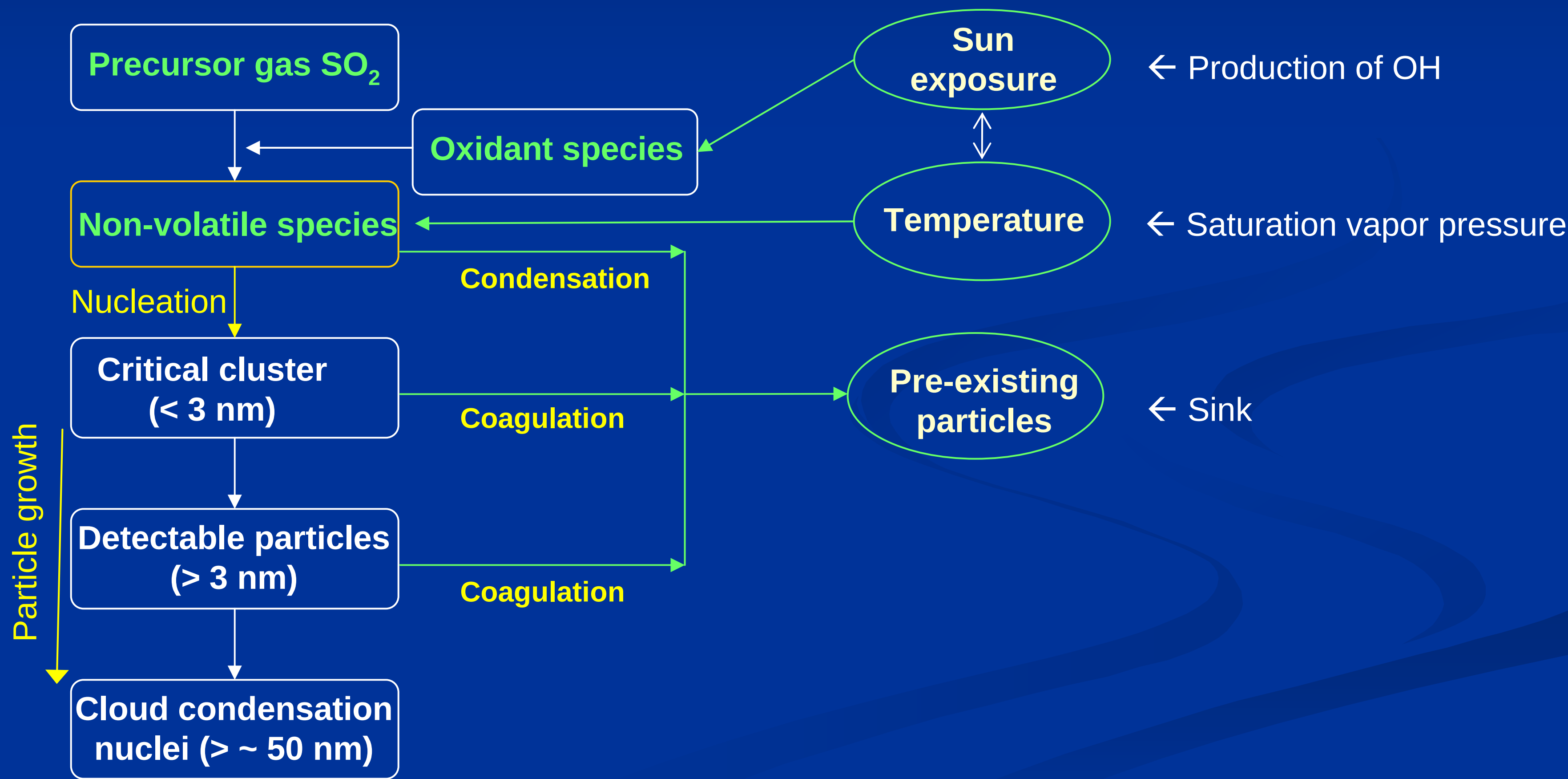
- NPF events are periods with elevated number conc of 4 – 8 nm particles (N_{4-8}) from background level

Background

- NPF events have been observed globally (rural, coastal, and urban area), but in-situ, size-resolved measurements in the upper troposphere and lower stratosphere (UT/LS) are still sparse
  - Most are ground-level measurements
  - Kulmala et al., 2004.
- UT/LS
  - Low temperature
  - Low pre-existing aerosol surface area conc
  - Hofmann, 1993; Brock et al., 1995;. Schröder and Ström, 1997; Nyeki et al., 1999; de Reus et al., 1998, 1999; Wang et al., 2000; Hermann et al., 2003; Lee et al., 2003.

Background

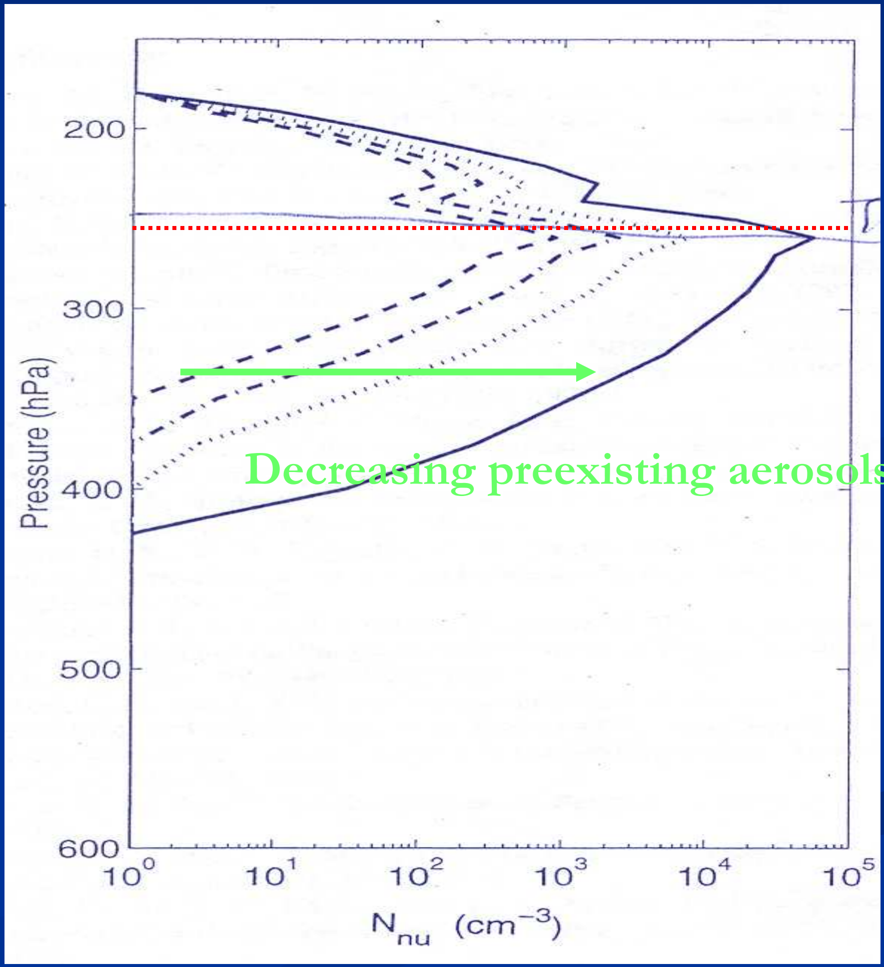
- NPF in the UT/LS



e.g., $SO_2 + OH \rightarrow HSO_3$ ---> $H_2SO_4 + H_2O \rightarrow$ new particles

Source and Sink

Model Simulation coupled with CN measurements



Above tropopause:
$N_{6\text{-}15}$ decreases with increasing altitude
Source-limited region (i.e., SO_2, H_2SO_4, etc.)

Below tropopasue:
$N_{6\text{-}15}$ increases with increasing altitude
Sink- limited region (i.e., pre-existing aerosols)
Max $N_{6\text{-}15}$ near tropopause

de Reus et al., 1998, *J. Geophys. Res.*

Source, Sink, and Sun Exposure

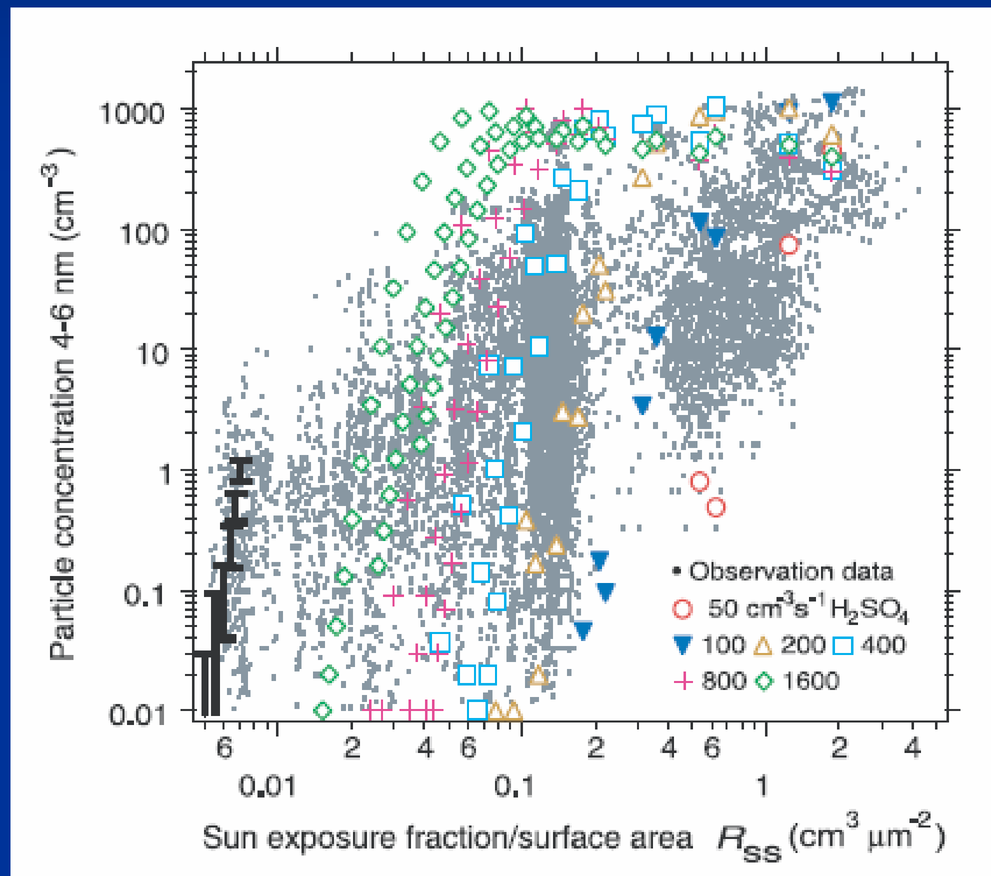
Model Simulation coupled with CN, OH, and SO_2 measurements in the UT/LS

N_{4-6} increases with increasing Rss

Higher sun exp.
→ higher H2SO4 production
→ stronger source strength

Lower particle surface area
→ weaker sink

Stronger NPF intensity
Agrees with ion-induced nucl model



Lee et al., 2003, *Science*

Atmospheric Mixing

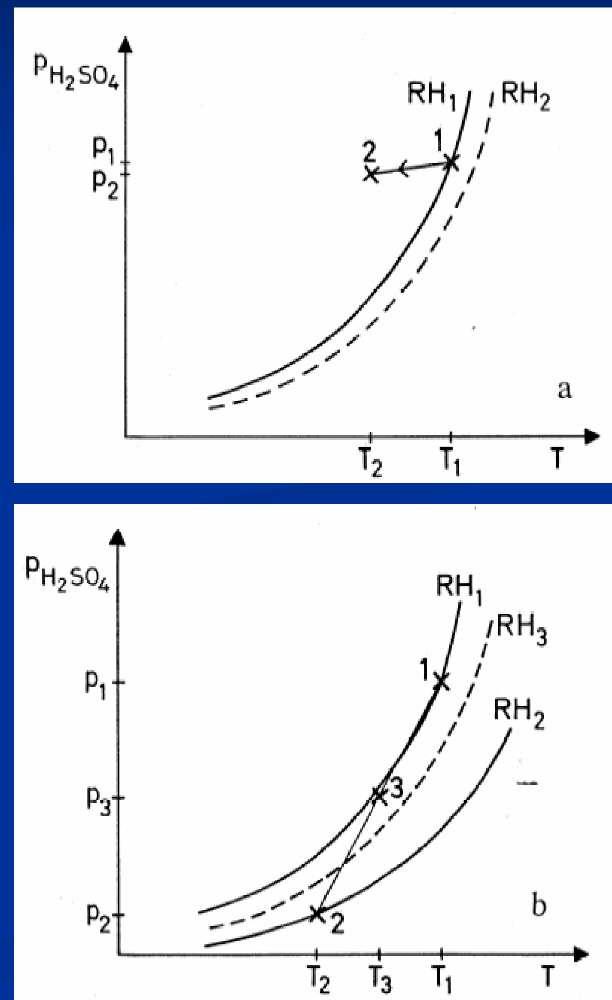
T and RH fluctuations as a result of atmos. mixing → enhanced NPF
(e.g., Easter and Peters, 1994; de Reus et al., 1999; Bigg, 1997, etc.)

Eddies, gravity waves, mountain waves.
Turbulence
Mixing of two air parcels with large T and RH diff.

A wave of 1 K amplitude → Nucl. rate increases 2 orders of magnitude

Top figure: Adiabatic cooling of ascending air decreasing T → vapor pressure (p) decreases and new equilibrium curve shifts to the right
→ supersaturation

Bottom: mixing of two air parcels
New state (3) → supersaturation



Nilsson and Kulmala, 1998, *J. Geophys. Res.*

Objectives

- Investigate the dependence of NPF on sun exposure, latitude, and altitude.

- Examine the influence of air mass exchange/mixing near tropopause fold on NPF.

Instruments

- High-performance Instrumented Airborne Platform for Environmental Research (HIAPER) – Gulfstream V

- Nuclei mode aerosol size spectrometer (NMASS)
  - 5 condensation nuclei counters: > 4, > 8, >15, > 30, and > 64 nm cumulative number concentration. (channel 1-channel 2 = N_{4-8})
  - 4 – 100 nm size range

- Focused cavity aerosol spectrometer (FCAS)
  - An optical spectrometer
  - 90 – 10000 nm size range

- Gas species (O_3, CO, H_2O), meteorological, and aircraft parameters

Flight Patterns

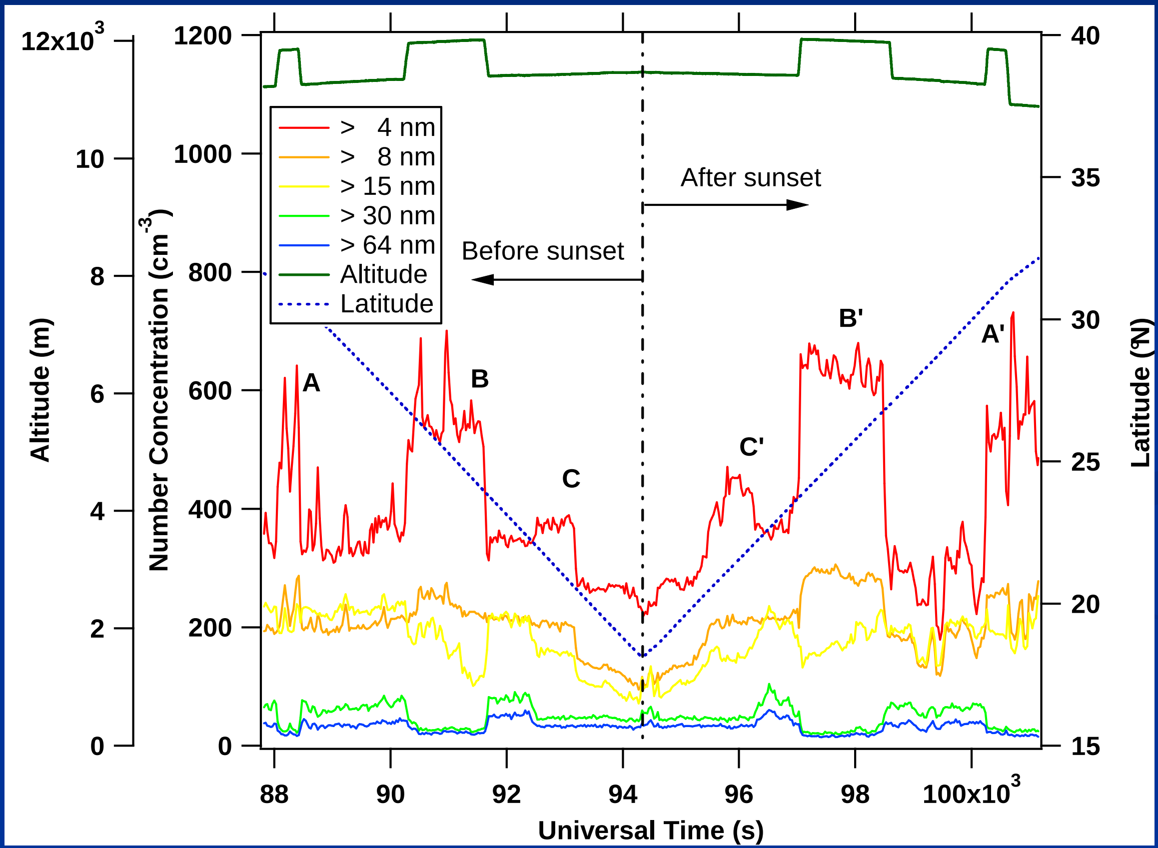
- 2005 Nov. and Dec.
  - Latitude: 18°N - 62°N
  - Altitude: up to 14 km
- Sun exposure experiments
  - Fly out prior to sunrise (or sunset) and fly in on the same flight path immediately after sunrise (or sunset)
- NPF near tropopause fold (Pan et al., START project)
  - Mid-latitude
  - Stacked horizontal flight legs

Aerosol Data Sets

RF #	Date	Experiment	NMASS	FCAS	Met. Parameters	Data Analysis
1	2005/12/01	START	√	√	√	In process
2	2005/12/02	NPF: Sunset	√	√	√	In process
3	2005/12/07	START	√	√	√	In process
4	2005/12/08	DOCIMS	√	√		To do
6	2005/12/12	NPF: Sunrise	√	√	√	In process
7	2005/12/13	CHAPS	√	√		To do
9	2005/12/16	CHAPS	√	√		To do
10	2005/12/19	NPF: Sunrise	√	√	√	In process

Sunset Experiment

2005/12/02



A time series plot of particle number concentrations of varying cut-off diameters, altitude, and latitude during a sunset experiment.
The vertical dash-dotted line indicates the **sunset** time.

Encountered three NPF events with elevated 4-8 nm particle number conc.

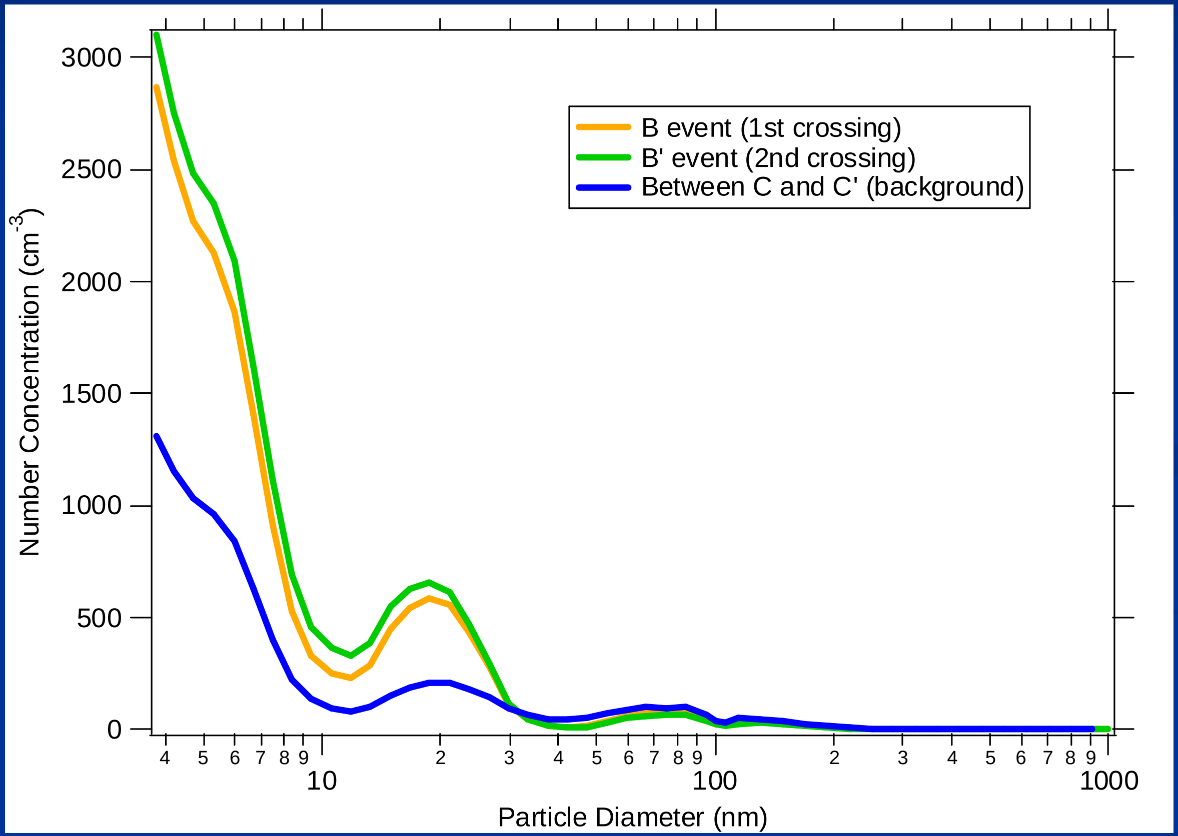
→ **1st crossing: A, B, C**

→ **2nd crossing: A', B', C'**

The absence of sunlight did not terminate the new particle formation instantaneously. (higher number conc during the 2nd crossing)

The spatial scales of observed new particle formation are in the range of 100 – 350 km.

Size Distribution



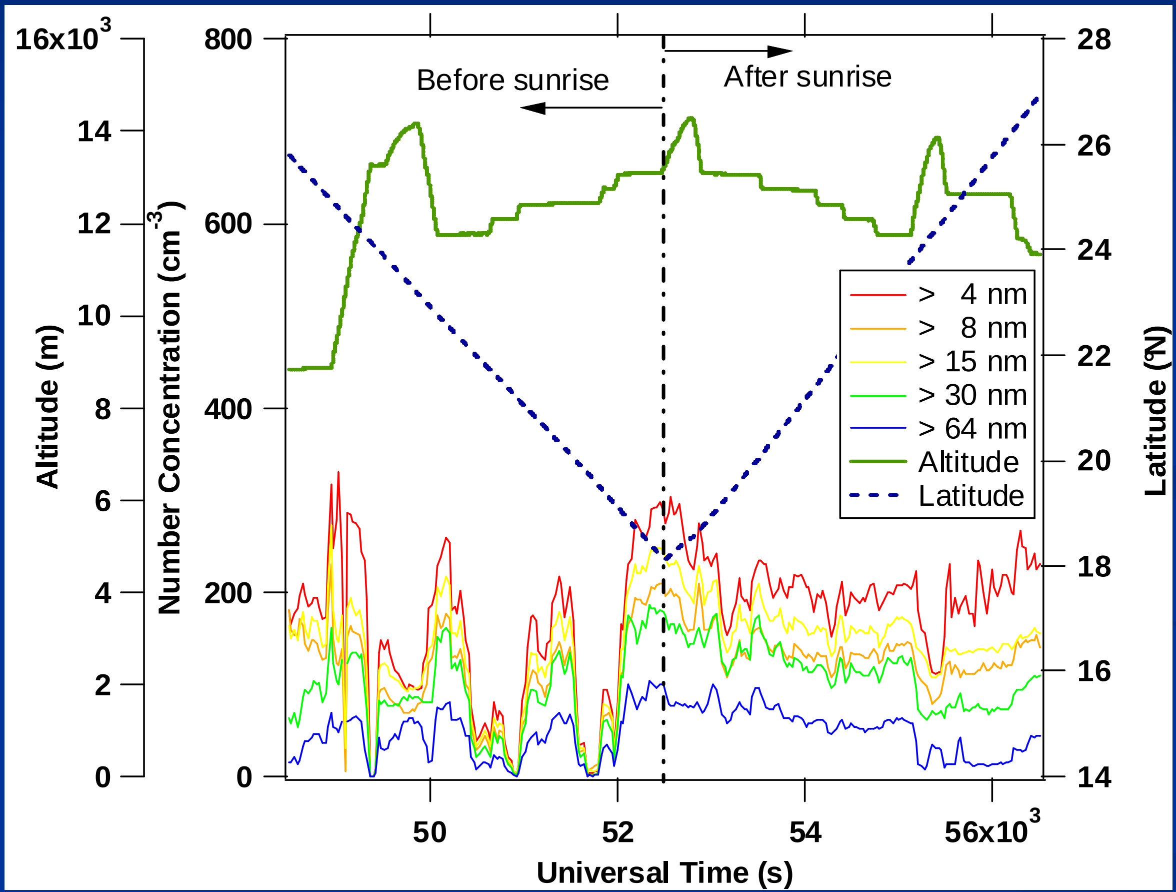
- ↑ number conc.
- Size distr. shift upward to the higher conc.
- Background: fresh particles are ubiquitous in the UT/LS

NMASS data

FCAS data

Sunrise Experiment

2005/12/12



The vertical dash-dotted line indicates the **sunrise** time.

The intensity of new particle formation after sunrise was not as strong as we have anticipated.

→ Weaker source strength?

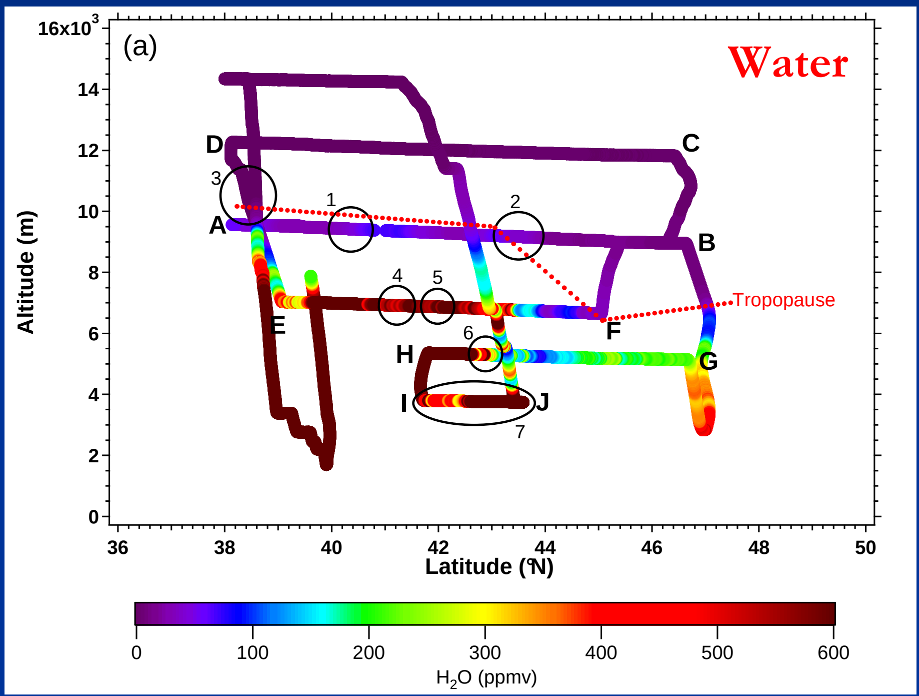
→ Stronger sink?

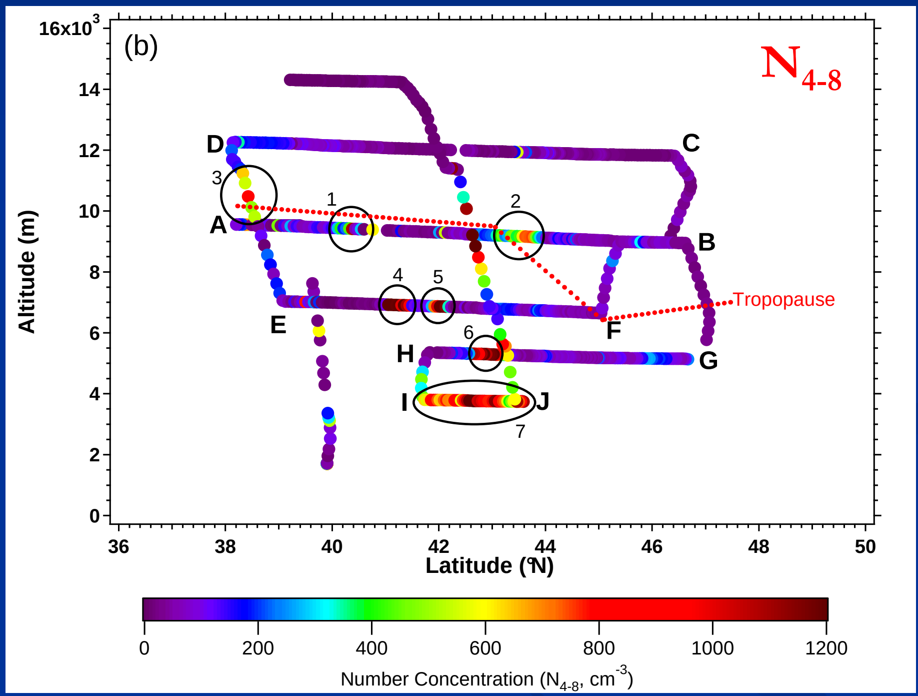
→ Not enough time for growth?

Nighttime new particle formation??

NPF near Tropopause Fold

2005/12/01

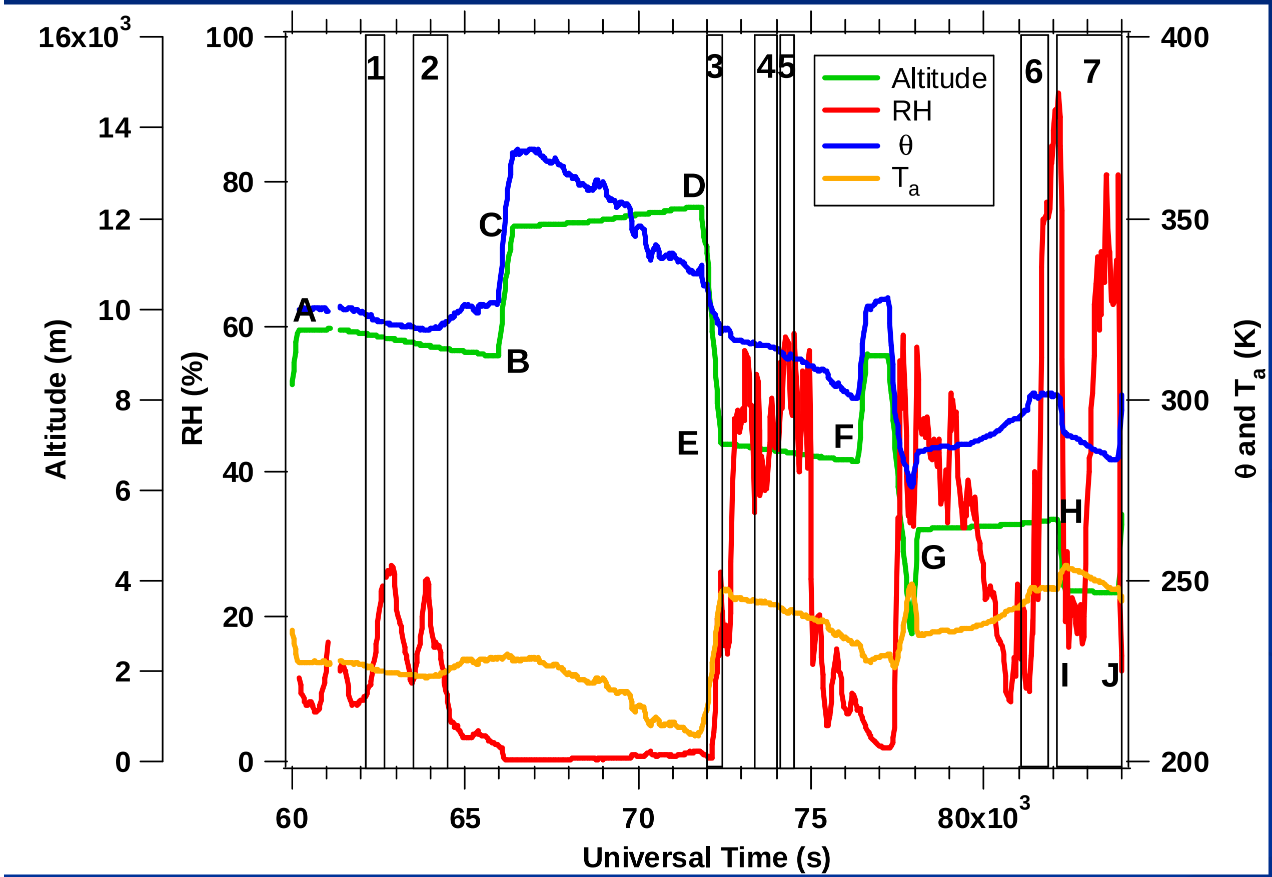




- < ~10 H_2O pmv → Stratospheric air
- H_2O gradient near tropopause → STE (mixing)

- Intense NPF events preferentially took place in regions with relatively larger H_2O gradient or fluctuation

T and RH



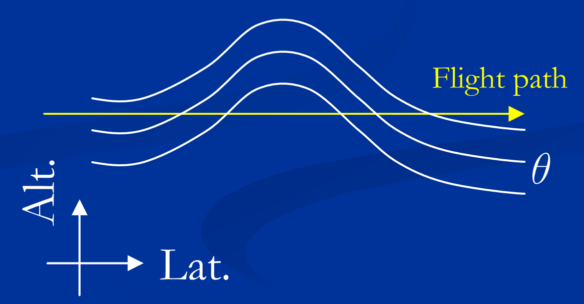
Ambient temperature (Ta)
Potential temperature (θ)

Favorable conditions:

↓ Ta and θ

↑ RH

Flight leg A-B:

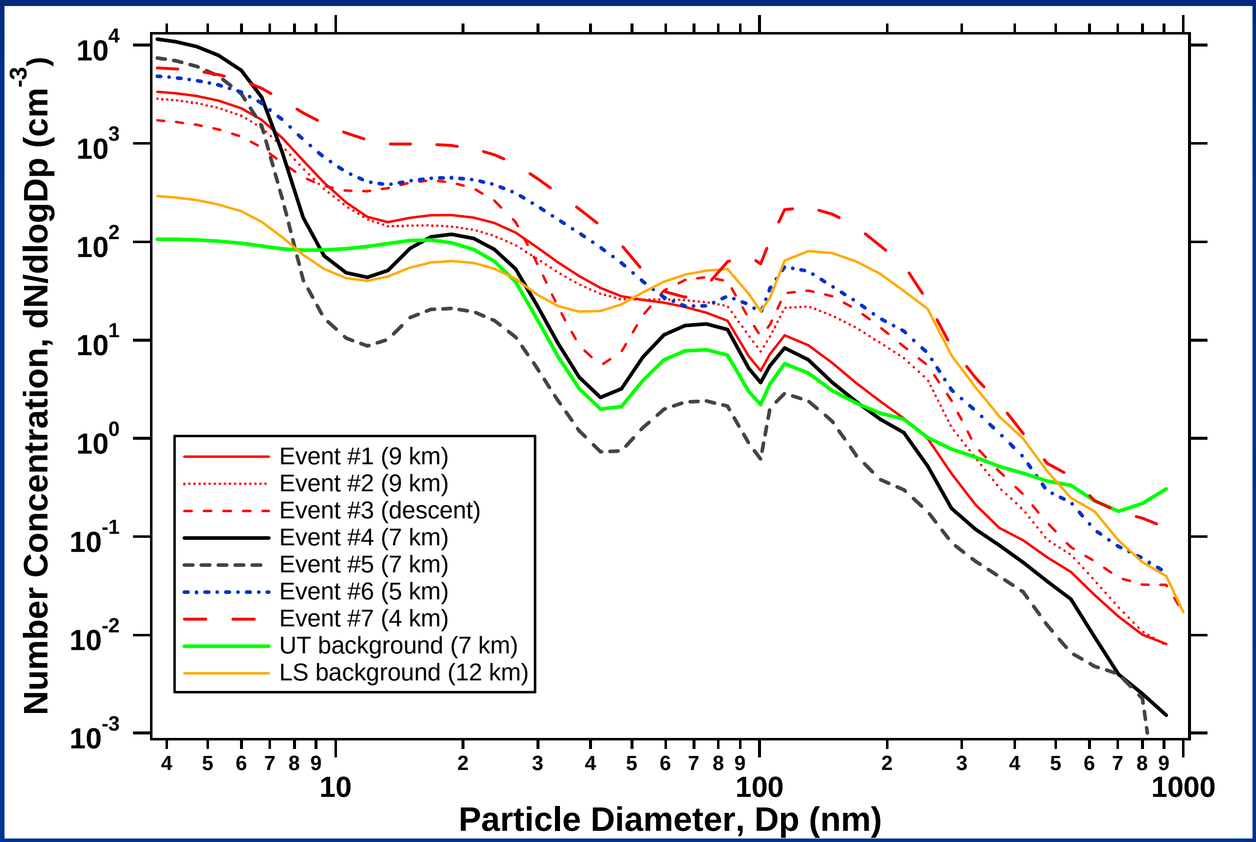


Adiabatic cooling → supersaturation → nucleation ?

Other flight legs:

Mixing b/w two air parcels with different Ta and RH
→ nucleation ?

Size Distribution



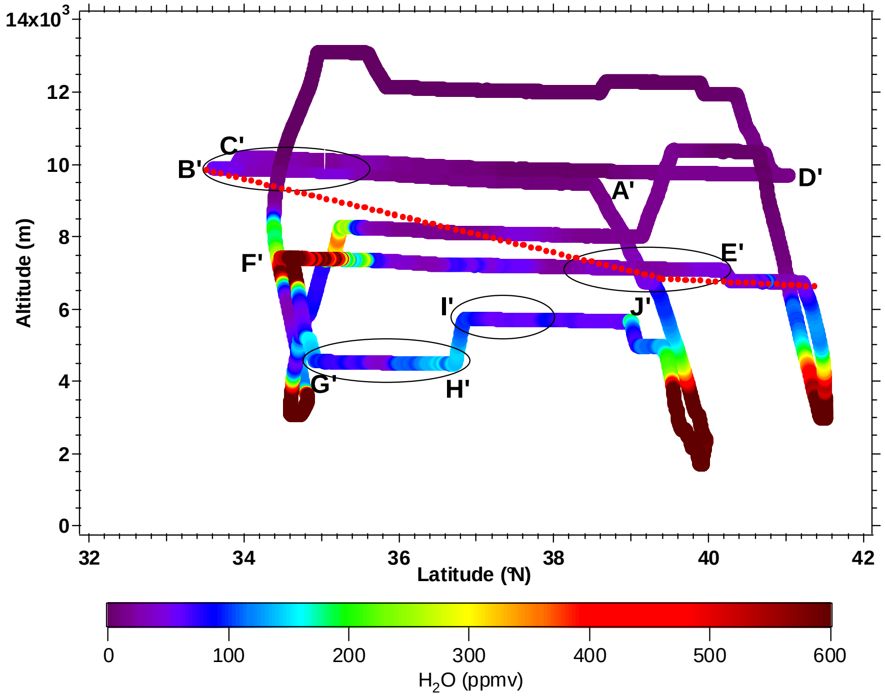
Average number size distributions

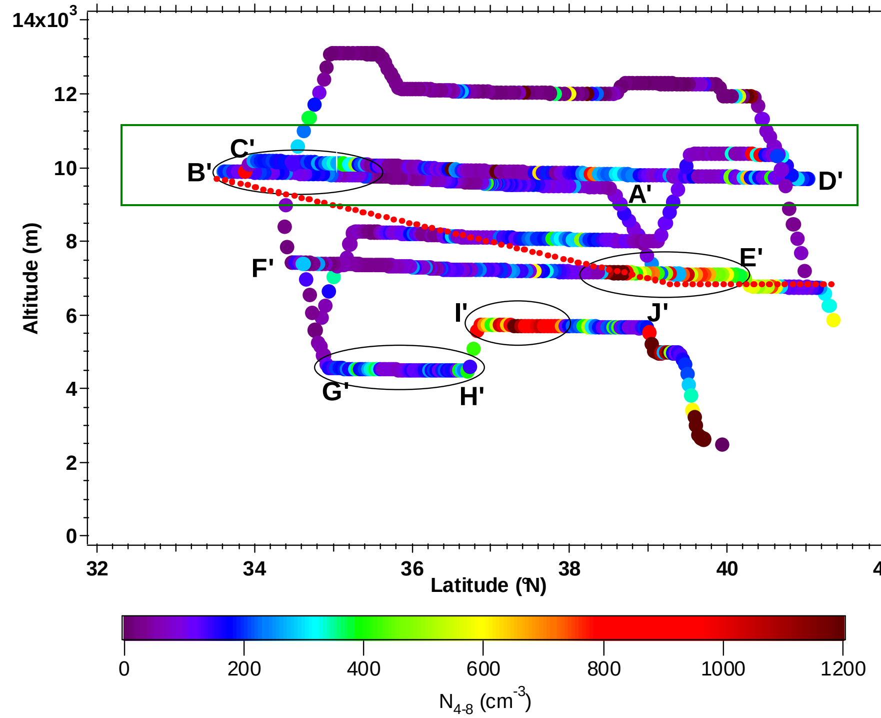
The UT and LS "background" aerosols represent non-event periods during a given altitude.

- Tri-modal size distr. (4, 20, 80 nm)
- The two most intense NPF events have lower > 10 nm particle number conc. ($N_{>10}$)
- $N_{>10}$ is higher in LS than in UT

NPF near Tropopause Fold

2005/12/07



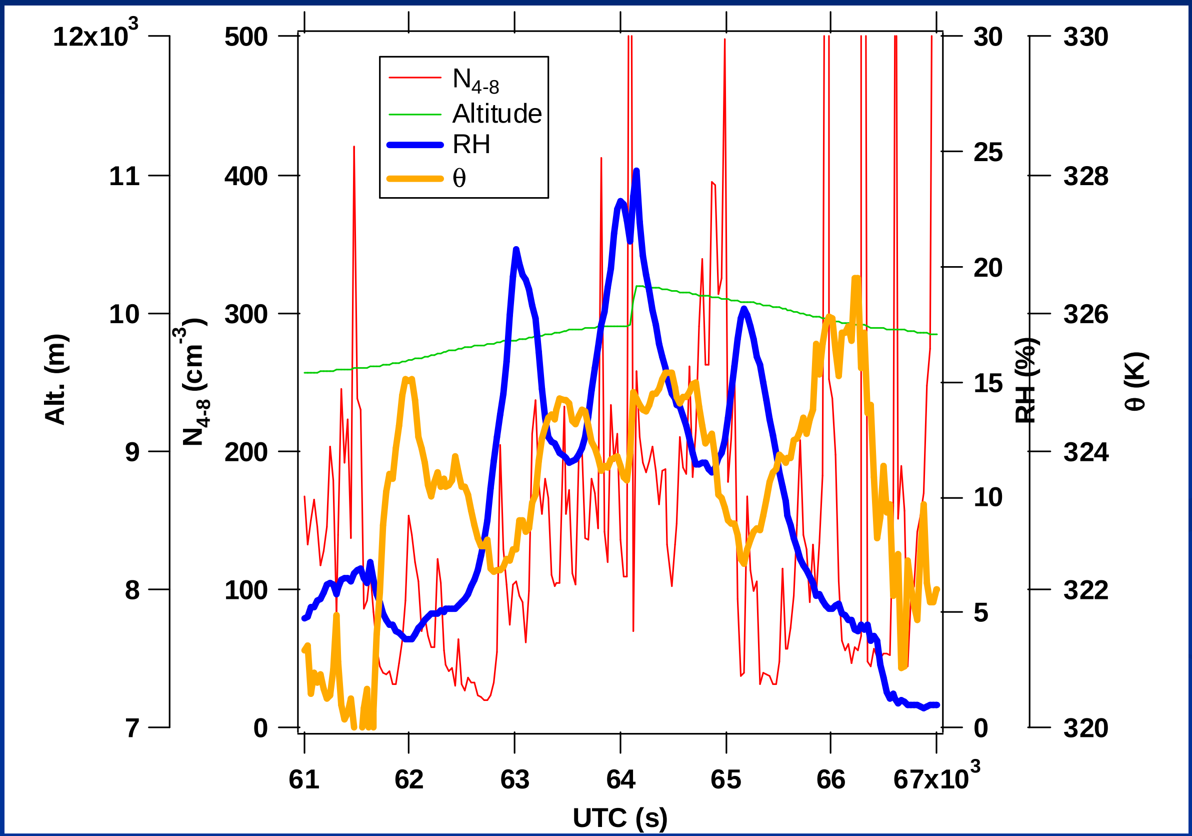


Elevated $N_{4\text{-}8}$ was considerably more prevalent on the flight track, <u>on both side of the tropopause</u>, than on Dec-1.

Intense NFP at the bottom of the jet; E-F (?)

(Pan et al., 2006, *J. Geophys. Res.*)

T and RH



9-10 km (near the tropopause)

Sporadic bursts of $N_{4\text{-}8}$

Anti-corr b/w RH and θ

Rapid changes of RH and θ in a relatively short and horizontal distance.

Mixing as a result of atmos. wave?

Summary of Findings

- New particle formation is ubiquitous in the upper troposphere and lower stratosphere.
- New particle formation and growth that occurred in the daytime can continue with the absence of sunlight.
- The intensity of new particle formation after sunrise appears to be not as strong as expected and requires further investigation.
- New particle formation appears to be significantly enhanced by mixing between two air parcels and/or RH and T fluctuation, consistent with model calculations by Nilsson and Kulmala (1998).

Scientific Questions

- Nighttime NPF events:
  - Thermodynamically stable clusters (TSCs) or critical clusters (1-3 nm) that are undetectable by instruments may be present in the nighttime air
  - Observation of NPF events (4-8 nm) then depends on both the availability of condensable species and atmospheric conditions (e.g., RH and T fluctuation, mixing, etc.)
  - Without OH → no new H_2SO_4 production; Then, what condensable species are involved in the nighttime growth process? NH_3, organics ?
  - Changes in atmos. conditions (e.g., T and RH) can result in superstaturated condensable species that favor growth of TSCs to detectable sizes

Scientific Questions

- NPF near tropopause fold
  - Many NPF events coincide with regions experiencing T, θ, and RH fluctuations.
  - What do those fluctuations imply in terms of atmospheric conditions and dynamics? Atmos. waves, turbulence, mixing?
  - How do we quantify the intensity of those fluctuations?

- Aircraft exhaust and contrail
  - How do we identify and exclude aerosols from exhaust and contrail?
  - NOx, CO_2, SO_2, CO, H_2O, size distribution ?

Future Work

- Examine the sun exposure history of air masses and its relationship with NPF.
  - Trajectory calculation
- Quantify the intensity of atmos. fluctuations and its relationship with NPF.
  - e.g., Temp, RH, wind speed variation, water mixing ratio gradient
- Compile all measurements for latitude and altitude dependence of NPF.

Future Work

- Compare observations with aerosol nucleation models
  - Binary HN vs. Ternary HN vs. Ion-induced N
- Inter-compare NMASS and SMPS measurements

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