On the fate of aerosols produced by new particle formation in the UTLS

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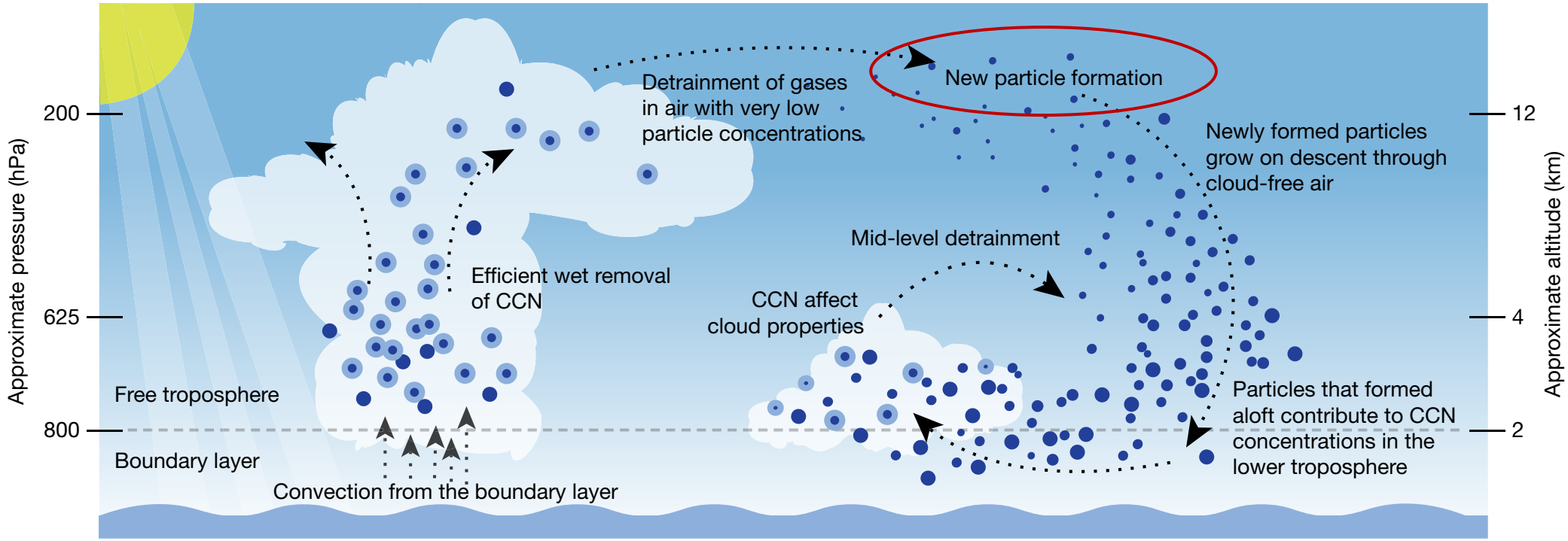
2024 SABRE and ACCLIP Joint Science Team Meeting

30 April – 3 May 2024

New particle formation in the upper troposphere

- New particle formation (NPF) is the process by which **gas phase species in the atmosphere come together to form new small (~2-3 nm) particles** that subsequently grow.
- NPF is a **source of cloud condensation nuclei** (Bianchi et al., 2016; Gordon et al., 2017; Williamson et al., 2019) and **aerosols** (Brock et al., 1995; Weigel et al., 2011; Andrea et al., 2018), which have important radiative effects.
- **Convection may contribute to NPF** by in-cloud scavenging of aerosols and by the direct supply of precursor gases, but evidence is mixed (Weigel et al, 2011, 2021; Williamson et al., 2019).



(Williamson et al., 2019)

Science Questions

1. Which **new particle formation (NPF) event particles** in the UTLS **ascend** (and therefore contribute to the stratospheric aerosol) or **descend** (thereby contributing to the tropospheric aerosol)?
    a) Where and under what conditions do stratosphere-relevant NPF events occur?
2. What is the **contribution of ultrafine particles** nucleated near the tropopause to **stratospheric aerosol abundance**?
3. Do NPF events in the UTLS predominantly occur in **air masses recently influenced by deep convection**?

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Our Approach

1. Identify NPF events in the UTLS

   - Use 0.1 Hz NMASS data from ACCLIP WB-57
   - NPF definition (based on Williamson et al., 2019):
     concentration in the smallest size channel (D = 3 nm) >> concentration in the next-largest channel
     * also require NMASS channel 1 concentration > 10 cm^{-3}

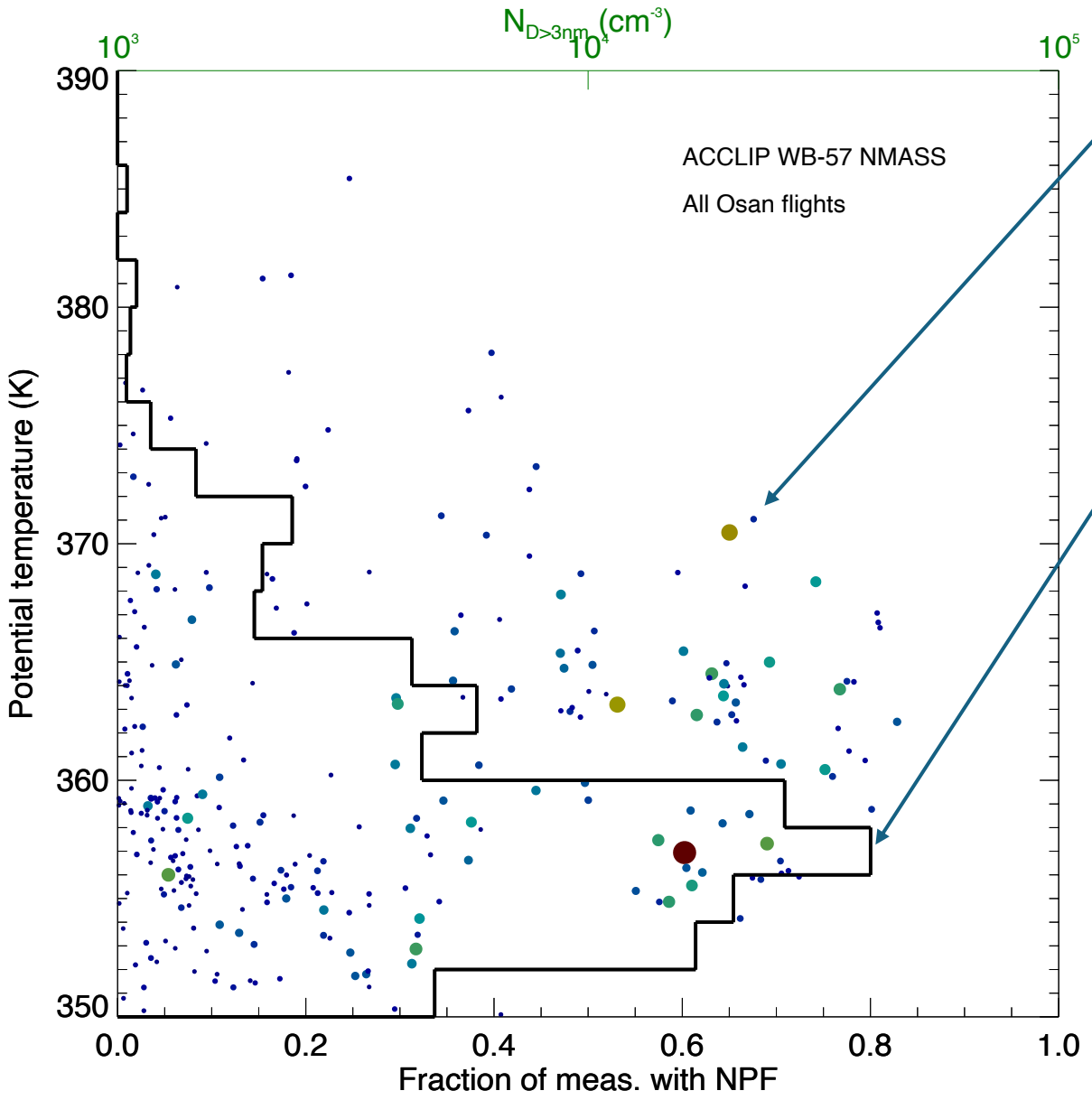
2. Calculate forward trajectories of NPF event particles

   - 30-d diabatic trajectories using ERA5 winds and heating rates (courtesy of Ben Clouser)
   - Trajectories every minute along the WB-57 flight track, each as a cluster of 75 points

3. Determine whether NPF event particles descend to the troposphere or ascend into the stratosphere

   - WMO lapse rate tropopause in ERA5 (Hoffmann & Spang, 2022)

NPF events from ACCLIP WB-57

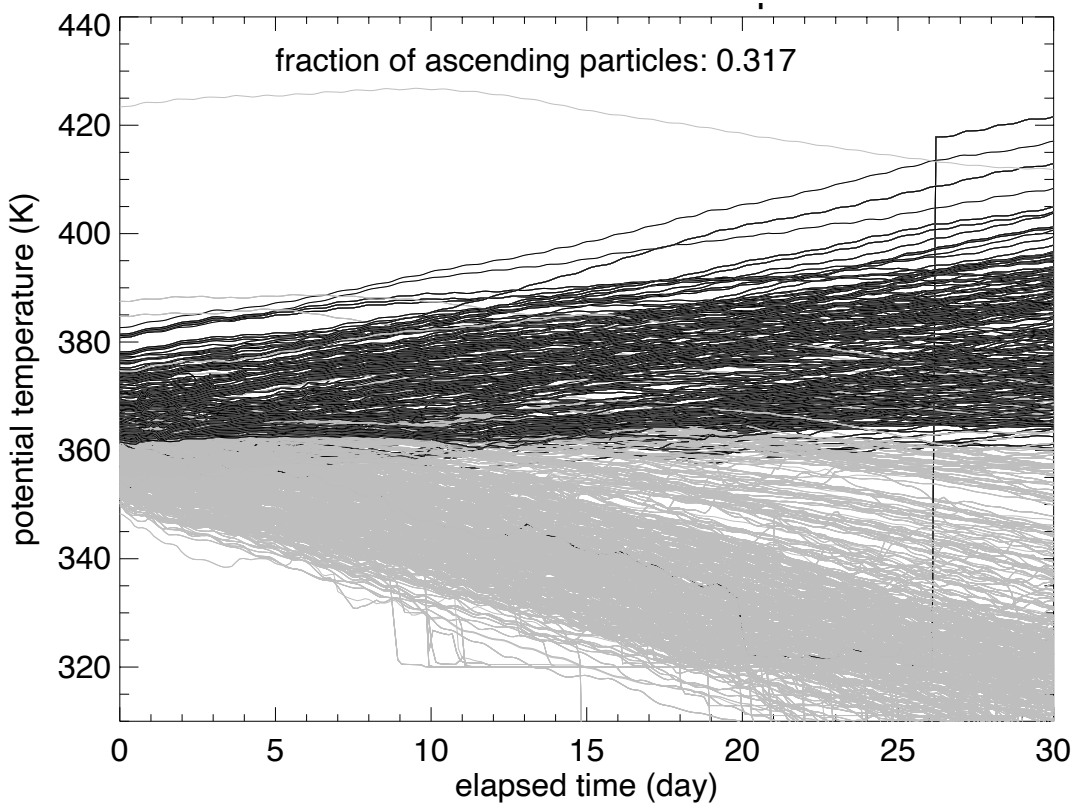


NMASS channel 1 aerosol concentration (D >3 nm) for each identified NPF events
(color/size of dots = duration of event)

Occurrence frequency of NPF events in 2K potential temperature bins (histogram)

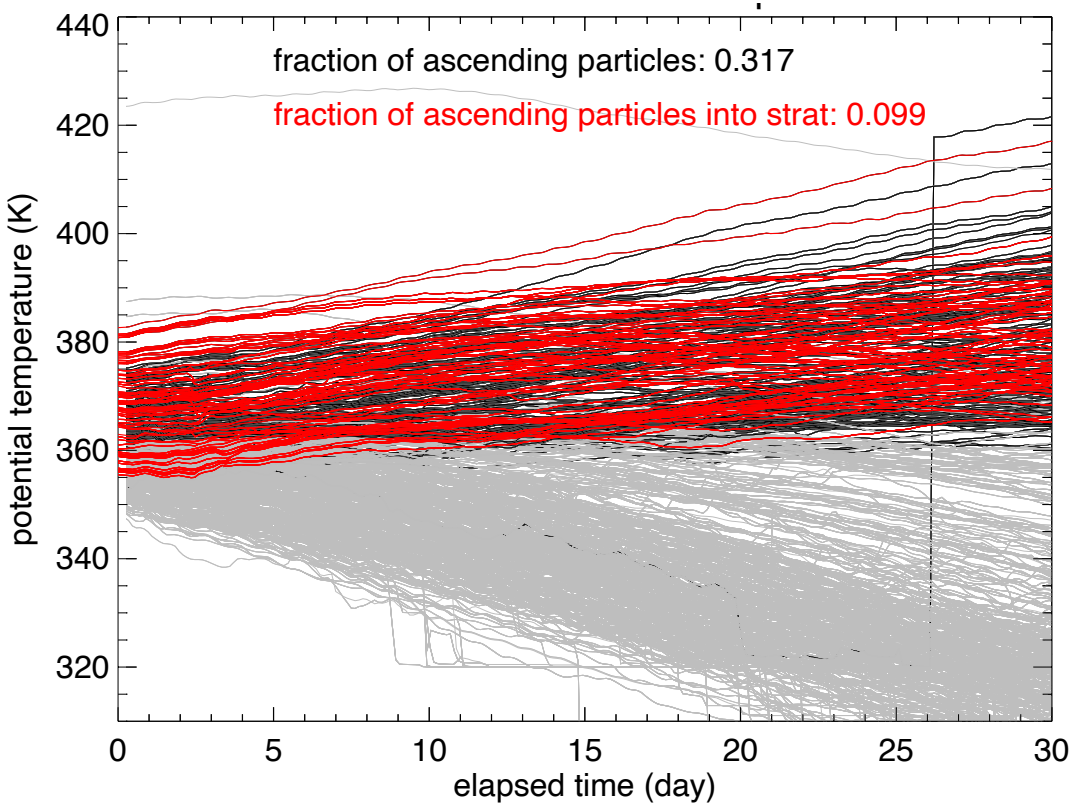
Most NPF events occur below 360K, but NPF events in the 360-370K range are not uncommon.

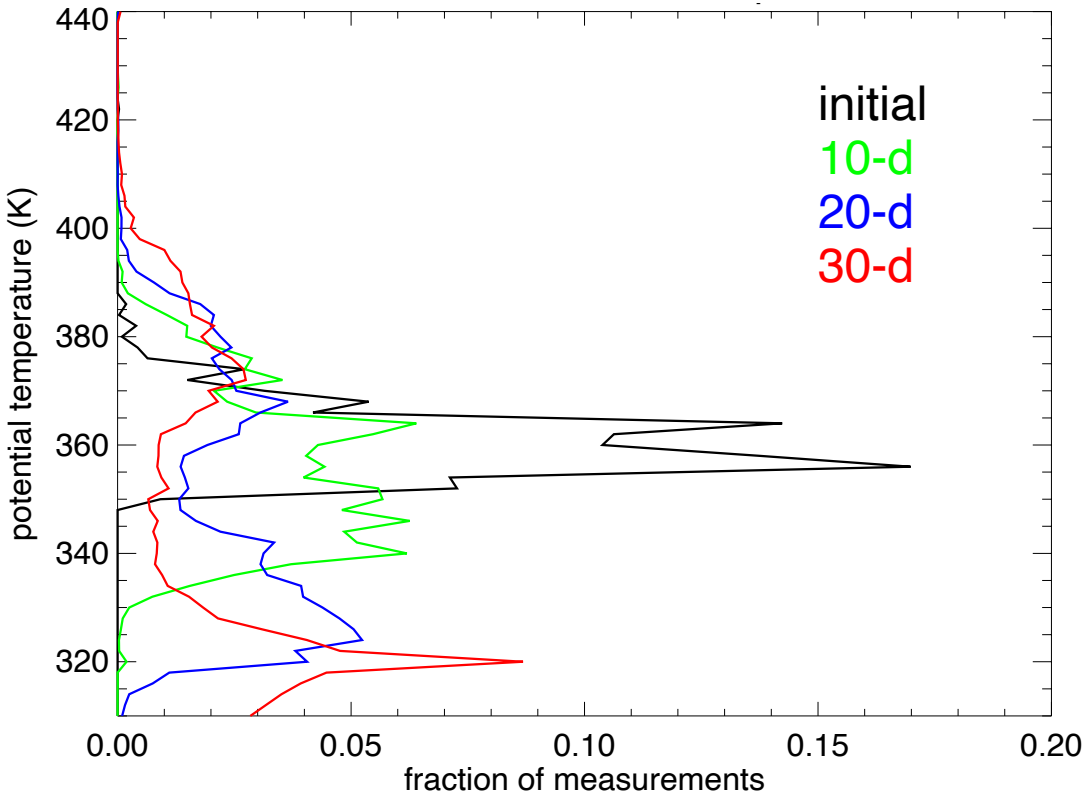
Fate of NPF event particles



- **~30% of all NPF event particles ascend** from their initial potential temperature level.

Fate of NPF event particles

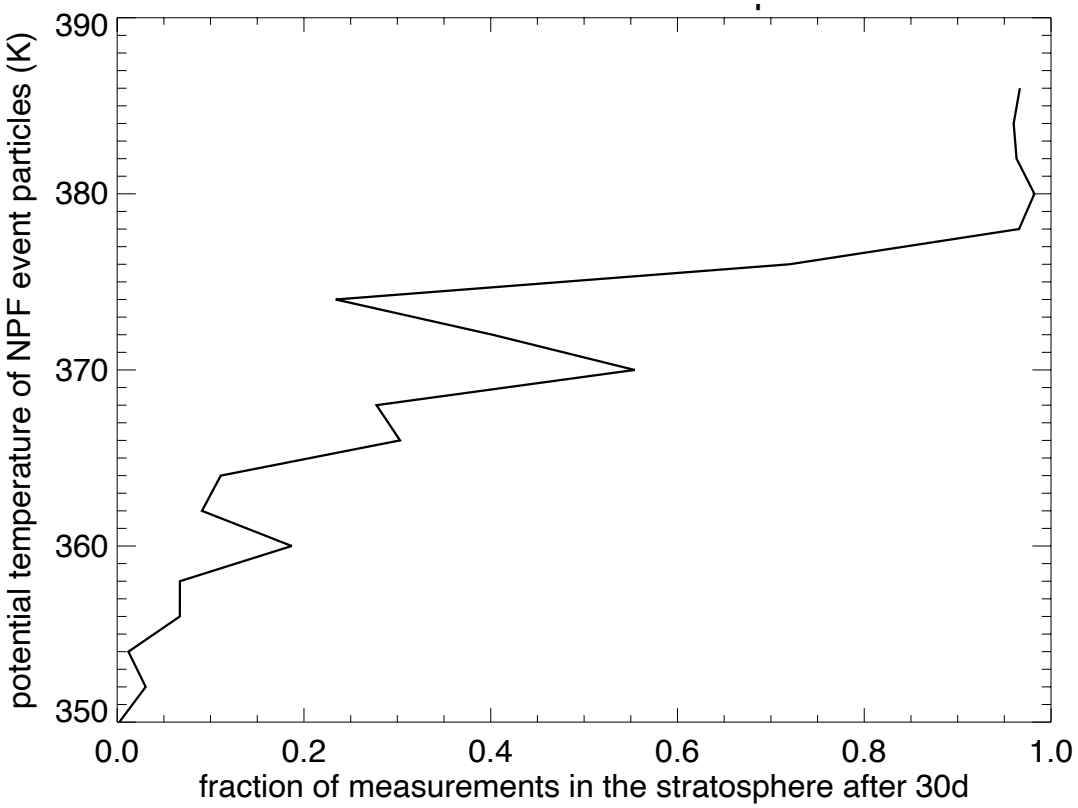




- **~30%** of all NPF event particles **ascend** from their initial potential temperature level.
- **~10%** of all NPF event particles **ascend into the stratosphere** within 30 days.
- Two population of (ascending vs. descending) trajectories emerge after ~20 days

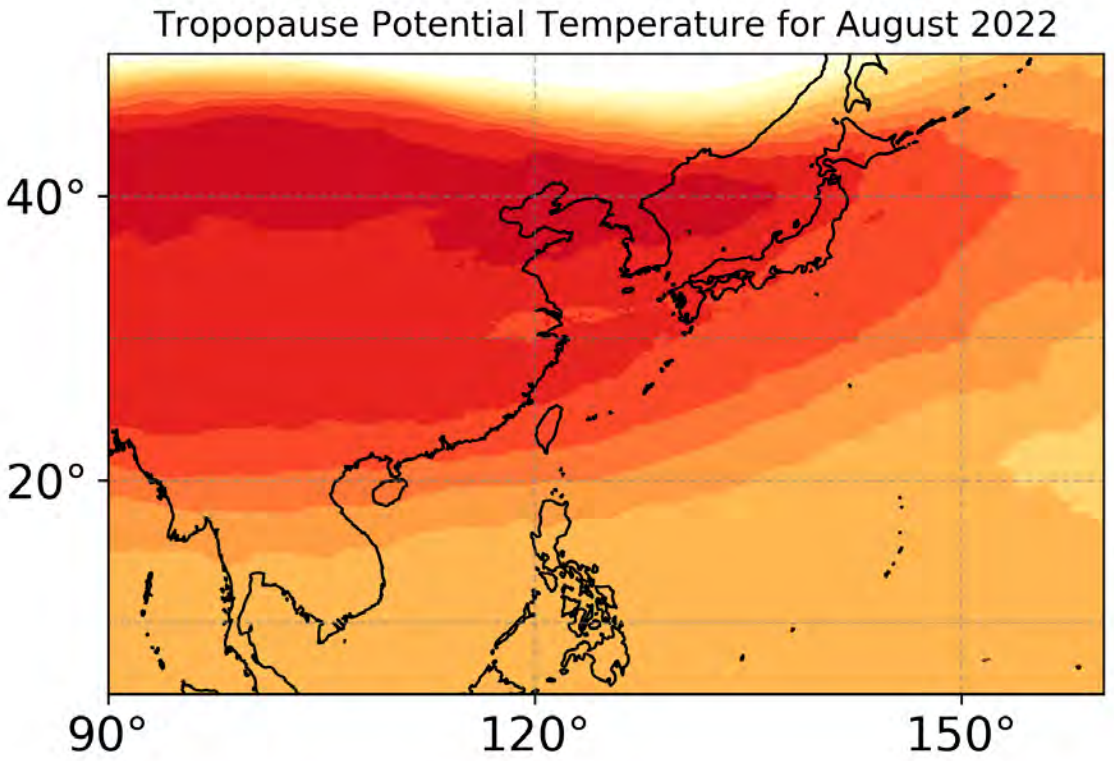
Fate of NPF event particles

Fraction of NPF event particles sampled at a given potential temperature level that ascend into the stratosphere within 30 days

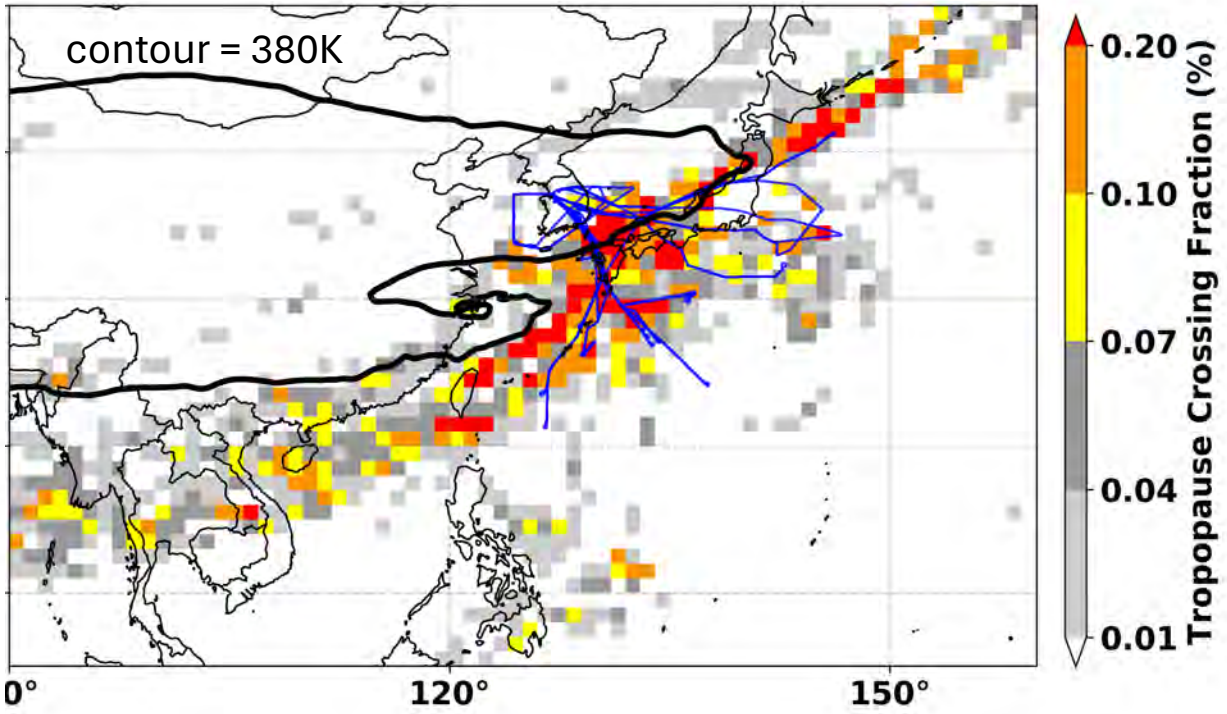


Majority of NPF event particles that form above ~375K will ascend into the stratosphere within 30 days and **contribute to the stratospheric aerosol budget**.

Tropopause over the Asian summer monsoon region

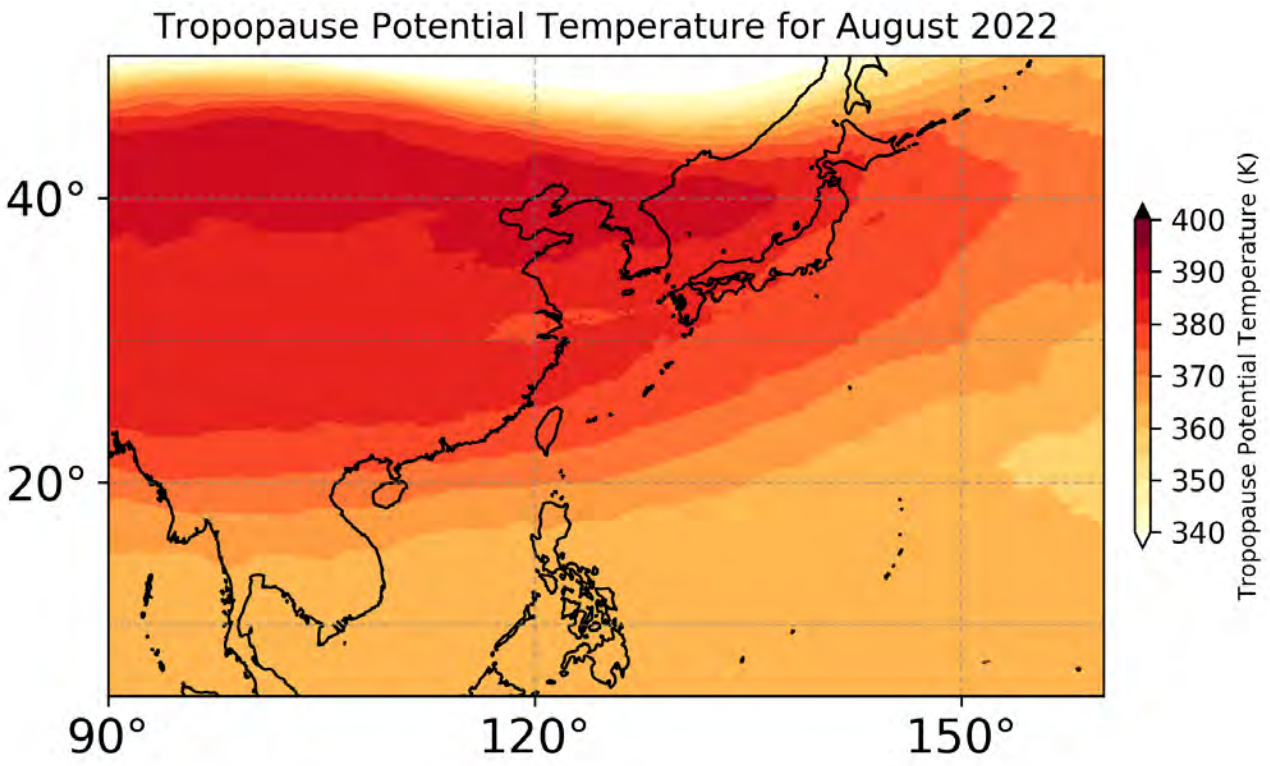


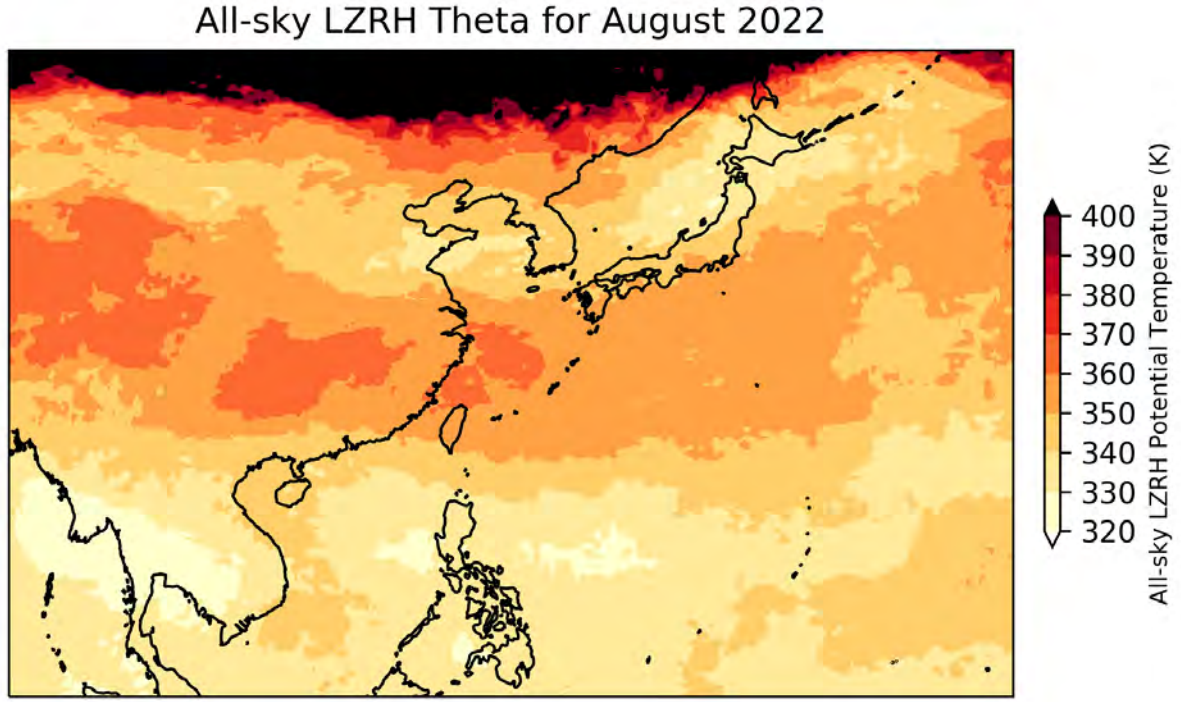
Tropopause-crossing locations of WB-57 sampled parcels (>350K)



- High (380-390K) “bulging” tropopause within the ASM anticyclone (ASMA)
- Tropopause over ACCLIP flight region ~360-385K
- ACCLIP sampled parcels ascend into the stratosphere along the edges of the ASMA: they **follow the anticyclonic flow to the tropical lower stratosphere** and to the **northern mid-to-high latitude stratosphere**.

Tropopause vs. level of zero radiative heating (LZRH)





- Whether NPF event particles ascend or descend depends on the **altitude of NPF event with respect to the level of zero radiative heating (LZRH)**, which is **typically below the tropopause**.
- LZRH over ACCLIP flight region is ~360-370K in ERA5, but **reanalysis heating rates are uncertain** (i.e., LZRH is generally low biased compared to observations).

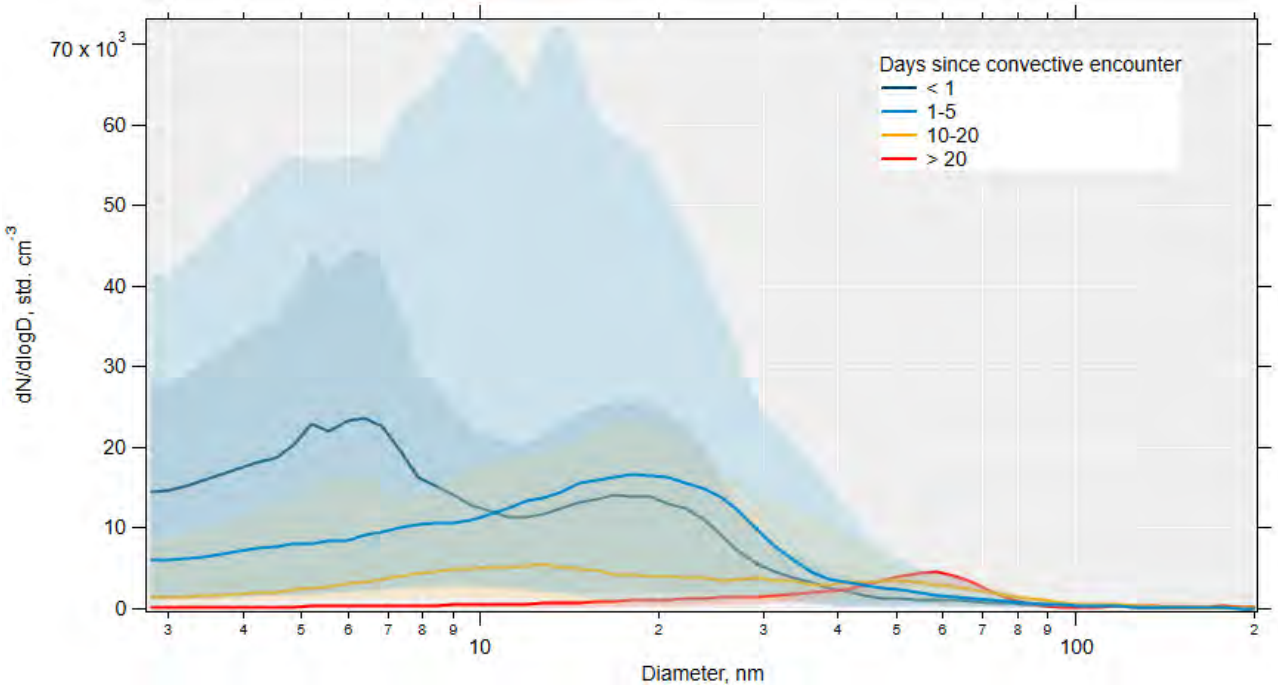
Summary

- Most of the NPF events sampled by WB-57 during ACCLIP occurred **below 360K**.
- About **30% of NPF event particles ascend** after formation, possibly contributing to the stratospheric aerosol budget.
  - **10% of NPF event particles ascend into the stratosphere** within 30 days.
- NPF events that occur **above 375K** over the Asian monsoon region will likely **contribute to the stratospheric aerosol budget**.

Next Steps

- Recalculate forward trajectories with **finer vertical grid** near the LZRH
- Examine the **geographic locations** of NPF events and their tropopause crossings
- Investigate the relationship between **convective influence** history and occurrence of NPF events
- Quantify the sensitivity to reanalysis **heating rates**
- Extend the analysis of NPF events from **other campaigns** (e.g., SABRE, StratoClim, CRAVE, TROCCINOX-2, SCOUT-O3, SCOUT-AMMA)

Size distribution shifts towards larger particles with convective influence age (based on ACCLIP GV data)



Courtesy of Christina Williamson