ACCLIP Science Discussion

In-Situ observation of New Particle Formation in the upper troposphere / lower stratosphere of the Asian Monsoon Anticyclone

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currently in Atmos. Phys. Chem. Discuss.

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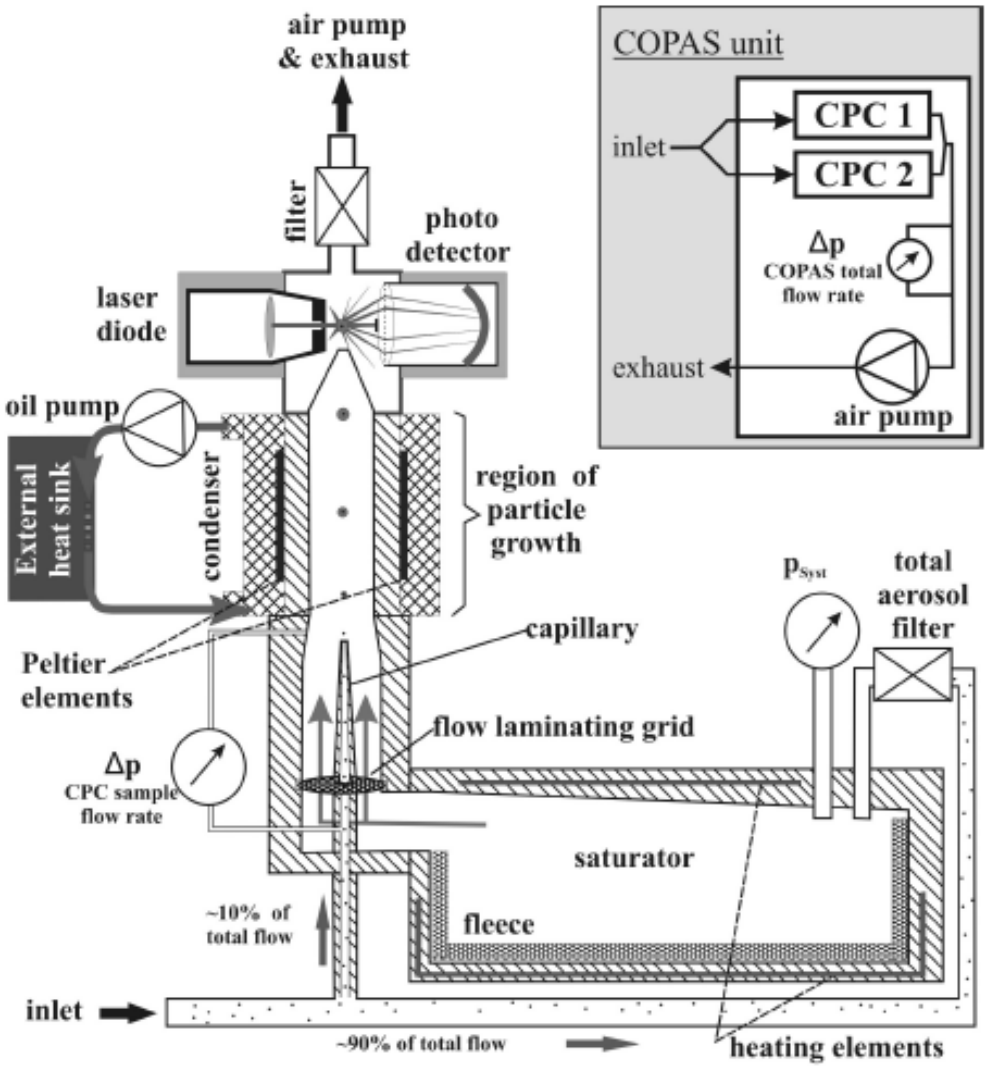
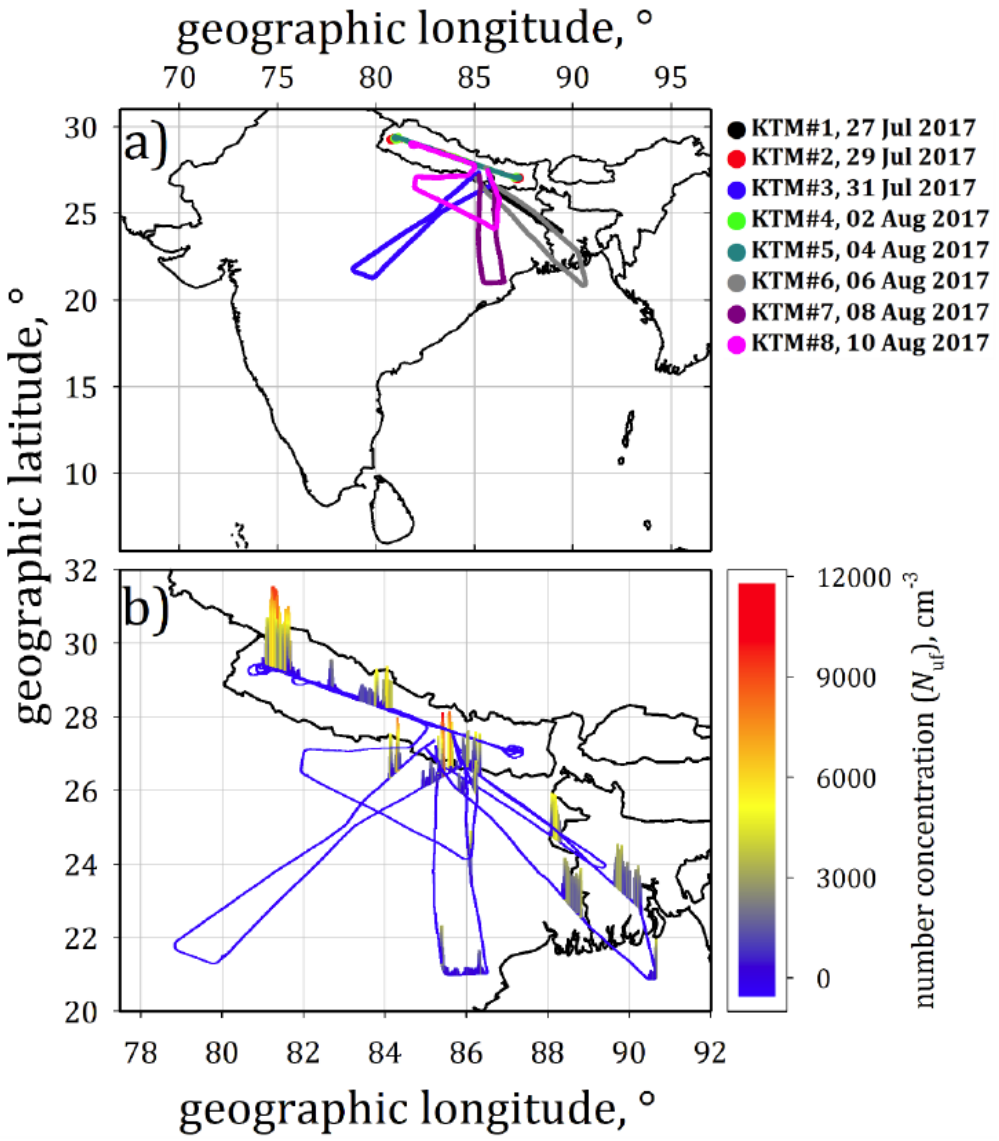
COPAS Measurements



Weigel et al., AMT, 2009

- Custom 2-channel CPC instrument designed for high altitude aircraft operation
- Two COPAS instrument flew on the M-55 *Geophysica* for StratoClim → 4 CPC channels
- Saturator/condensor temperatures set to produce cut-points of
  - 6 nm (n_6) and 15 nm (n_{15})
  - 10 nm (n_{10}) and 10 nm after a 270°C heated inlet ($n_{10,nv}$)
- Combining the channels produces two variables
  - $n_{uf} = n_6 - n_{15}$
  - $f_{nv} = n_{10,nv} / n_{10}$

StratoClim Flights



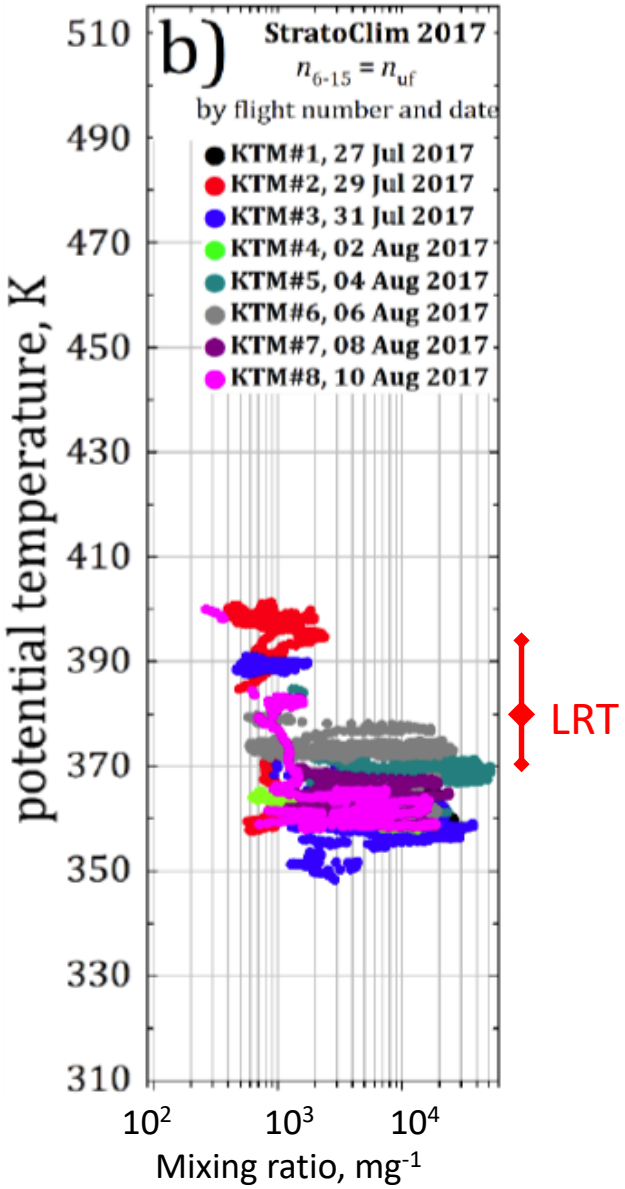
- 8 science flights in July / August 2017
- M-55 *Geophysica* based in Kathmandu, Nepal
- Flights within the Asian summer monsoon anticyclone over Nepal, Bangladesh and northern India
- New particle formation (NPF) was observed during 2.63 hours out of 22.5 total flight hours above 10 km, or nearly 12% of the time
  - Significantly higher frequency than in previous high altitude TTL missions (TROCCINOX, SCOUT-O3, SCOUT-AMMA)
  - n_{uf} mixing ratios up to 5 x 10^4 mg^{-1} were observed

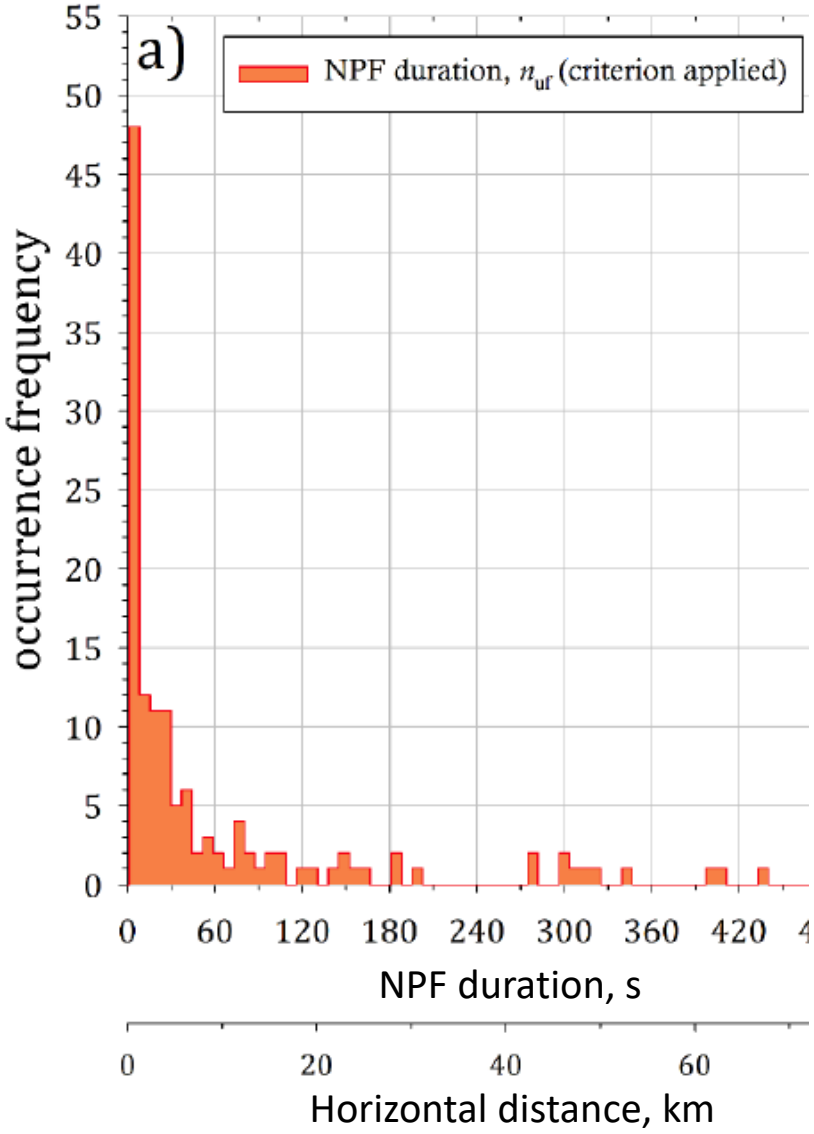
Principal Conclusions

- Highest n_{uf} mixing ratios were observed in air masses with CO > 80 ppb, but n_{uf} not correlated with CO above that level
- Trajectory analysis did not reveal a strong link between observed NPF and
  - Boundary layer source region
  - Rapid transport from the boundary layer
  - Time since convection
- Modeling of particle size distribution indicates rapid coagulation and short (few hours) lifetime of detectability of NPF events by measurement of [ultrafine/nucleation mode] aerosols

- Delay (days) in NPF event from direct convective outflow due to chemical conversion time (e.g. $SO_2 \rightarrow H_2SO_4$, $NO_x \rightarrow HNO_3$, organic oxidation)?
- Gravity wave induced cooling responsible for producing conditions for NPF?
  - Spatial extent of NPF events
  - Weigel et al 'New particle formation inside ice clouds: In-situ observations in the tropical tropopause layer of the 2017 Asian Monsoon Anticyclone', ACPD, reports that ~1/2 of StratoClim NPF observations occurred within cirrus clouds
- NPF in the AMA UT/LS may contribute significantly to the establishment and maintenance of the ATAL

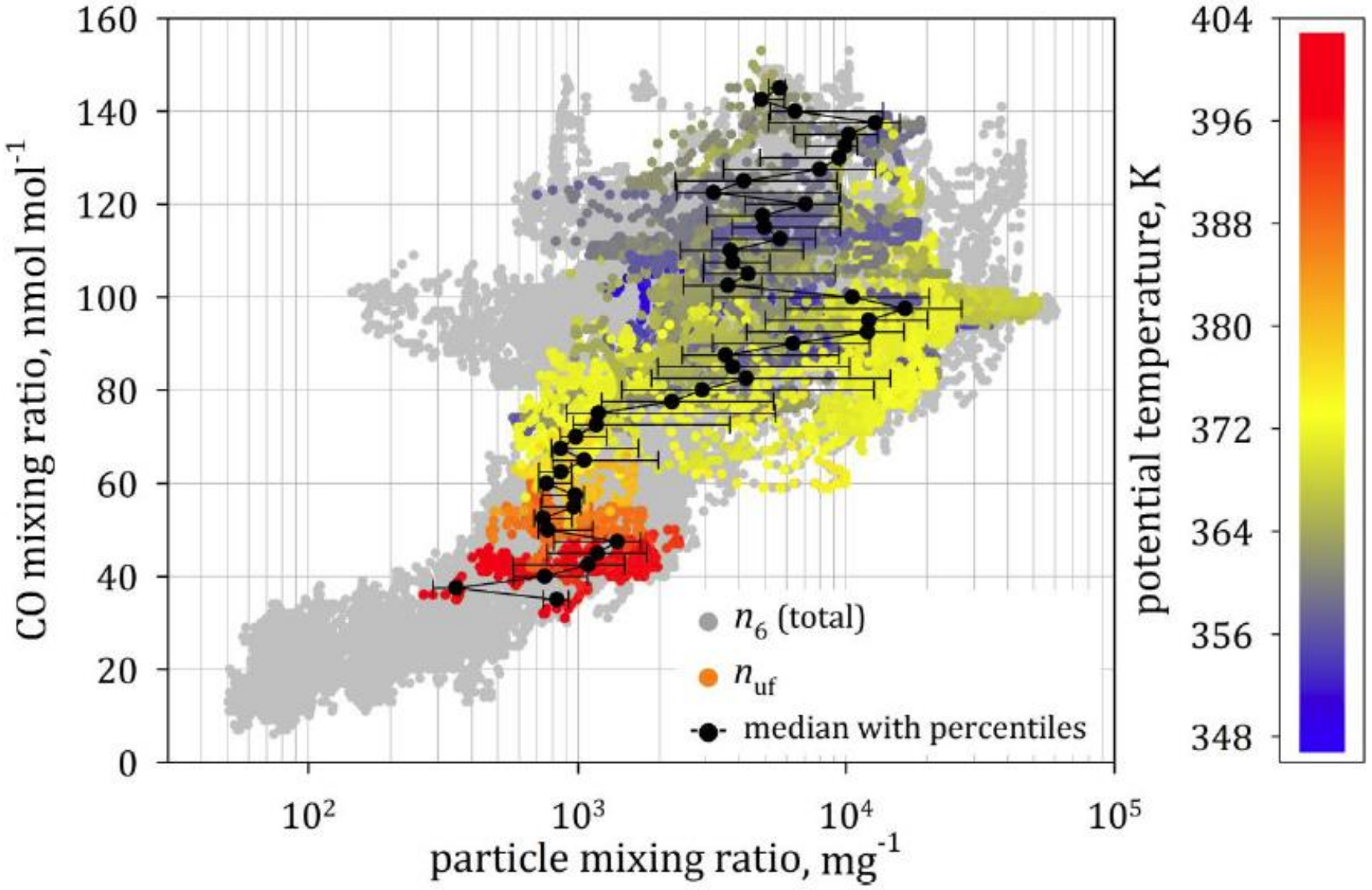
StratoClim AMA NPF





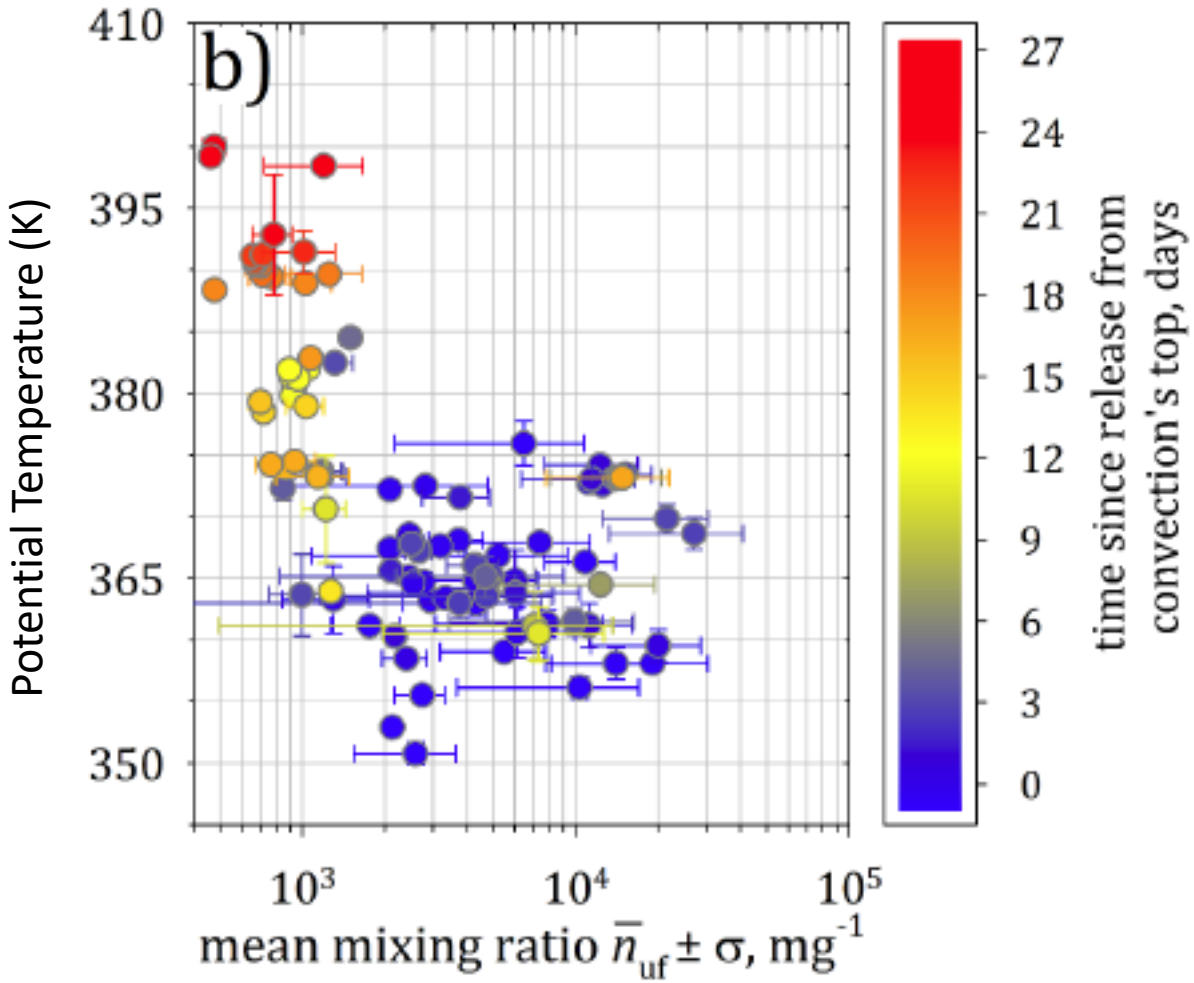
- Highest intensity (concentration) NPF events observed just below the lapse rate tropopause (LRT)
- Lower intensity NPF observed up to 400 K
- Most NPF events had spatial scales < 10 km, but a few were more extensive

Carbon Monoxide



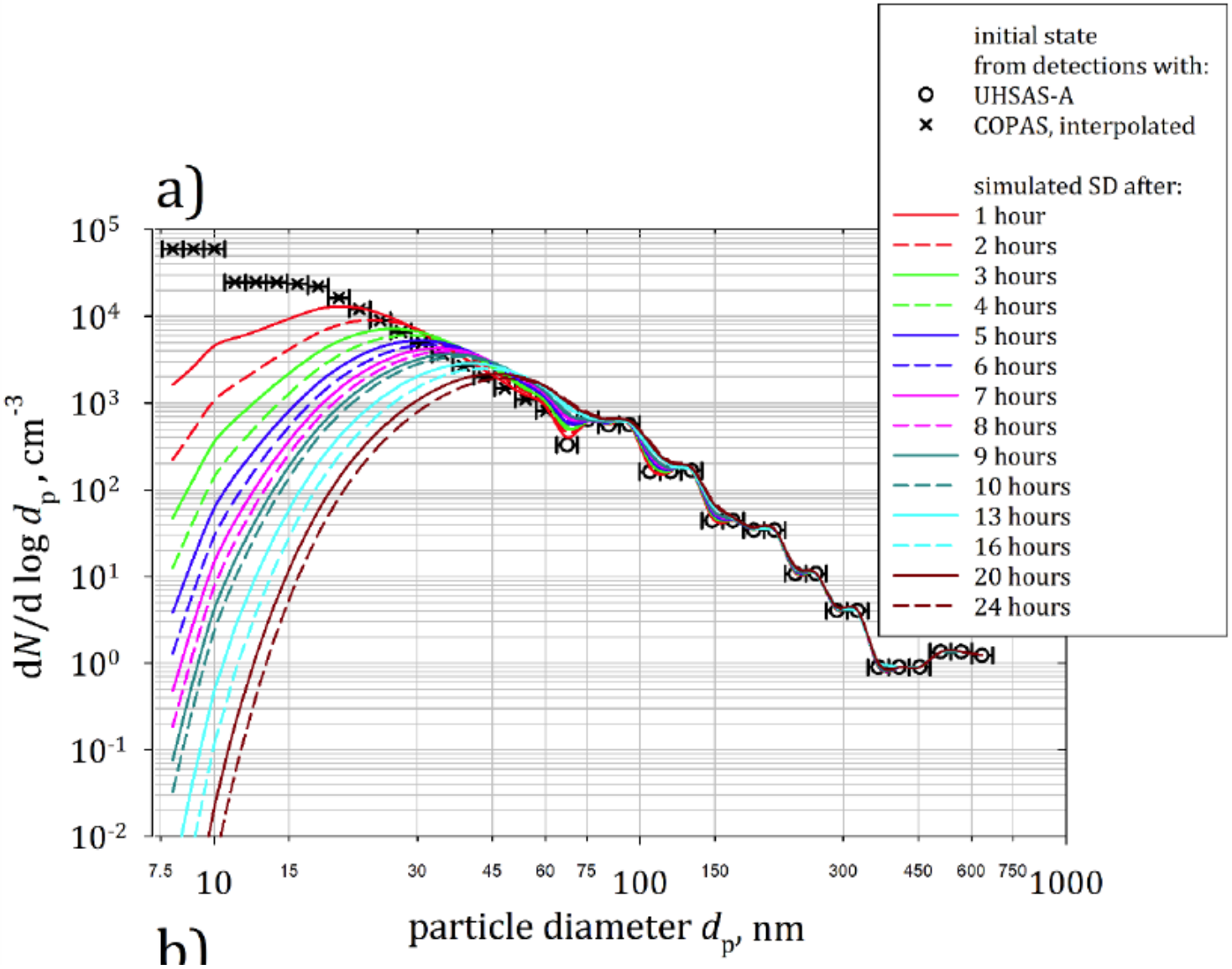
- UT mixing ratios of CO in the AMA 70 – 150 ppb
- Mixing ratios decrease slightly above the level of convective outflow, more significantly above the LRT
- NPF intensity in the UT does not appear to be correlated with CO

Convective Influence



- AMA UT experiences frequent deep convection
- Below the LRT, NPF time since convective influence appears to be typically < 4 days, but with an extended tail to as many as ~15 days
- Above the LRT, NPF events have lower n_{uf}, but these events are seen at times out to > 20 days since convective influence
- LRT-relative coordinates might sort intermediate Θ points into shorter and longer times since convection

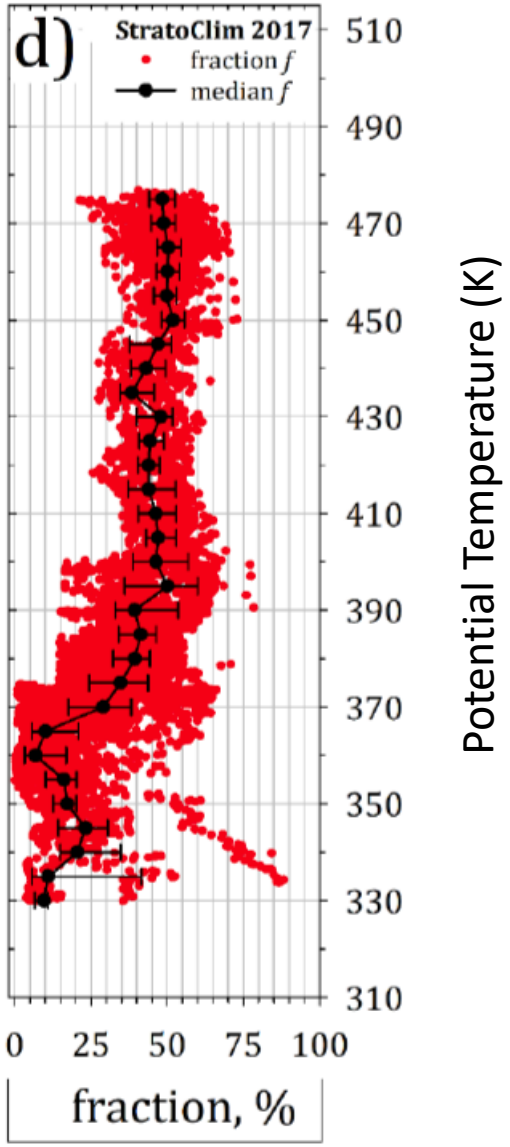
Modeling NPF Evolution



- PSD (6 – 1000 nm) measured by COPAS + UHSAS
- Case from 4 Aug: 26 s duration, P = 110 hPa, T = 196 K
- SOCOL model discretized coagulation
- n_{uf} drops rapidly due to coagulation and identification of a NPF event from n_{uf} observations possible for only a few hours

➢ StratoClim observations of NPF frequency and short detection time imply that NPF events are occurring frequently in the AMA

ATAL Aerosol Volatility



- Above 390 K (above the LRT), f_{nv} is typically (median) 45-50%
- f_{nv} lower in the UT
- Between 350 – 370 K, level of deep convective outflow and most frequent intense NPF, f_{nv} very low (15 nm particles don't tend to leave 10 nm non-volatile cores)
- SO_4 and NO_3 likely quantitatively removed by 270°C inlet (residence time?)
- For mixed organic-nitrate or organic-sulfate particles, sufficient organics likely survive the heated inlet to provide a 10 nm non-volatile core
- Would be interesting to see $n_{10,nv}$ / n_{60} (UHSAS) and compare with single particle composition observations

Implications for ACCLIP

- Transport time of air from the interior of the AMA (where NPF was observed) to the ACCLIP operations region likely means little to no NPF expected to be observed in ACCLIP in the absence of additional convective influence during shedding/transport
  (not surprising)

- To the extent that NPF in the AMA UT indicates the presence of aerosol mass pre-cursors, StratoClim observations suggest that there will not necessarily be a CO – aerosol correlation within the AMA shed airmasses
  (interesting to compare obs to model CO – aerosol correlations…)