CONTRASTING THE INFLUENCE OF EAST AND SOUTH ASIA ON THE CHEMICAL COMPOSITION OF THE ATAL DURING ACCLIP

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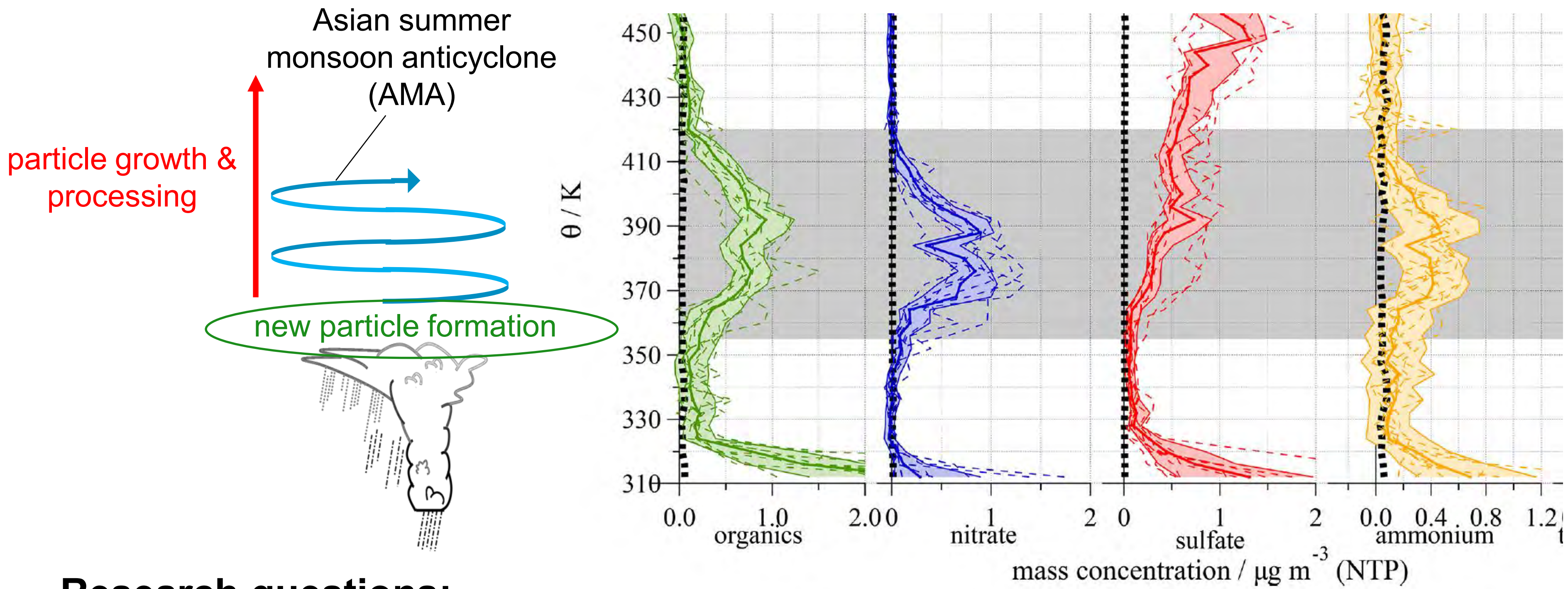
[4]NASA Ames Research Center, Mountain View, CA, USA

MOTIVATION



Appel et al., ACP, 2022;
Höpfner et al., Nat. Geosc., 2019

Asian tropopause aerosol layer (ATAL)

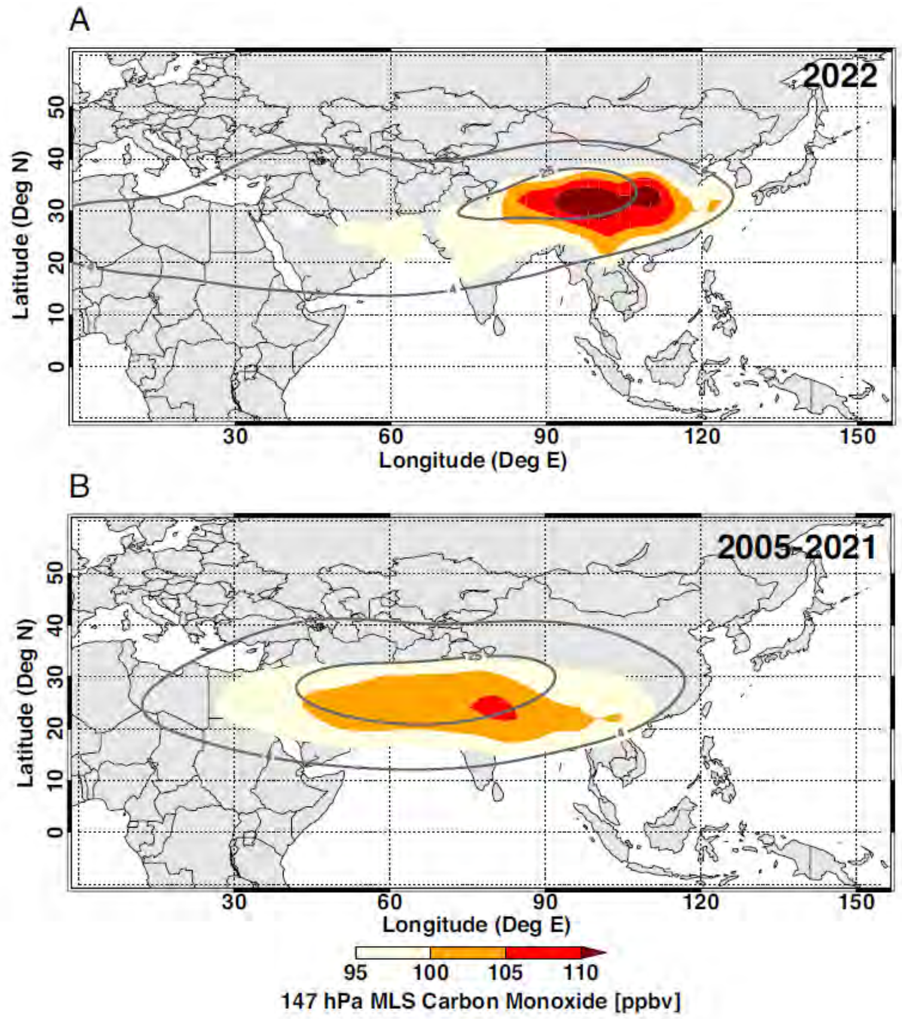
Research questions:

- **What is the chemical composition of the ATAL in the Western Pacific outflow region?**

MOTIVATION

Special monsoon situation for ACCLIP 2022, eastward-shift of the AMA:

- Large fraction of polluted BL air from East China flooding the AMA
- High CO values in the Asian UT, along with record-breaking Cl-VSLS, e.g., CH_2Cl_2 (Pan et al., 2024)



Pan et al., PNAS, 2024

Research questions:

- **What is the chemical composition of the ATAL in the Western Pacific outflow region?**
- **Does the different monsoon situation affect the ATAL composition?**

ERICA HYBRID MASS SPECTROMETER

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ERC Instrument for Chemical composition of Aerosols (ERICA)

Combination of:

- Laser ablation mass spectrometer (ERICA-LAMS)
  - ➢ Single particle composition including non-refractory & refractory compounds
  - ➢ Data output: Size and fraction of different particle types
  - ➢ Size range: ~180 nm – 3 µm

ERICA-rack inside the HIAPER-GV aircraft

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- Flash vaporization/ electron impact ionization mass spectrometer (ERICA-AMS)
  - Composition of particle ensembles (non-refractory species)
  - Data output: Mass concentrations of particulate nitrate, sulfate, ammonium and organics
  - Size range: ~110 nm – 3 µm

ERICA-rack inside the HIAPER-GV aircraft

ACCLIP 2022 CAMPAIGN

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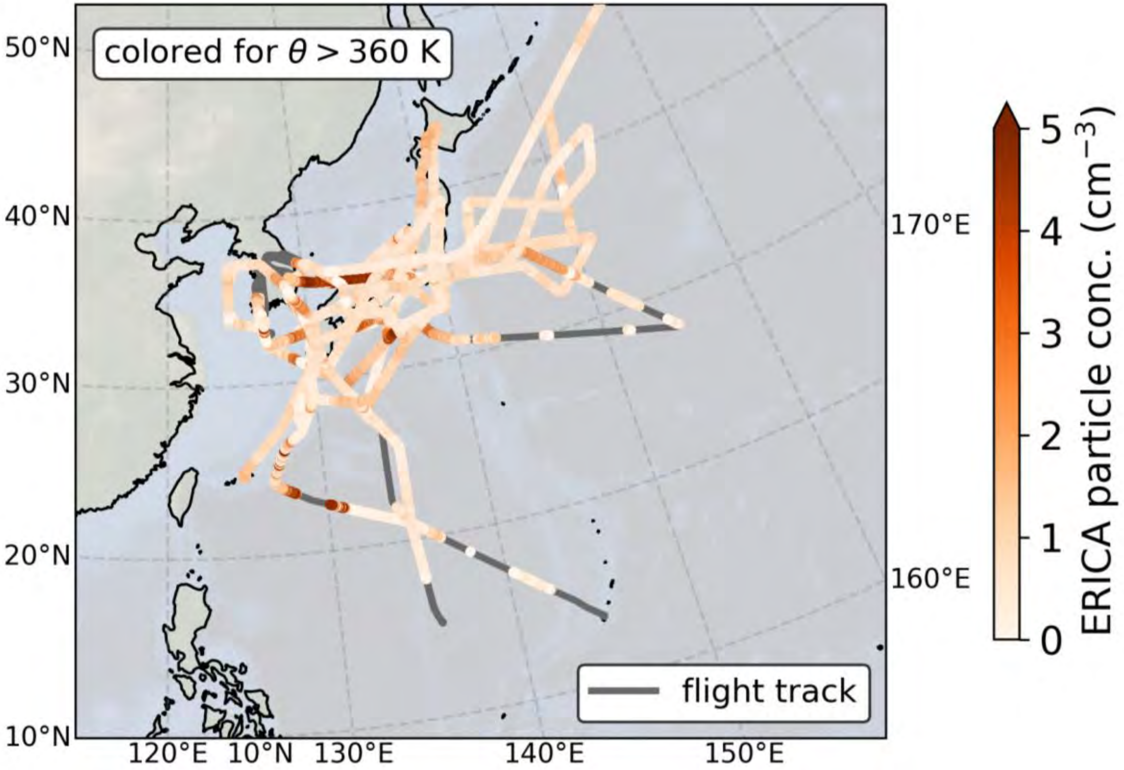
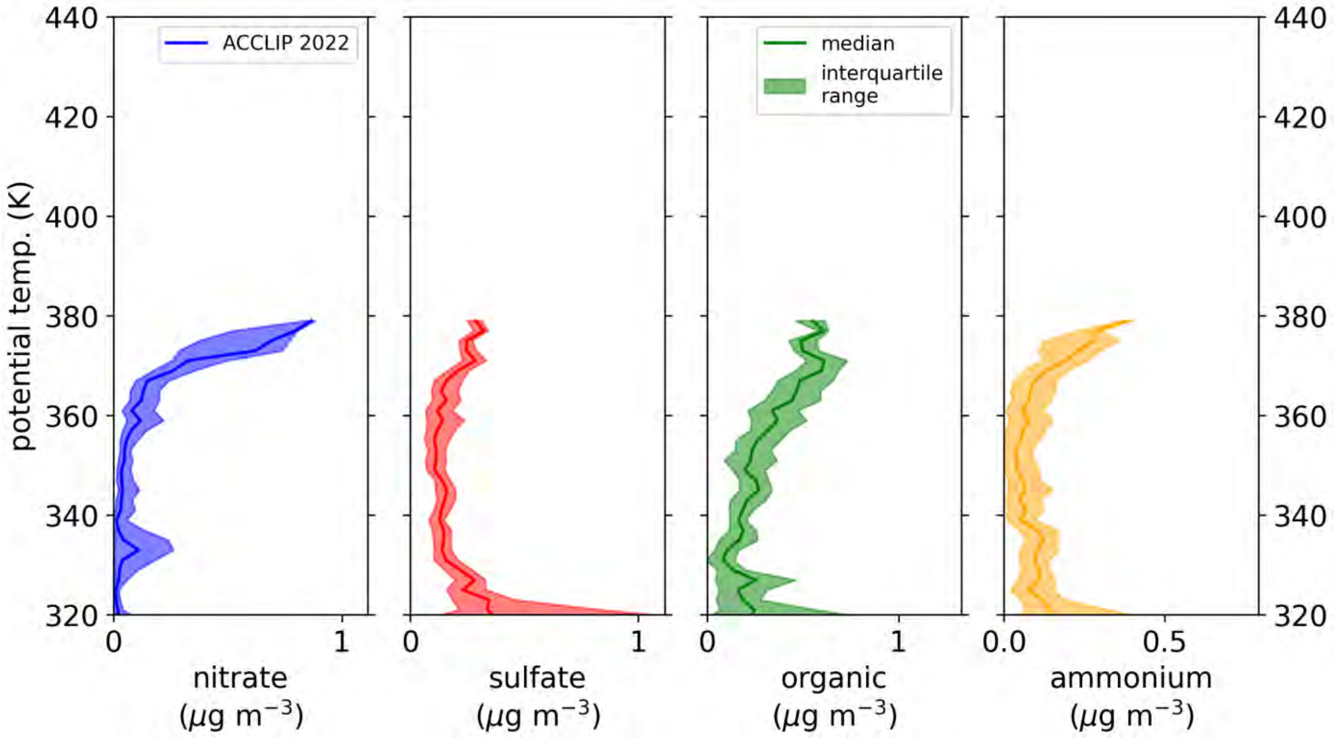
- ERICA onboard of HIAPER-GV aircraft
- 12 local research flights in the Western Pacific region
- Features of elevated aerosol concentrations detected by ERICA (accumulation mode) above 360 K
  - ➢ Indications of sampling periods within the ATAL

HIAPER GV aircraft



Map of HIAPER GV flights during ACCLIP 2022

ATAL COMPOSITION DURING ACCLIP



Median and interquartile range of ERICA-AMS species

ATAL COMPOSITION DURING ACCLIP

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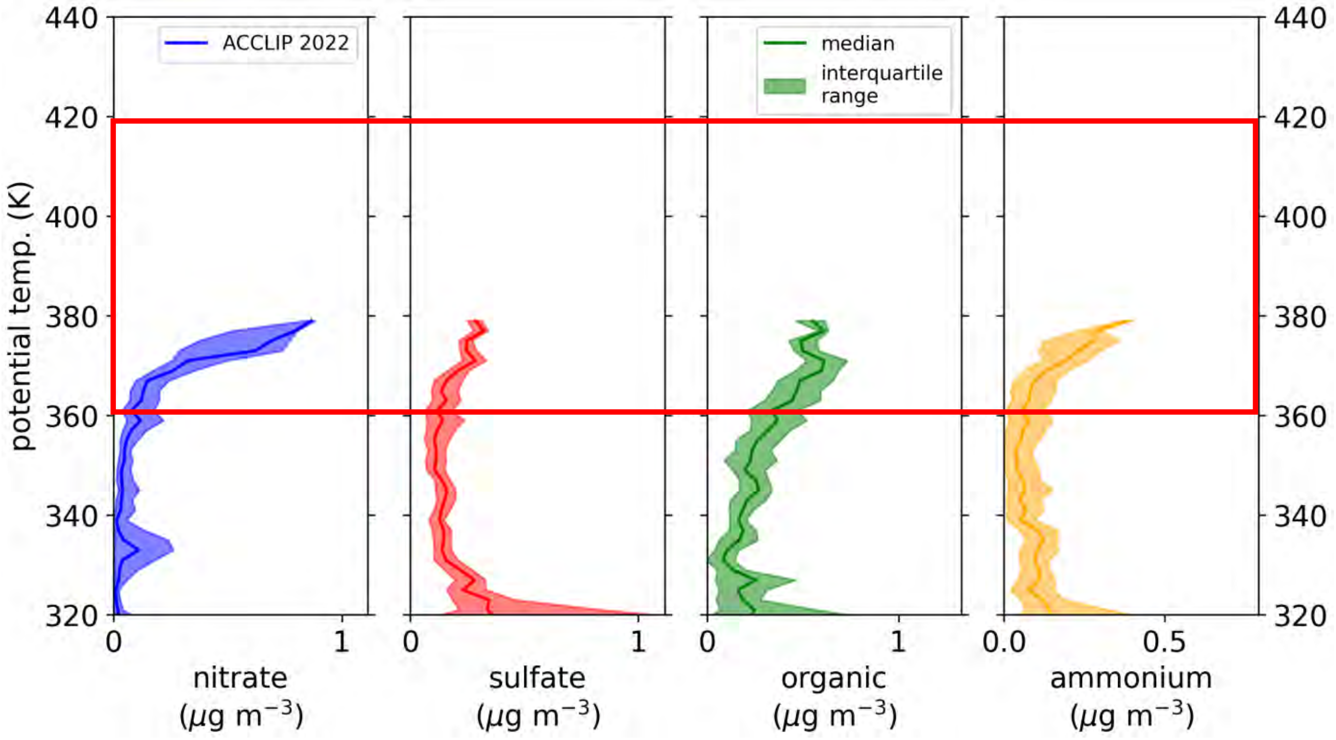
- Increase of non-refractory aerosol mass above ~360 K
- Indications of the ATAL (or its lowermost part)



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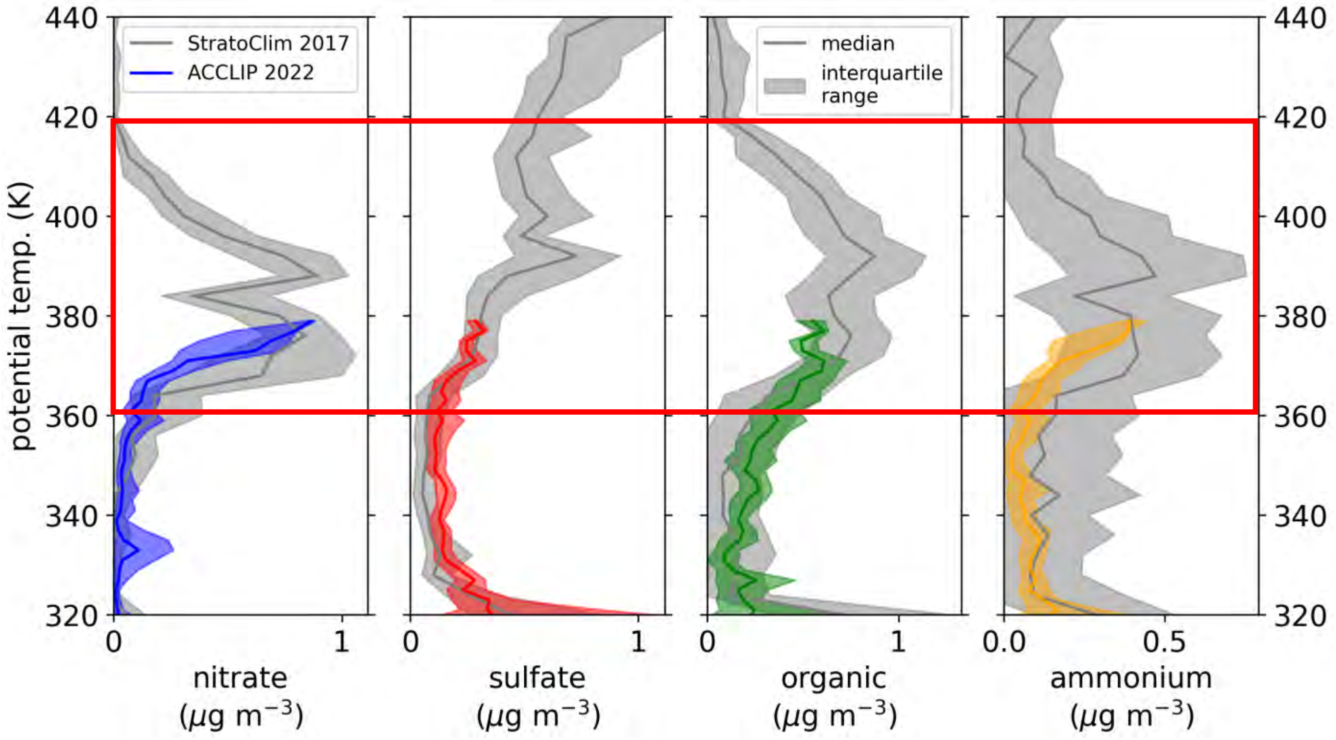

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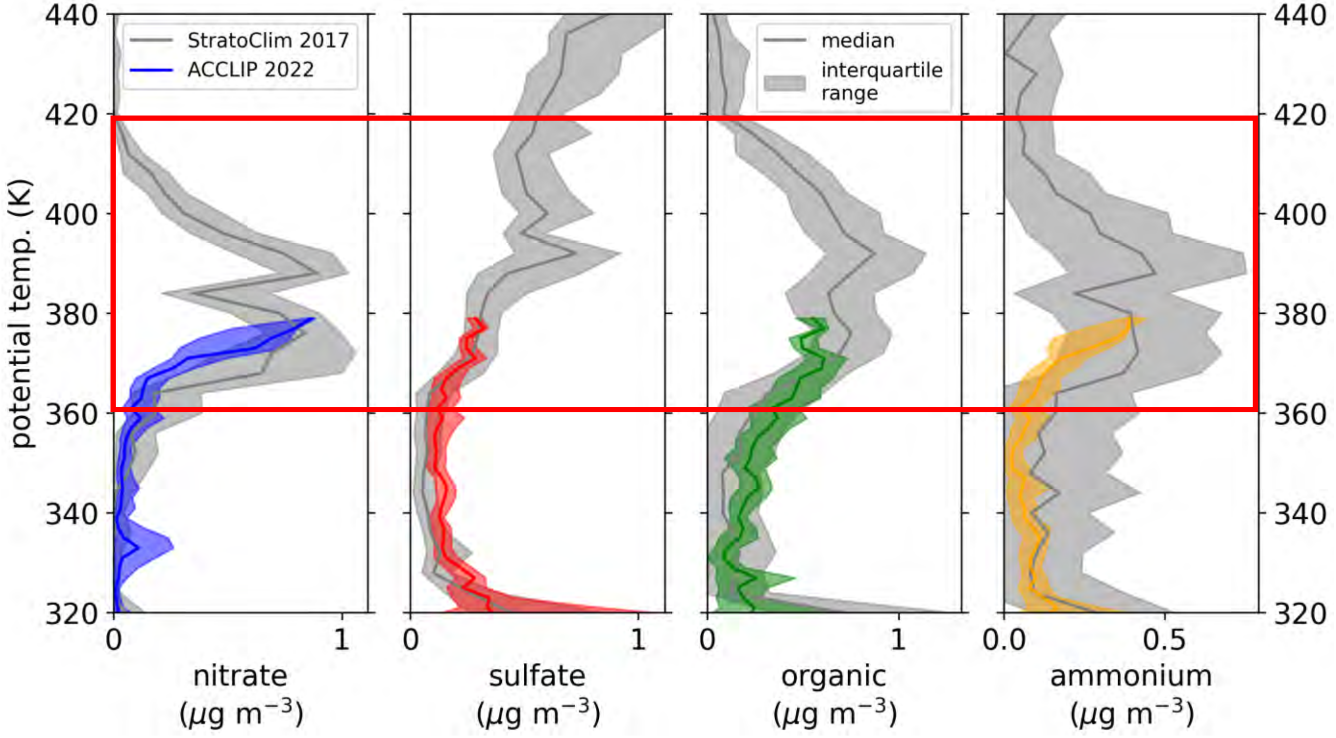
Different monsoon situation in 2022 (Pan et al, 2024),

effect on the aerosol composition?



Median and interquartile range of ERICA-AMS species

MOST RECENT CONVECTIVE INFLUENCE

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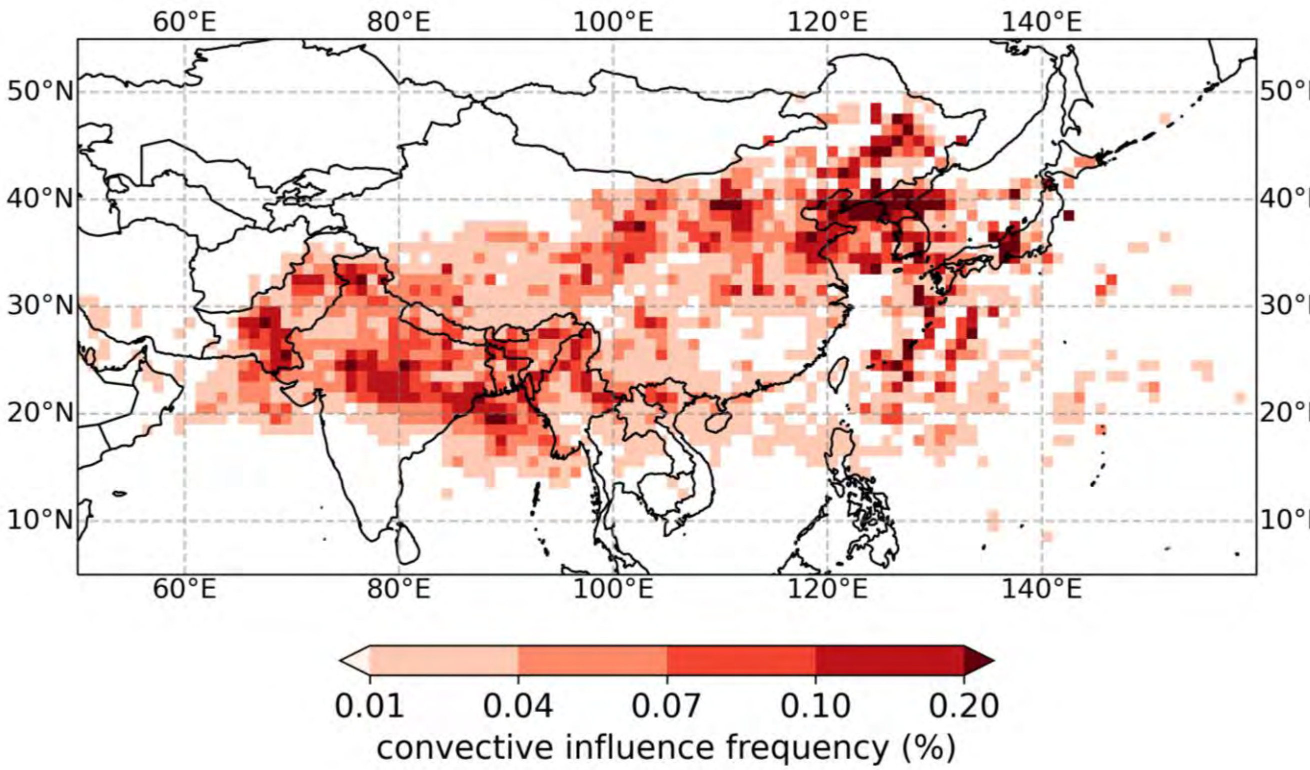
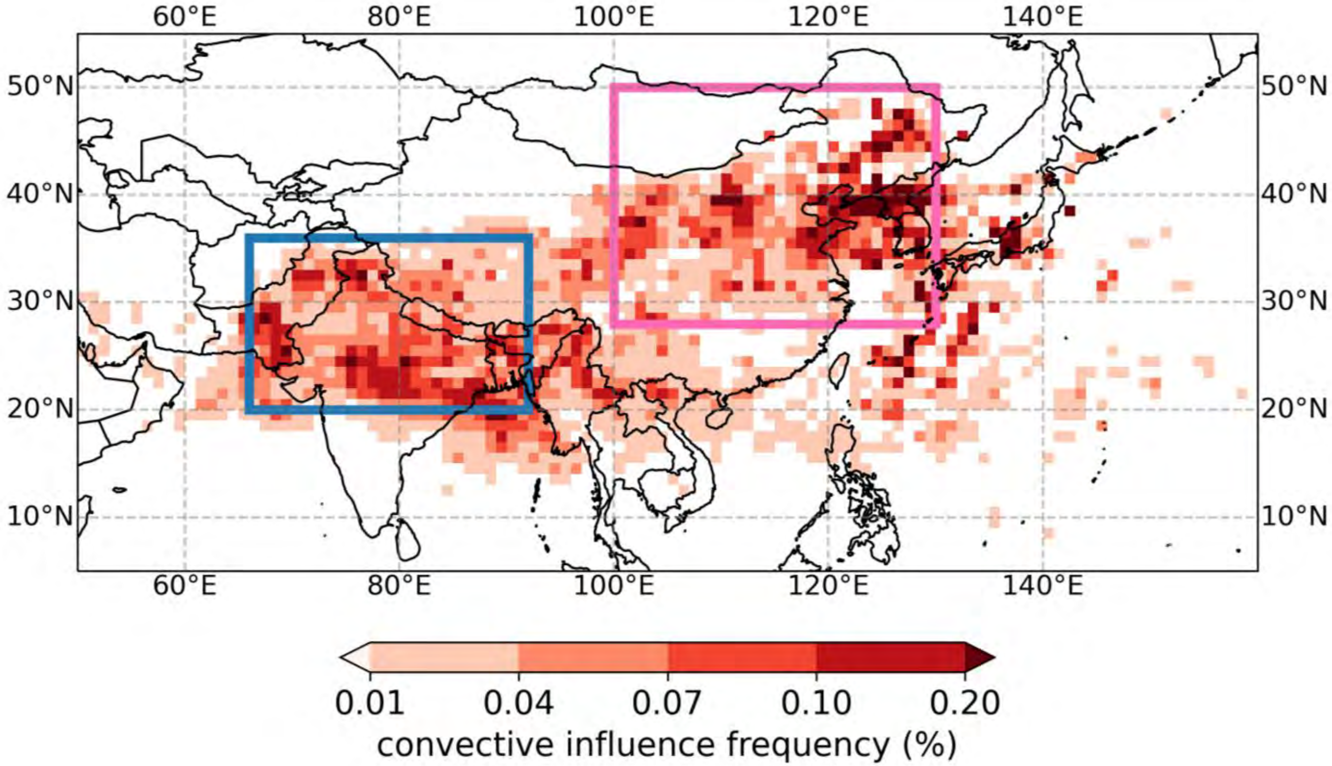
- Use trajectories-based data product to separate air mass origin
- Two major origin regions identified for air masses sampled above 350 K



Recent convective influence locations derived from back-trajectory product (TRAJ3D, satellite-derived convective cloud tops)

MOST RECENT CONVECTIVE INFLUENCE

- Use trajectories-based data product to separate air mass origin
- Two major origin regions identified for air masses sampled above 350 K
- Selection of two boxes:
  - South Asia
  - East Asia



Recent convective influence locations derived from back-trajectory product (TRAJ3D, satellite-derived convective cloud tops)

COMPARISON OF NON-REFRACTORY SPECIES

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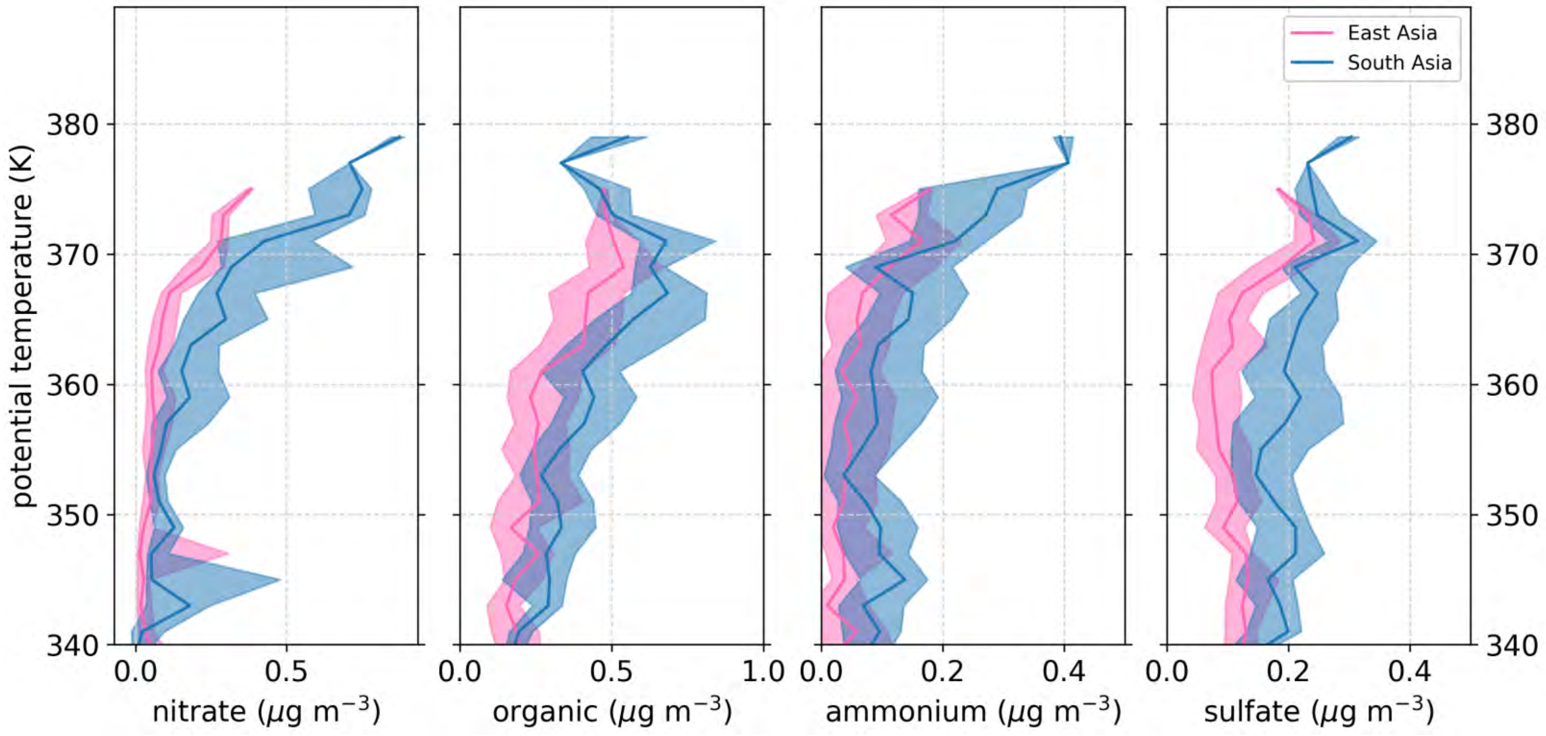
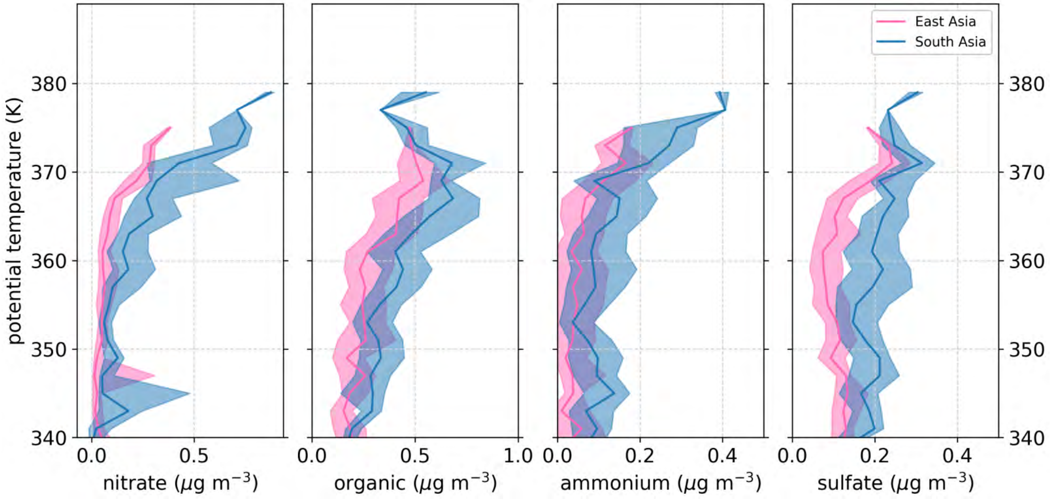
- South Asia: More contribution to aerosol mass concentrations
- East Asia: Increase of mass concentrations occurred at higher potential temperatures

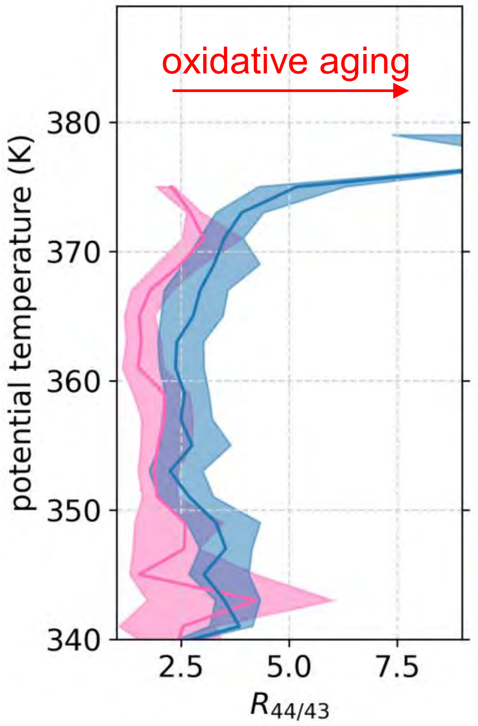


Median and interquartile range of ERICA-AMS species

COMPARISON OF NON-REFRACTORY SPECIES

- Oxidative aging marker $R_{44/43}$: Less oxidized organics from East Asian convection

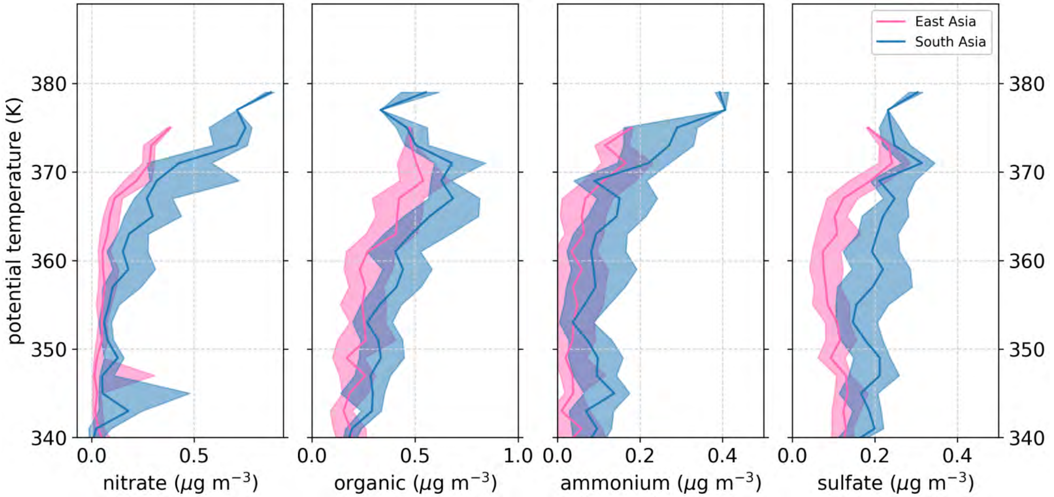


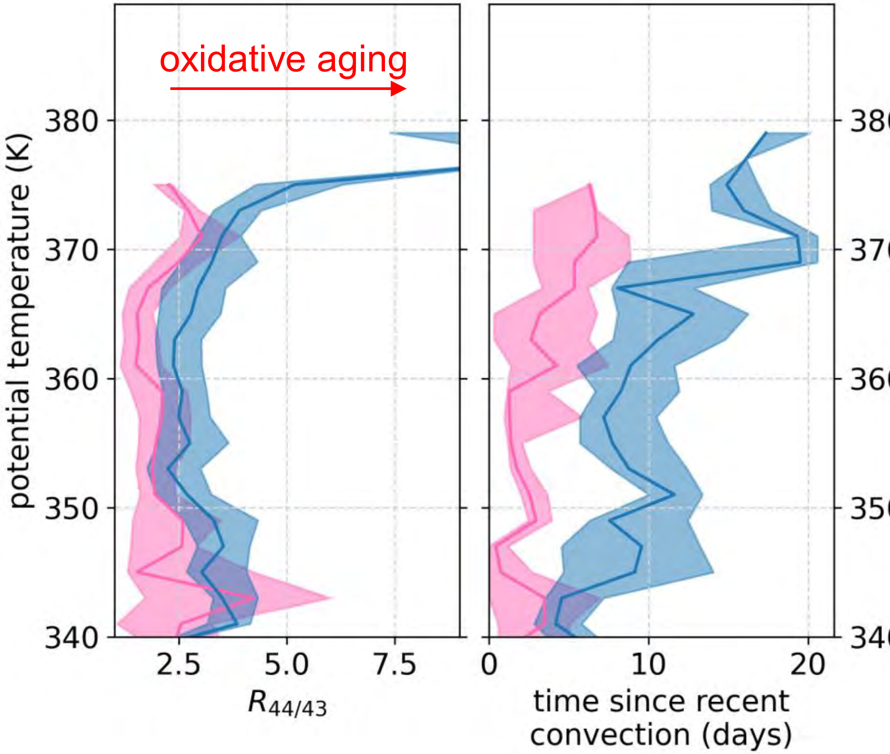


Vertical profile: median and interquartile range

COMPARISON OF NON-REFRACTORY SPECIES

- Oxidative aging marker $R_{44/43}$: Less oxidized organics from East Asian convection
- Further supported by trajectory time since recent convective encounter





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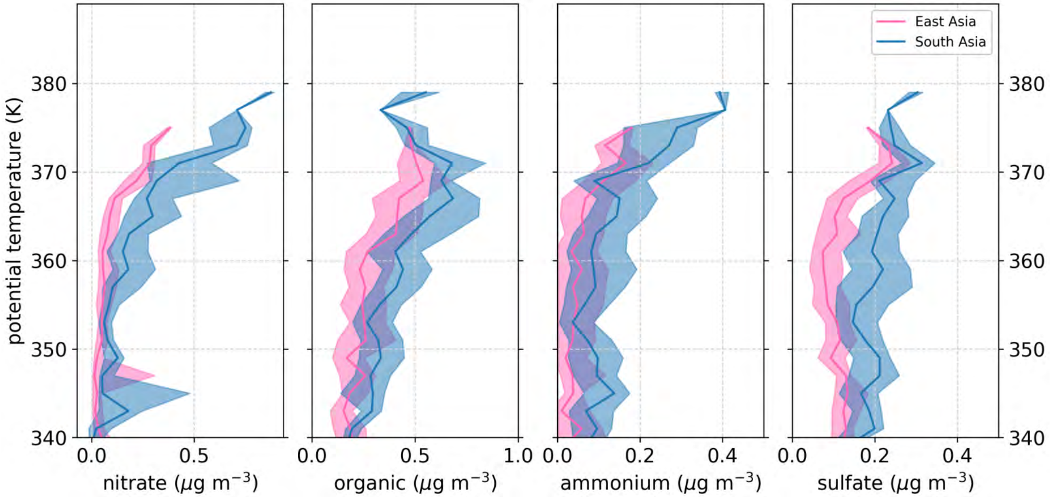

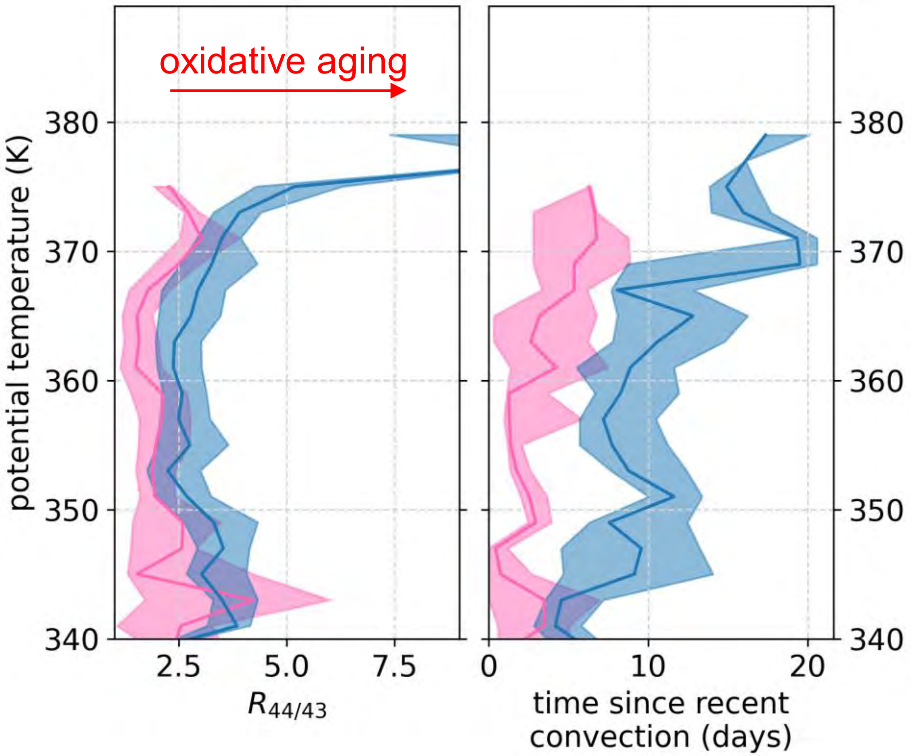

longer transport times from South Asia

→ more time for growth & processing

Vertical profile: median and interquartile range

NO-RICH PARTICLE TYPES FROM ERICA-LAMS

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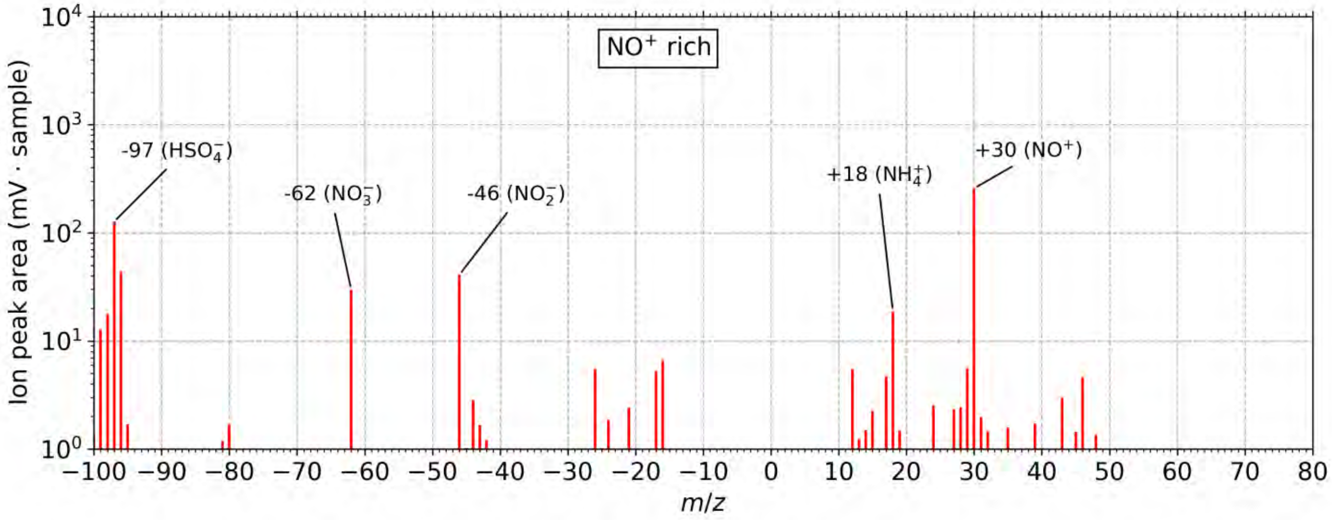
Two clusters identified in the single-particle measurements by ERICA-LAMS associated with the ATAL (Appel et al., 2022):

- NO^+ rich:

    nitrate, sulfate and ammonium signals



Mean mass spectrum for NO^+ rich cluster

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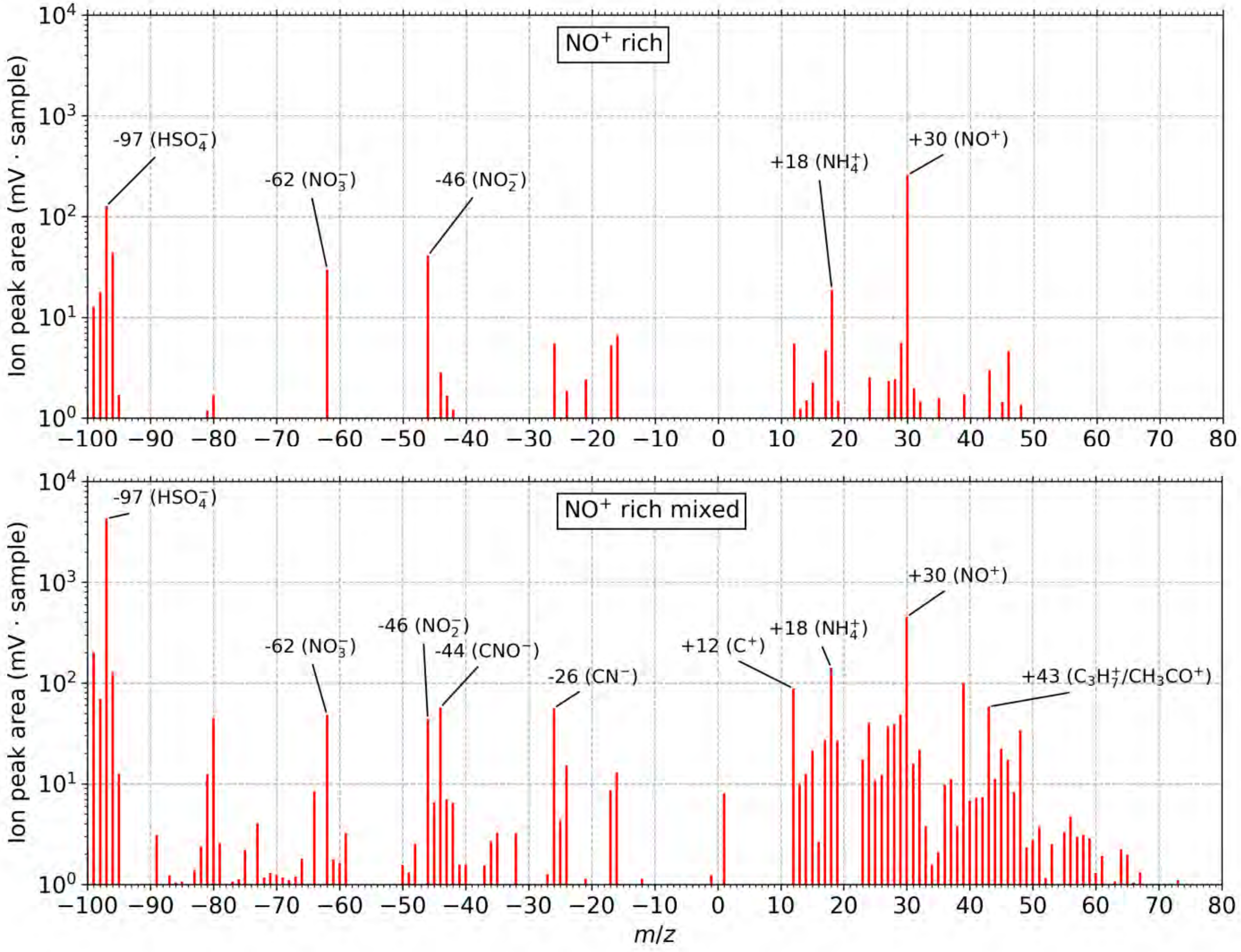
- NO^+ rich mixed:

  similar to NO^+ rich, but significantly larger sulfate and organics signals



Mean mass spectrum for NO^+ rich cluster (top) and NO^+ rich mixed cluster (bottom)

COMPARISON OF NO-RICH TYPES

- Now: Considering **7 – 10 days** since recent convective encounter

COMPARISON OF NO-RICH TYPES

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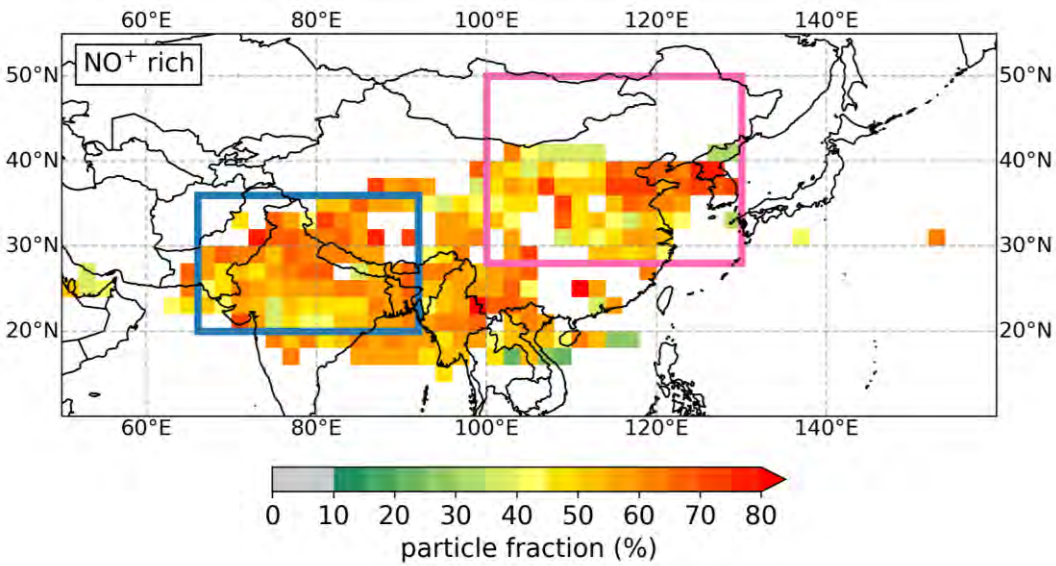

Map with particle fraction from different convective influence region (filtered by 7 – 10 days since recent convection)

COMPARISON OF NO-RICH TYPES

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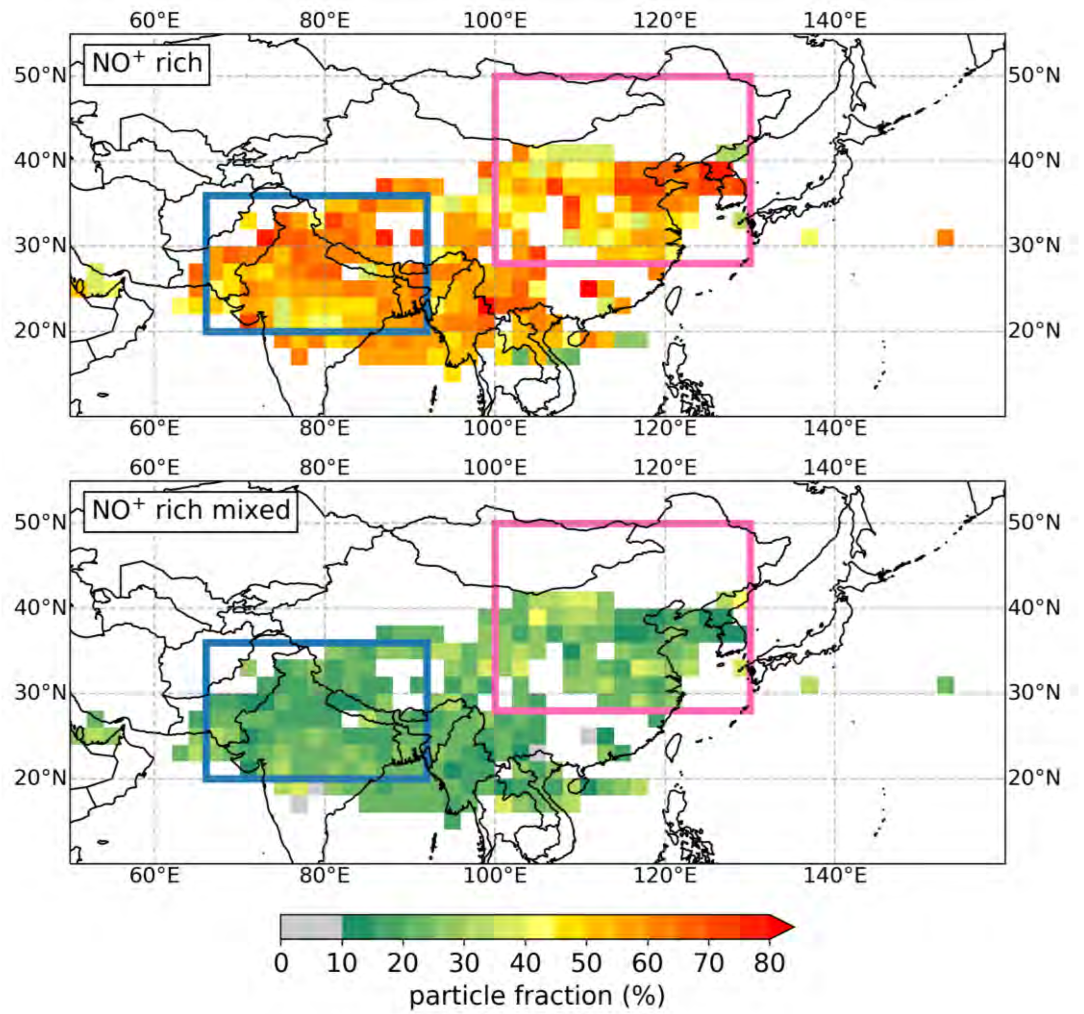
- Two hotspots for NO^+ rich particles (Northern India, East Asian subtropical front convection)
- Mixed type:
    - Predominantly from North/East China
    - Minimum in Northern India, in contrast to more 'pure' NO^+ rich type



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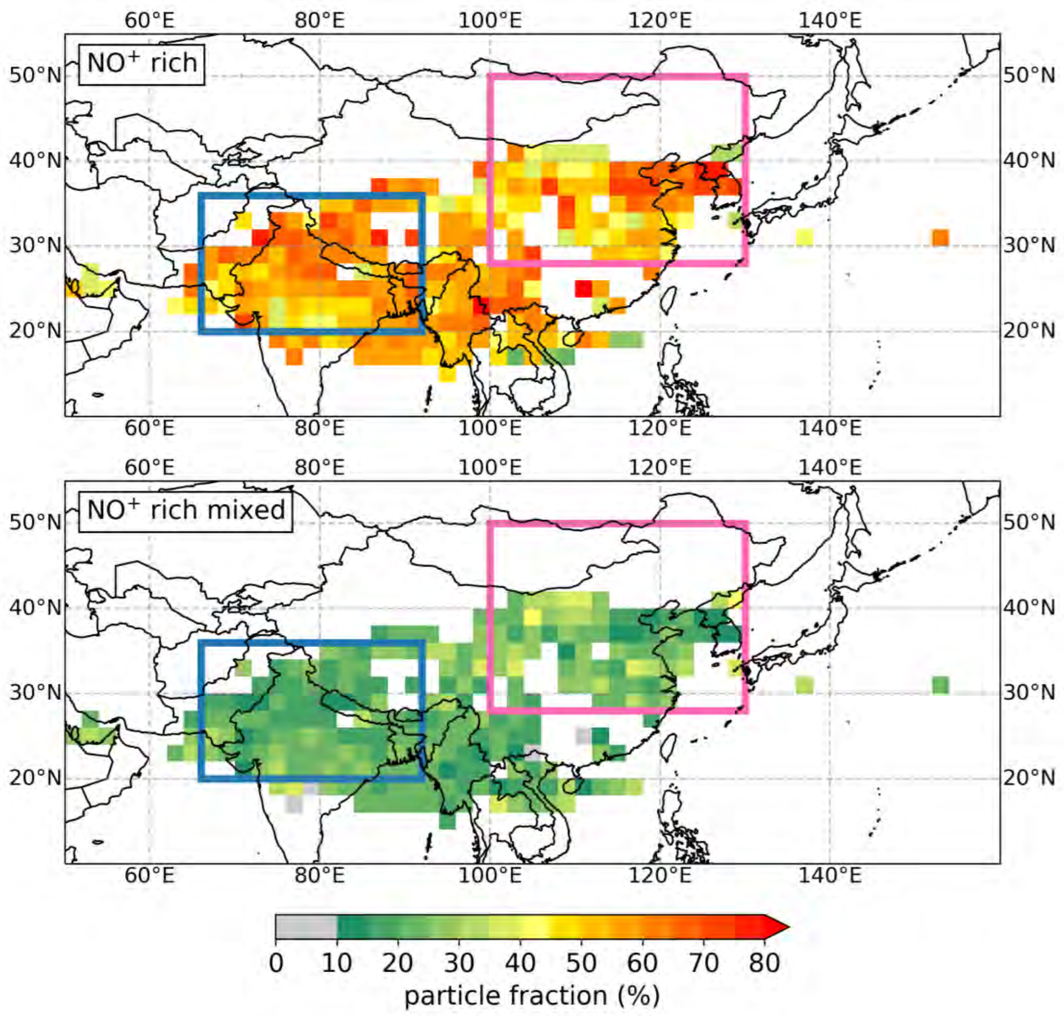
Indications of increased organic and sulfate contribution from East Asia



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EDDY SHEDDING OF AEROSOL LAYER

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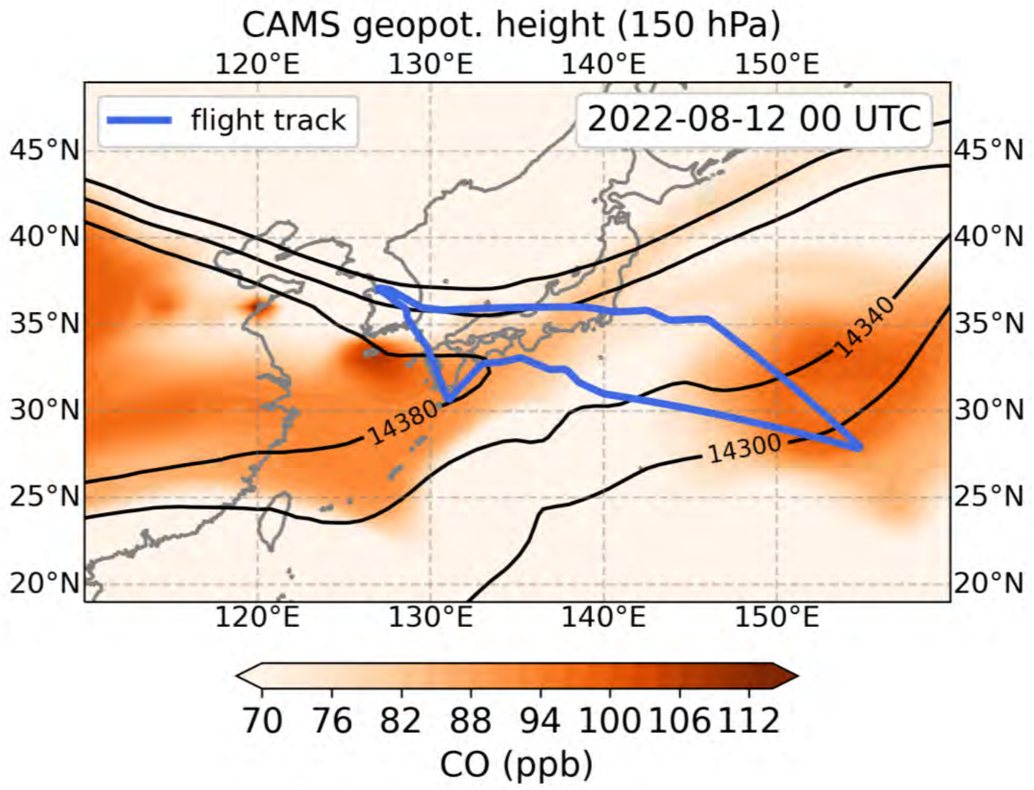
- Exemplary flight: RF05 (8/12/2022)
- Transecting two separate shed air masses



Geopotential height (contour lines; in gpdm) and CO mixing ratio for RF05 from CAMS global reanalysis (EAC4) from Copernicus.

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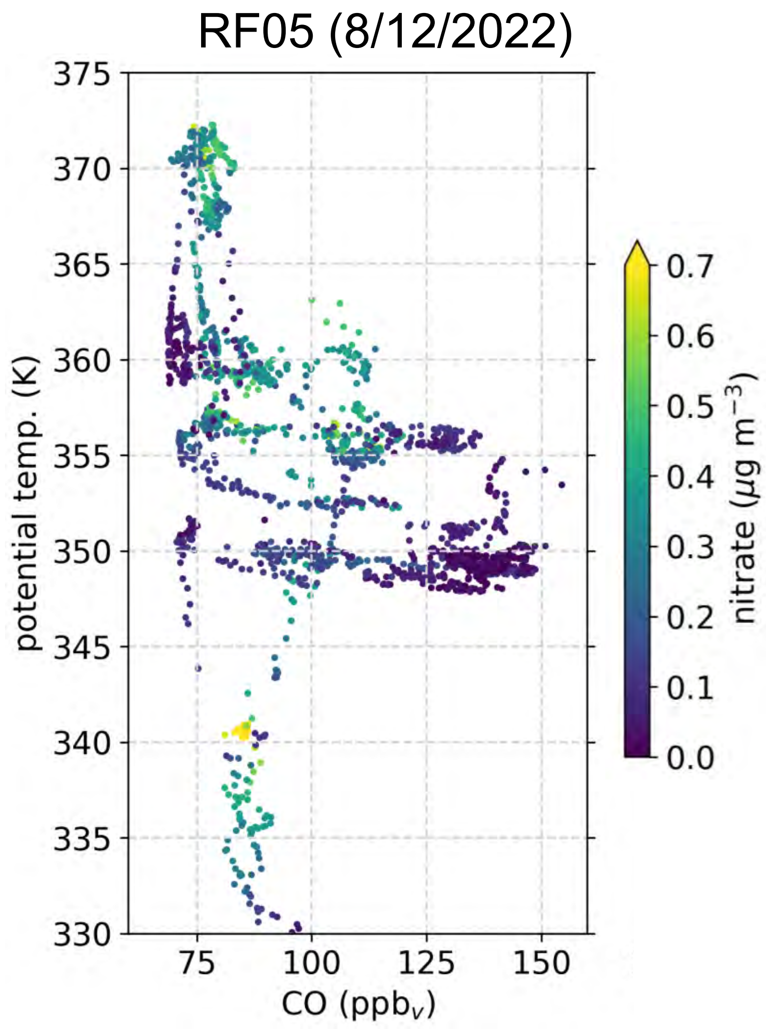
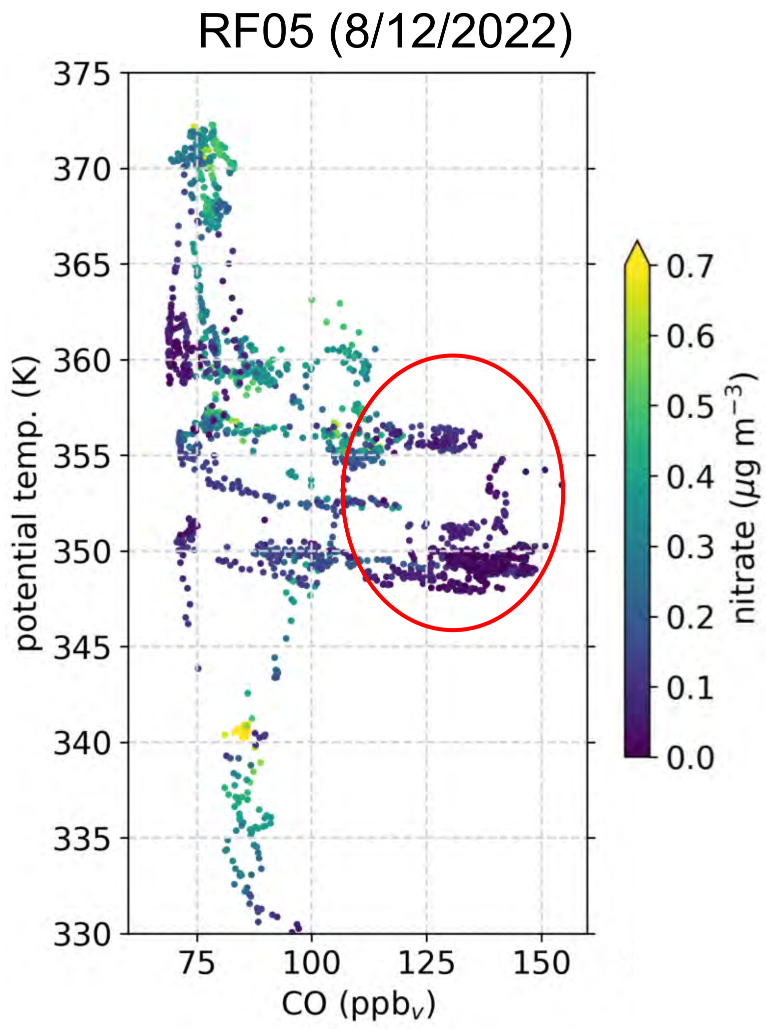


CO data from Aerodyne CS-108 miniQCL (courtesy of T. Campos; NCAR)

EDDY SHEDDING OF AEROSOL LAYER

- Main shedding occurring from 348 to ~355 K, without significant nitrate mass



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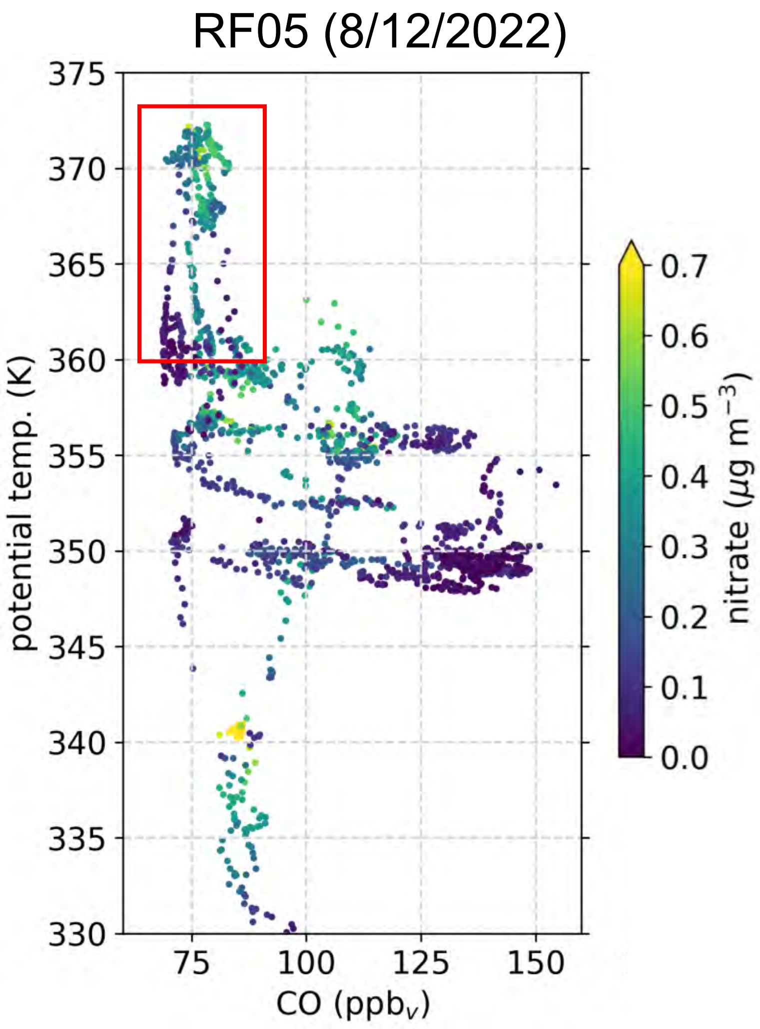

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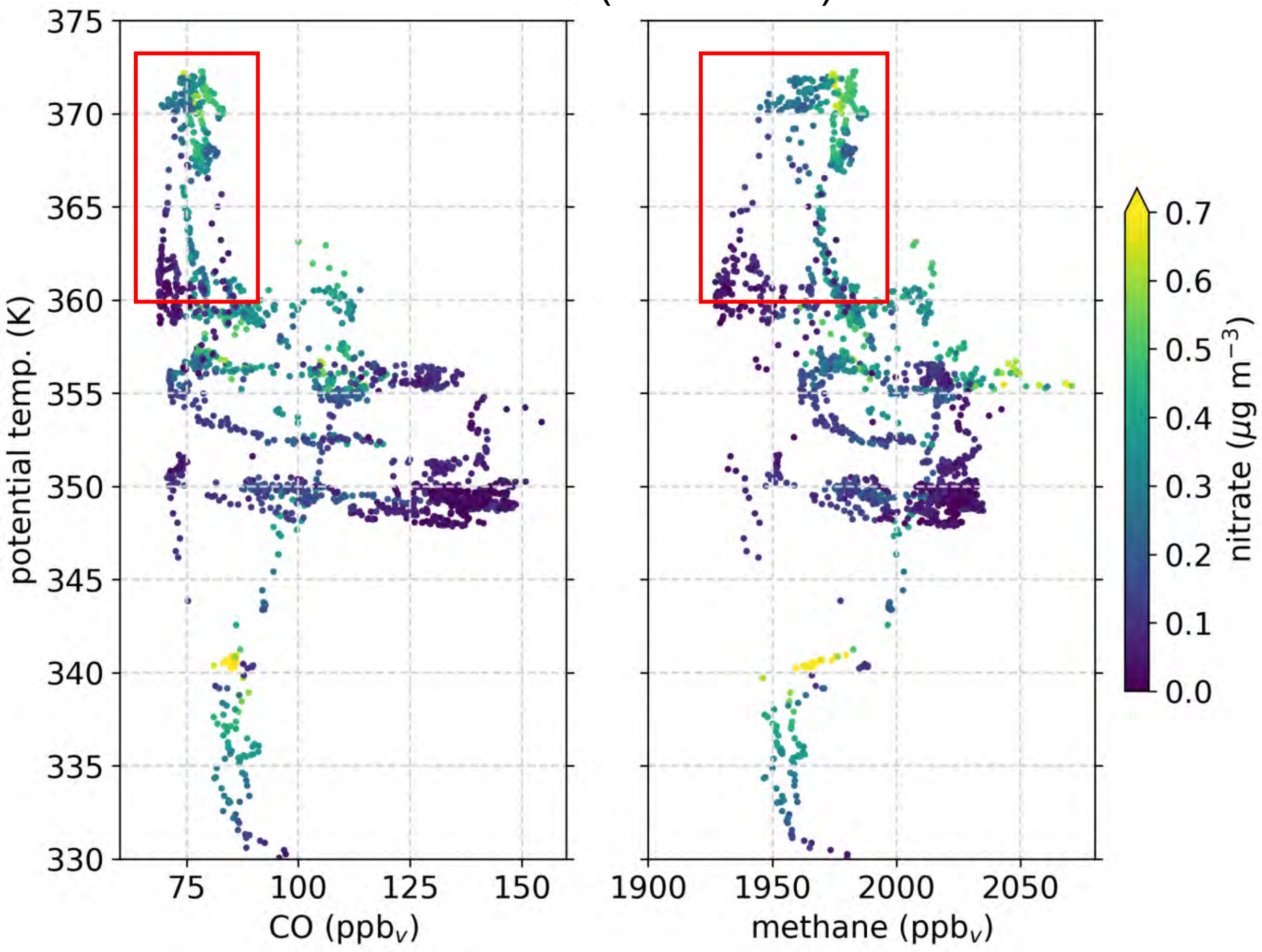

CO data from Aerodyne CS-108 miniQCL; CH_4 data from Picarro G2401 Analyzer (courtesy of T. Campos; NCAR)

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CH4 more suitable monsoon tracer for AMA influence in ATAL altitudes

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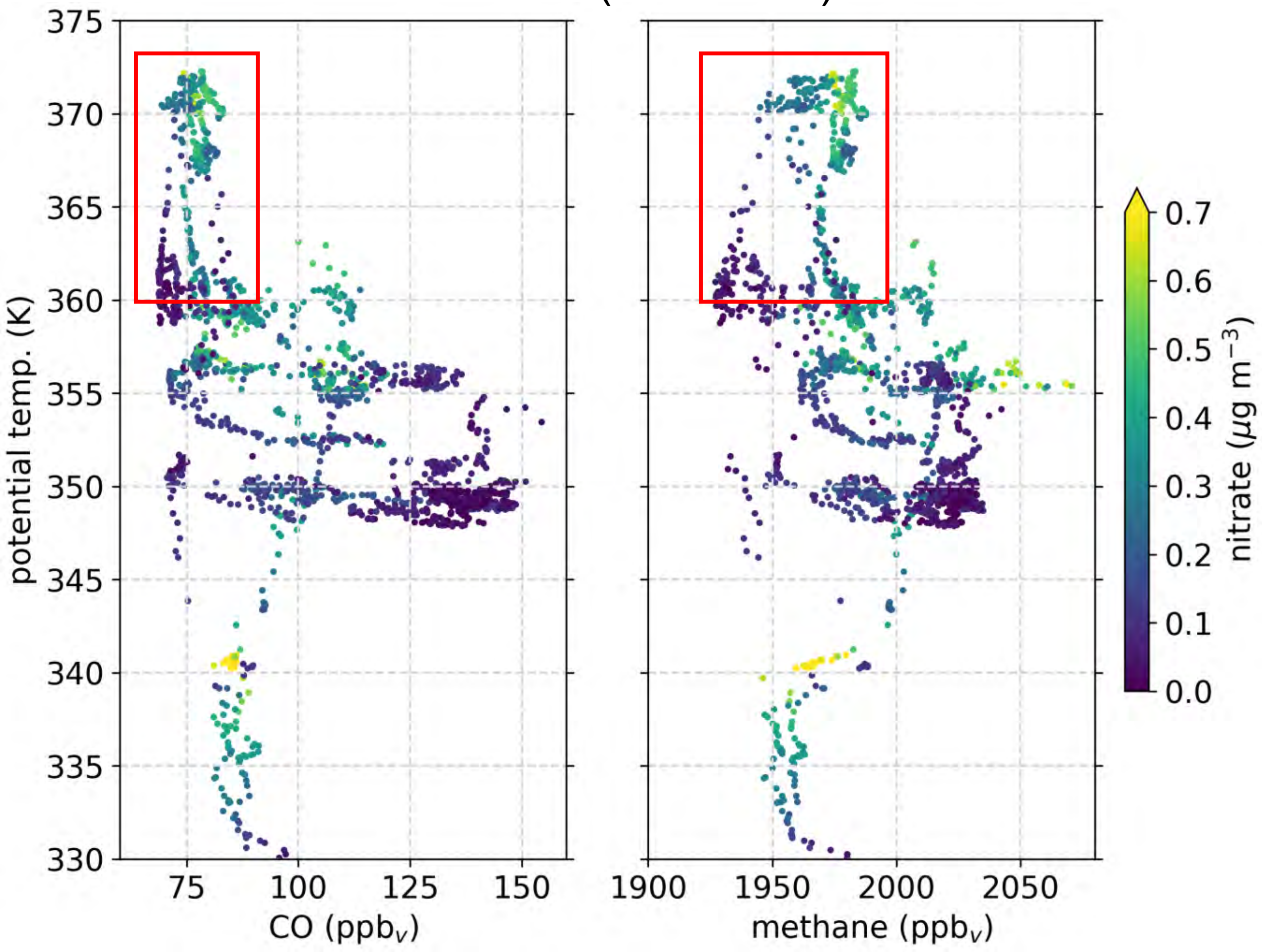

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CONCLUSIONS & OUTLOOK

- Observation of the ATAL in the monsoon outflow region over the Western Pacific at $\theta > 360$ K
- Special monsoon scenario in 2022: AMA flooded by fresh convection from polluted East Asian BL
    - Low aerosol mass concentrations from East Asia due to short transport times
    - Highest mass concentrations still from South Asia
    - But: Single-particle analysis indicating higher contribution of sulfate and organics from East Asia
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Further steps:

- Analysis of particle processing during transport from region
- Include tracers for East Asian influence (e.g. CH_2Cl_2, …)
- Comparison of fresh convection from East China with results from StratoClim
- Model study with ECHAM/MESSy model (in collaboration with M. Kohl; MPIC)

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Thank you!

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