In-situ measurements of aerosols within the Asian Monsoon Anticyclone and the ATAL: Particle physical properties and chemical composition

Stephan Borrmann ...

University of Mainz

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4th ACAM Workshop – Universiti Kebangsan

Bangi, Malysia – 28*.6.2019*

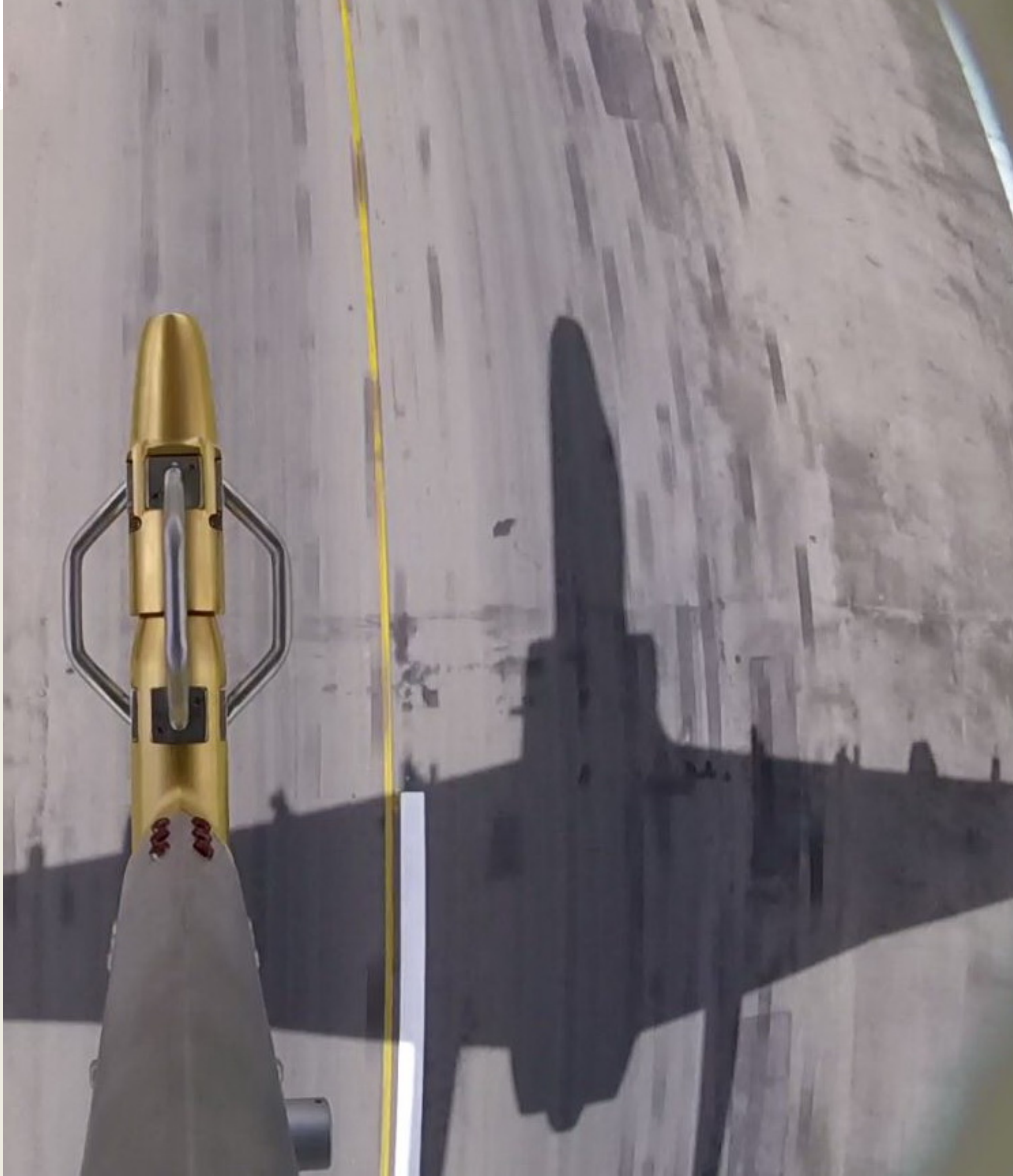
… and Investigators:

Ralf Weigel, Oliver Appel, Antonis Dragoneas, Sergej Molleker, Anneke Batenburg, Oliver Schlenczeck, Andreas Hünig, Christoph Mahnke, Max Port, Johannes Schneider, Frank Drewnick, Thomas Klimach, ***the entire Particle Chemistry Dept.,*** Martina Krämer (FZ Jülich), Jean-Paul Vernier (NASA), Francesco Cairo (CNR) ....

PLUS the coordinators

Markus REX (AWI)

Fred STROH (FZ Jülich)



Atmospheric setting

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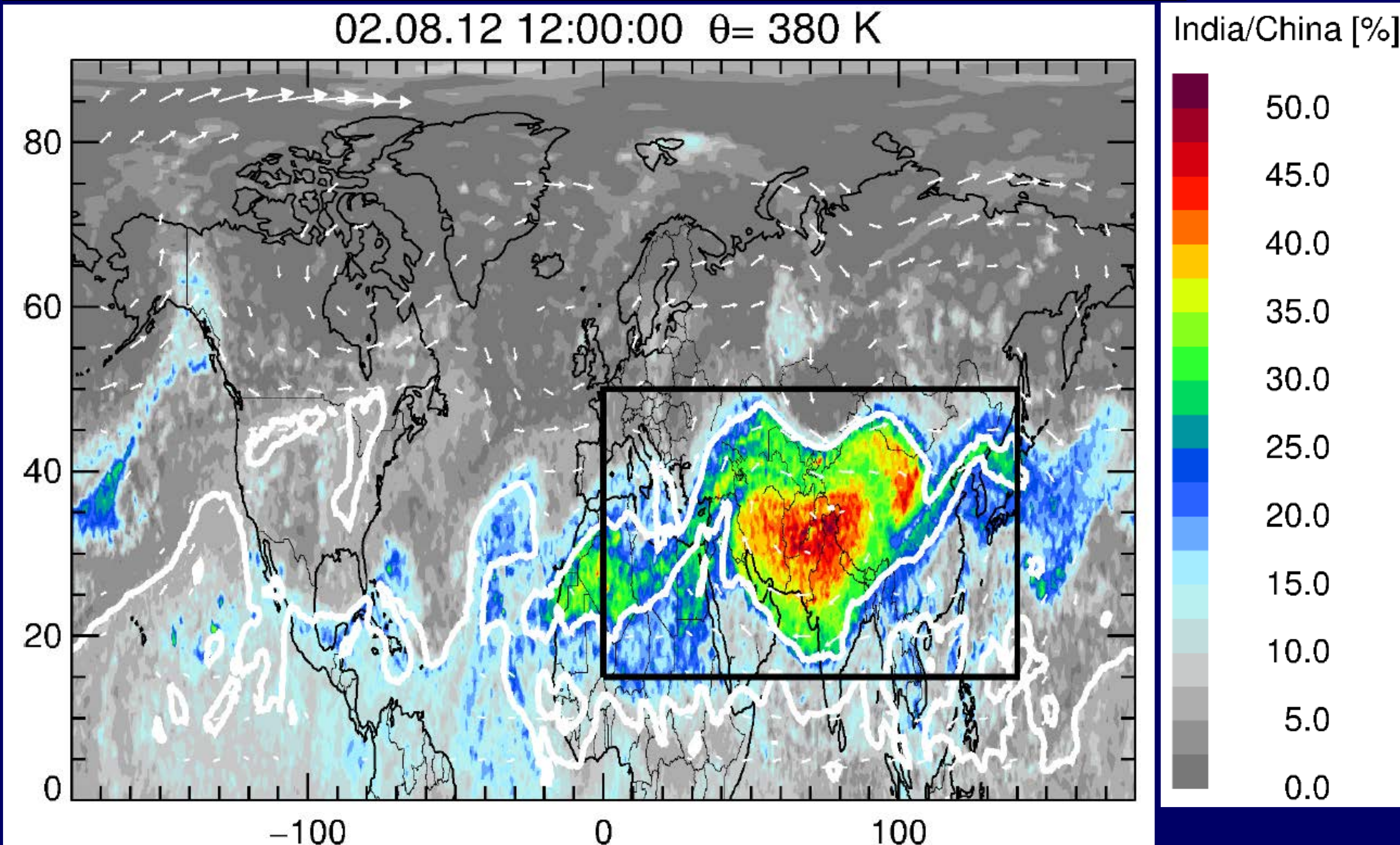
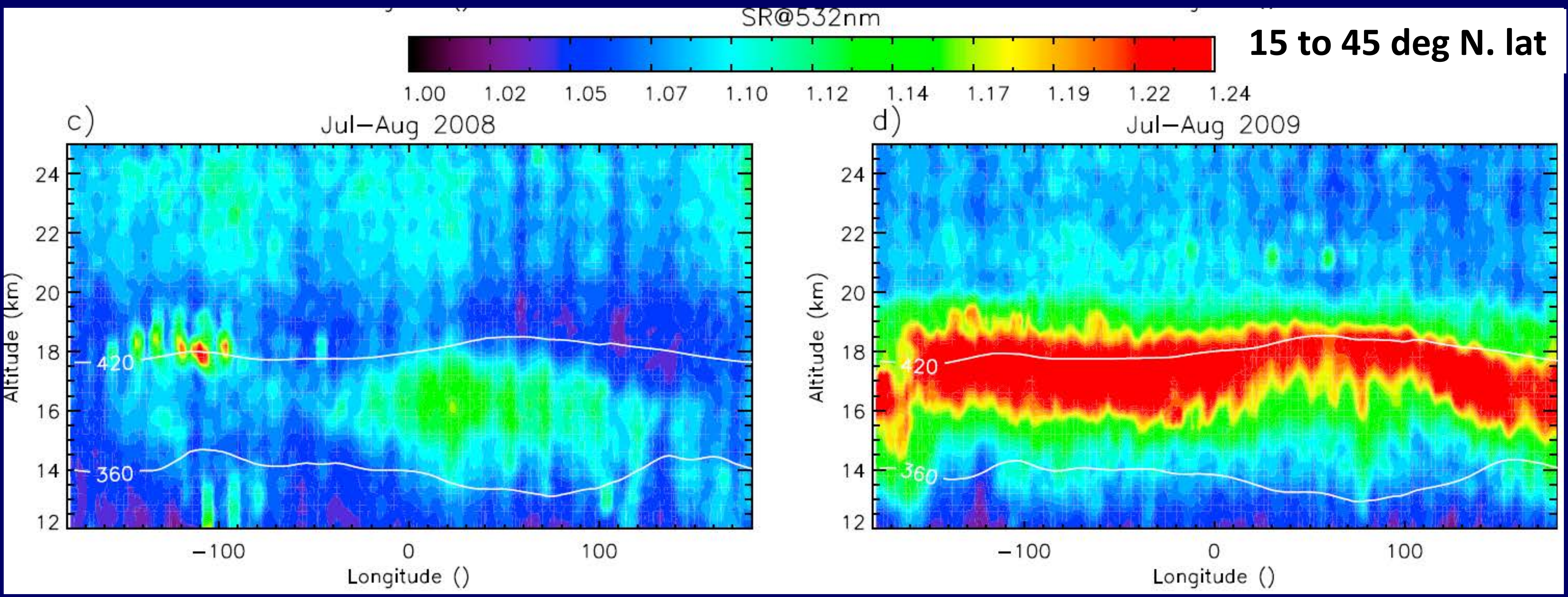
CLaMS model simulations: Fraction of air masses inside the AMA originating from BL in India/China



Vogel, B., et al., ACP, 2015

Asian Tropopause Aerosol Layer - ATAL



* ***Aerosol layer*** seen from CALIOP lidar extends from Eastern Mediterranean to Western China as far South as Thailand

* ***Enhancement of background aerosol*** – at tropopause ***only*** during the AMA monsoon time.

Vernier, J.-P., et al., GRL, 2011

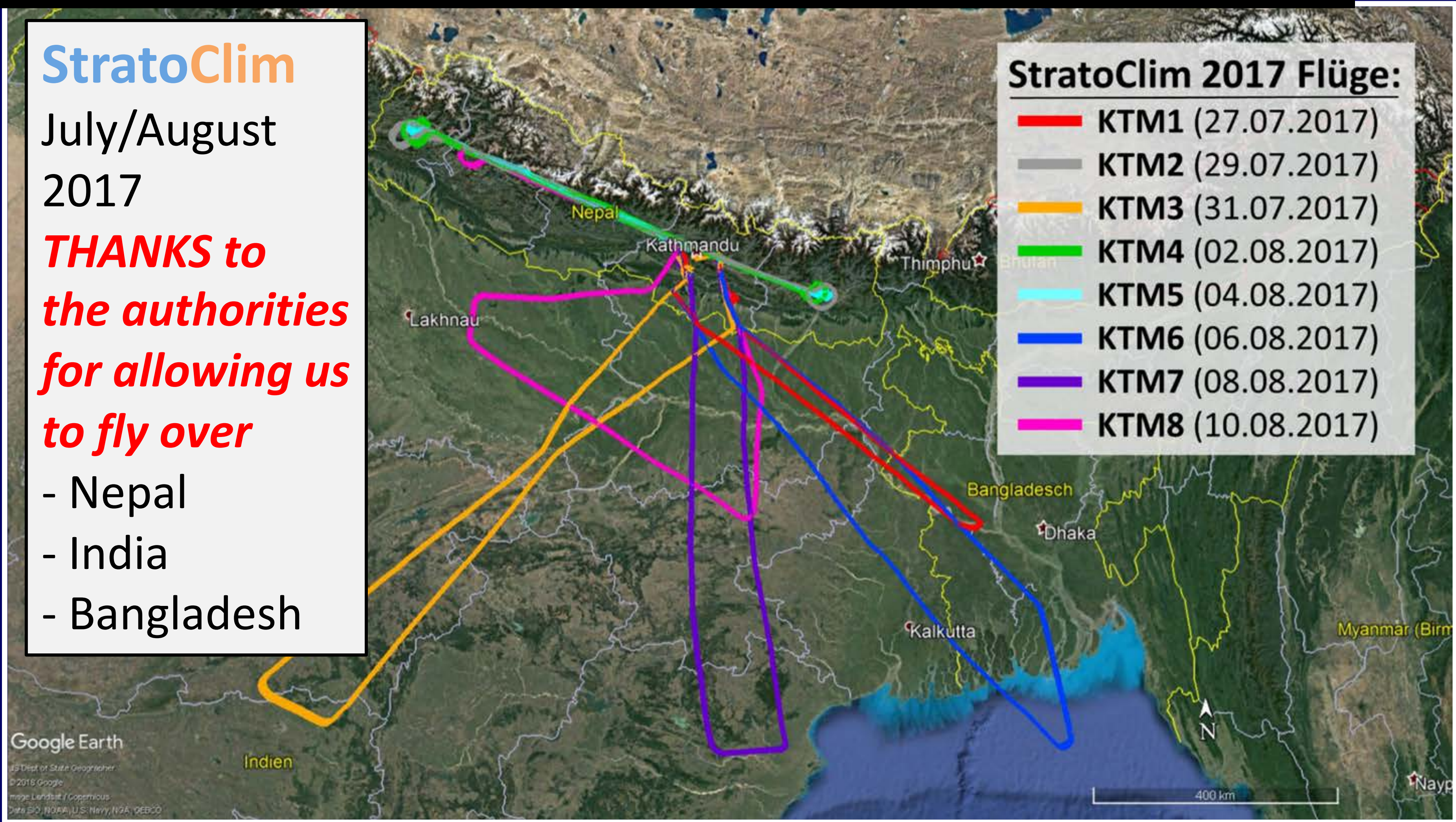
ATAL and AMA – setting for research flights

StratoClim

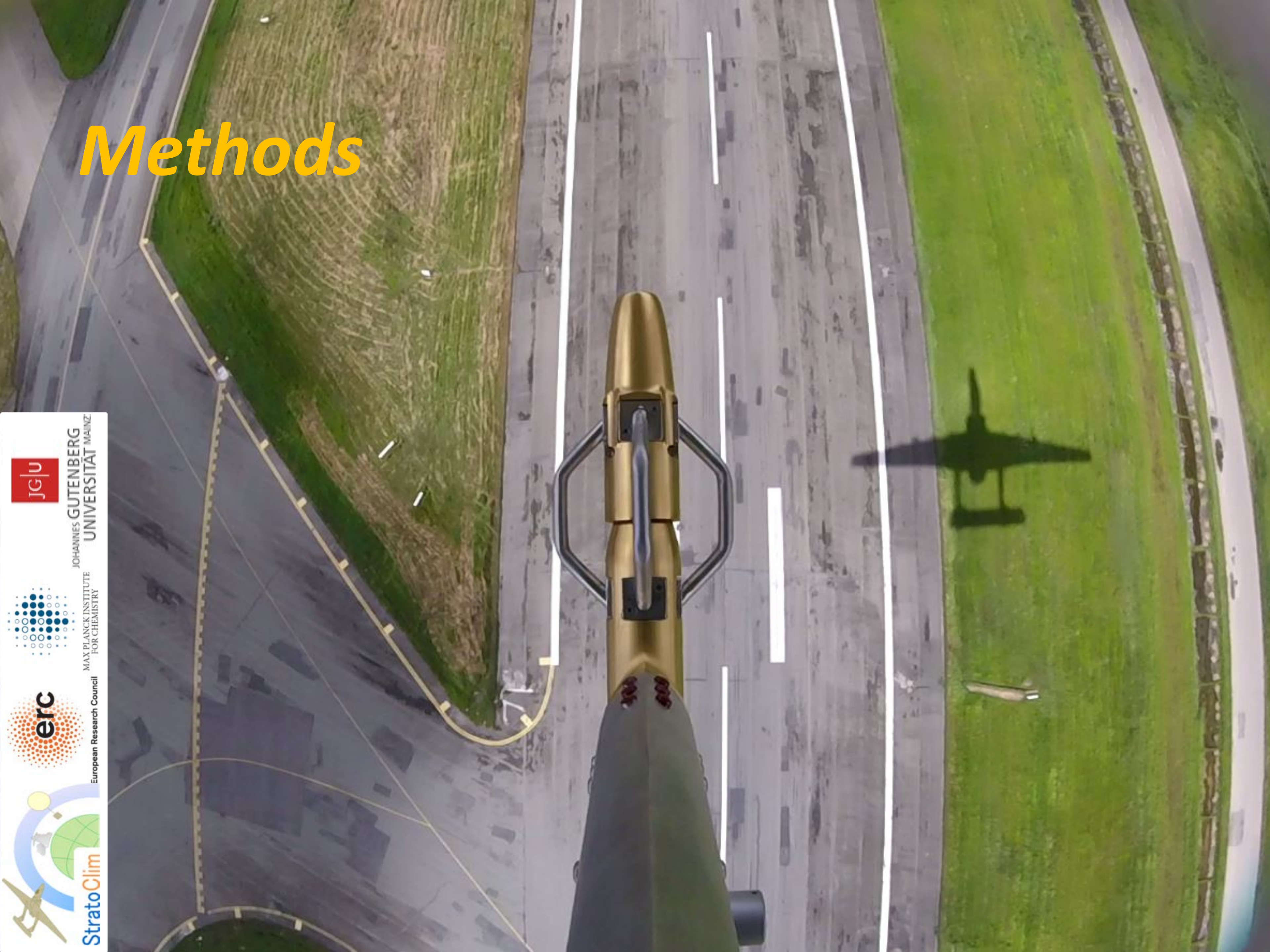
July/August 2017

THANKS to the authorities for allowing us to fly over

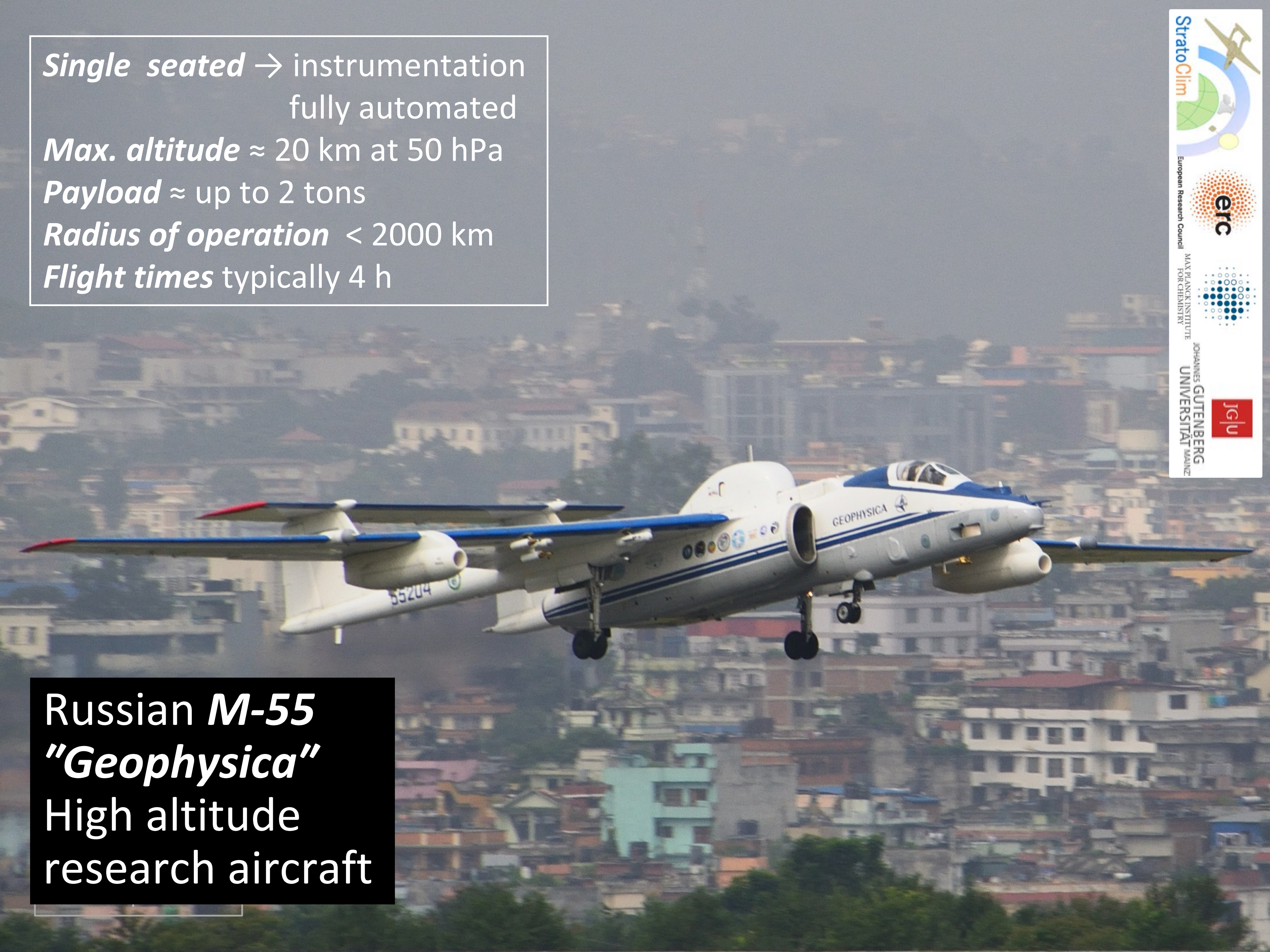
- Nepal
- India
- Bangladesh



Methods



Single seated → instrumentation fully automated
Max. altitude ≈ 20 km at 50 hPa
Payload ≈ up to 2 tons
Radius of operation < 2000 km
Flight times typically 4 h



Russian ***M-55***
"Geophysica"
High altitude
research aircraft

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ERICA

ERC Instrument for the the Chemical Composition of Aerosols

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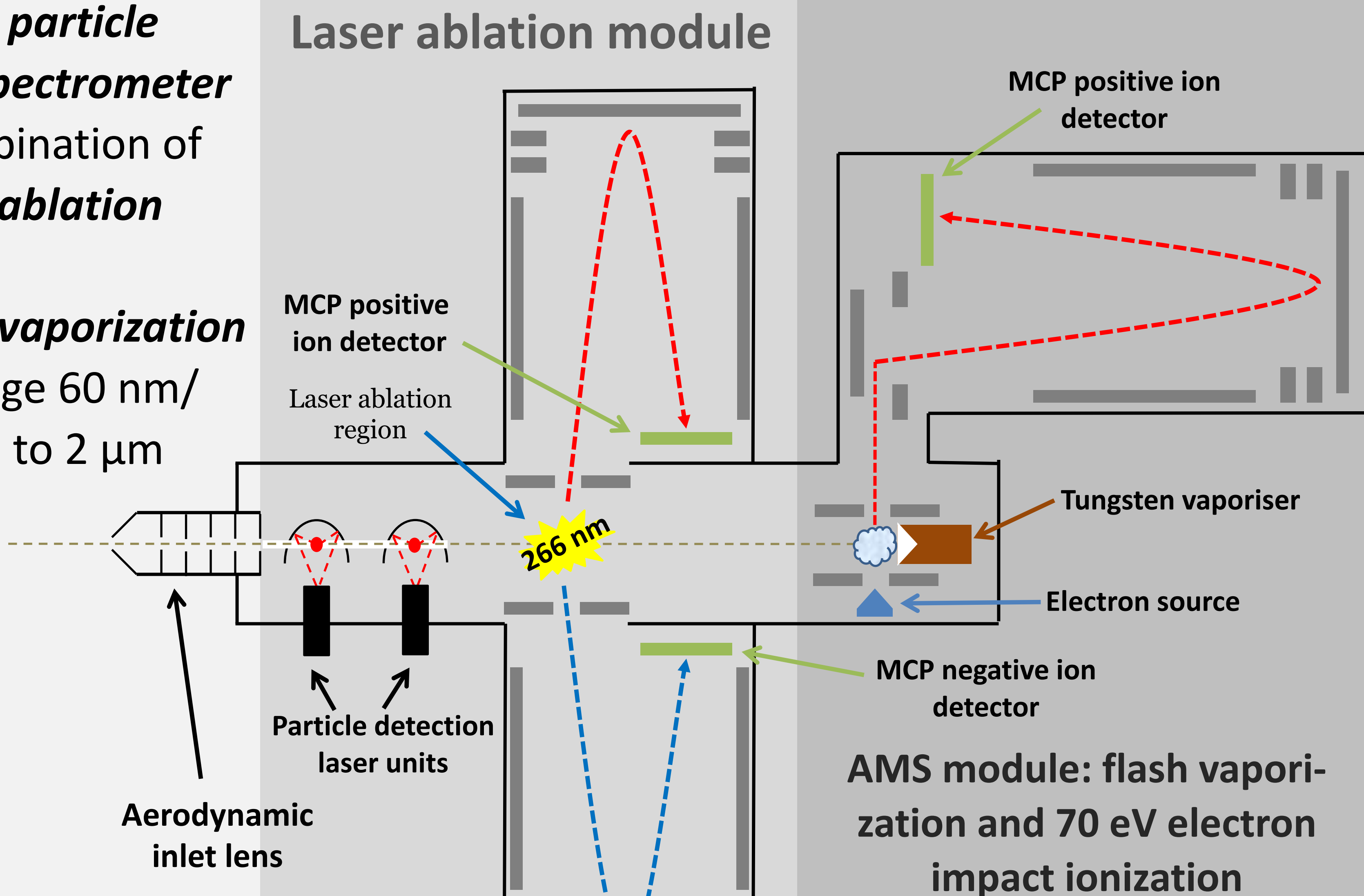
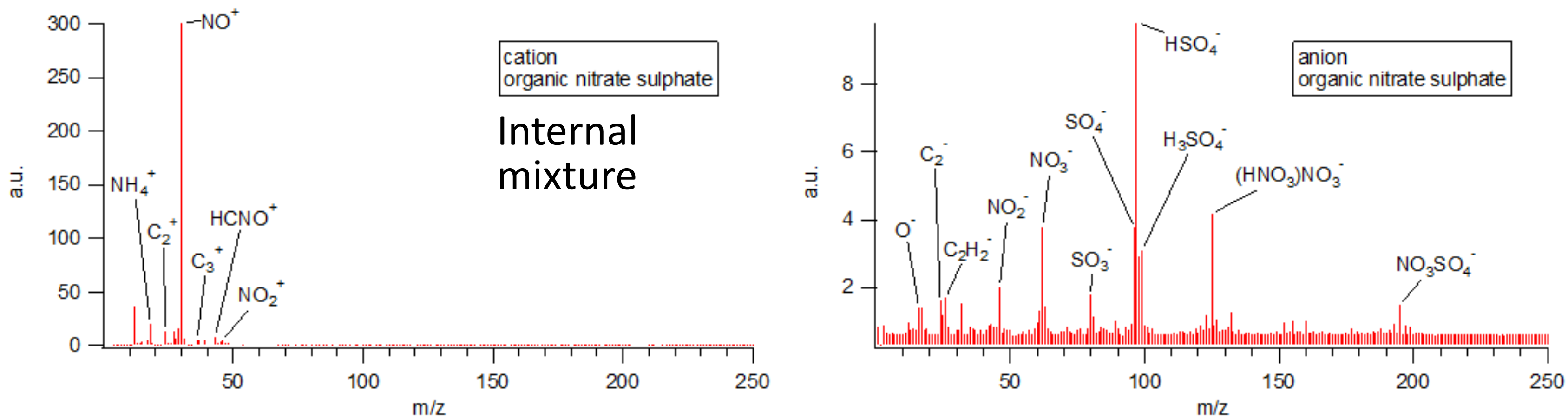
Unique particle mass spectrometer
as combination of
– laser ablation
with
– flash vaporization
size range 60 nm/ 150 nm to 2 µm



Example: single particle mass spectra



Organic, nitrate, sulphate

Note:
* ERICA measured the first stratospheric mass spectra with ***both*** polarities.
* Particle ***sizes*** have been retrieved,
* … spectra sorted into seven categories.
* ALL have ***sulfate in the anion*** spectra

Important for ***talk by Michael Höpfner***

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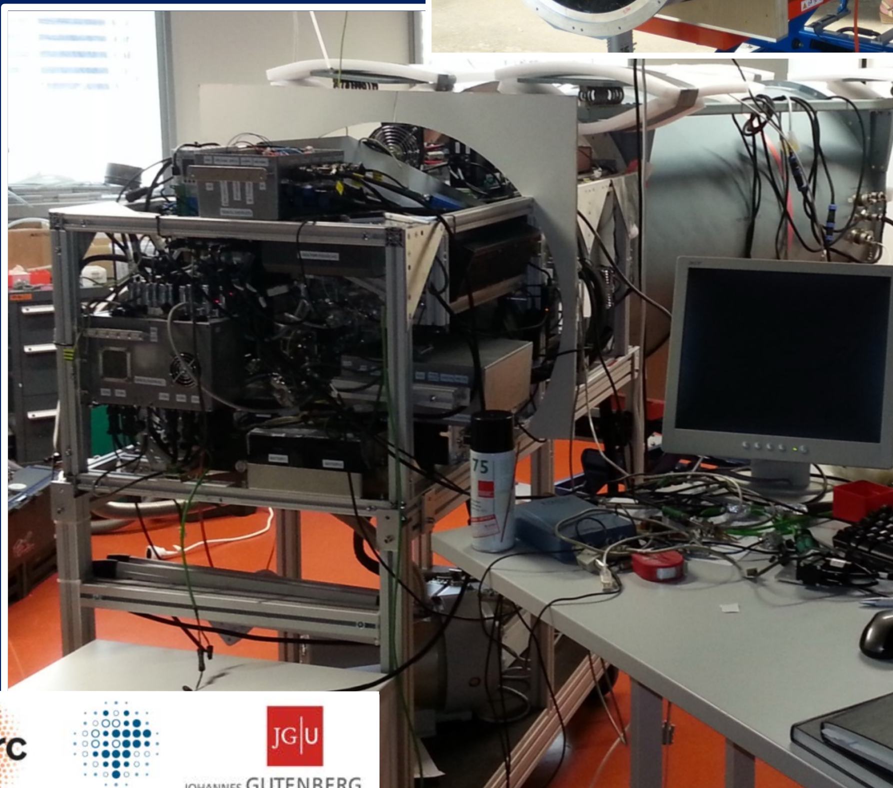
Design properties:

* pressurized barrel
* ≈ 360 kg, ≈ 1.5 kW
* fully automated
* IRIDIUM remote link
* ≈ 2000 parts
* IPA/MPIC in-house developed/built

ERICA



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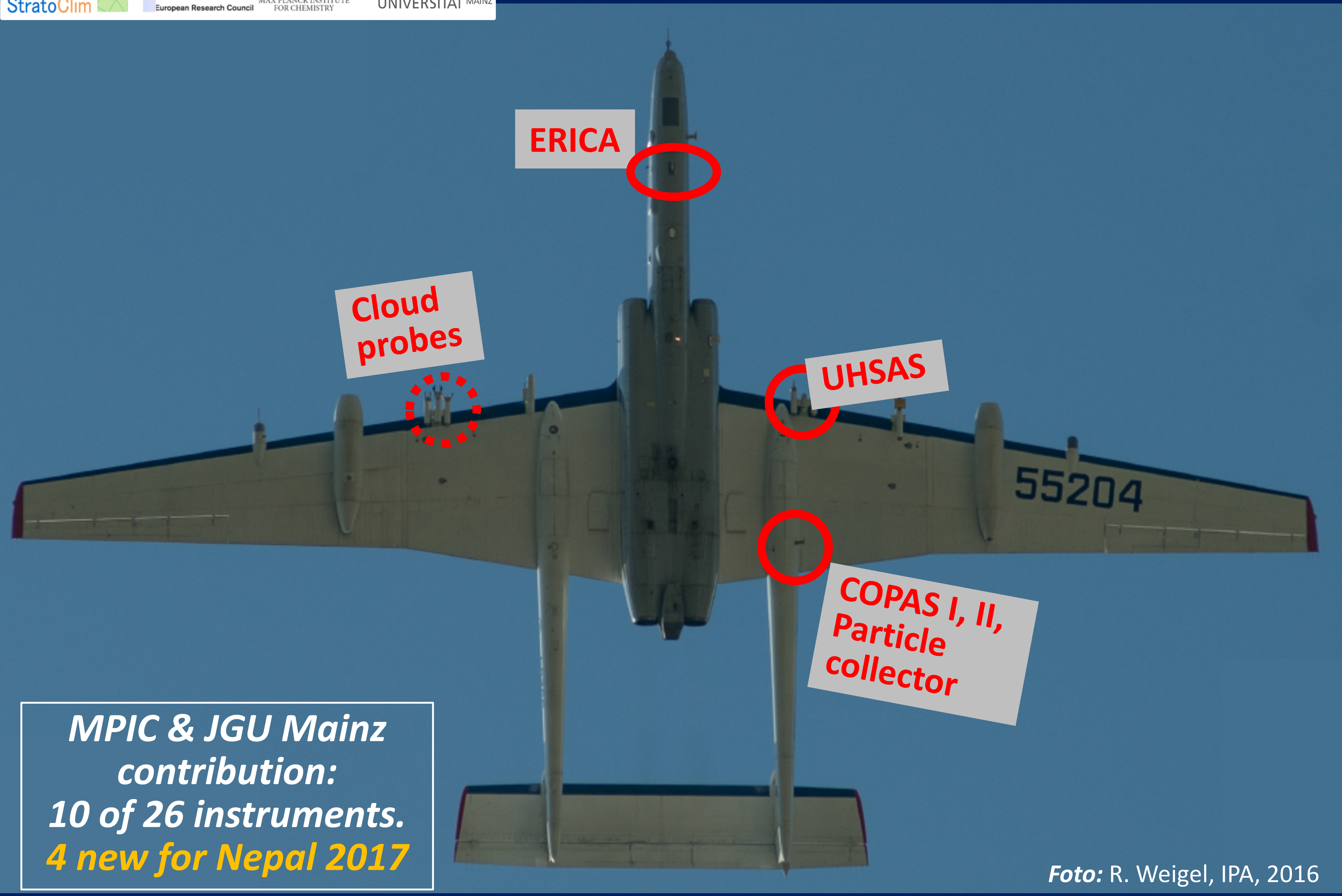


MPIC & JGU Mainz contribution:
10 of 26 instruments.
4 new for Nepal 2017

Foto: R. Weigel, IPA, 2016

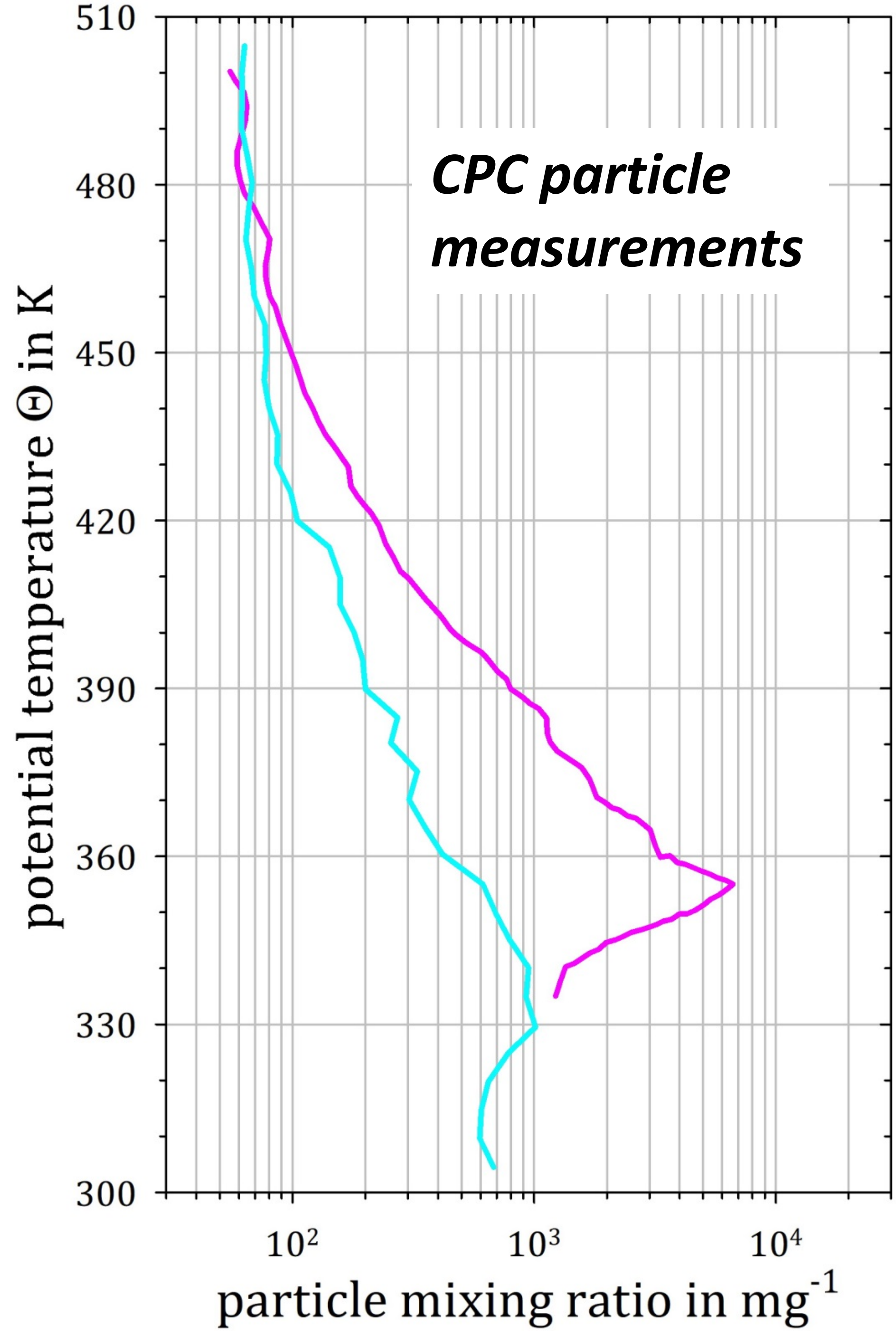
Results on aerosol physical properties

Tropical vertical profiles 1987-1994
(Hawaii, ER-2):

Particles 10 nm-1 µm

Brock et al. (1995): Tropics
Brock et al. (1995): Ex-Tropics

Brock et al., Science, 1995



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Tropical vertical profiles 2005-2006

(Russian M-55 „Geophysica“ high altitude resarch aircraft):

Particles 10 nm-1 µm

Brazil, ***Australia***,

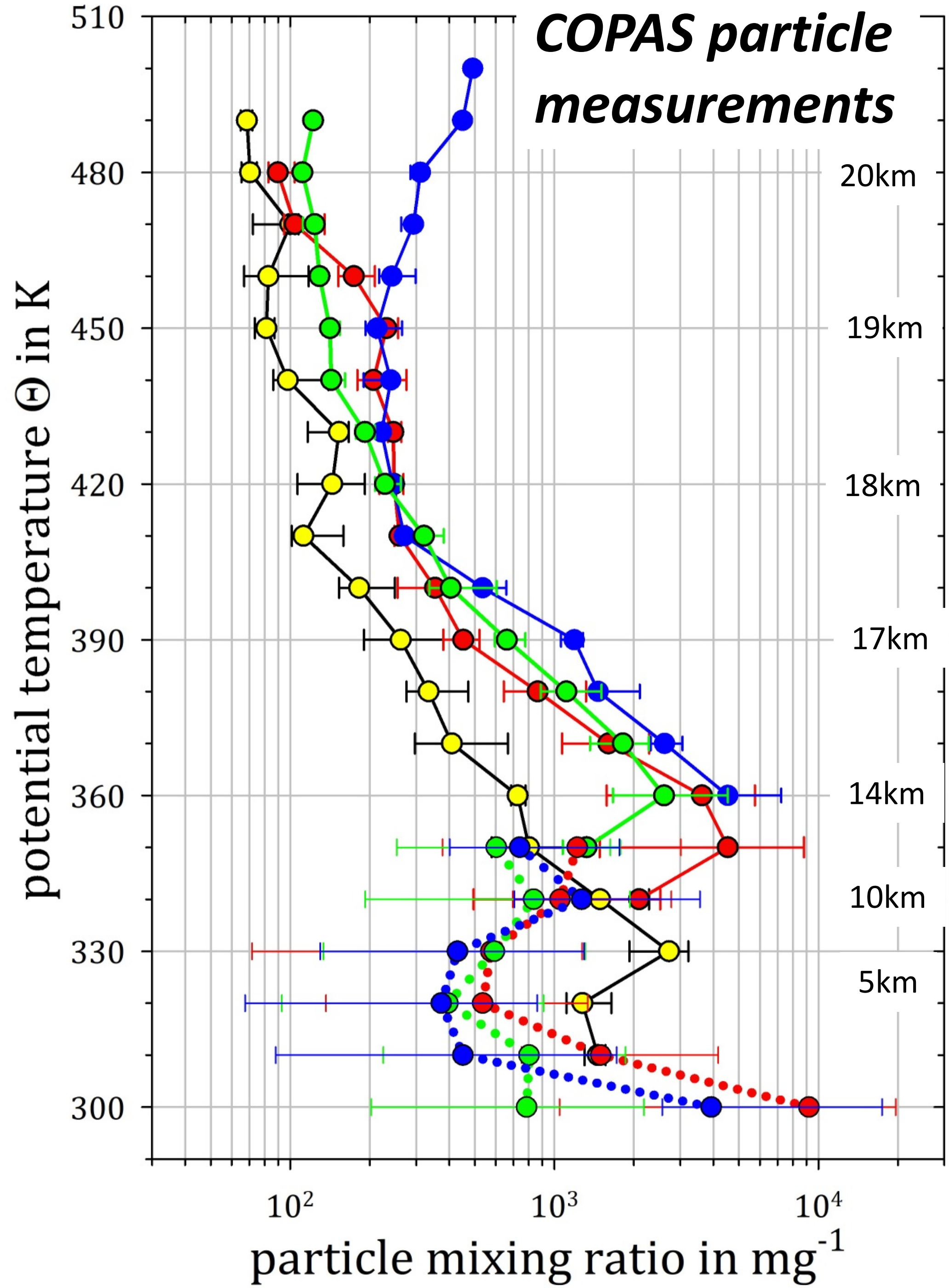
West Africa

- Median N_{10}: mid-latitudes (Forli, 2002)
- Median N_6: Tropics (TROCCINOX, 2005)
- Median N_{10}: Tropics (SCOUT-O_3, 2005)
- Median N_{10}: Tropics (SCOUT-AMMA, 2006)

CN measurements by DLR (Falcon 20)

- Median N_{13}: Tropics (TROCCINOX, 2005)
- Median N_5: Tropics (SCOUT-O_3, 2005)
- Median N_{10}: Tropics (SCOUT-AMMA, 2006)

Borrmann et al., ACP, 2010

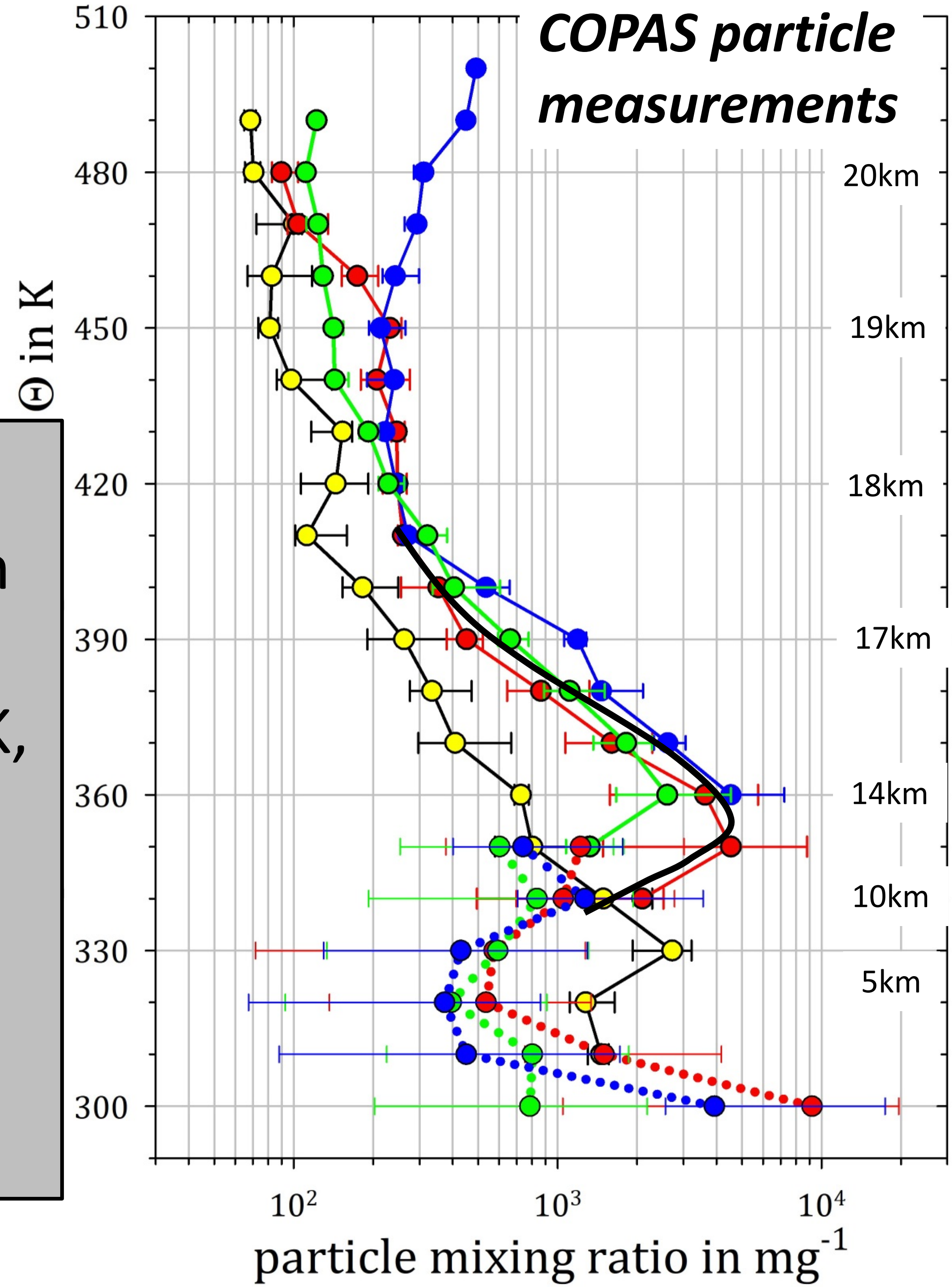


Tropical vertical profiles 2005-2006
(Russian M-55 „Geophysica“ high altitude resarch aircraft):
Particles 10 nm-1 µm
Brazil, Australia,

Interpretation:

* ***global*** layer of submicron particles in the **tropical belt** betw. 340K and 400K, i.e. ***not only in ATAL***

* has maximum inside and below the TTL (***Tropical Transition Layer***)



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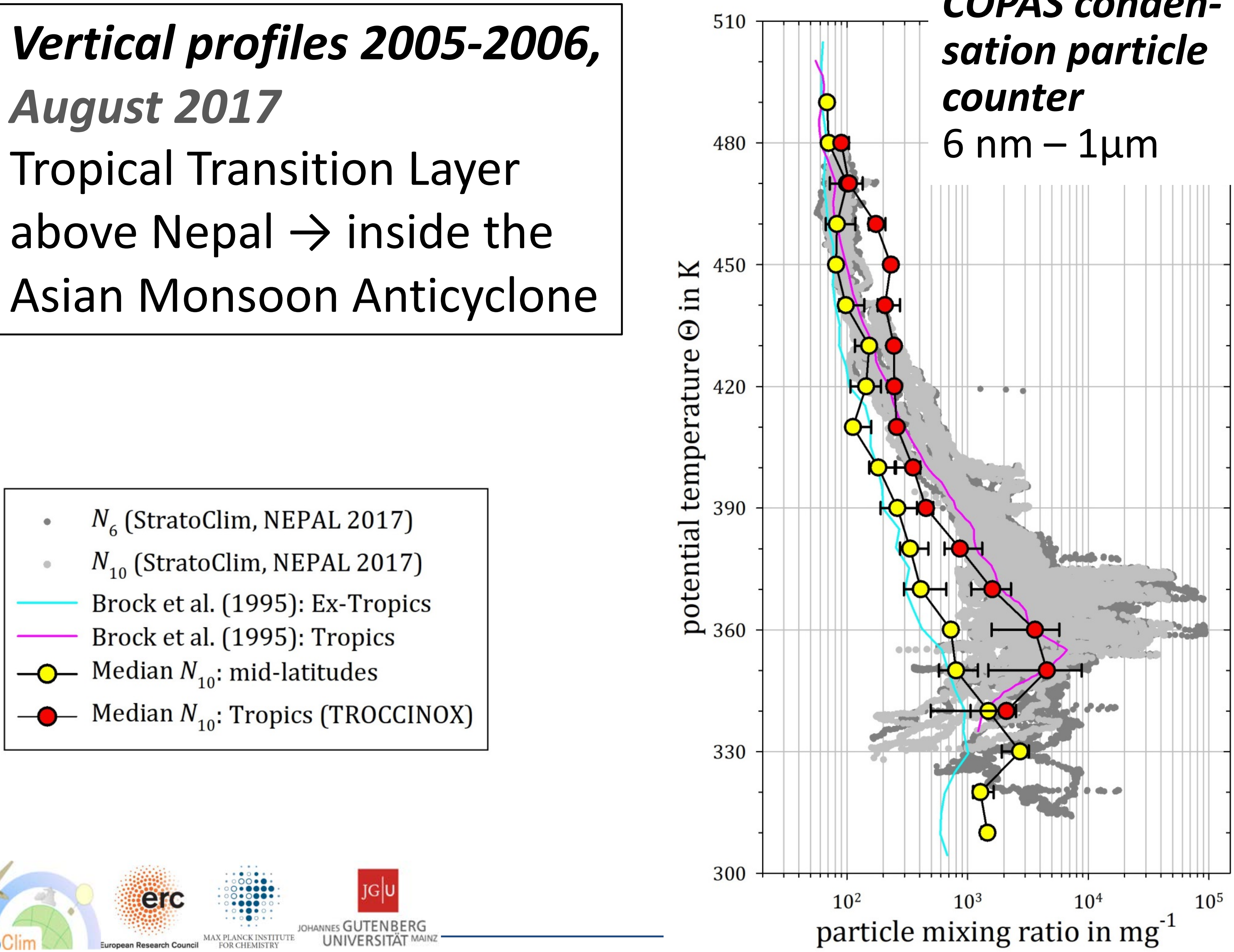
Vertical profiles 2005-2006,
August 2017
Tropical Transition Layer above Nepal → inside the Asian Monsoon Anticyclone

COPAS condensation particle counter
6 nm – 1µm



Analysis: R. Weigel, IPA

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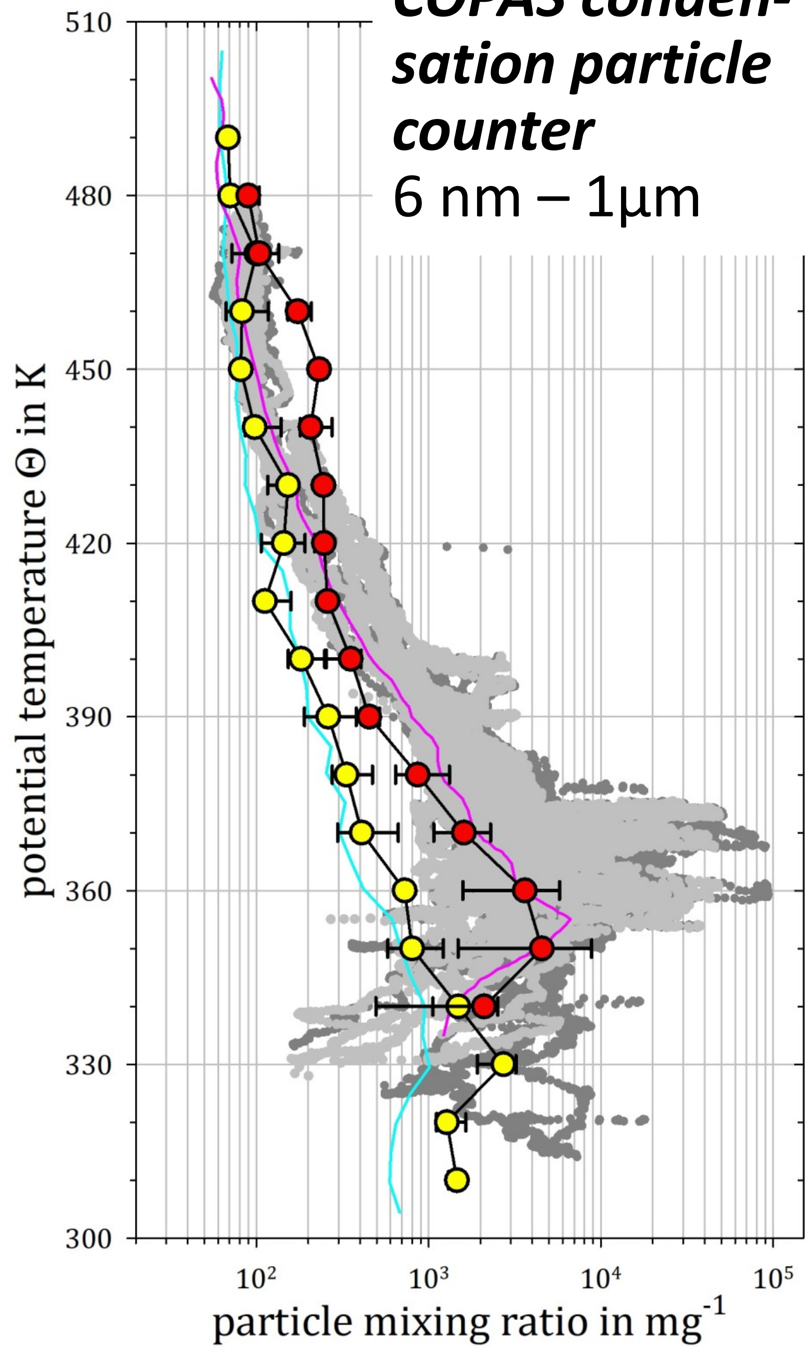
Vertical profiles 2005-2006,
August 2017
Tropical Transition Layer above Nepal → inside the Asian Monsoon Anticyclone

Smallest particles [nm]:

* highest particle mixing ratios ever seen are inside AMA
* peak altitude 5 – 10 K higher than elsewhere in the tropics
* influence of AMA visible in the particle data up to 420K

COPAS condensation particle counter
6 nm – 1µm



Analysis: R. Weigel, IPA

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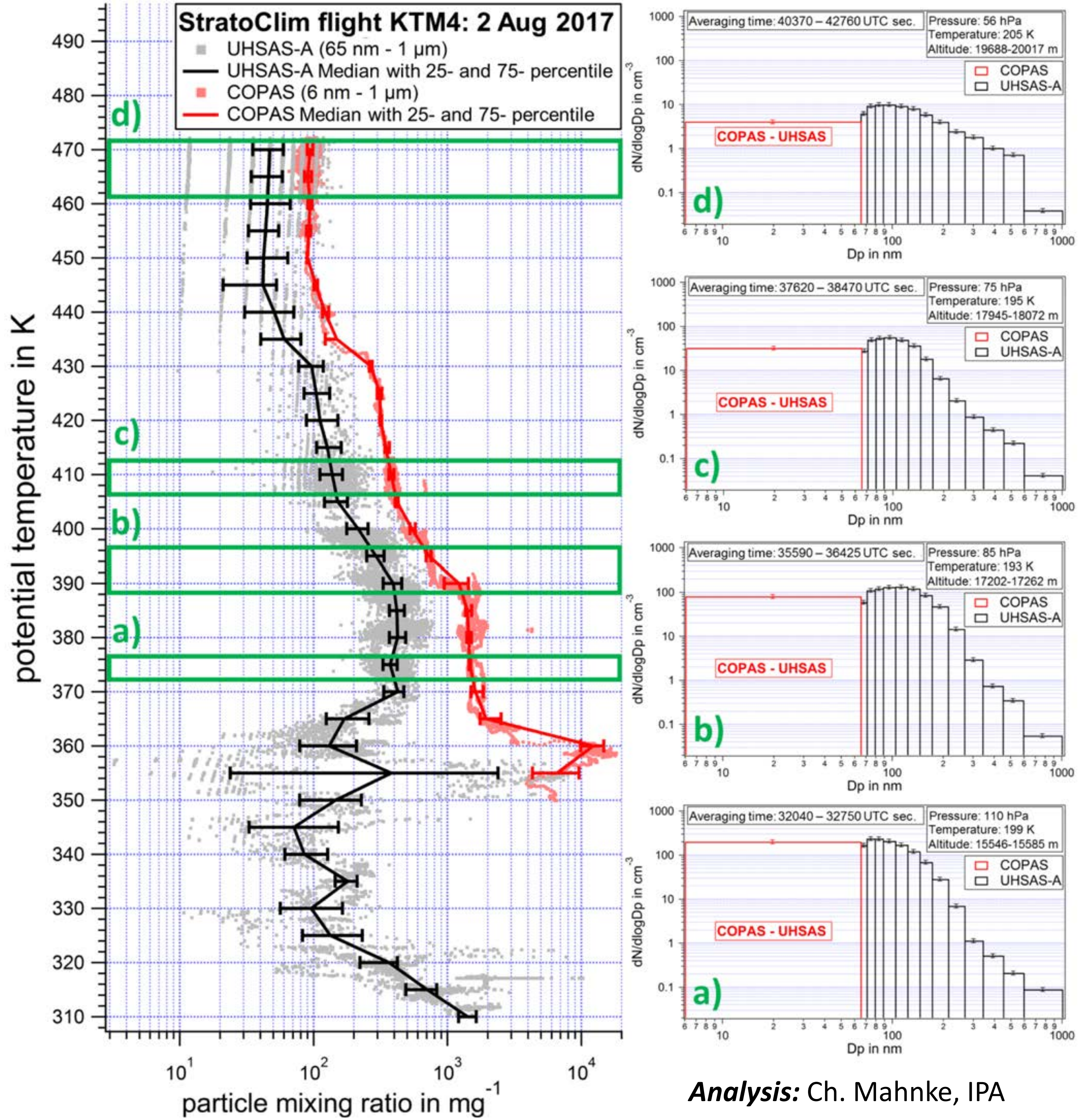
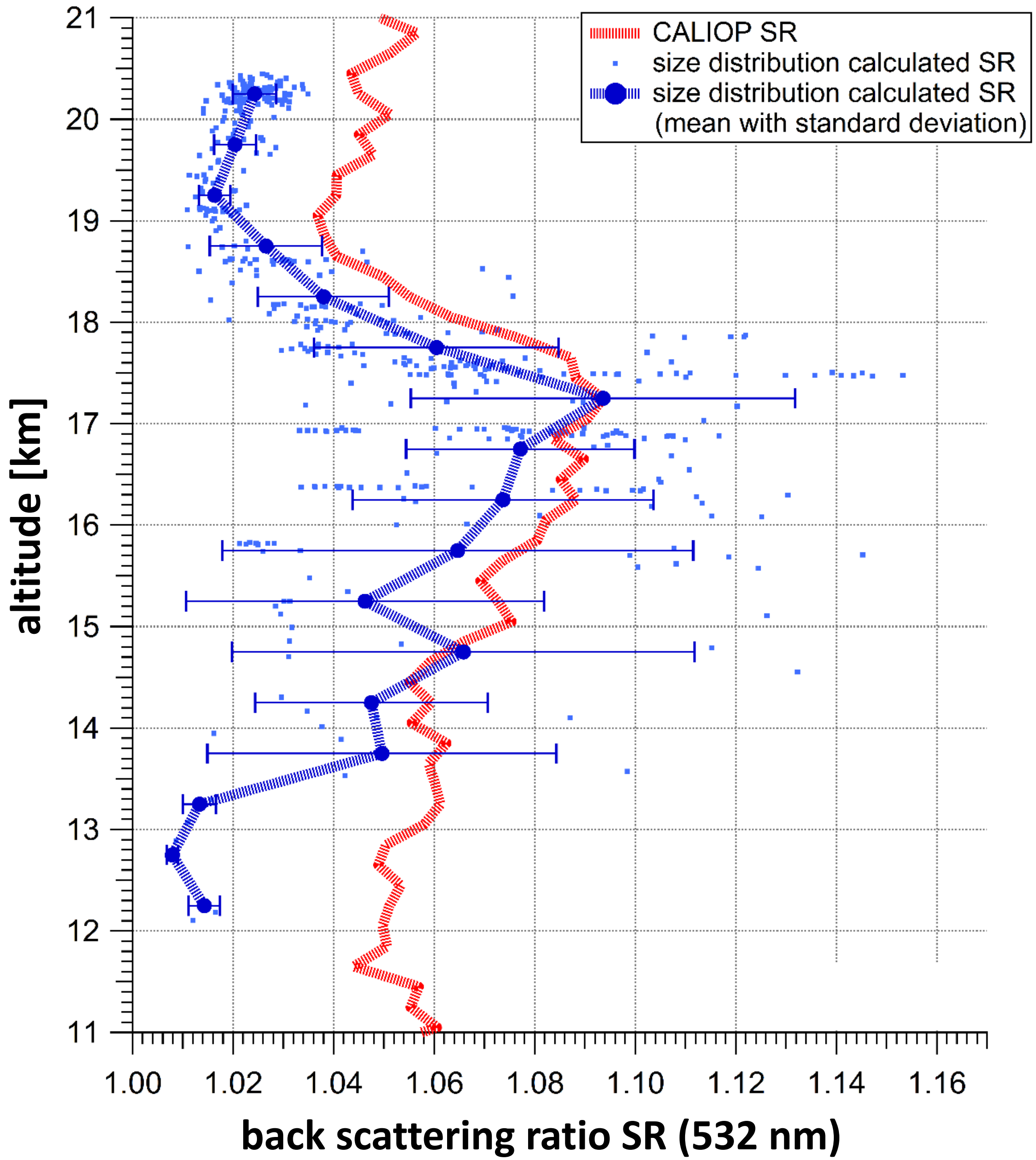
Larger, submicron particles

from MPIC modified ***optical particle counter*** UHSAS 65 nm – 1.0 µm



Analysis: Ch. Mahnke, IPA

ATAL vertical profile of the aerosol particle SR



From Mie calculation on measured size distribution n(r):

$$\beta = \int_0^\infty n(r) \cdot \pi \cdot r^2 \cdot Q(r) \cdot dr$$

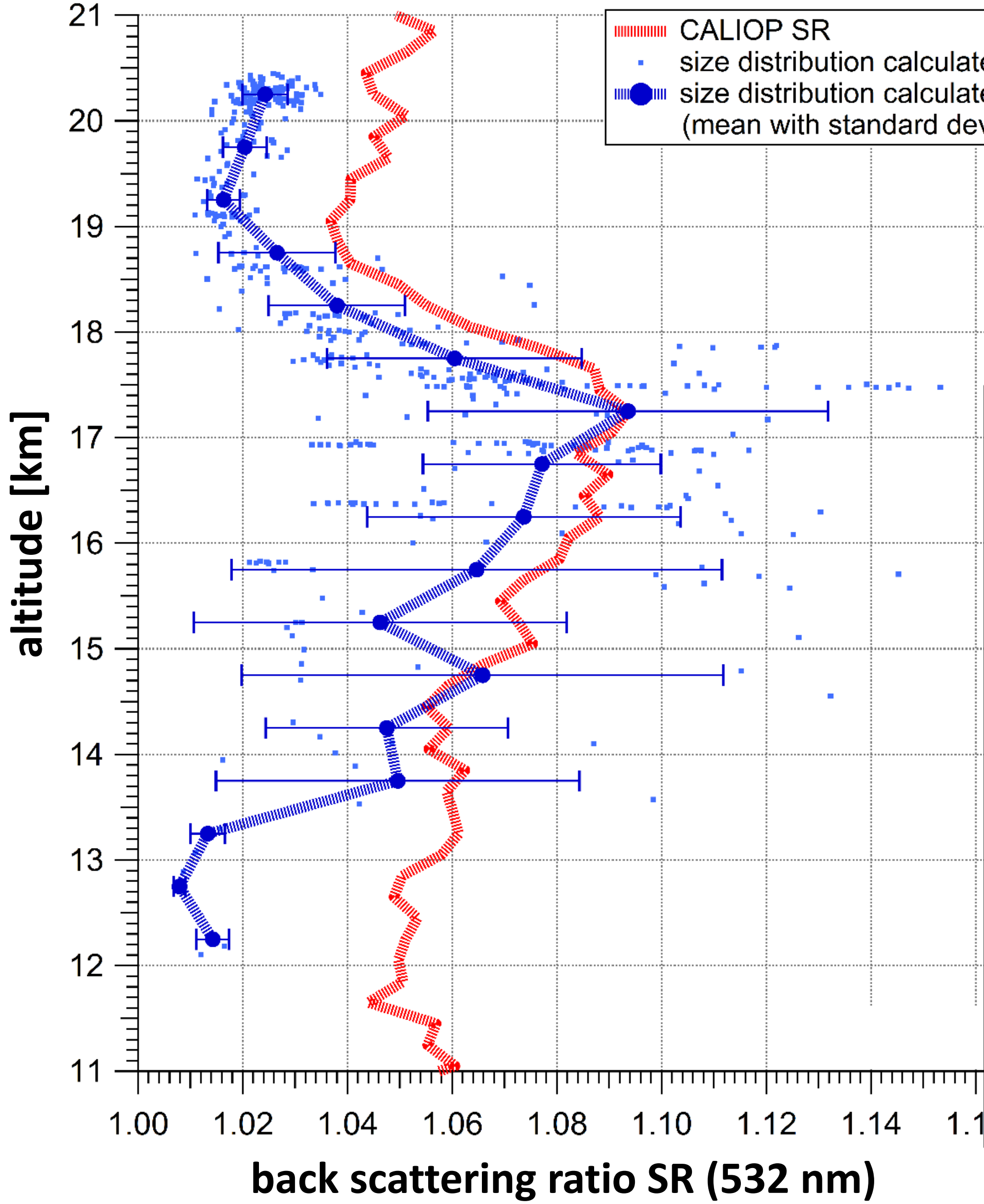
$$\mathrm{SR} = \frac{\beta_a + \beta_R}{\beta_R}$$ ***Backscatter ratio SR***

<u>CALIOP SR averaged:</u>
4 - 31 August 2017
15N - 45N; 70E - 100E

<u>SR from size distributions :</u>
COPAS: 10 – 65 nm
+
UHSAS-A: 65 – 1000 nm
+
NIXE-CAS: 1000 – 3000 nm

Analyses by J.-P. Vernier, F. Cairo, and C. Mahnke.

ATAL vertical profile of the aerosol particle SR



From Mie calculation on measured size distribution n(r):

$$\beta = \int_0^\infty n(r) \cdot \pi \cdot r^2 \cdot Q(r) \cdot dr$$

$$SR = \frac{\beta_a + \beta_R}{\beta_R}$$

Backscatter ratio SR

CALIOP signal of 2017 ATAL well retraced by in-situ particle size distribution measurements

Results on aerosol chemical composition

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Chemical composition: ERICA deliverables

* Single particle mass spectra – simultaneous
positive and negative ions

* ***qualitative*** – metals, soot, mineral dust, meteoric

* AMS – Flash vaporization/e^- impact ionization spectra

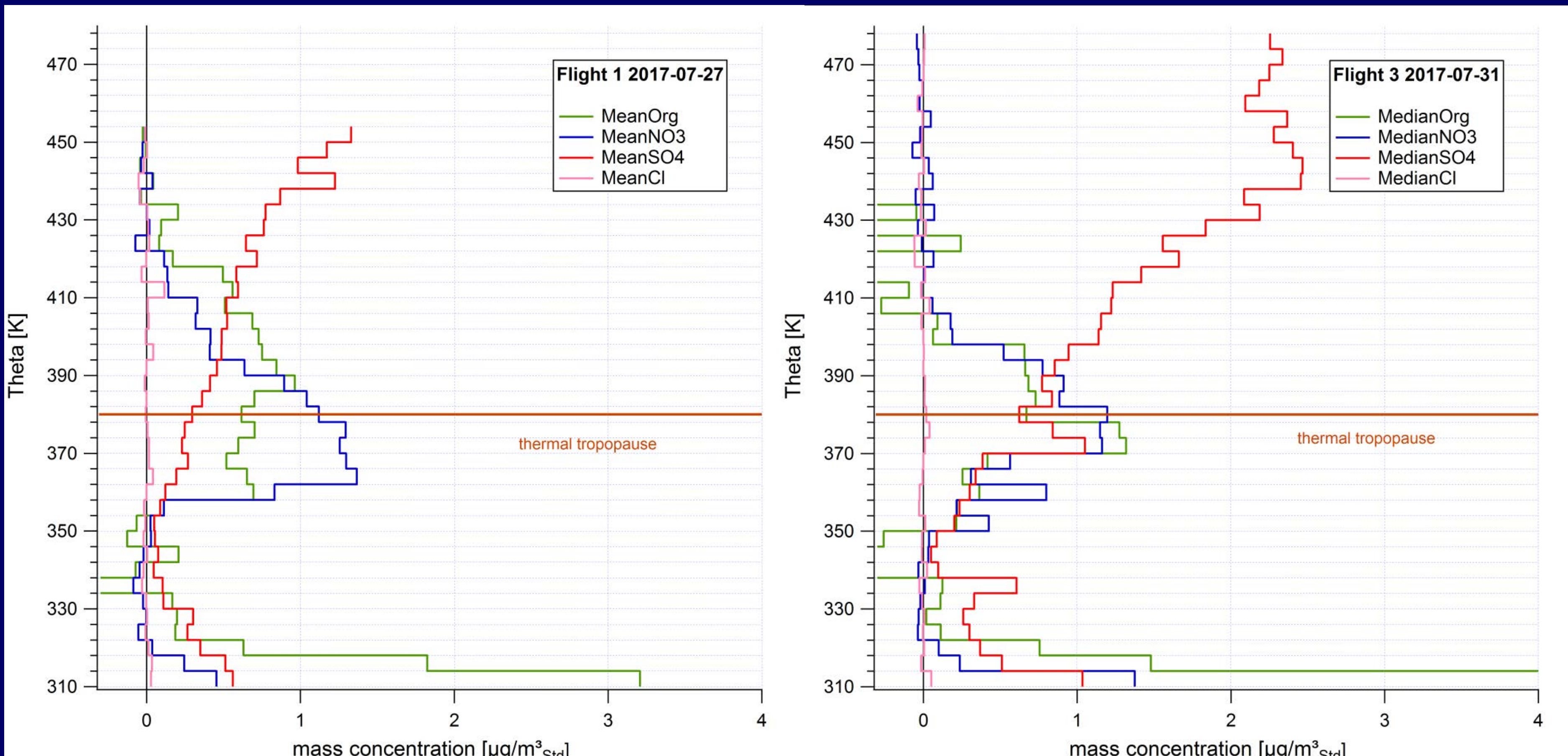
* ***quantitative*** – sulfate, nitrate, ammonia,
organics, chloride

* ***size range*** – from 60 nm (AMS), 100 nm (single
particle ms) to 2 µm

* particle collection – on electron microscope grids
and boron plates via COPAS II

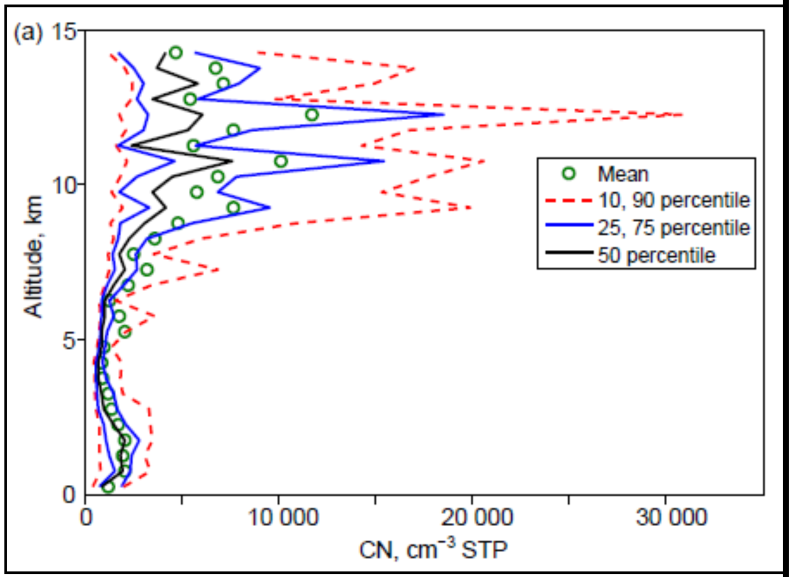
Delivered ≈ 140 000 single particle mass spectra in
Nepal data good for size determination

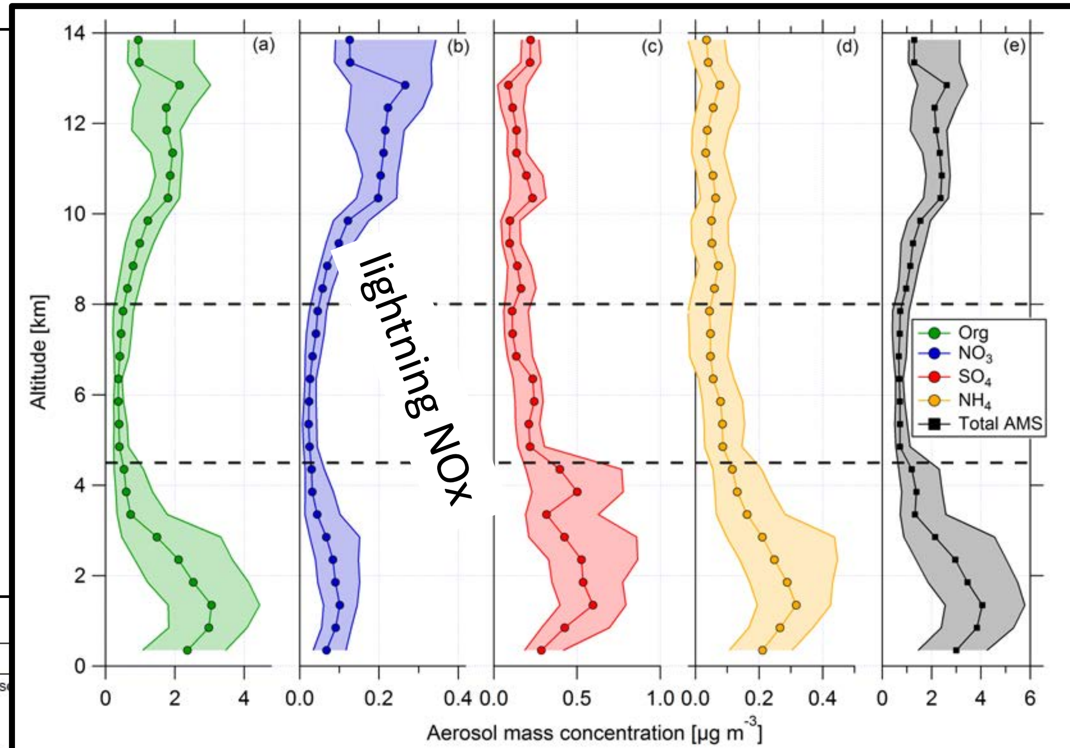
ERICA AMS part – Nepal Flights 1 and 3

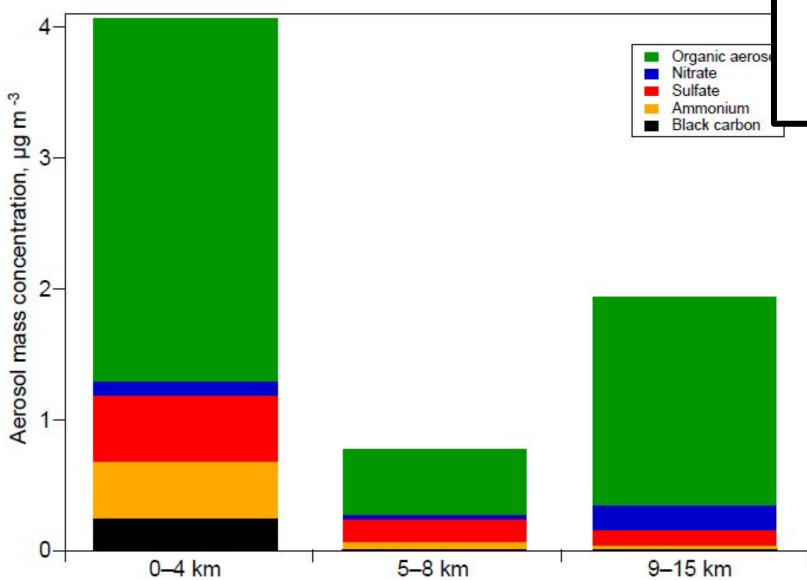


Analysis: O. Appel, MPIC

Tropical UT Amazonia, Brazil



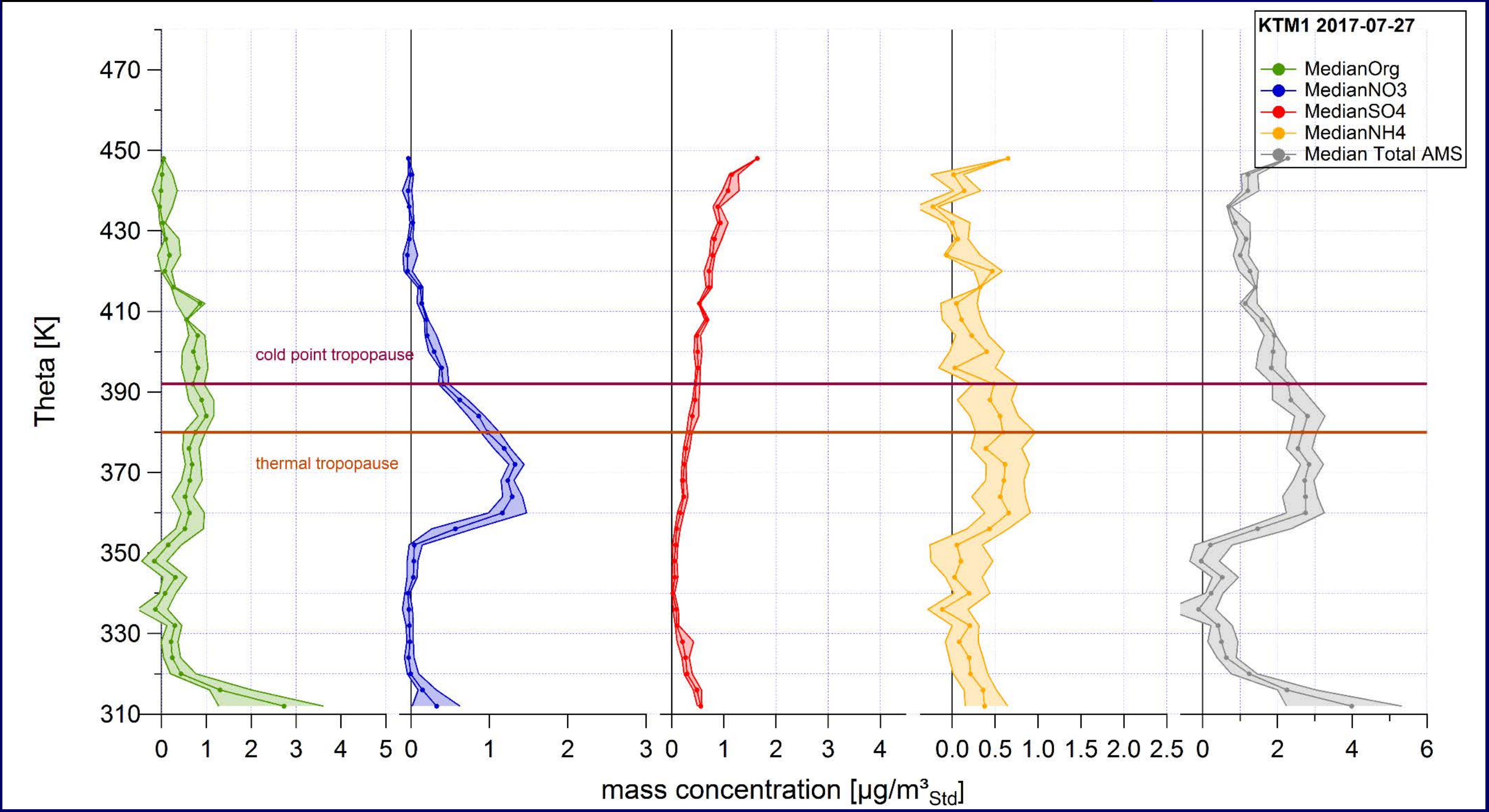




ACRIDICON-CHUVA, Manaus, Sept./Oct. 2014

Source: Andreae et al., ACP, 2018 ↑; ↗ Schulz Ch., Dissertation 2019, ACP 2019

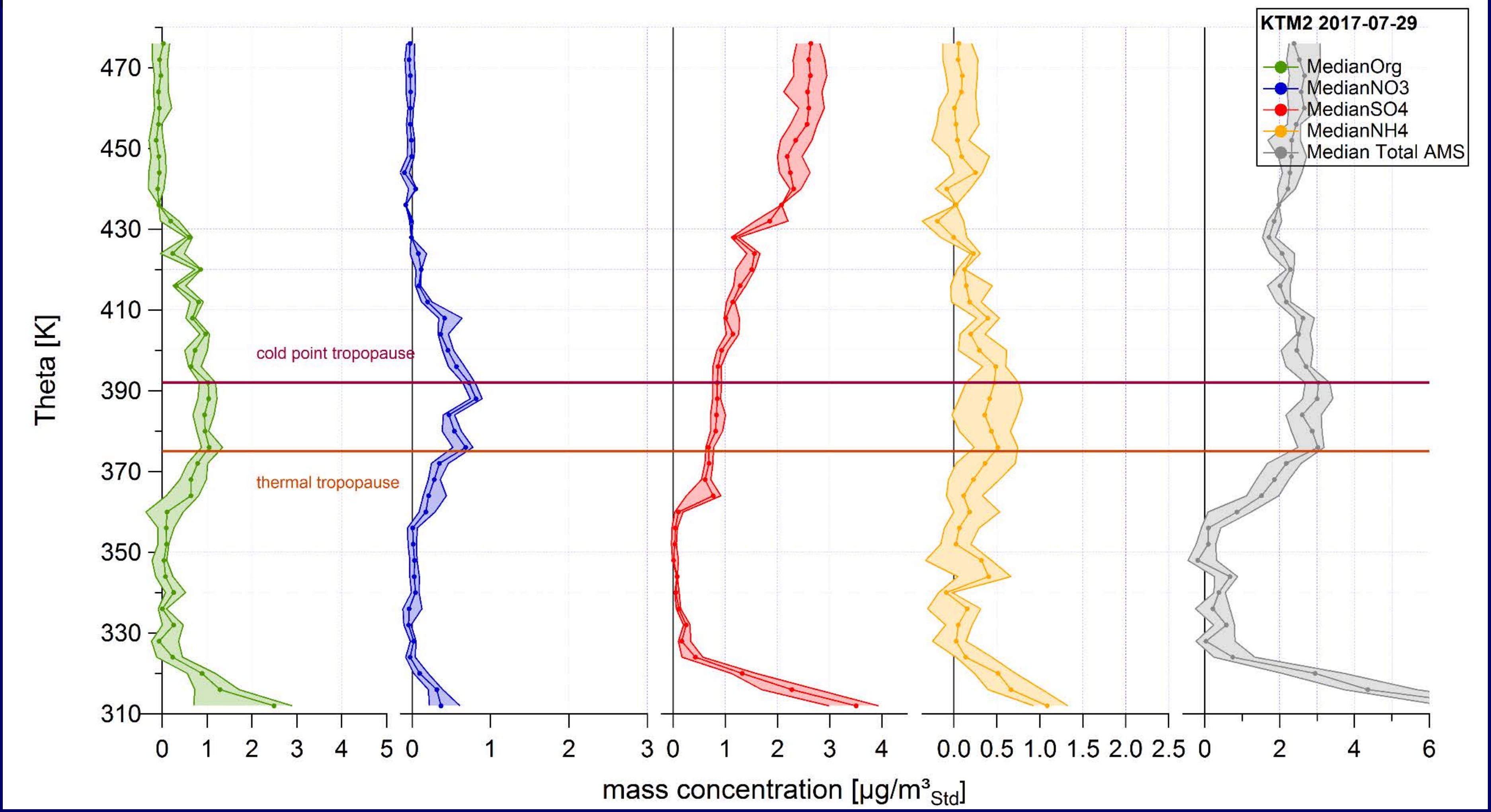
ERICA AMS part – Flights KTM01



500 m ***bin means***
and ***25/75 percentiles***

Analysis: O. Appel, MPIC

ERICA AMS part – Flights KTM02



500 m ***bin means***
and ***25/75 percentiles***

Analysis: O. Appel, MPIC

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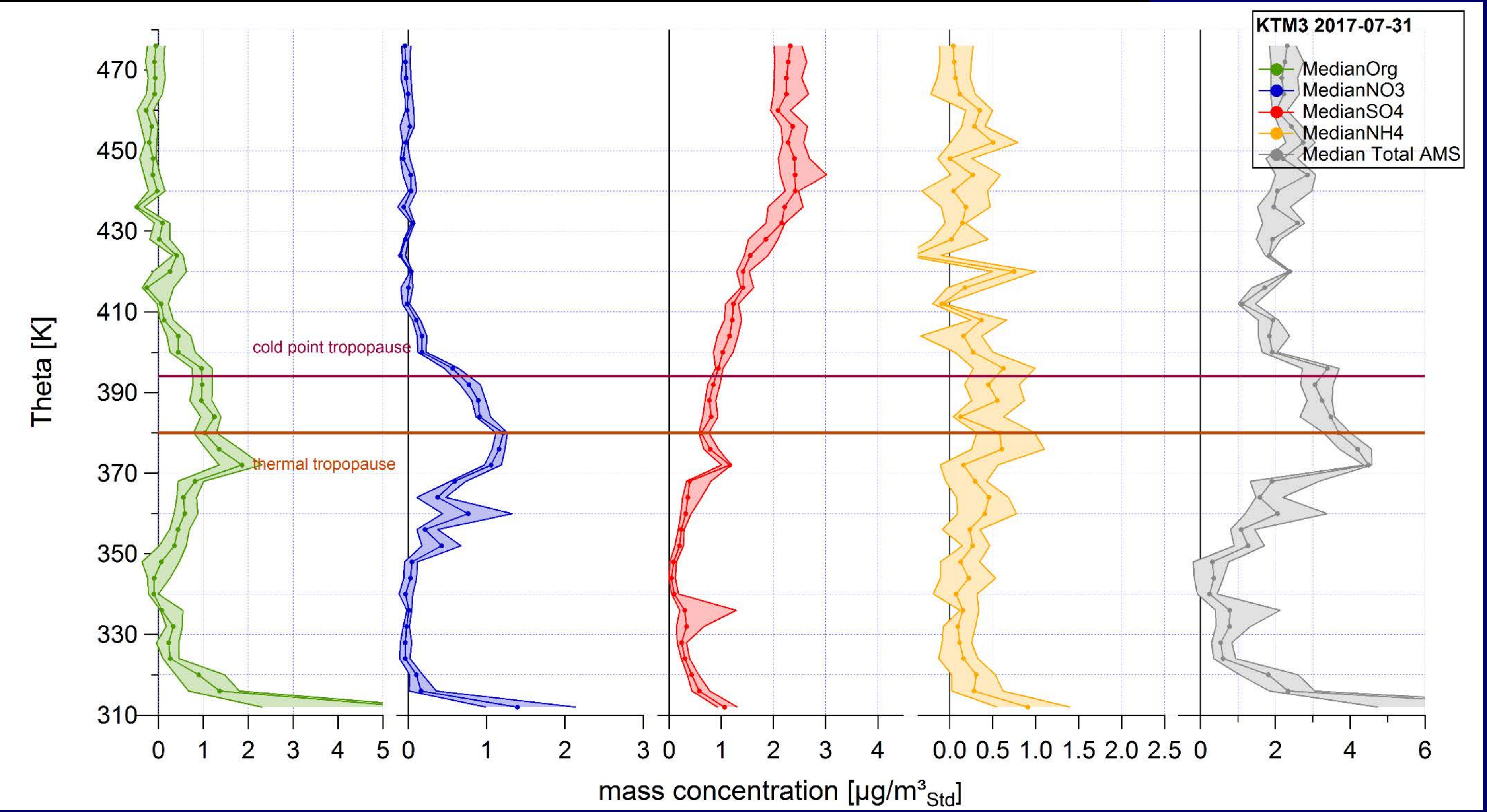
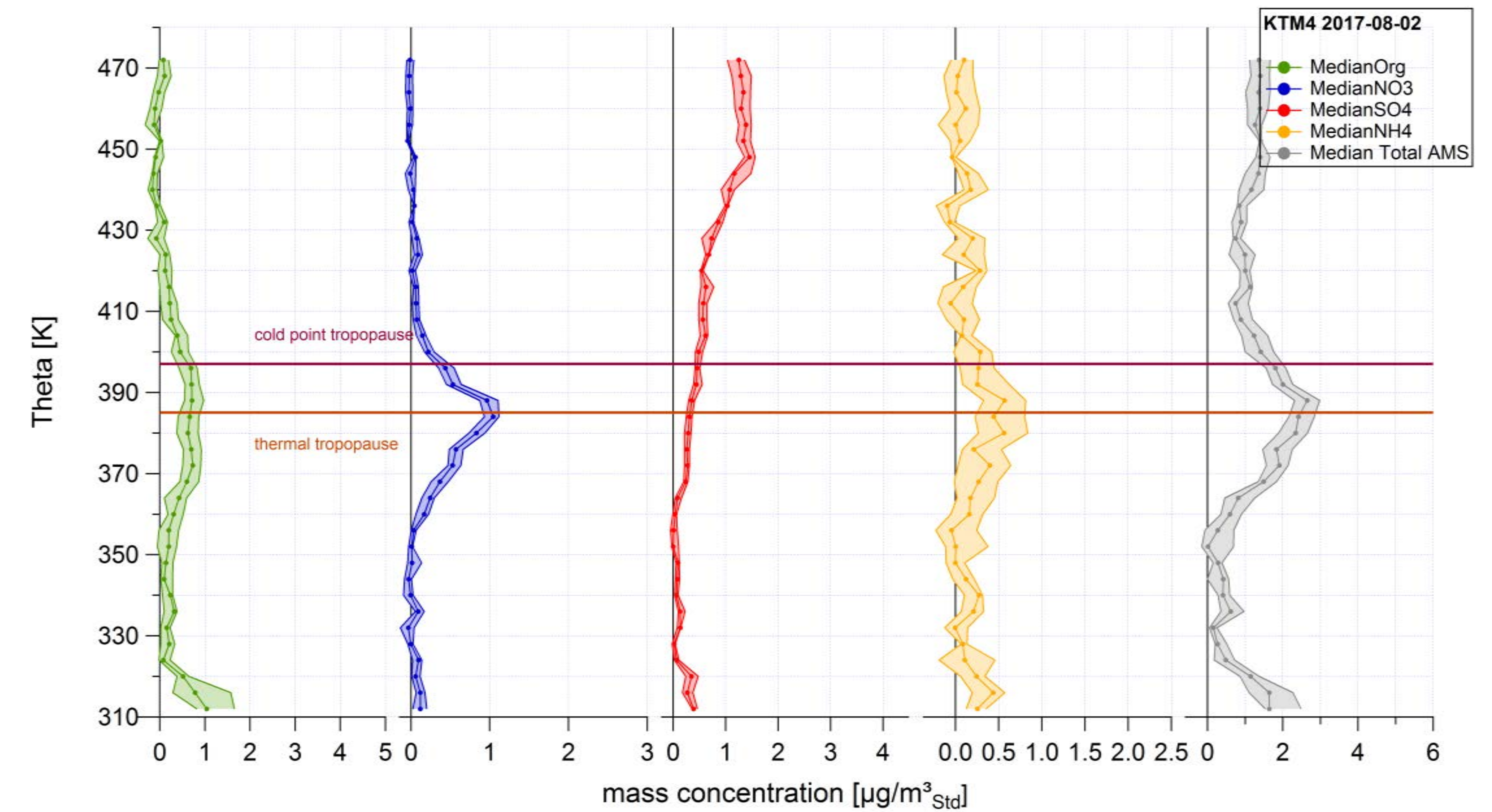
ERICA AMS part – Flights KTM03



500 m ***bin means***
and ***25/75 percentiles***

Analysis: O. Appel, MPIC

ERICA AMS part – Flights KTM04



500 m ***bin means***
and ***25/75 percentiles***

Analysis: O. Appel, MPIC

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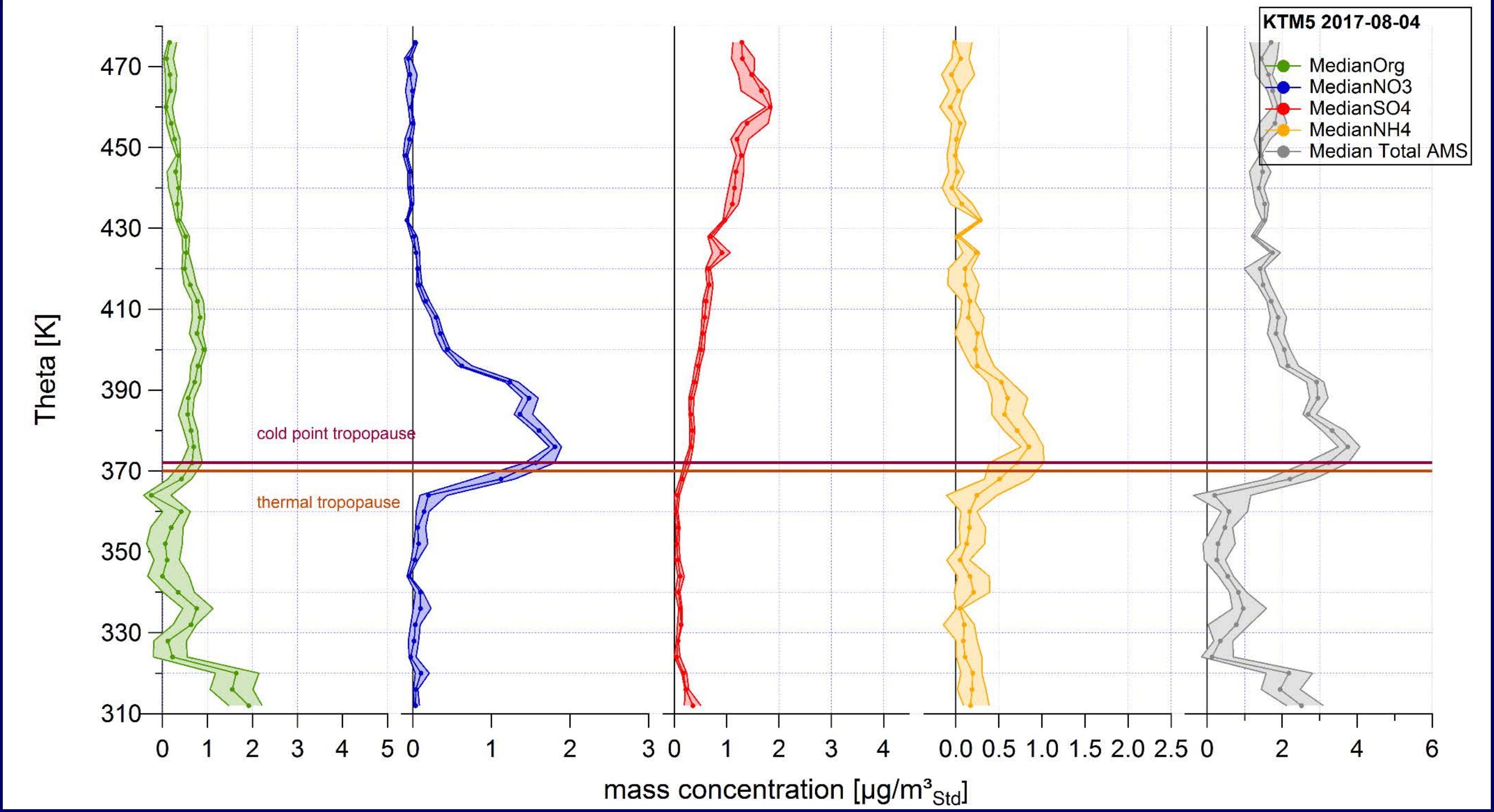
ERICA AMS part – Flights KTM05



500 m ***bin means***
and ***25/75 percentiles***

Analysis: O. Appel, MPIC

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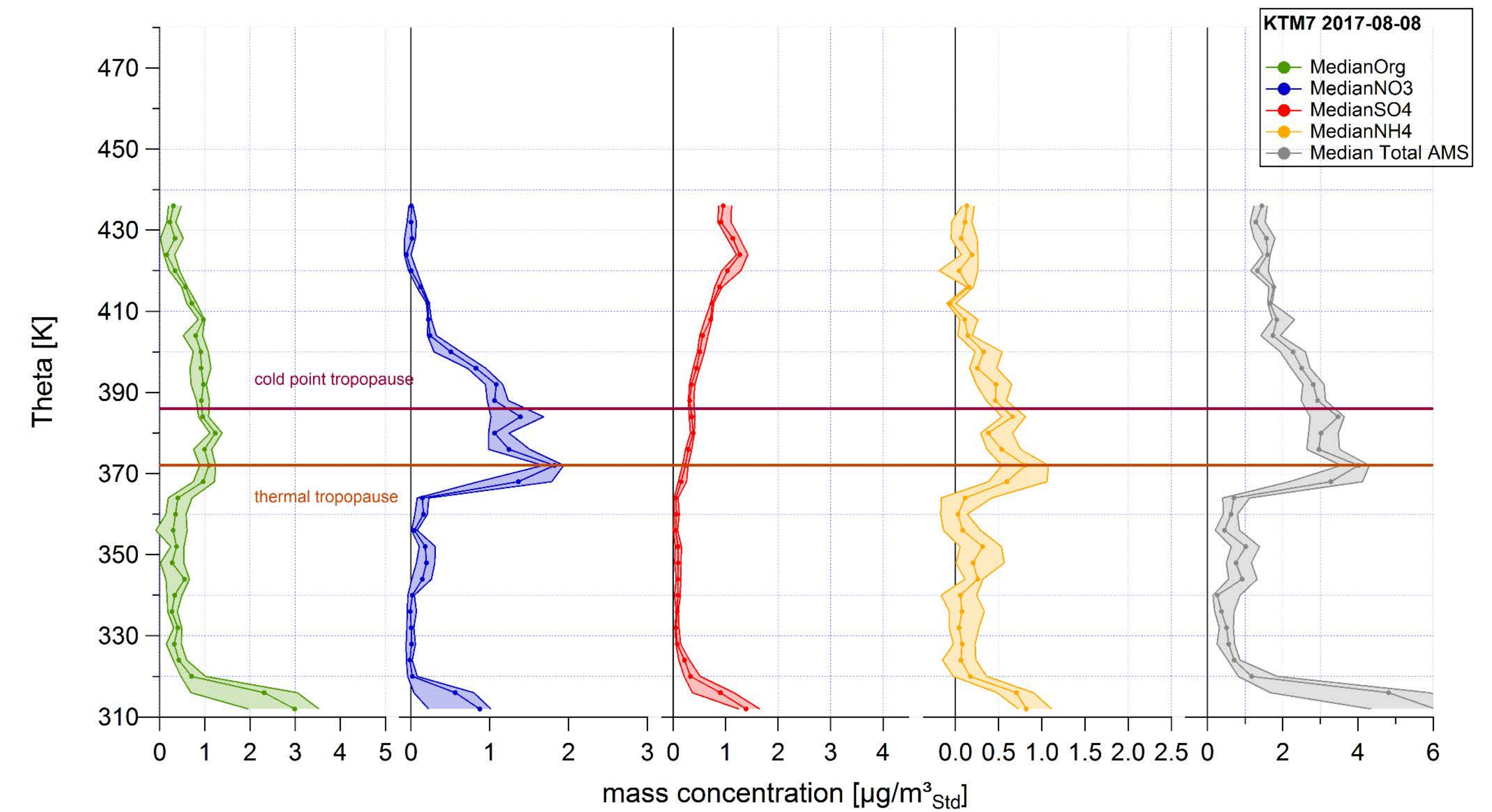
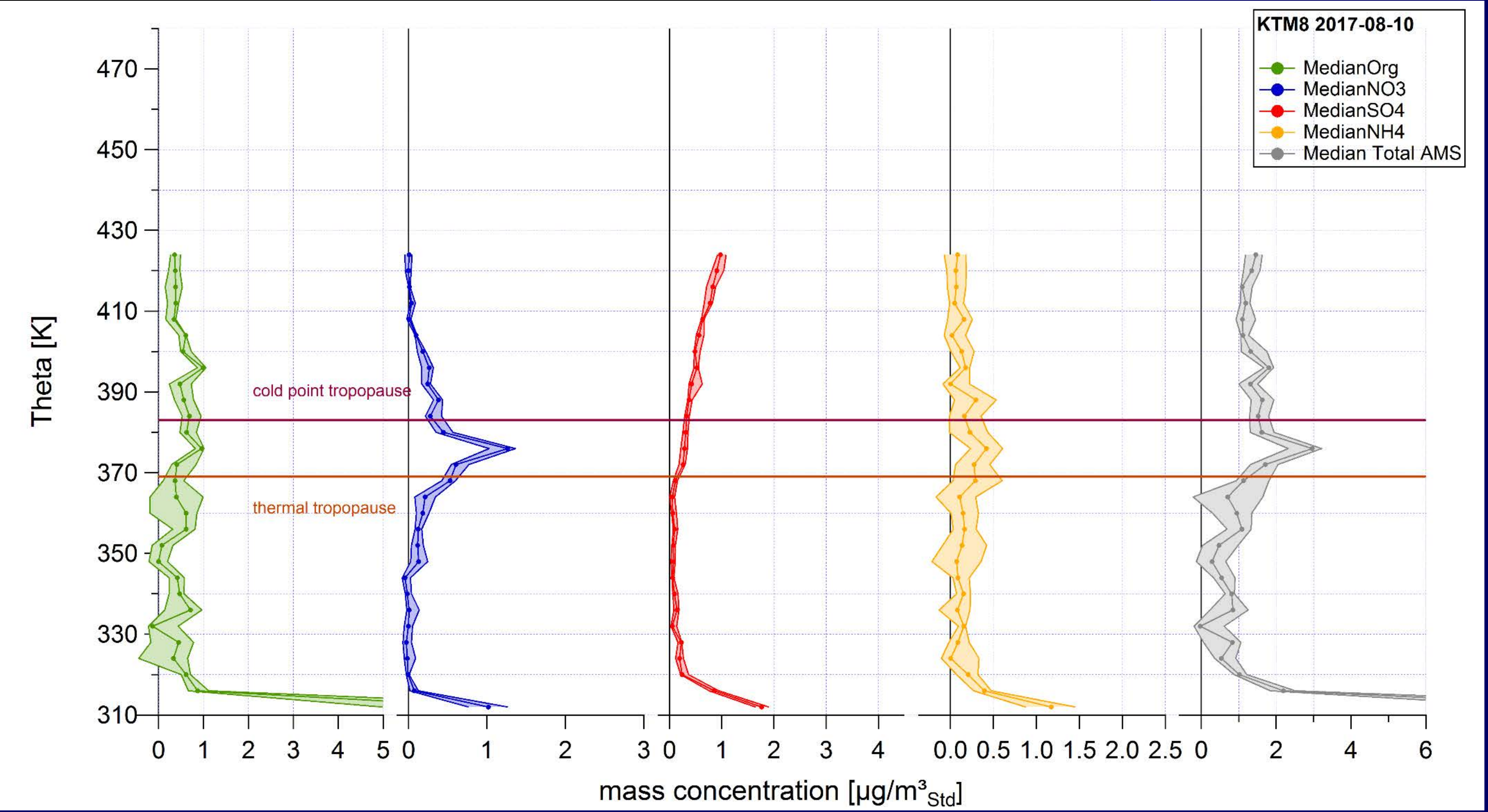
ERICA AMS part – Flights KTM07



500 m ***bin means***
and ***25/75 percentiles***

Analysis: O. Appel, MPIC

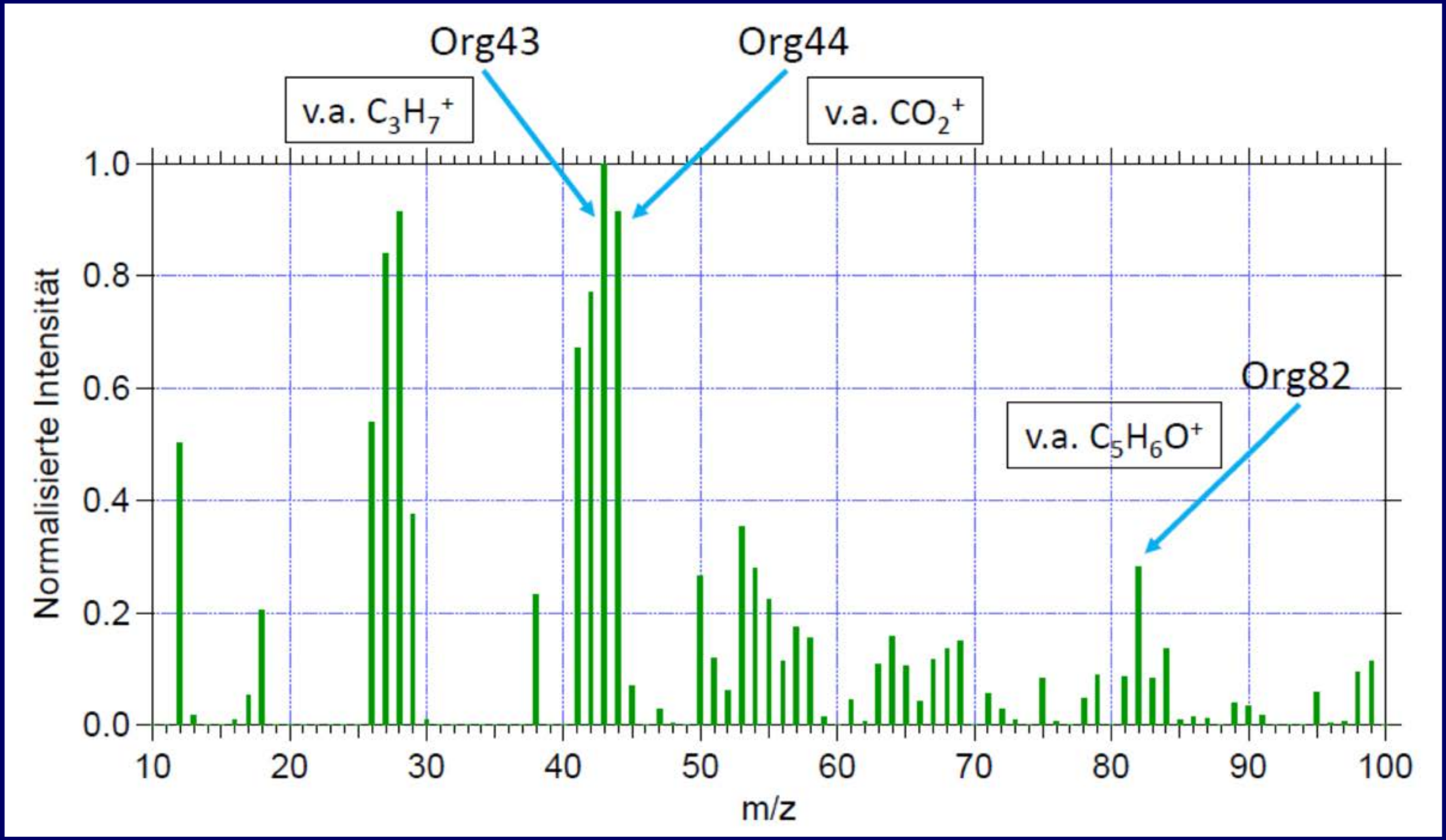
ERICA AMS part – Flights KTM08



500 m ***bin means***
and ***25/75 percentiles***

Analysis: O. Appel, MPIC

Level of aerosol particle oxidation



$$f_{43} = \frac{Org43}{Organik}$$

$$f_{44} = \frac{Org44}{Organik}$$

Signal of mass line 43 (or 44) divided by total signal of all organics mass lines

Analysis: O. Appel, MPIC

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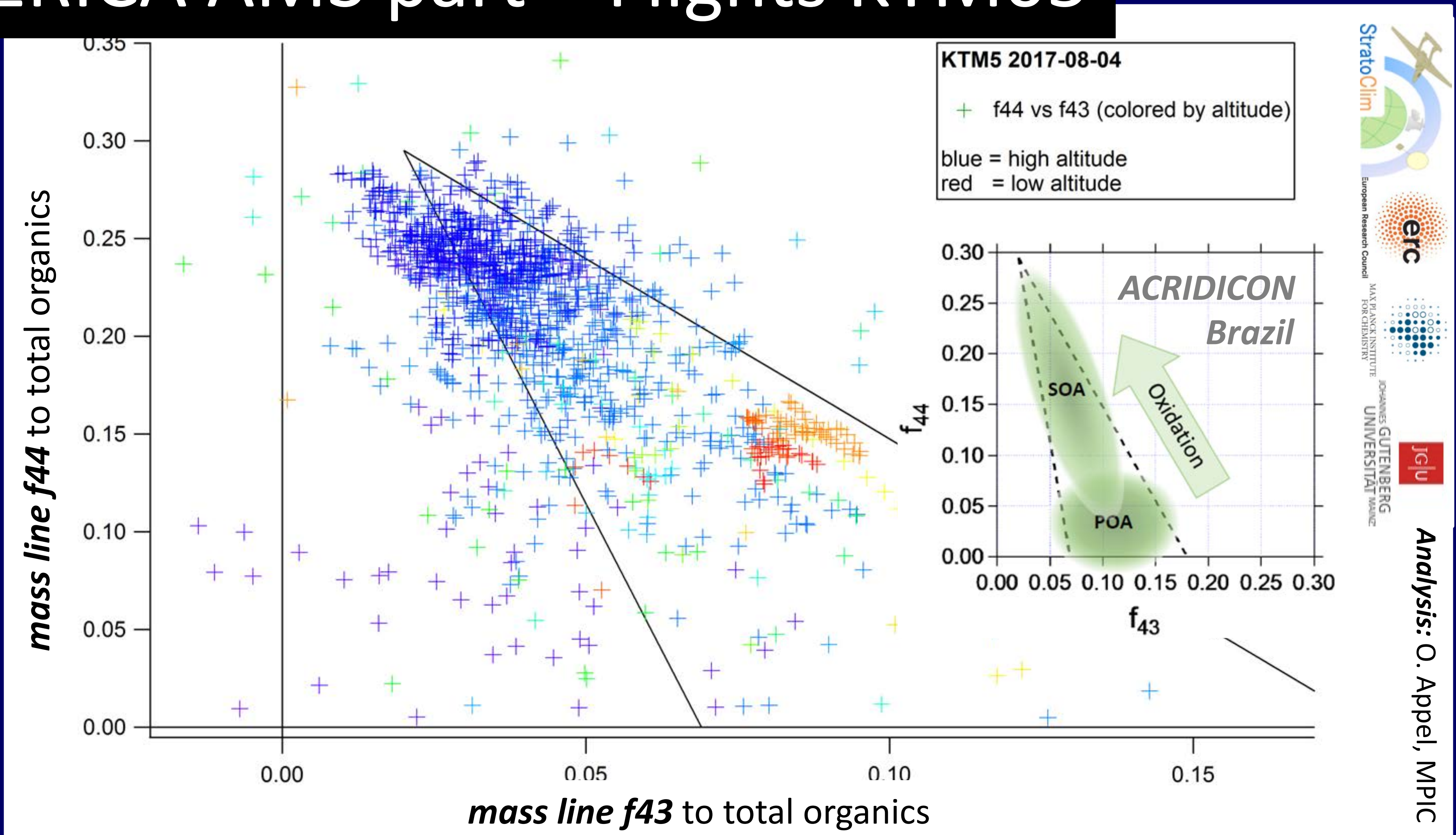
ERICA AMS part – Flights KTM05



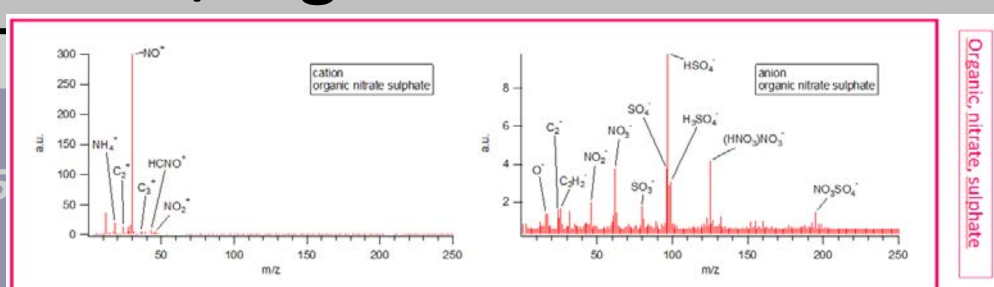
- high fraction of m/z=44 indicates ***aged aerosols*** (Ng et al., 2010)
- StratoClim: medium fresh organics in lower troposphere,
- and aged organics in upper troposphere.

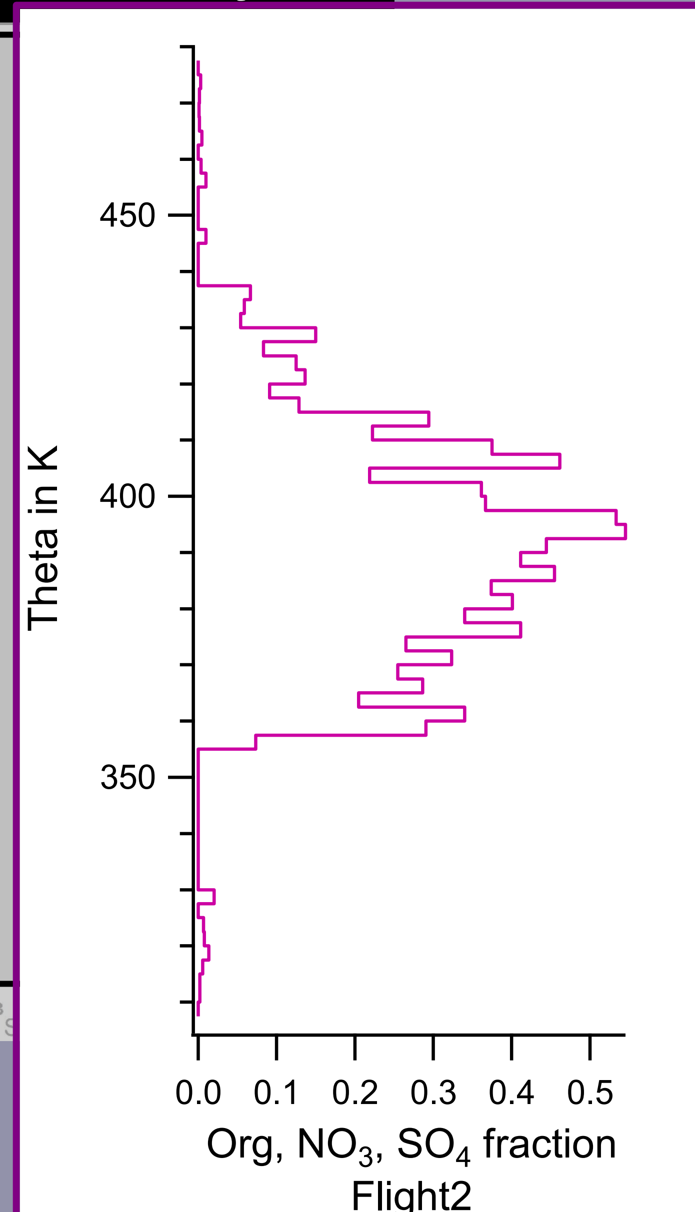
Particle phase in ATAL nitrate layer

- * ***AIDA chamber experiments*** at KIT show ammonium-nitrate particles are solid ONLY IF traces of sulfate are inside the particles
- * ERICA ***single particle*** in situ measurements indicate internal mixtures of nitrate/sulfate/organics in the ATAL.

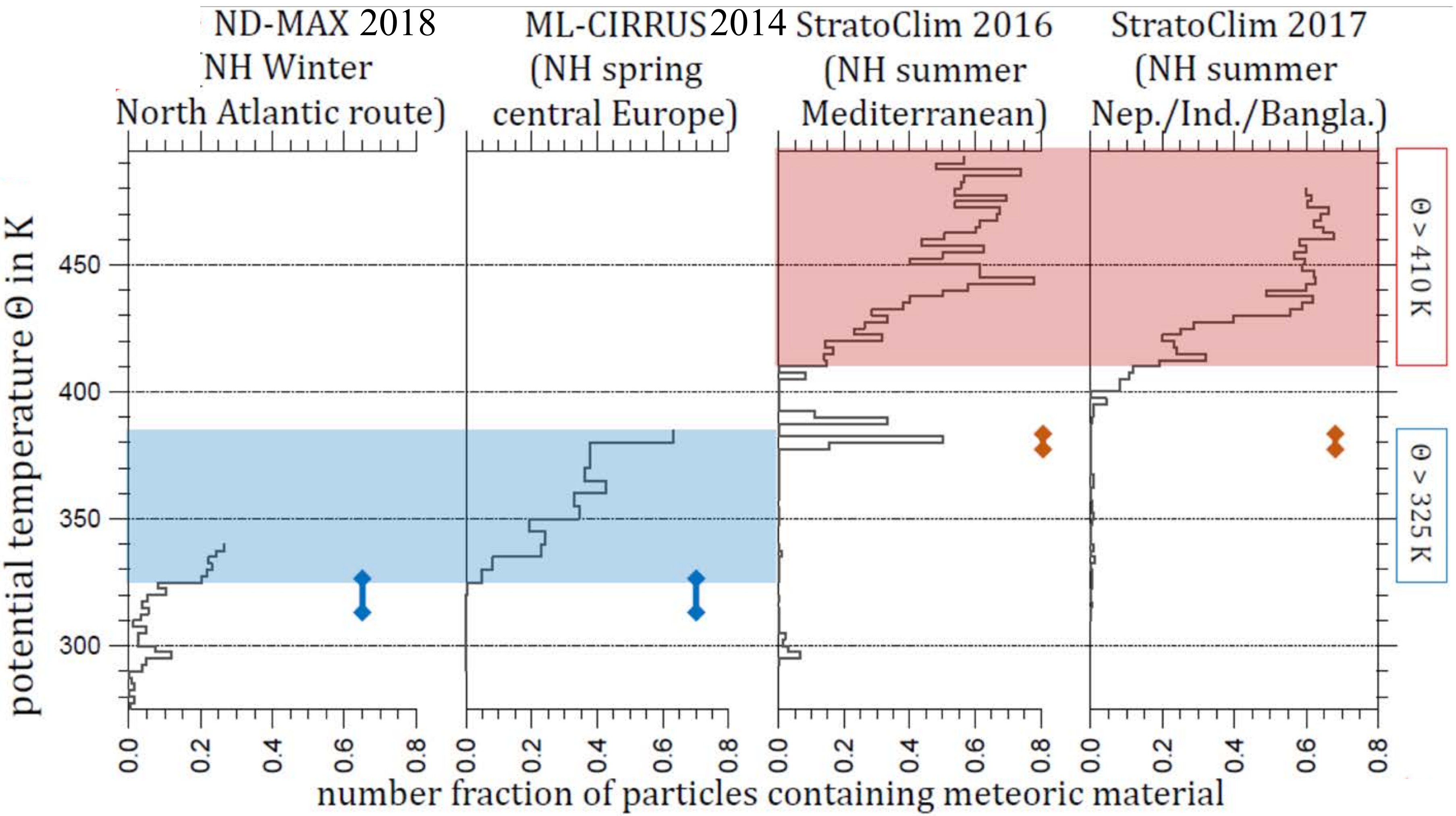
500 m

and ***25***





Meteoric dust from HALO, DC8, and Geophysica



(Cation identification scheme of Cziczo et al., 2001)

Analyses: R. Weigel, J. Schneider, A. Hünig, O. Appel, IPA&MPIC, Mainz

Summary

Findings

* ***Nitrate layer*** consistent feature of the ATAL.
* ATAL particles are mostly ***ammonium nitrate***.
* In-situ measured particulate ammonia sufficient for ***neutralization***.
* Nitrate particles are ***solid, internally mixed*** with sulfate, and organics.
* Particulate ***organics*** significant, having a ***maximum also*** in the AMA/ATAL.
* ATAL aerosol origin probably a mix of NPF and upward transport of ready made particles.
* Detailed info on ***chemical composition*** and ***phase*** of the ATAL gained from StratoClim.
* ***CALIPSO*** and in-situ ***particle size distribution*** data can be compared.

… and Investigators:

Ralf Weigel, Oliver Appel, Antonis Dragoneas, Sergej Molleker, Anneke Batenburg, Oliver Schlenczeck, Andreas Hünig, Christoph Mahnke, Max Port, Johannes Schneider, Frank Drewnick, Thomas Klimach, ***the entire Particle Chemistry Dept.,*** Martina Krämer (FZ Jülich), Jean-Paul Vernier (NASA), Francesco Cairo (CNR) ....

PLUS the coordinators

Markus REX (AWI)

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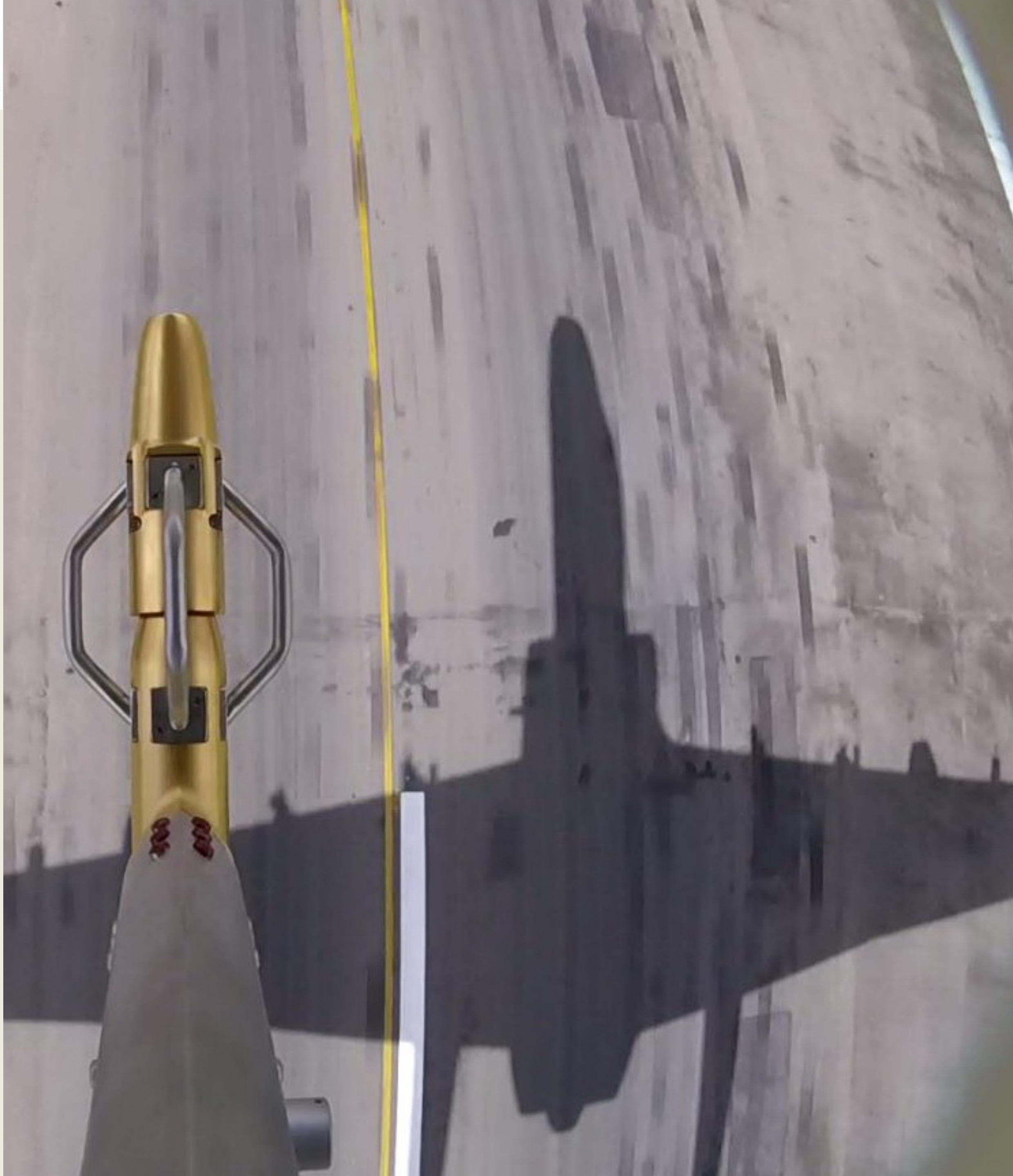
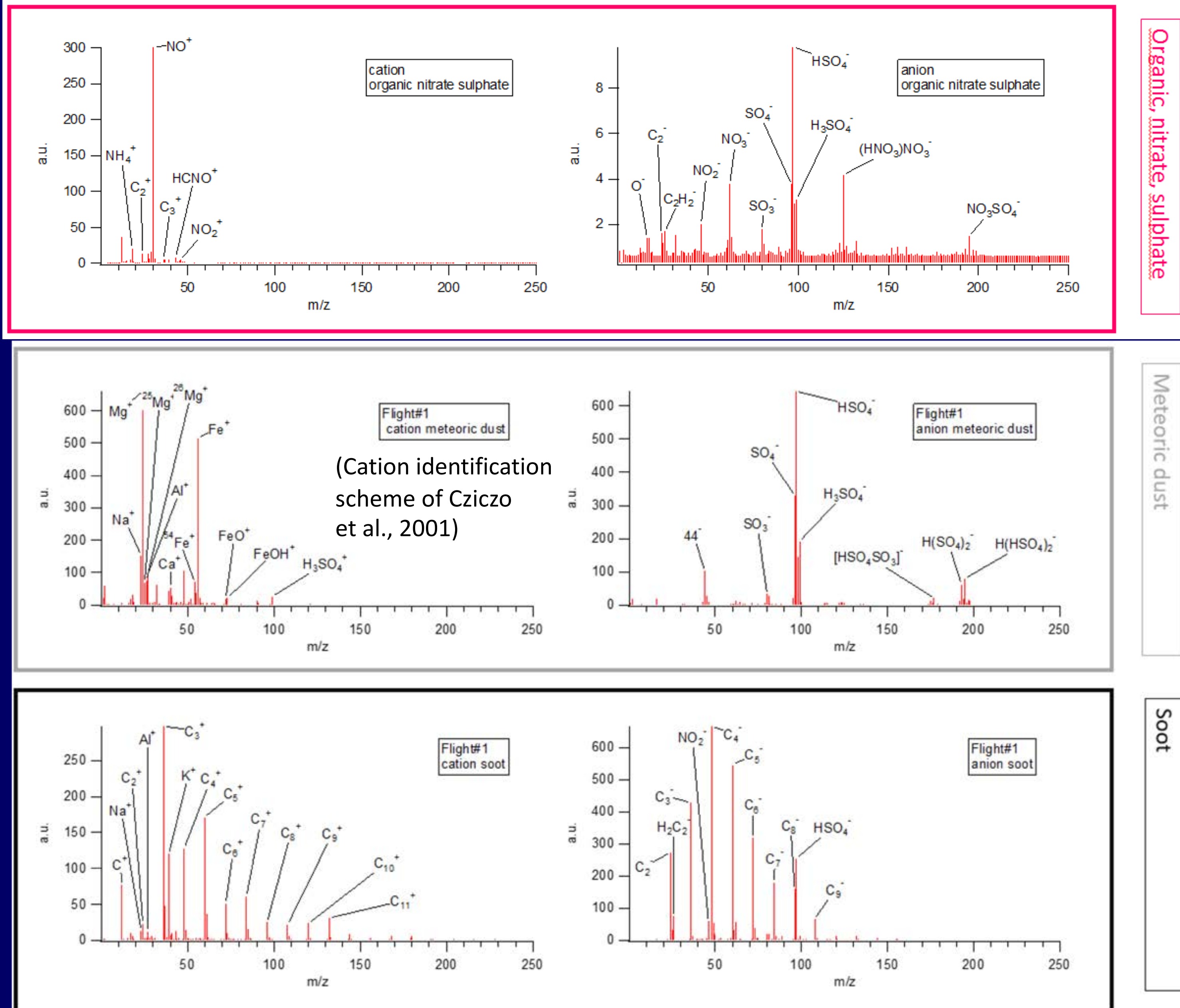


Classification of spectra into 7 particle types: 3 examples



(Cation identification scheme of Cziczo et al., 2001)

Graphics: A. Hünig