The tropical upper troposphere and lower stratosphere: Source region for stratospheric aerosols

Stephan Borrmann ...
University of Mainz
Max-Planck-Institute for Chemistry

AGU Chapman Conference on
Stratospheric Aerosols
Tenerife, *19 March 2018*

… and Investigators:

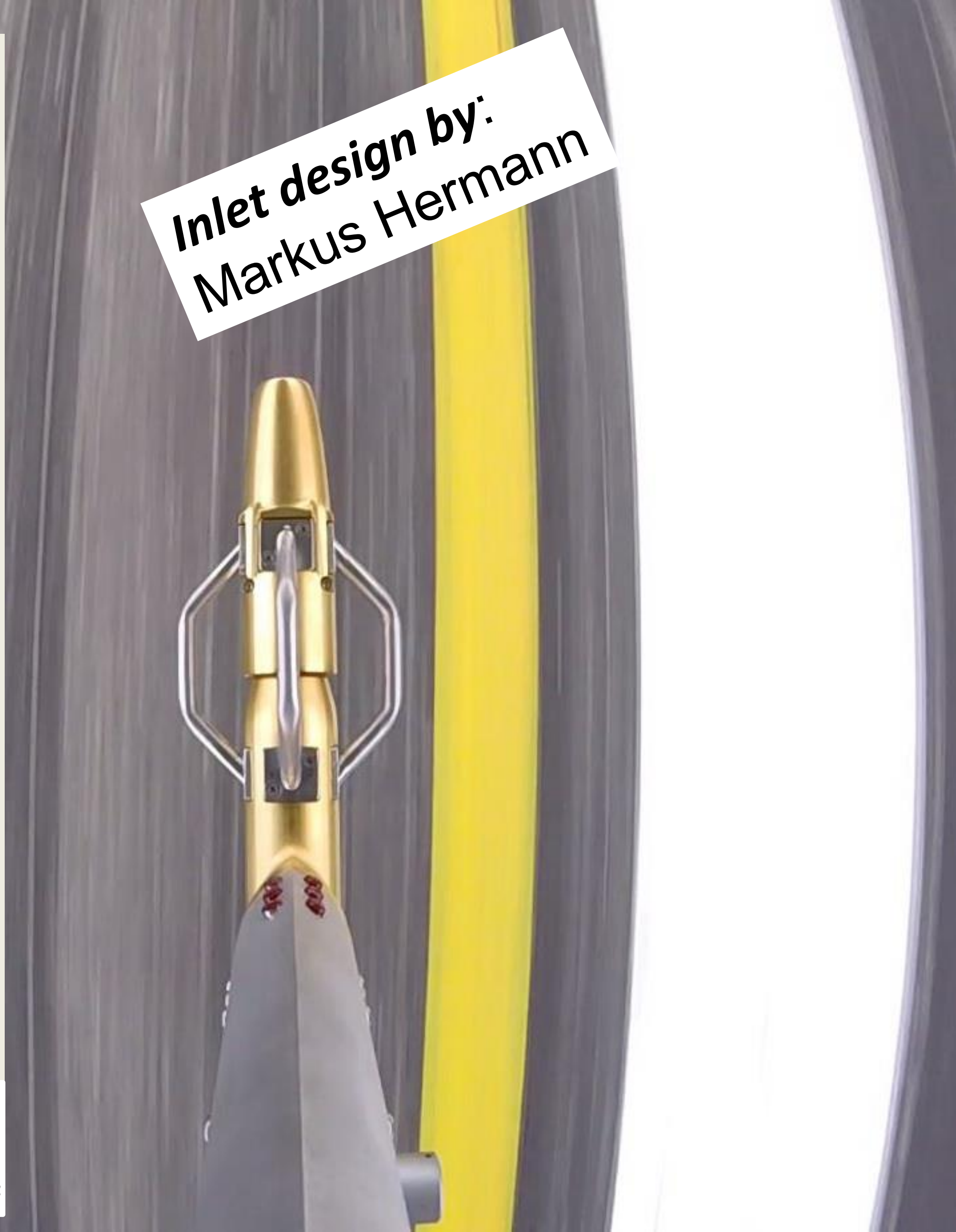
Ralf Weigel, Jacob Fugal, Antonis Dragoneas, Oliver Appel, Sergej Molleker, Anneke Batenburg, Oliver Schlenczeck, Andreas Hünig, Christoph Mahnke, Johannes Schneider, Christiane Schulz, Max Port, our instrument design team, the ***StratoClim*** team, Silvia Vicani (CNR) et many al.

PLUS the coordinators

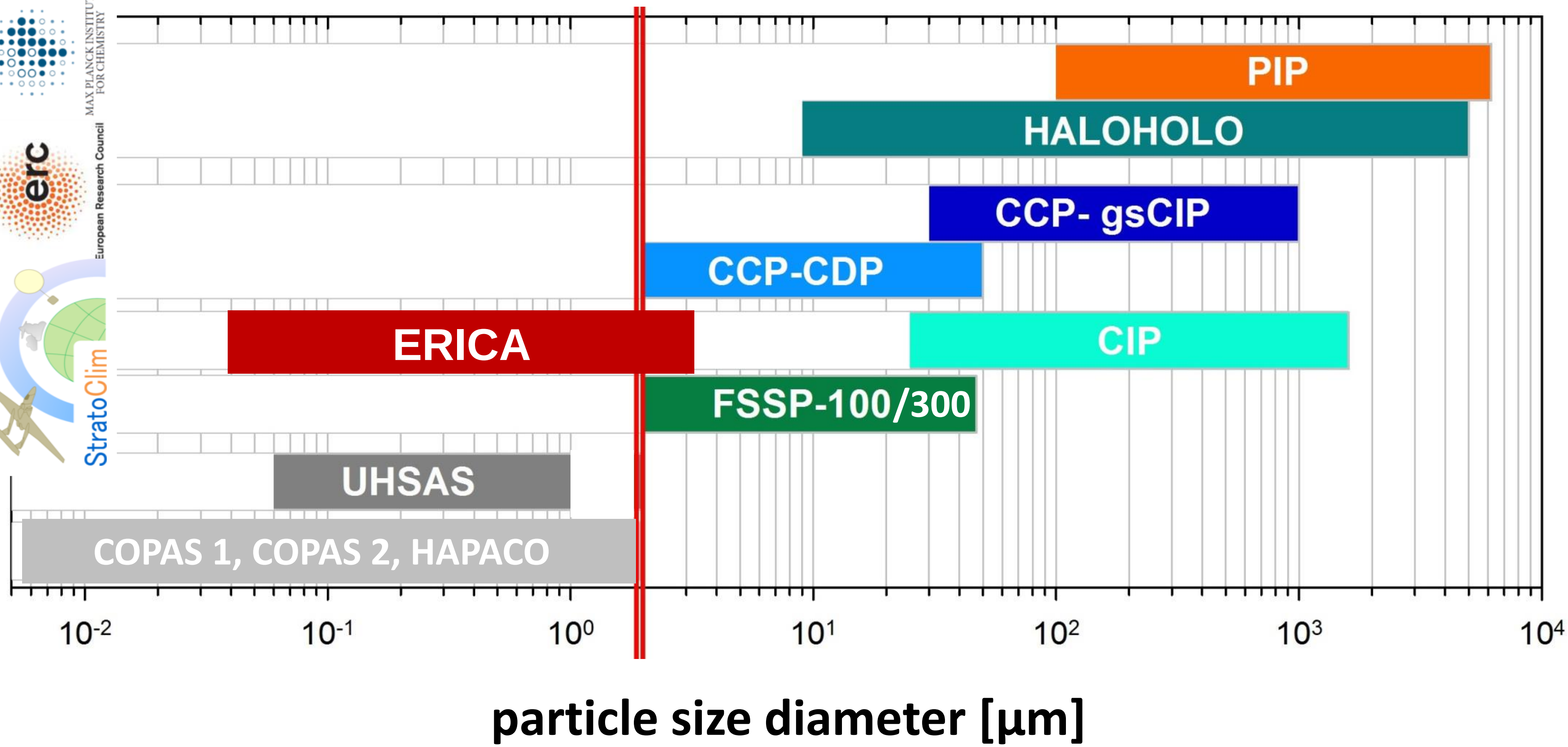
Markus REX (AWI)

Fred STROH (FZ Jülich)

StratoClim — erc European Research Council — MAX PLANCK INSTITUTE FOR CHEMISTRY — JG|U JOHANNES GUTENBERG UNIVERSITÄT MAINZ

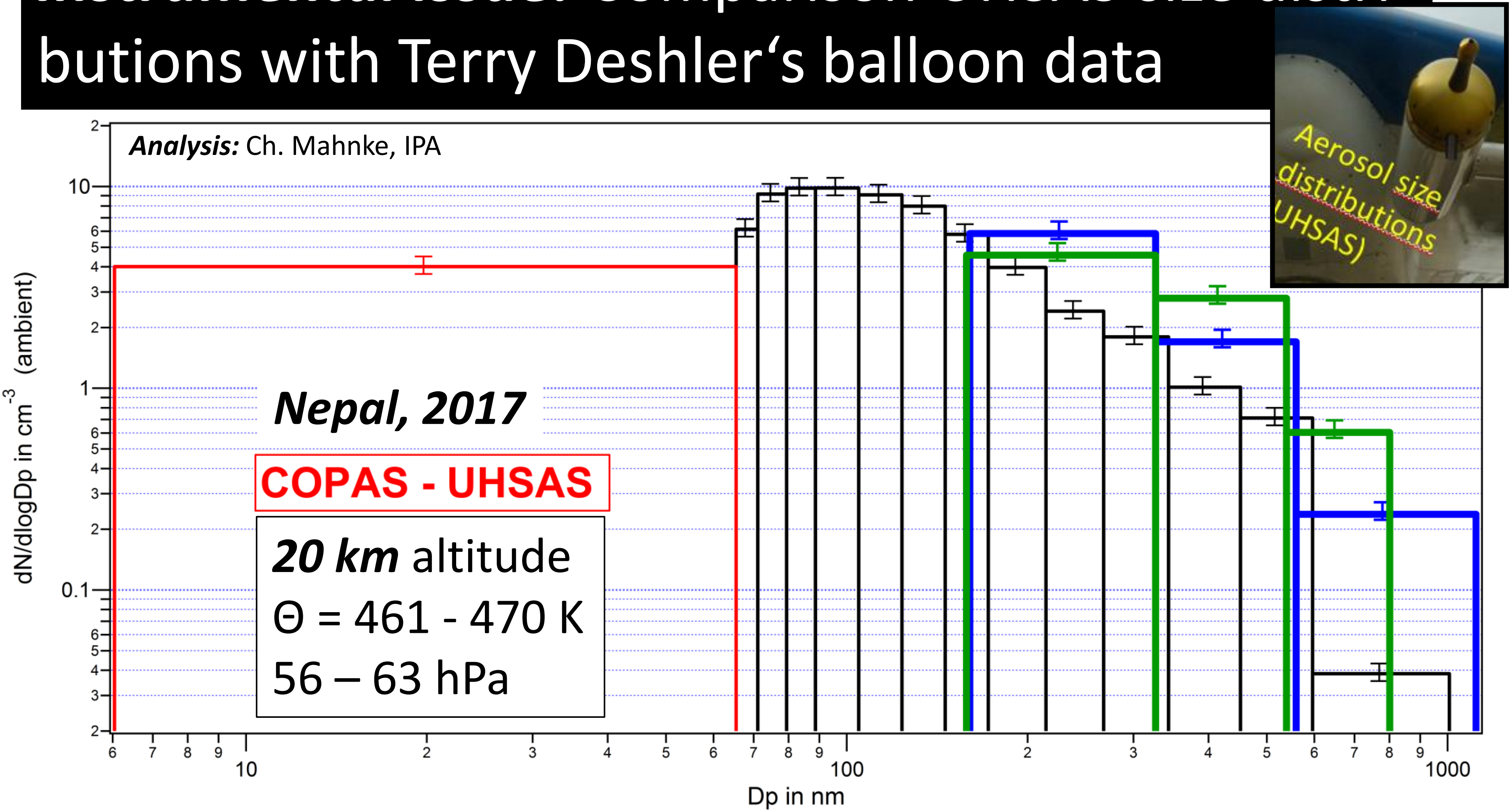


Overview: AEROSOL instruments on "Geophysica"
Particles from 6 nm to 6 mm (Mainz)



Techniques: Condensation particle counting, sampling/collecting (SEM, EDX), light scattering, shadow casting, holographic imaging, laser ablation mass spectrometry, thermodesorption mass spectrometry

Instrumental issue: Comparison UHSAS size distributions with Terry Deshler's balloon data



- ***Hyderabad (India), August 2013***
- ***Wyoming, August 2013***

ERICA – ERC Instrument for the the Chemical Composition of Aerosols

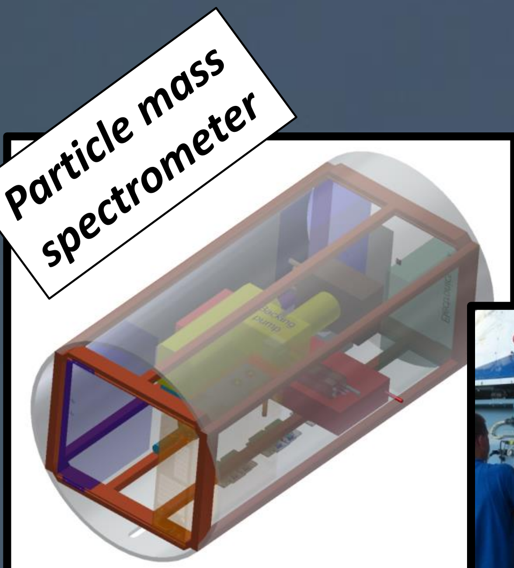
JOHANNES GUTENBERG UNIVERSITÄT MAINZ

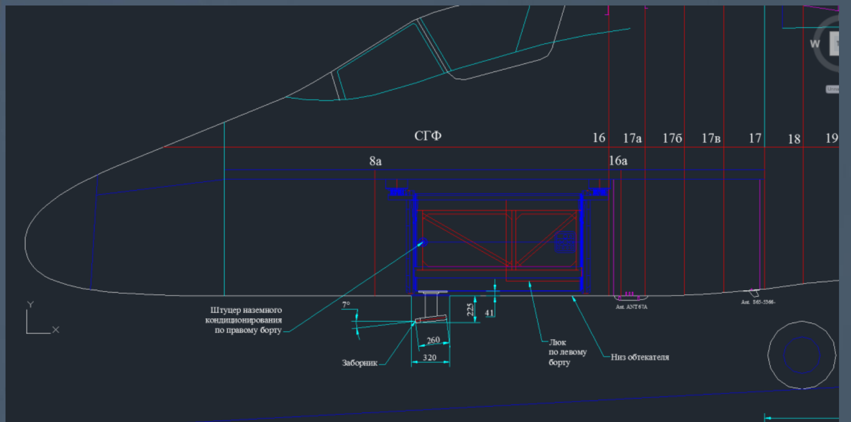
European Research Council

MAX-PLANCK-INSTITUT FÜR CHEMIE

EU funded StratoClim campaigns:

- Kalamata, Greece, 2016
- Nepal July/Aug 2017
- *(Jan/Feb 2018 on NASA DC-8)*

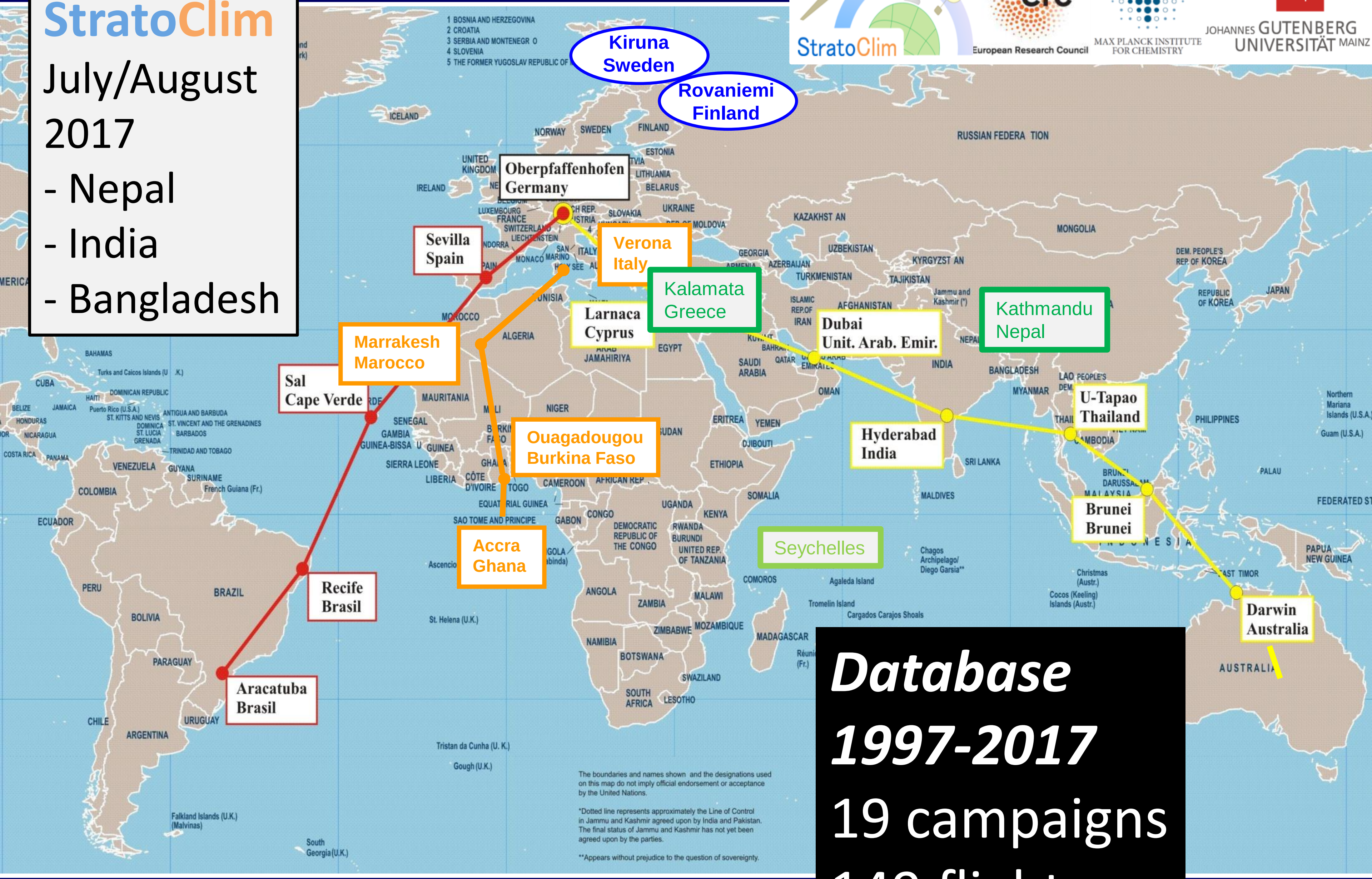




StratoClim

July/August 2017

- Nepal
- India
- Bangladesh

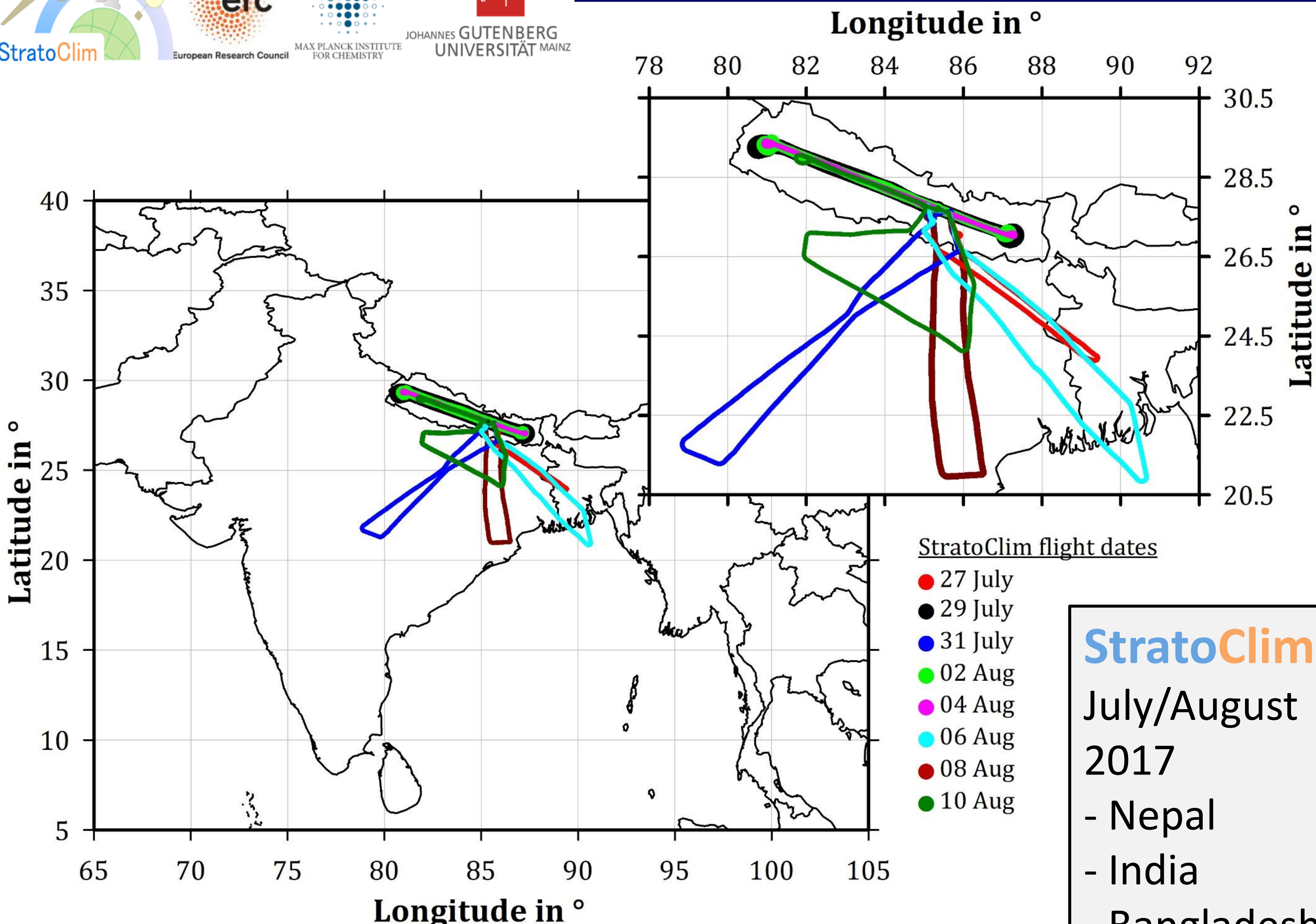


Database

1997-2017

19 campaigns

149 flights



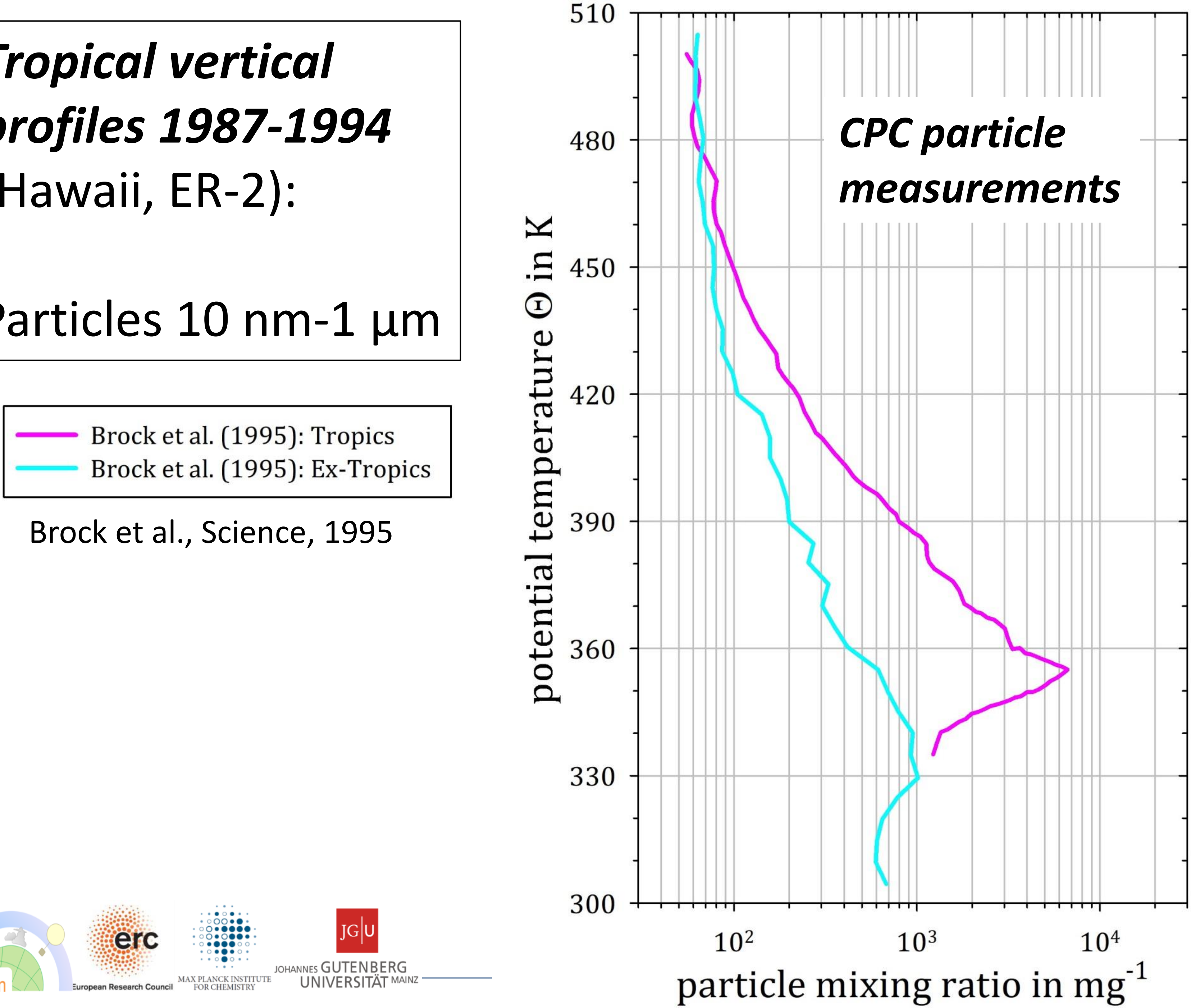
StratoClim

July/August 2017

- Nepal
- India
- Bangladesh

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

Particles 10 nm-1 µm



Brock et al., Science, 1995

StratoClim European Research Council MAX PLANCK INSTITUTE FOR CHEMISTRY JOHANNES GUTENBERG UNIVERSITÄT MAINZ

Tropical vertical profiles 2005-2006

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

Particles 10 nm-1 µm

Brazil, ***Australia***,

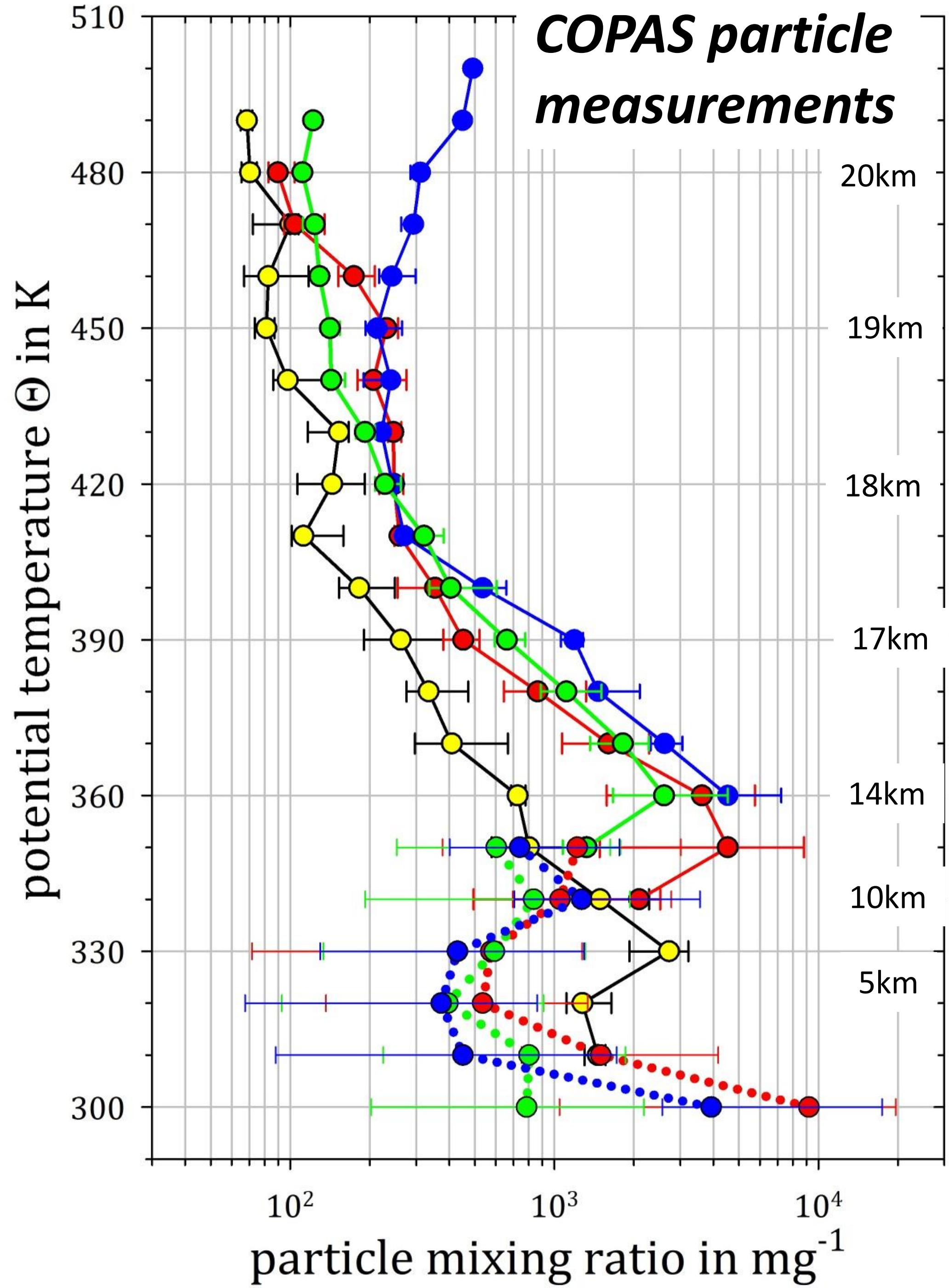
West Africa

- 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



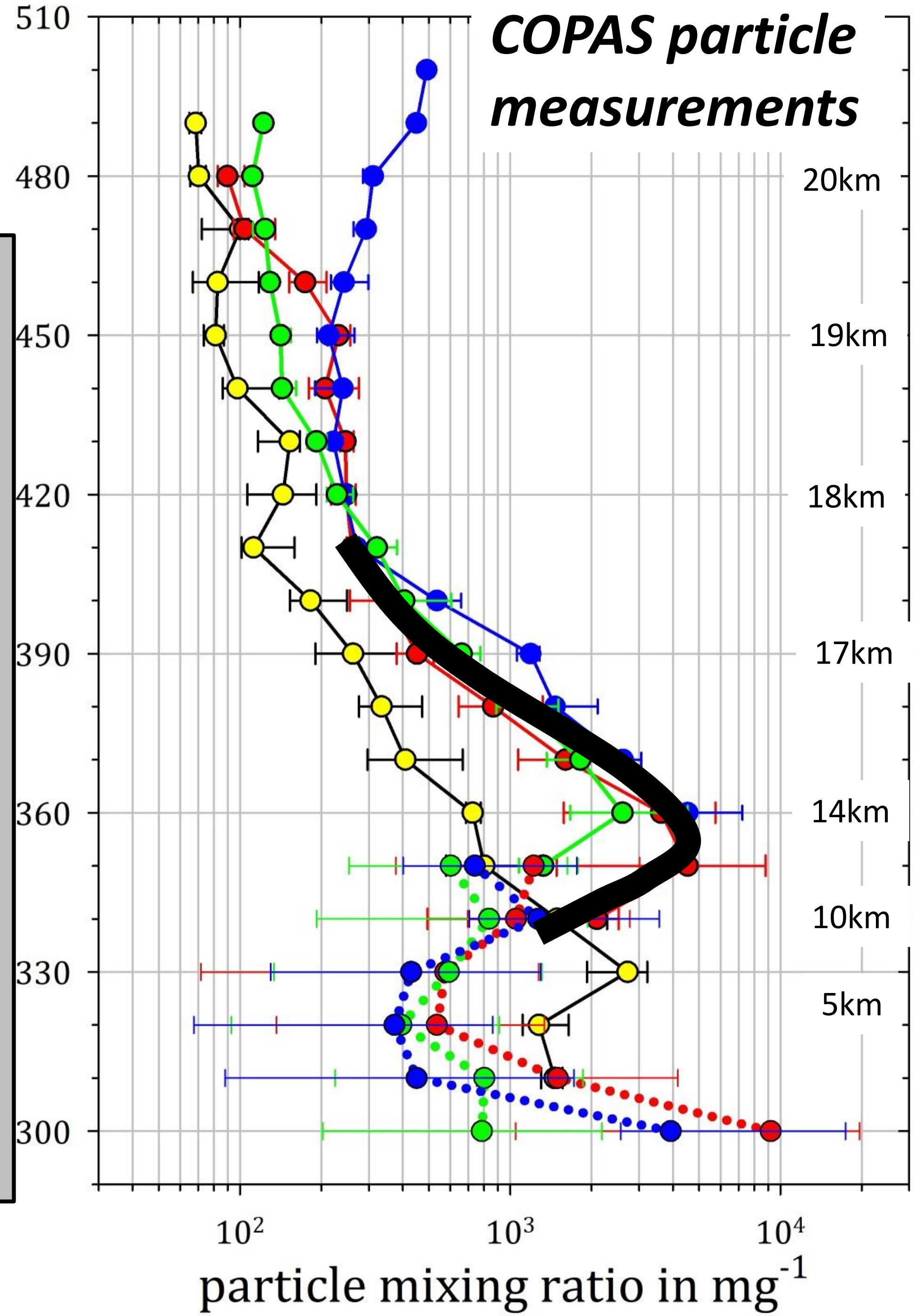
StratoClim
erc
European Research Council
MAX PLANCK INSTITUTE FOR CHEMISTRY
JG|U
JOHANNES GUTENBERG UNIVERSITÄT MAINZ

Tropical vertical profiles 2005-2006

(Russian M-55 „Geophysica“

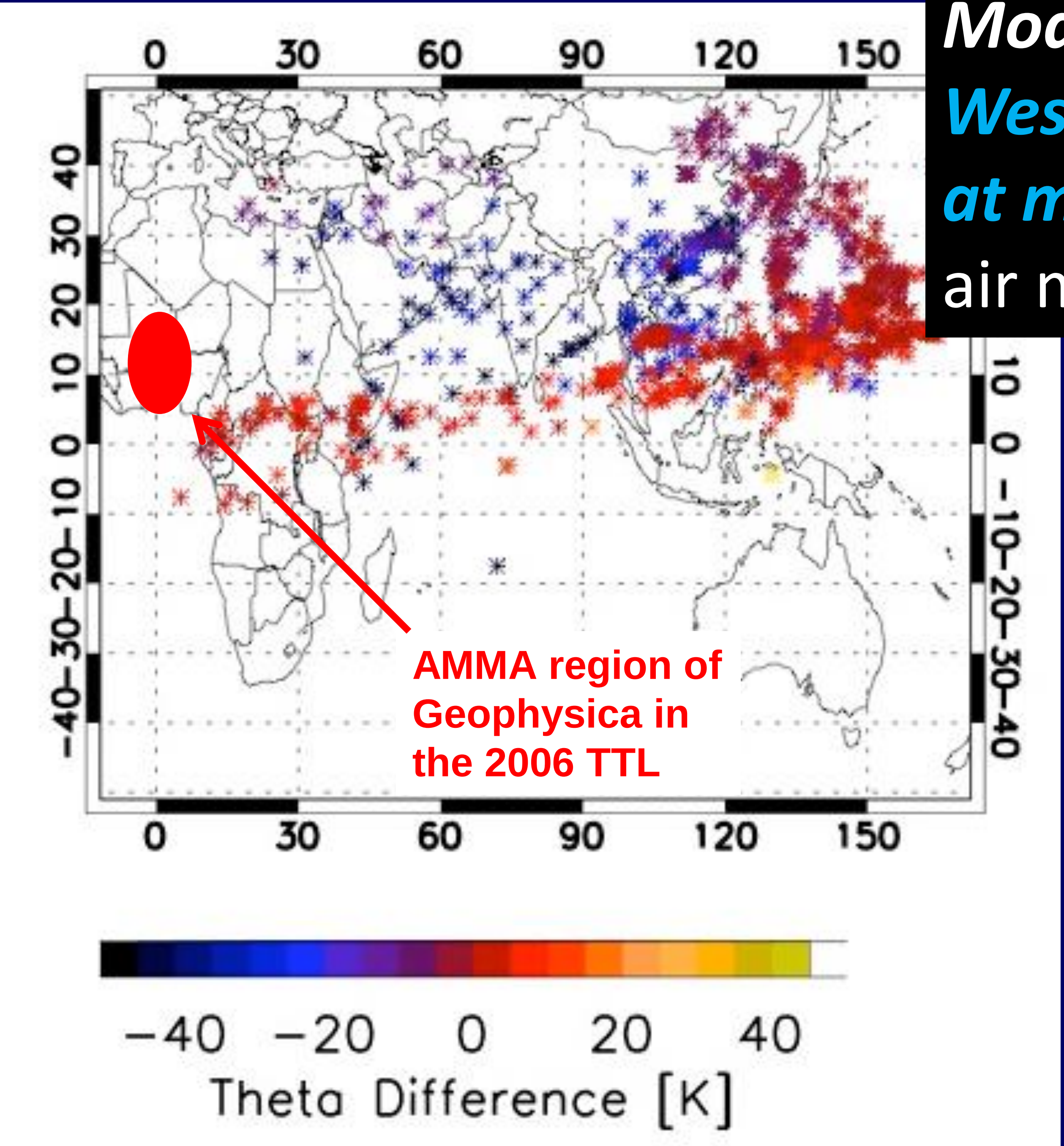
Interpretation:

* global layer of submicron particles in the **tropical belt** betw. 340K and 400K
* in analogy to Junge layer
* has maximum inside and below the TTL (***Tropical Transition Layer***)
* sources/processes unclear
* potential ***source for SP particles*** (Brock et al., 1995)



StratoClim
European Research Council
MAX PLANCK INSTITUTE FOR CHEMISTRY
JG|U JOHANNES GUTENBERG UNIVERSITÄT MAINZ

Model simlations:
West African TTL at monsoon times
air mass origin?



Domain filling trajectories:

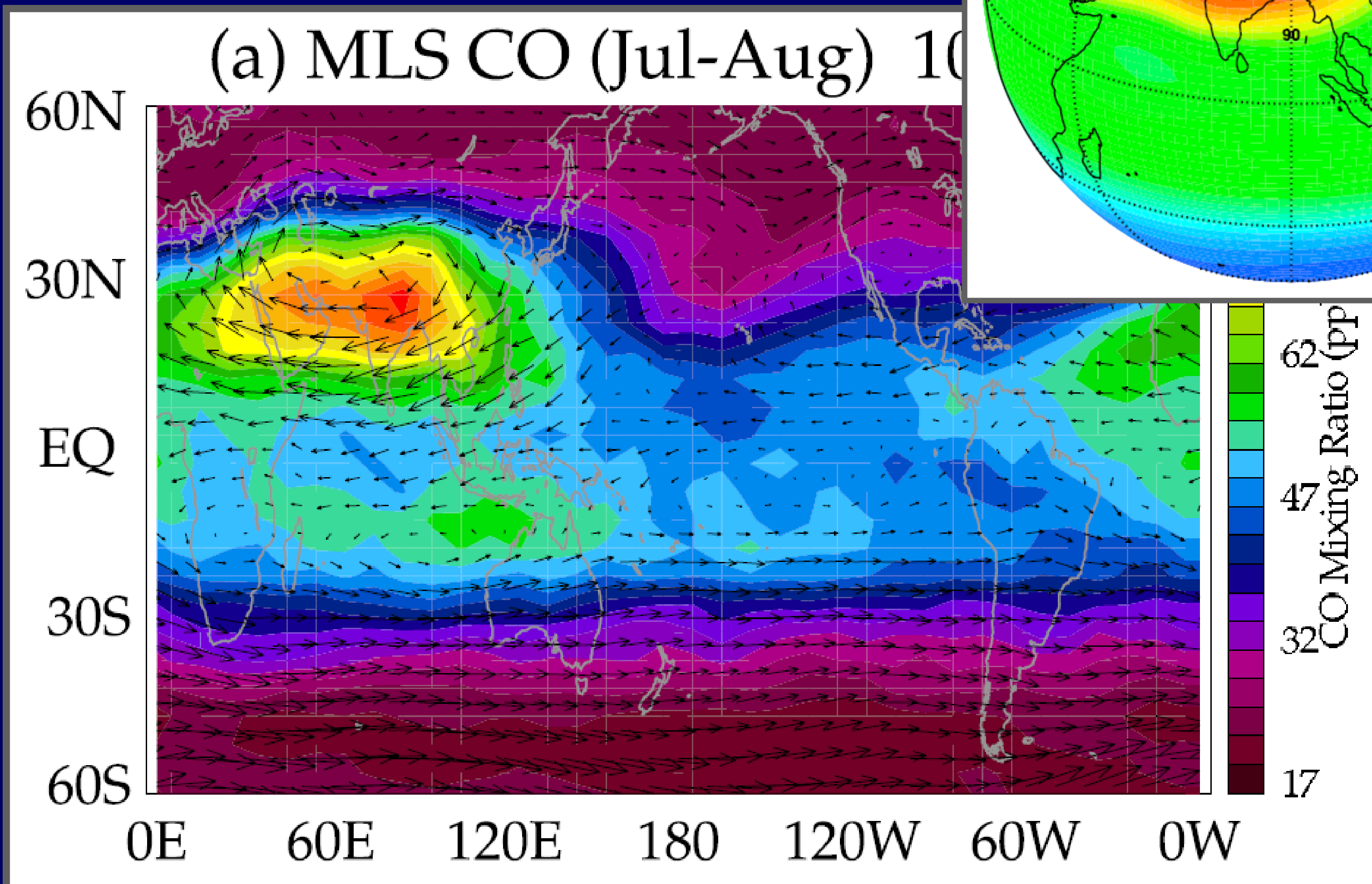
*** Residence time inside TTL ≈ 10 dy**

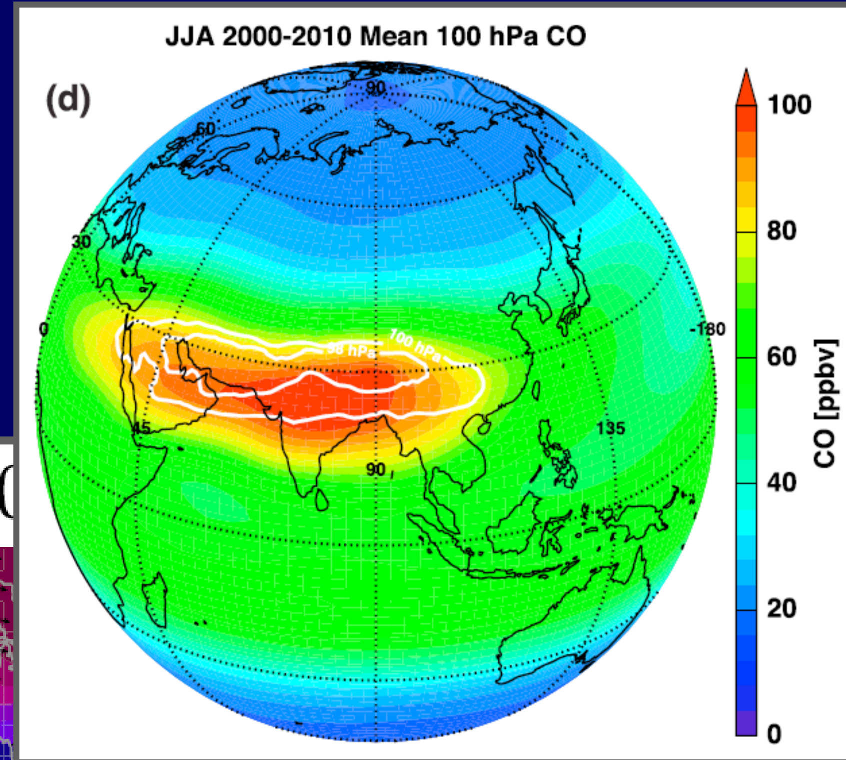
*** 39% of trajectories contain signatures from Asia, India**

*** few point to BMB**

Law et al., ACP, 2010
Fierli et al., ACP, 2011

Asian Monsoon Anticyclone

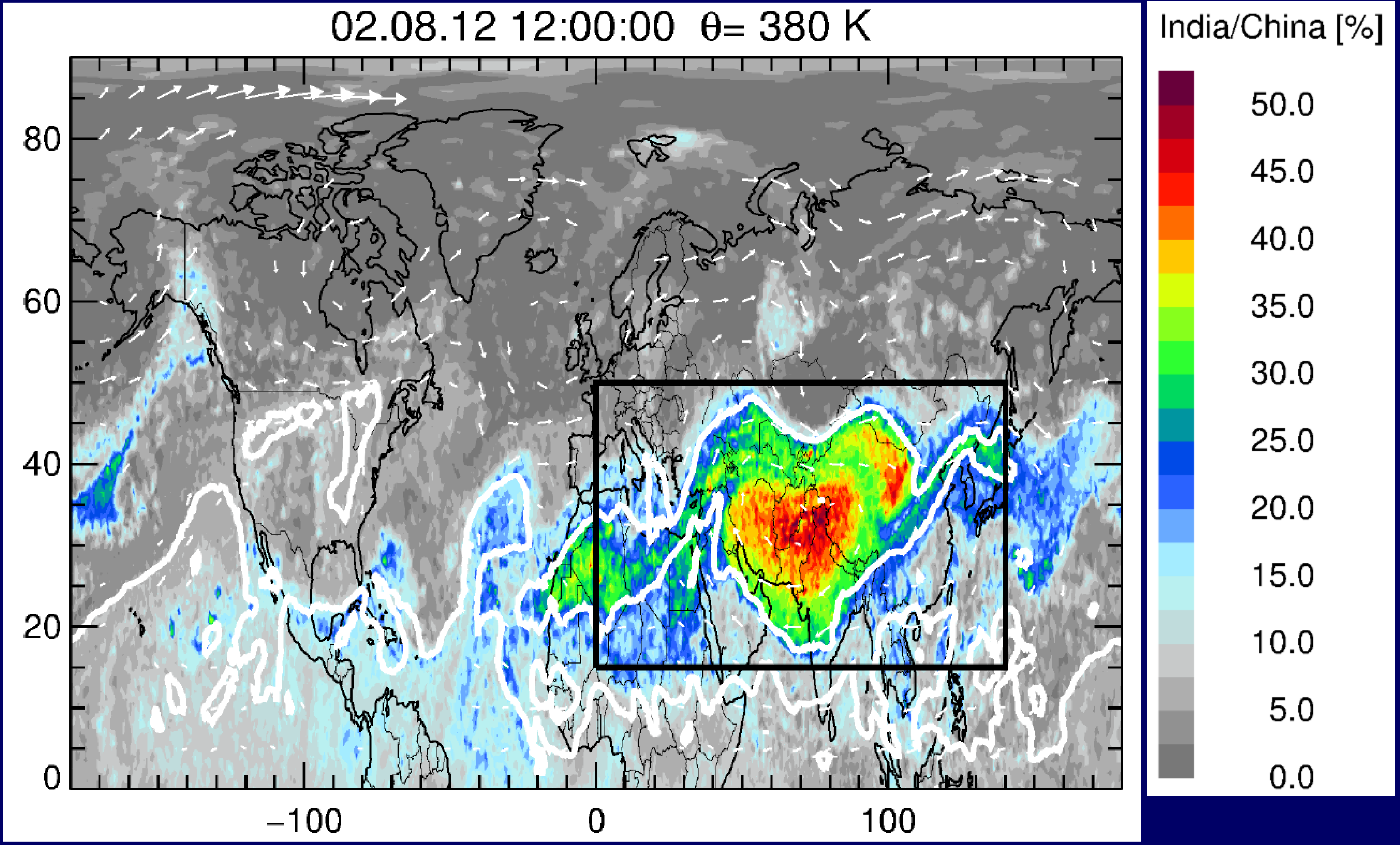




Pan et al., JGR, 2016

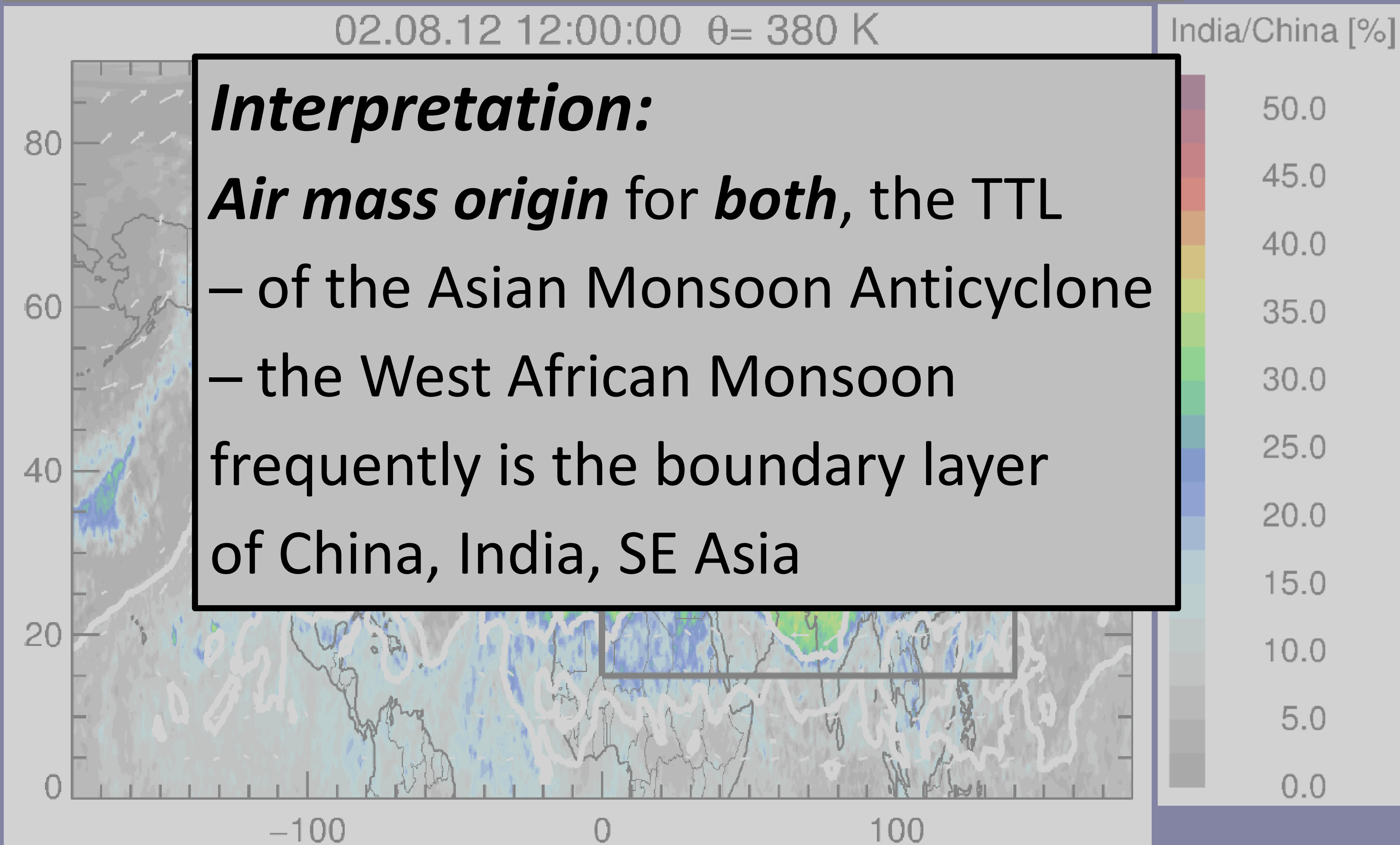
Park et al., JGR, 2007

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



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

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



Interpretation:

Air mass origin for ***both***, the TTL

– of the Asian Monsoon Anticyclone

– the West African Monsoon

frequently is the boundary layer

of China, India, SE Asia

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

COPAS particle measurements

Tropical vertical profiles 2005-2006

New: August 2017

TTL above Ka

Nepal → insic

Asian Monso

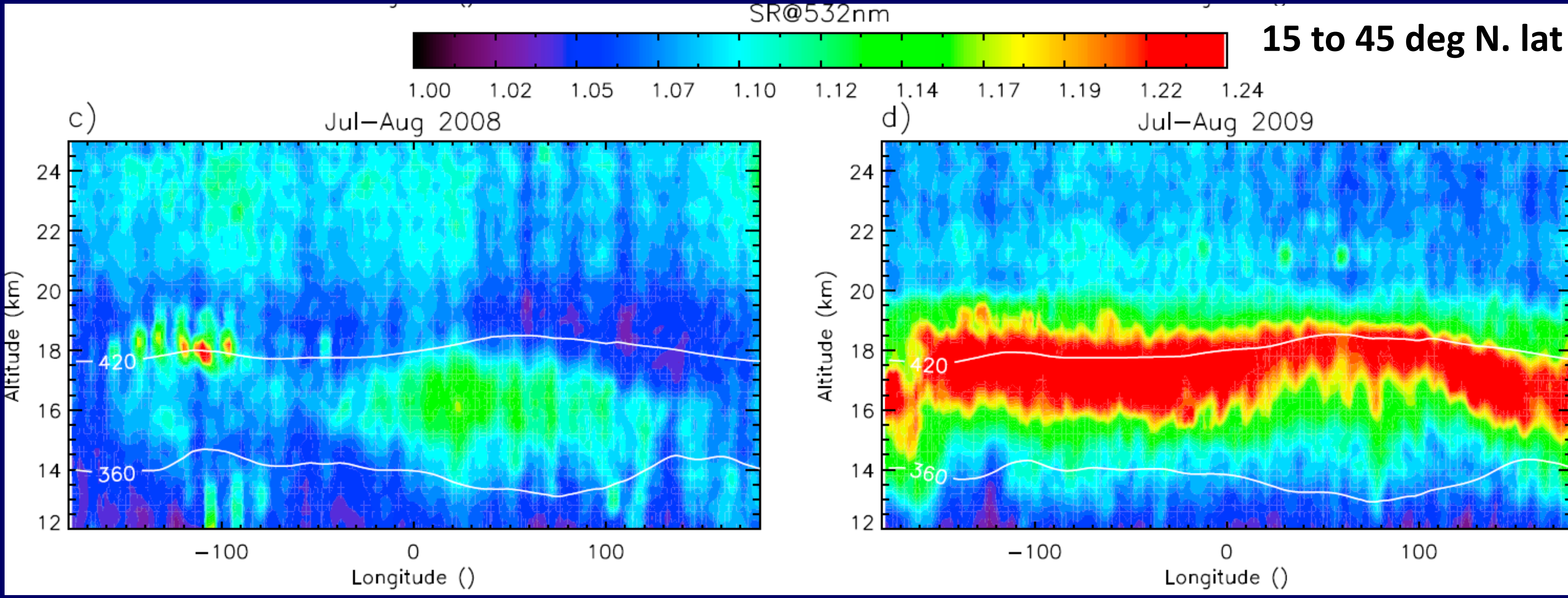
Anticyclone

Interpretation:

* highest particle mixing ratios ever seen are inside AMA
* peak altitude 5 – 10 K higher than elsewhere
* Influence of AMA visible in the particle data up to 420K
* How much is this NPF a ***source*** for the tropical peak and the LS aerosol?

Analysis: R. Weigel, IPA

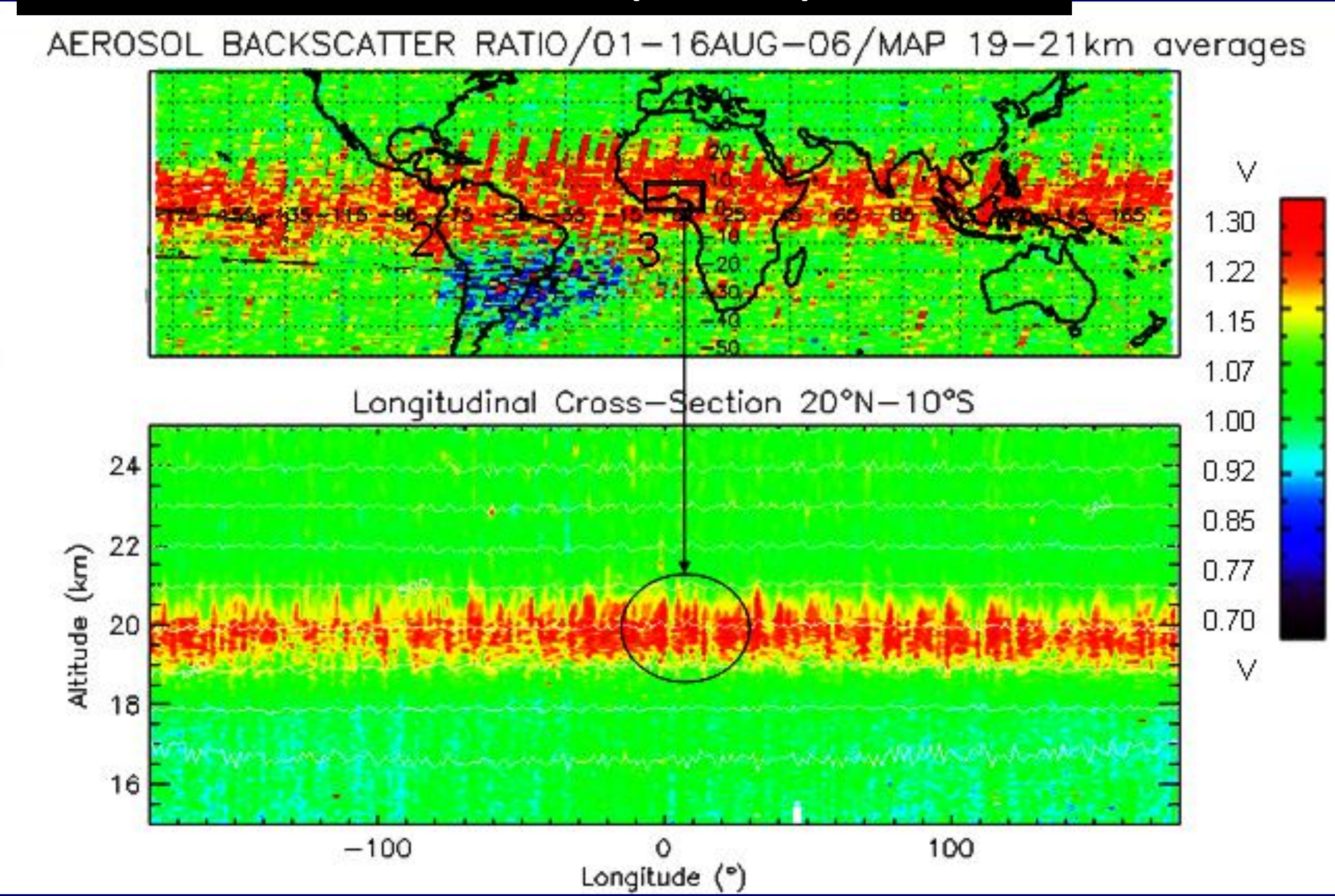
Asian Tropopause Aerosol Layer - ATAL



* ***Aerosol layer*** seen from CALIPSO lidar from Eastern Mediterranean to Western China as far South as Thailand
* ***Enhancement of background aerosol***, not much by volcanic eruptions
* At tropopause associated with ***monsoon circulation***.
* Particles most likely ***uplifted by convection*** from BL and thus heavily influenced by ***anthropogenic sources***

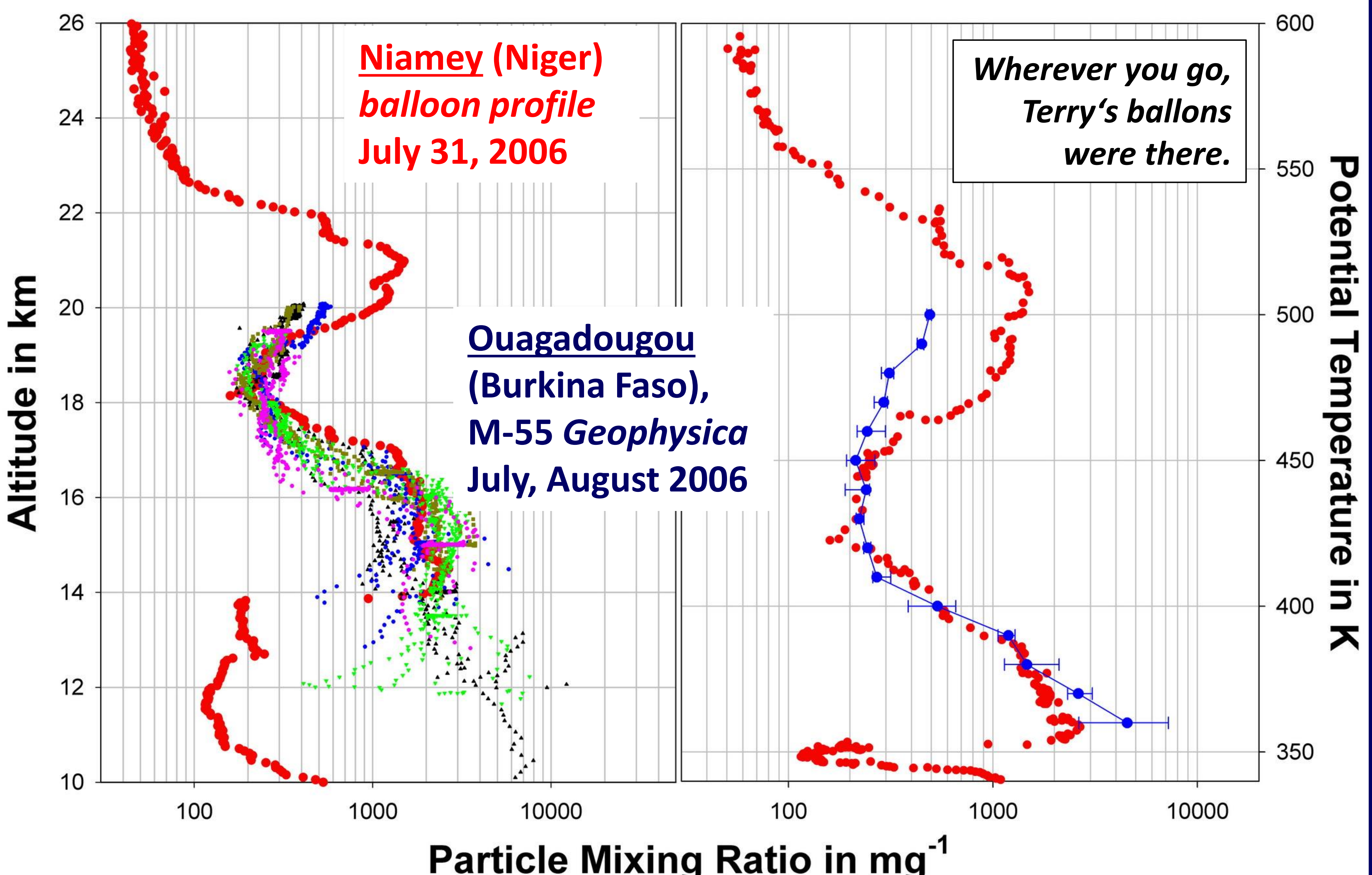
Vernier, J.-P., et al., GRL, 2011

La Soufriere Hills eruption plume….

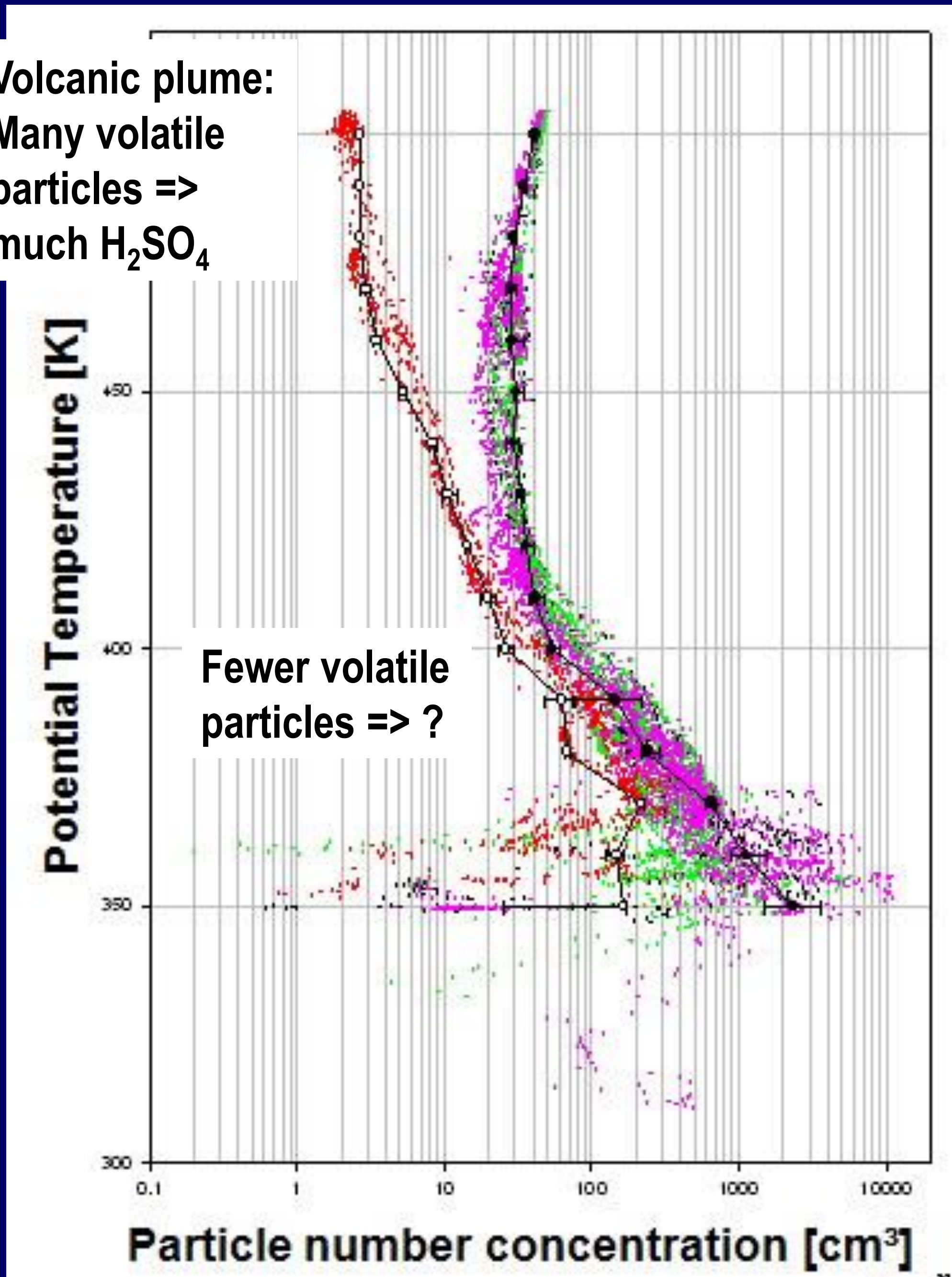


CALIOP LIDAR, image adapted from Vernier et al. (2008).

… appears in West-African profiles: Geophysica vs. balloon



Source: Borrmann et al., ACP, SCOUT&AMMA Special Issues, 2010

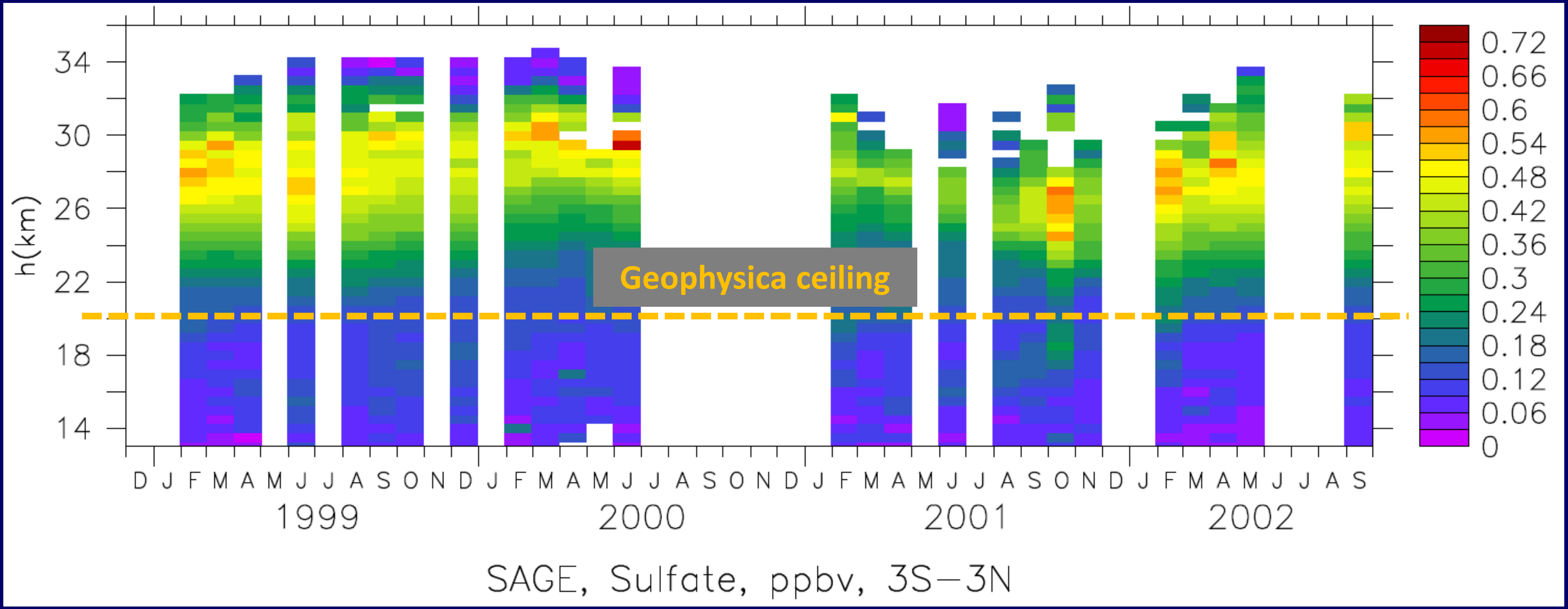


Increase > 420 K due to eruption of Soufrière Hills in May 2006.

Volatility data indicate H_2SO_4/H_2O droplets

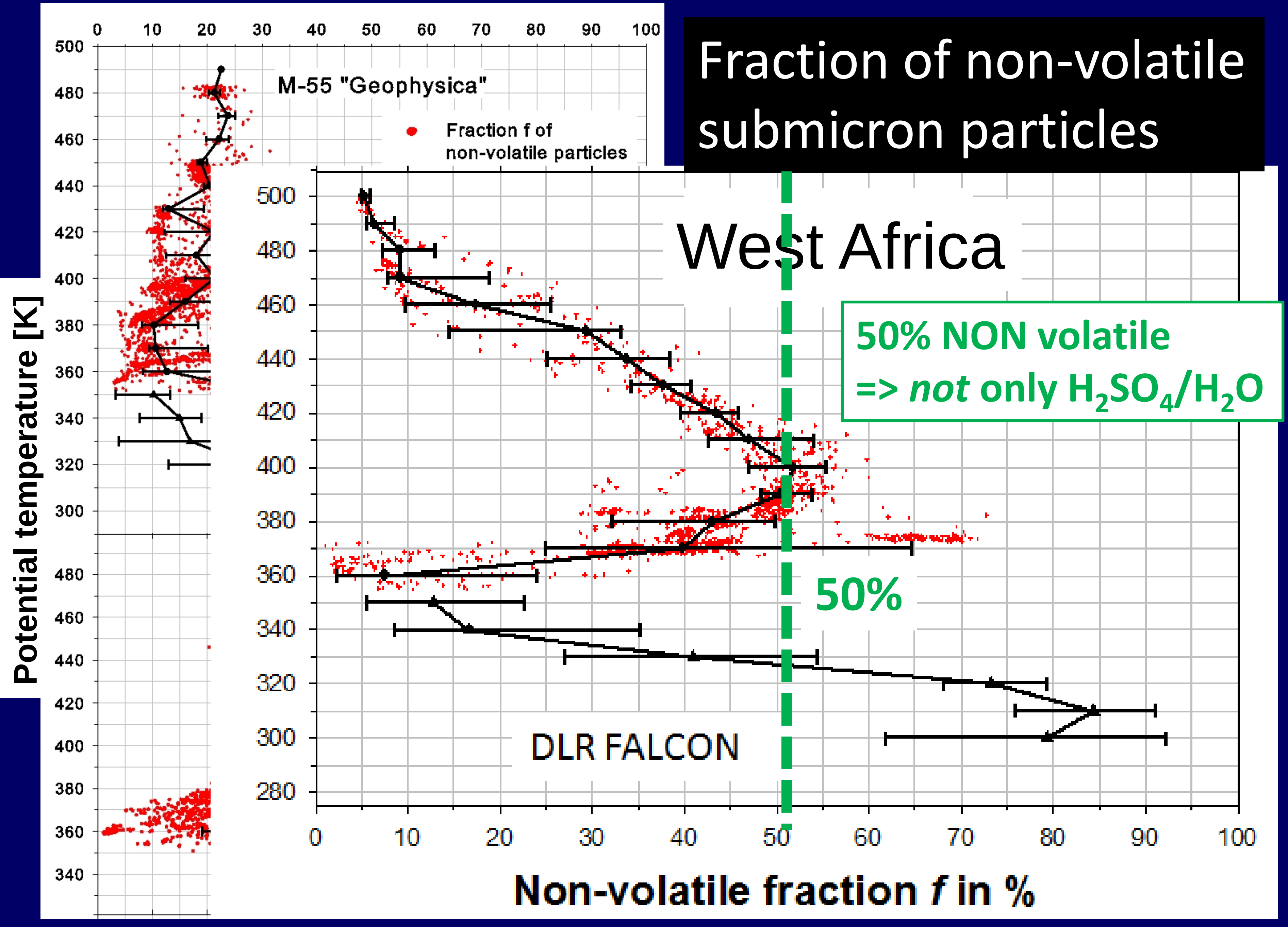
Source: Borrmann et al., ACP, SCOUT&AMMA Special Issues,2010

Stratospheric Junge sulfate aerosol layer in the tropics



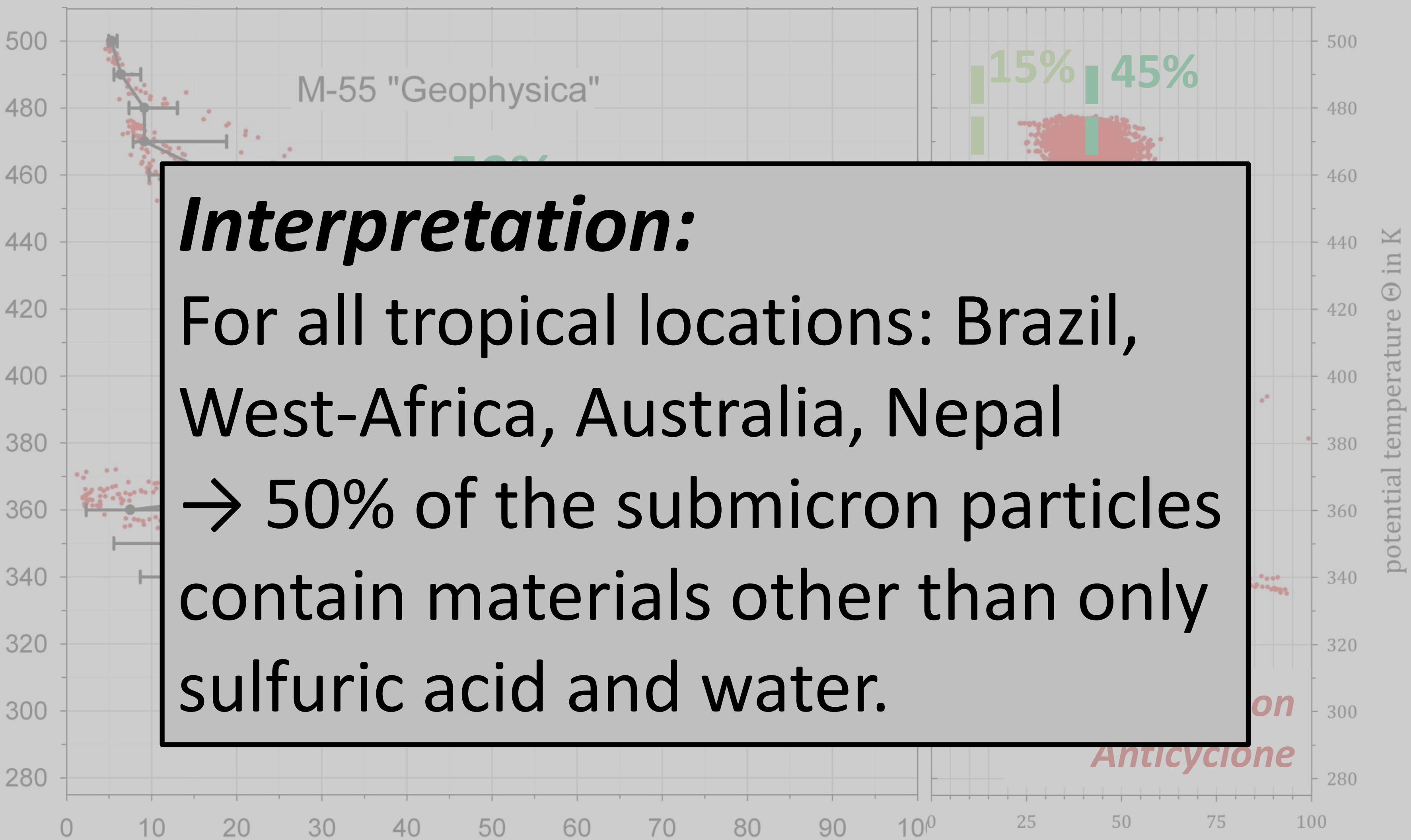
Source: Brühl et al., ACP, 2012

Fraction of non-volatile submicron particles



Borrmann et al., ACP, 2010

Non volatile fraction of submicron aerosol (at 250 °C)

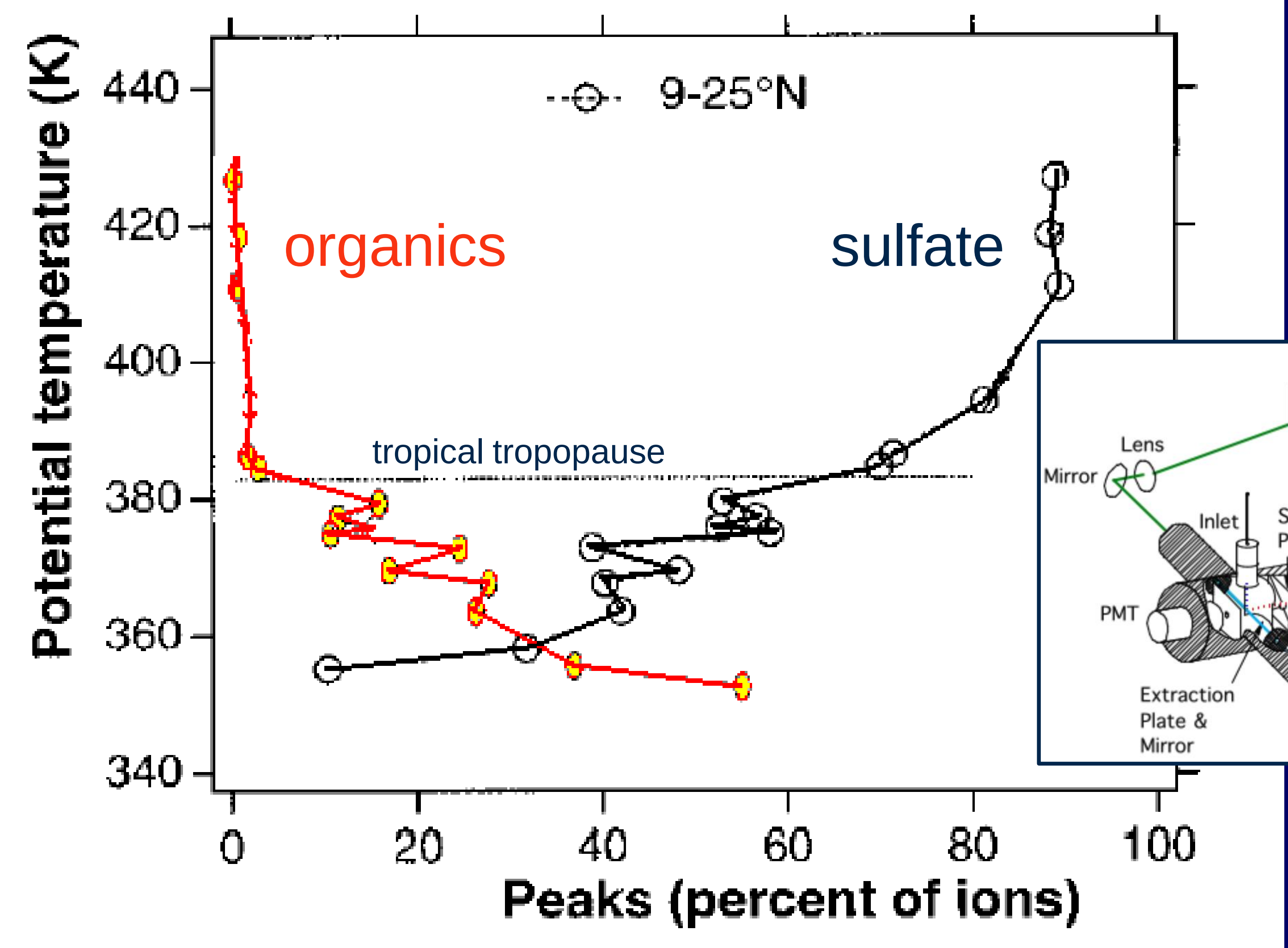


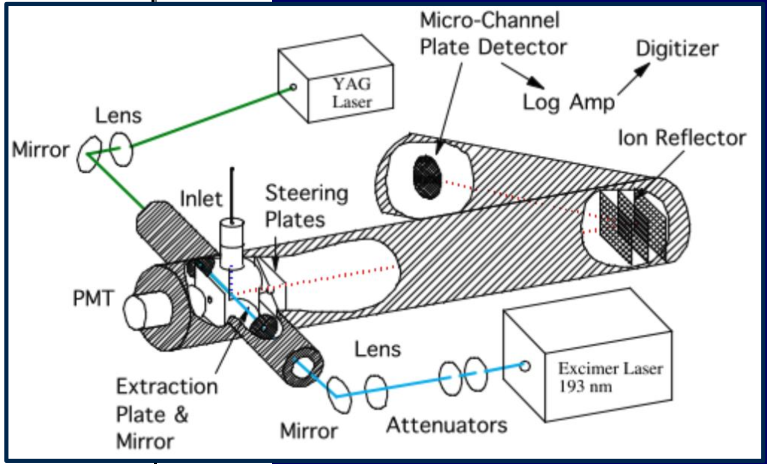
Interpretation:

For all tropical locations: Brazil, West-Africa, Australia, Nepal

→ 50% of the submicron particles contain materials other than only sulfuric acid and water.

Aerosols in the Tropical Transition Layer (TTL) from PALMS laser ablation mass spectrometer





Source: *Murphy et al., Science 1998*

Aerosols in the Tropical Transition Layer (TTL) from PALMS laser ablation mass spectrometer

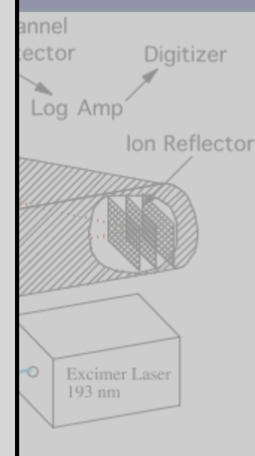
Findings:

* „almost **all particles** contain organics“
  (Murphy et al., 1998)

* „residues of **cirrus** and **subvisual cirrus** consist of
  neutralized sulfate and some organics.“
  NO dust, NO metals. (Froyd et al, 2009)

* ice cloud forming particles **(IN)** contain **metals**
  (Cziczo et al., 2009; Murphy et al. several papers)

* Results HALO aircraft 2014: Meteoric dust
  plays a role (see Ralf Weigel‘s talk tomorrow)



Source: *Murphy et al., Science 1998*

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

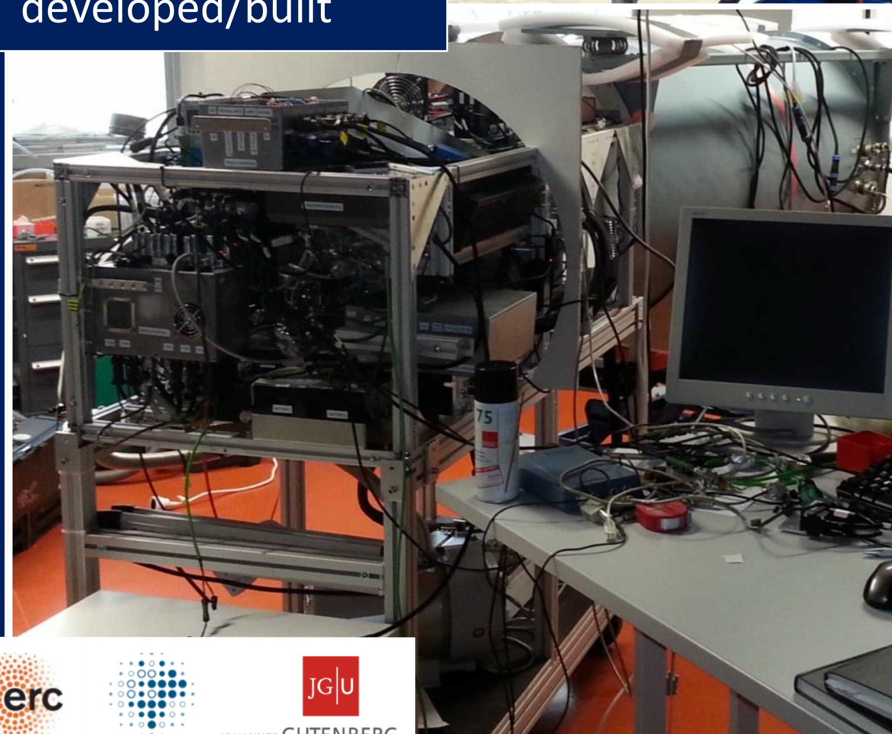
* ***sampling*** – via modified CARIBIC inlet, gold coated

* 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

Properties:
* pressurized barrel
* ≈ 360 kg, ≈ 1.5 kW
* fully automated → 15000 lines of code 12 LabView virtual instruments
* IRIDIUM remote link
* ≈ 2000 parts
* IPA/MPIC in-house developed/built

ERICA



European Research Council
MAX PLANCK INSTITUTE FOR CHEMISTRY
JOHANNES GUTENBERG UNIVERSITÄT MAINZ

Atmos. Chem. Phys., 16, 6641–6663, 2016
www.atmos-chem-phys.net/16/6641/2016/
doi:10.5194/acp-16-6641-2016

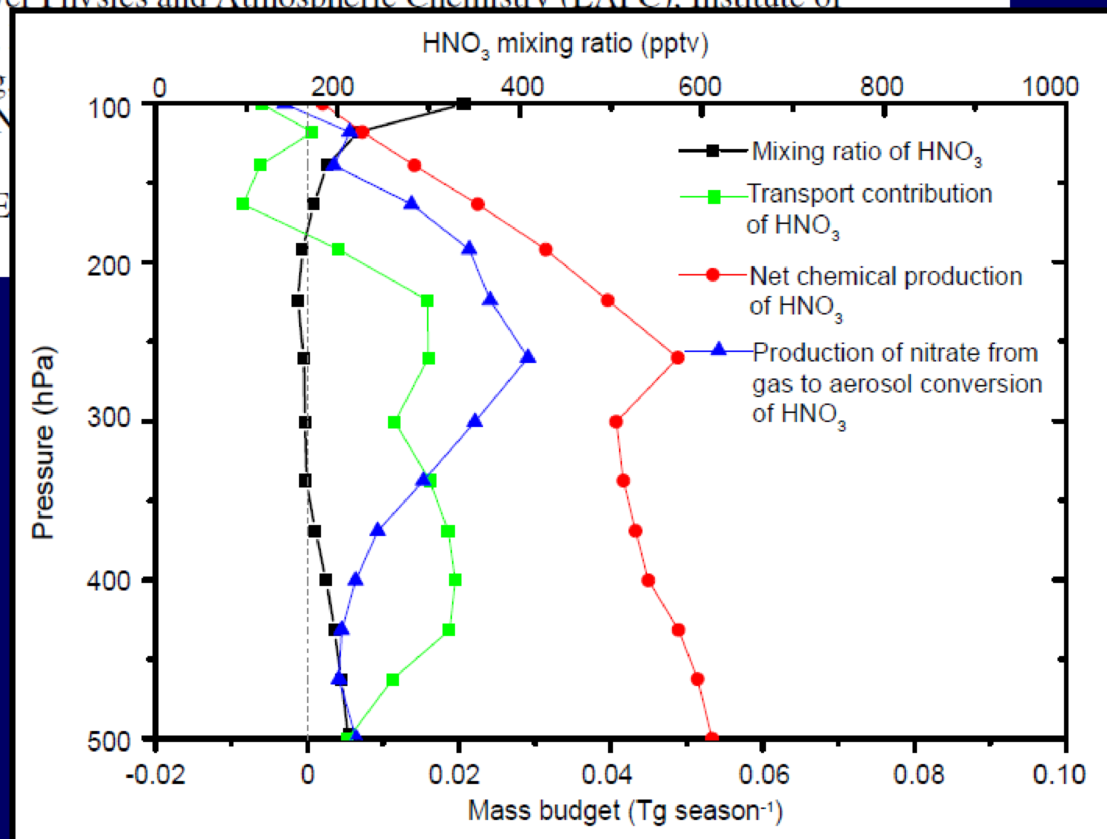
Atmospheric Chemistry and Physics
Open Access
EGU

Summertime nitrate aerosol in the upper troposphere and lower stratosphere over the Tibetan Plateau and the South Asian summer monsoon region

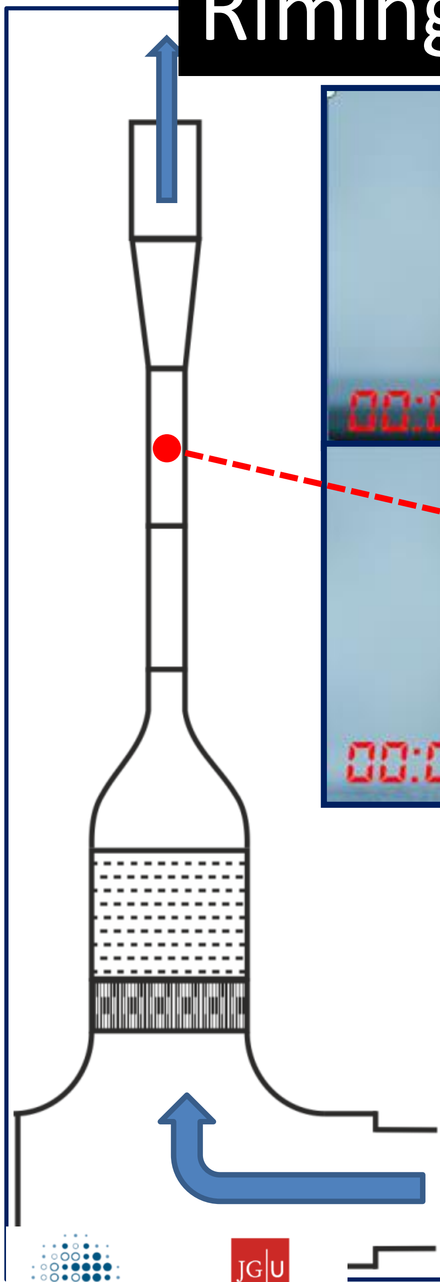
Yixuan Gu[1,2], Hong Liao[3], and Jianchun Bian[4]

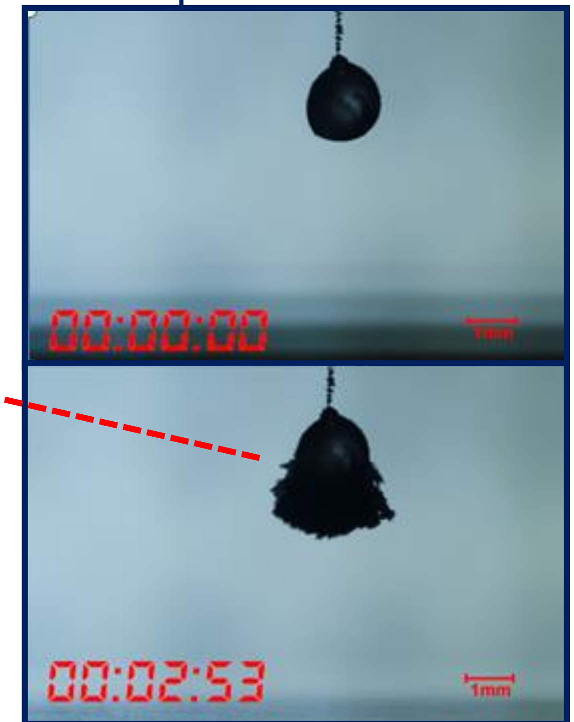
[1]State Key Laboratory of Atmospheric Boundary Layer Physics and Atmospheric Chemistry (LAPC), Institute of Atmospheric Physics, Chinese Academy of Sciences,
[2]University of Chinese Academy of Sciences, Beijing,
[3]School of Environmental Science and Engineering, N
Nanjing, China
[4]Key Laboratory of Middle Atmosphere and Global E
Chinese Academy of Sciences, Beijing, China

Application of GEOS-CHEM



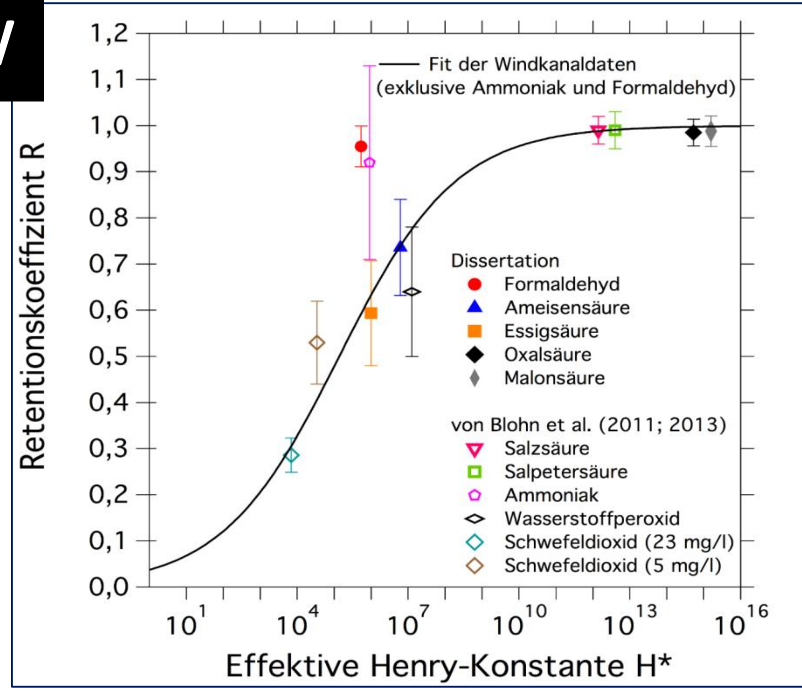
Riming and Cb outflow

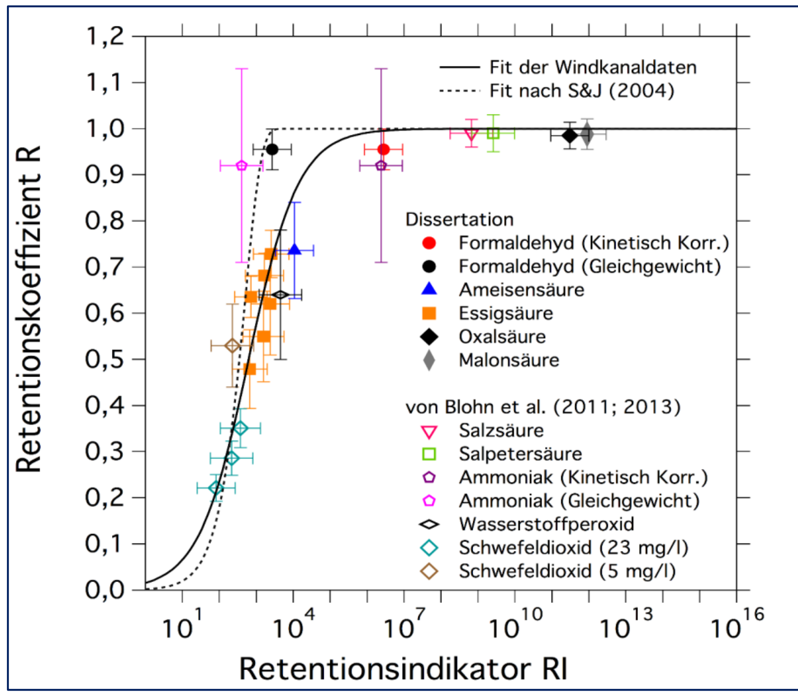




Source: A. Jost, Dissertation, ACP, 2017

- ***Supercooled riming droplets*** import species onto ***glaciated hydro-meteors***
- release into UTLS upon evaporation
- ***retention determines transport efficiency***





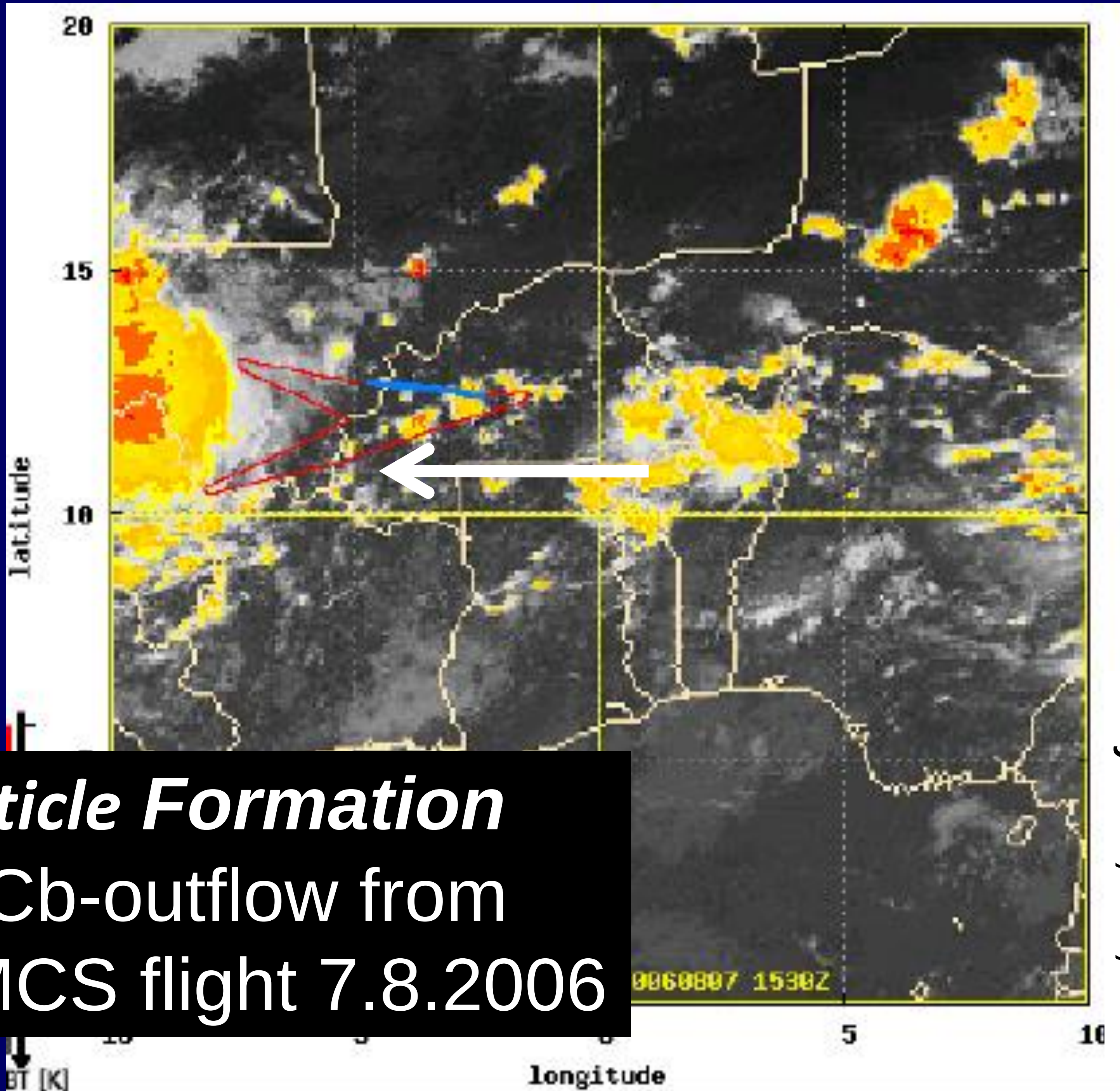
MAX PLANCK INSTITUTE FOR CHEMISTRY

JOHANNES GUTENBERG UNIVERSITÄT MAINZ

KTM08: 180810 09:15 UTC to 10:30 UTC – cloud particle number concentrations vs CO

Interpretation:

Cb outflow ice particles correlated with CO as f(Θ) → evaporating ice particles will release ”retained”
chemical species from rimed hydrometeors.

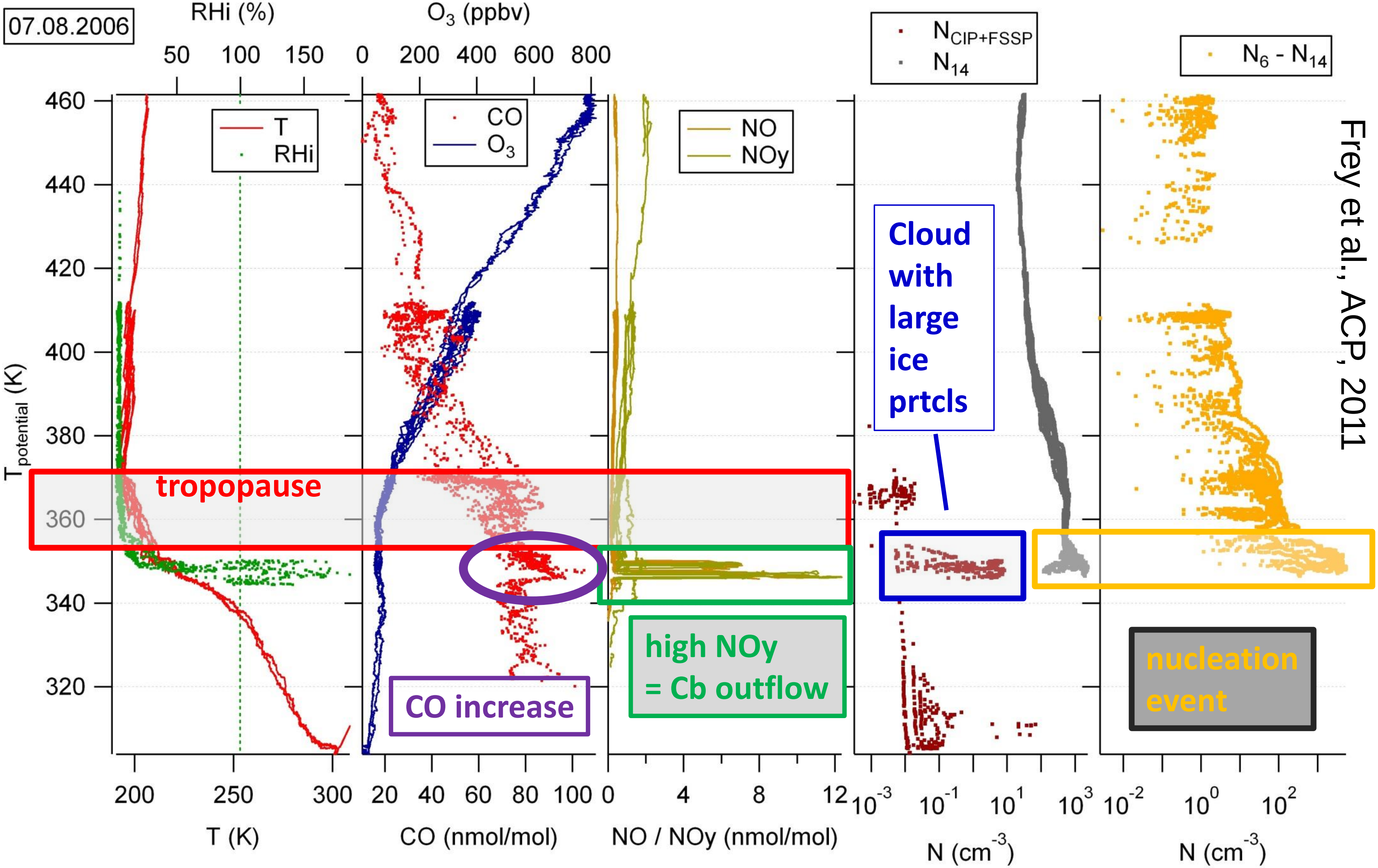


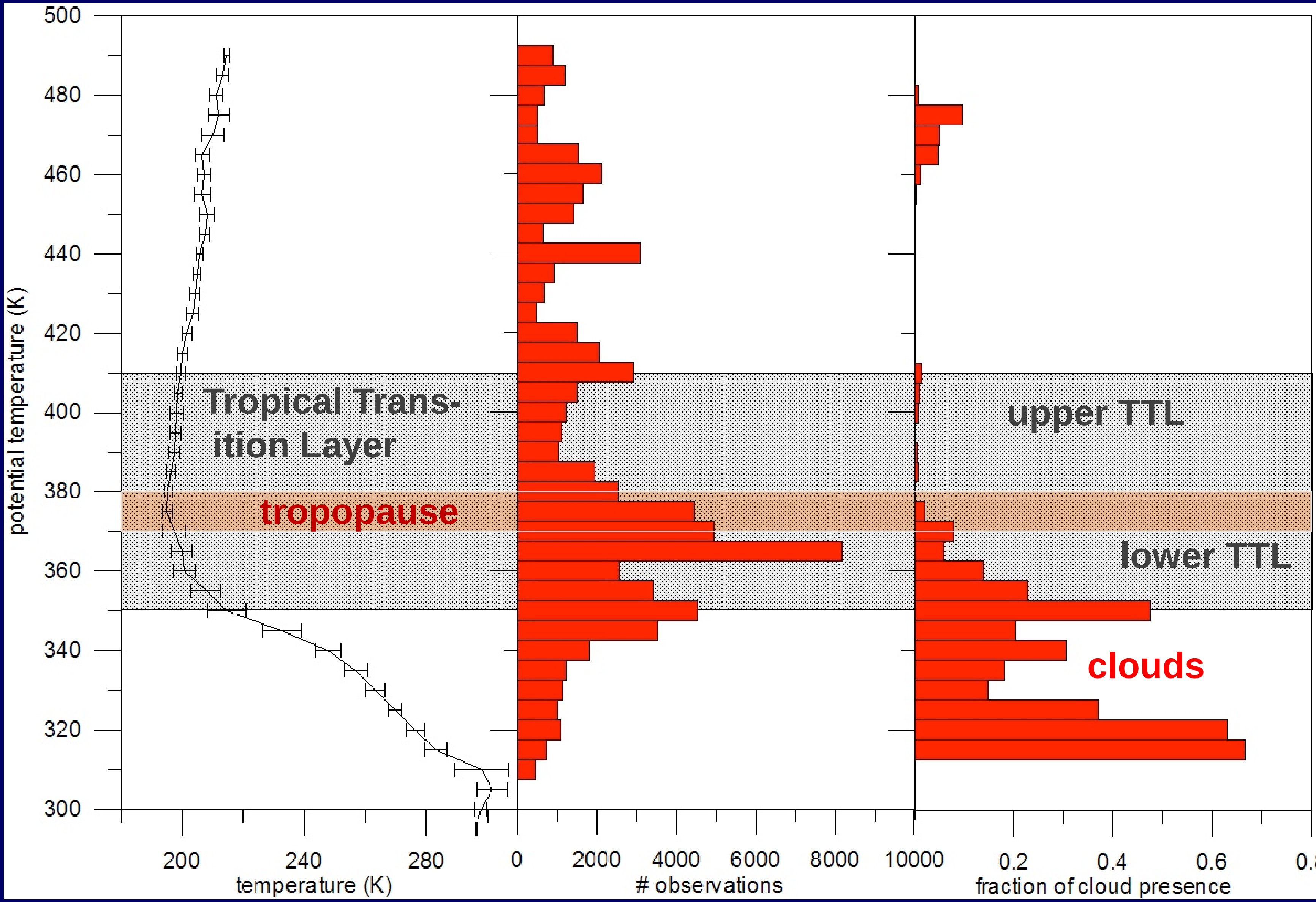
New Particle Formation

INSIDE Cb-outflow from AMMA MCS flight 7.8.2006

Frey et al., ACP, 2011

West African MCS outflow scenario





Law et al., ACP, 2010

Cloud presence inside the west African TTL

West African MCS outflow scenario

Interpretation:

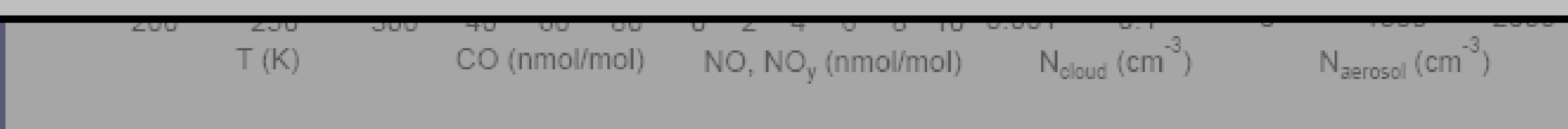
In-cloud New Particle Formation

* West-African MCS outflows make ultrafine particles at the TTL bottom, many and often.

* Probably binary solution droplets (H_2SO_4/H_2O)

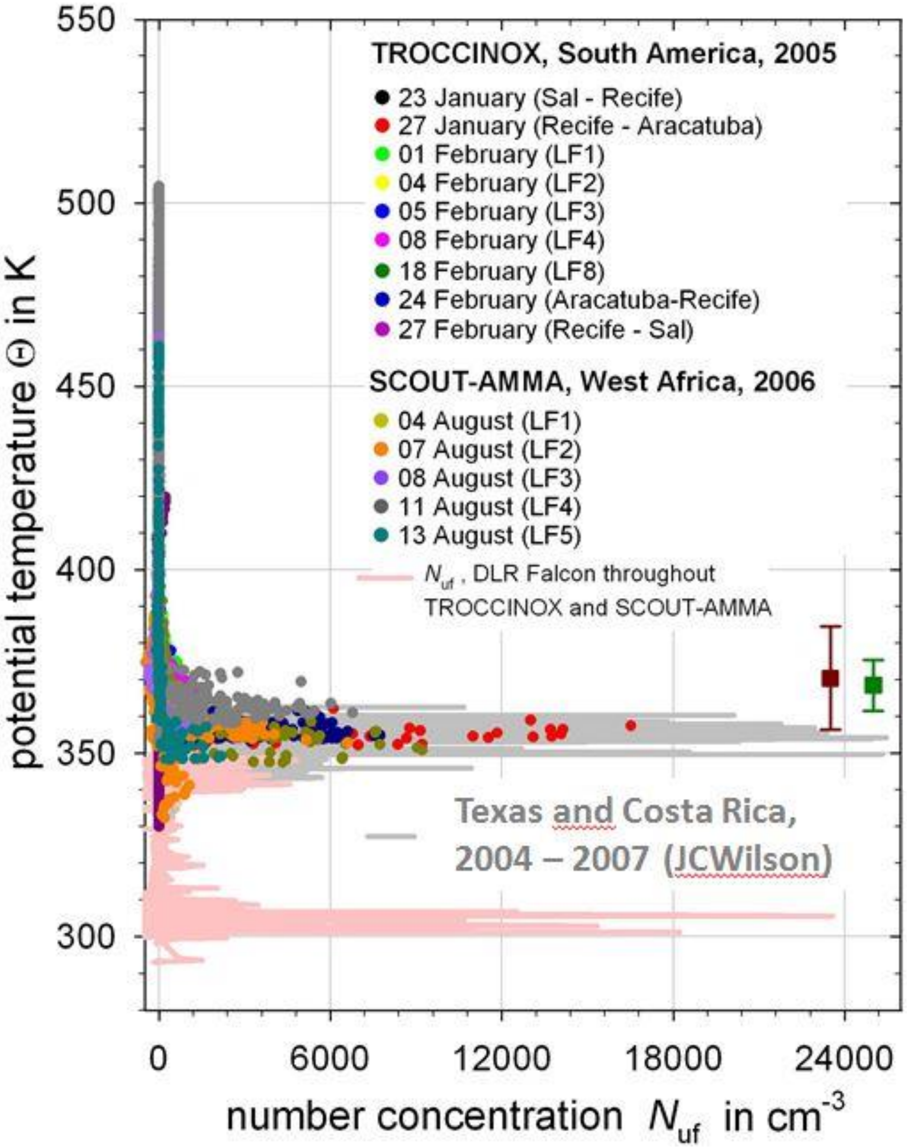
* ***Scale ≈ km to tens of km. Clear Air NPF cases at scales of hundreds of km.***

* If cloud particles >2 µm are more than 2 cm^{-3}, then NPF "quenched" by coagulation and competing condensation.



Frey et al., ACPD, 2011

New particle formation in the UTLS 2005-2006, 2017



Interpretation:

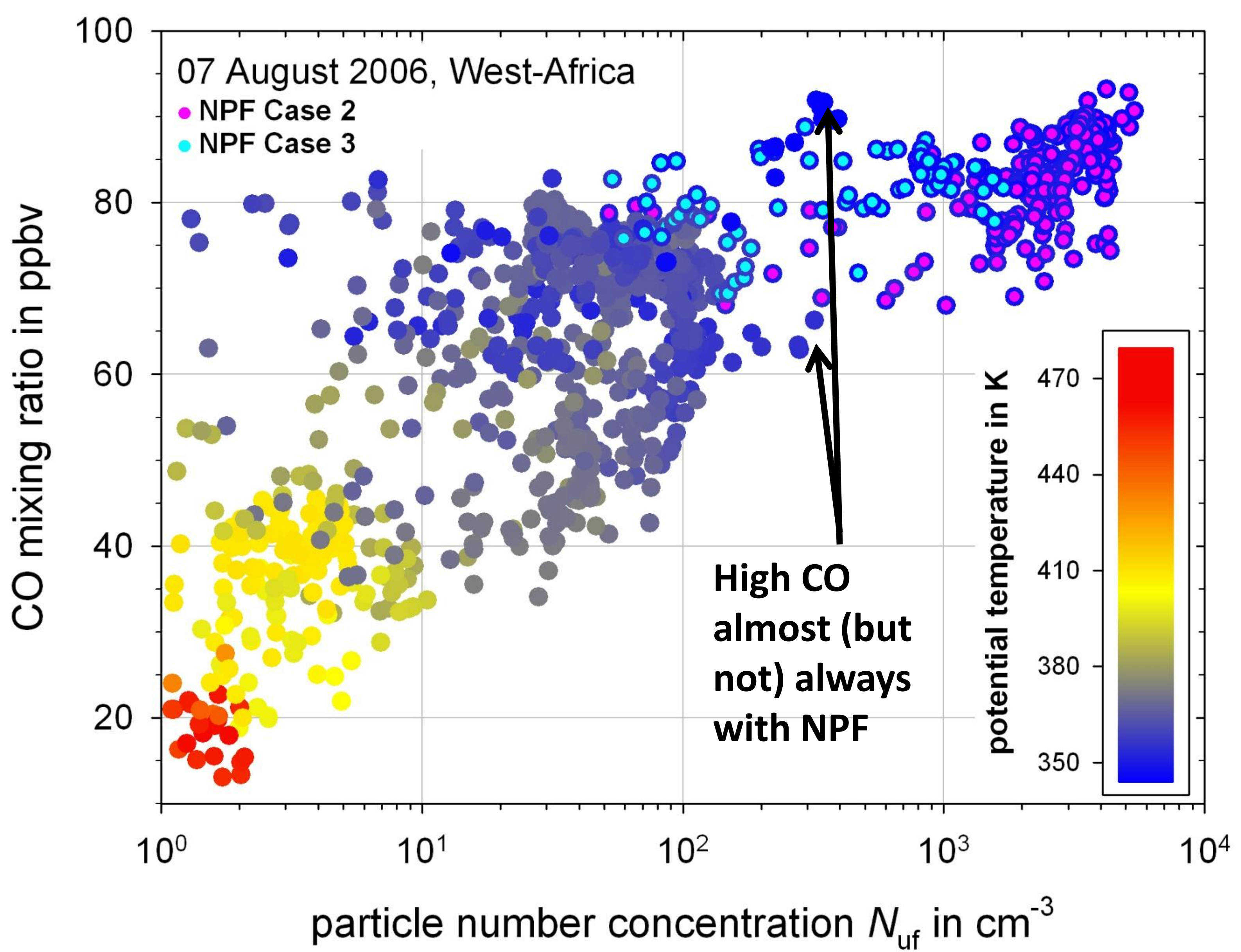
* New Particle Formation events encountered almost during each flight in tropical UTLS.

* Often associated with high CO levels. (Next slide)

* What role do organics play in NPF ?

Weigel et al., ACP, 2011

Role of organics?



Weigel et al., ACP, 2011

Suggested by many studies, e.g.: Ekman et al., 2008; Schmale et al., 2010

Summary

* Global submicron ***aerosol layer in the tropical belt*** between 340K and 400K, in Asian monsoon anticyclone (AMA) somewhat higher.
* ***highest particle mixing ratios*** ever seen are inside AMA
* Influence of AMA visible in the particle data ***up to 420K.***
* ***New Particle Formation events*** frequently occur in tropical UTLS, even inside clouds.
* Importance of ***clear air NPF vs in-cloud NPF*** unclear.
* For all tropical locations→ ***50% of the submicron particles do/do not fully evaporate*** at 250 °C.
* Within AMA there is a ***layer with dominating nitrate***, → detailed analyses just started

Summary

* ***Cb anvil outflows*** → CO shows BL contact for monsoons in West Africa and AMA. Air mass origin frequently in India, China, SE Asia

* ***ATAL still is elusive***; which particles exactly cause the faint CALIPSO signals?

* ***Which processes*** contribute to the AMA aerosol load ***to what extent?***
- clear air New Particle Formation
- in-cloud NPF
- convective transport of „already made“ aerosols
- role of organic and inorganic precursor gases

* Issue of influence of these ***aerosols on cloud formation***