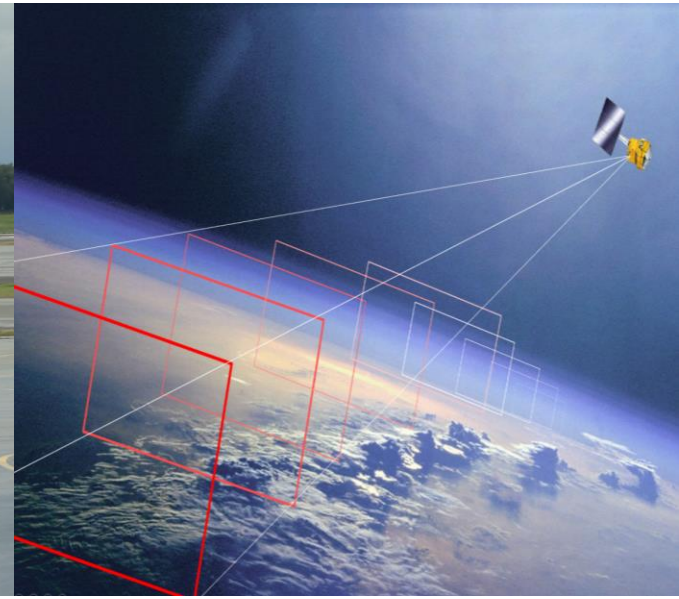
Karlsruhe Institute of Technology

Composition, origin, and fate of the Asian Tropopause Aerosol Layer – a view from aircraft and satellite by infrared remote sounding

Michael Höpfner, Institute of Meteorology and Climate research (IMK-ASF), KIT, Germany





The ATAL

CALIPSO detection of an Asian tropopause aerosol layer

J.-P. Vernier,[1] L. W. Thomason,[1] and J. Kar[2]

GRL, 2011, 10.1029/2010GL046614

Increase in upper tropospheric and lower stratospheric aerosol levels and its potential connection with Asian pollution

J.-P. Vernier[1,2], T. D. Fairlie[2], M. Natarajan[2], F. G. Wienhold[3], J. Bian[4], B. G. Martinsson[5], S. Crumeyrolle[6], L. W. Thomason[2], and K. M. Bedka[2]

JGR, 2014, 10.1002/2014JD022372

- **What is the composition and phase of ATAL particles?**
- **What is their source?**
- ***Can ATAL particles influence nucleation of cirrus clouds and, thus, have an indirect radiative effect possibly larger than the direct one?***
- ***What is their fate (e.g. entering the lower stratosphere, influencing ozone)?***

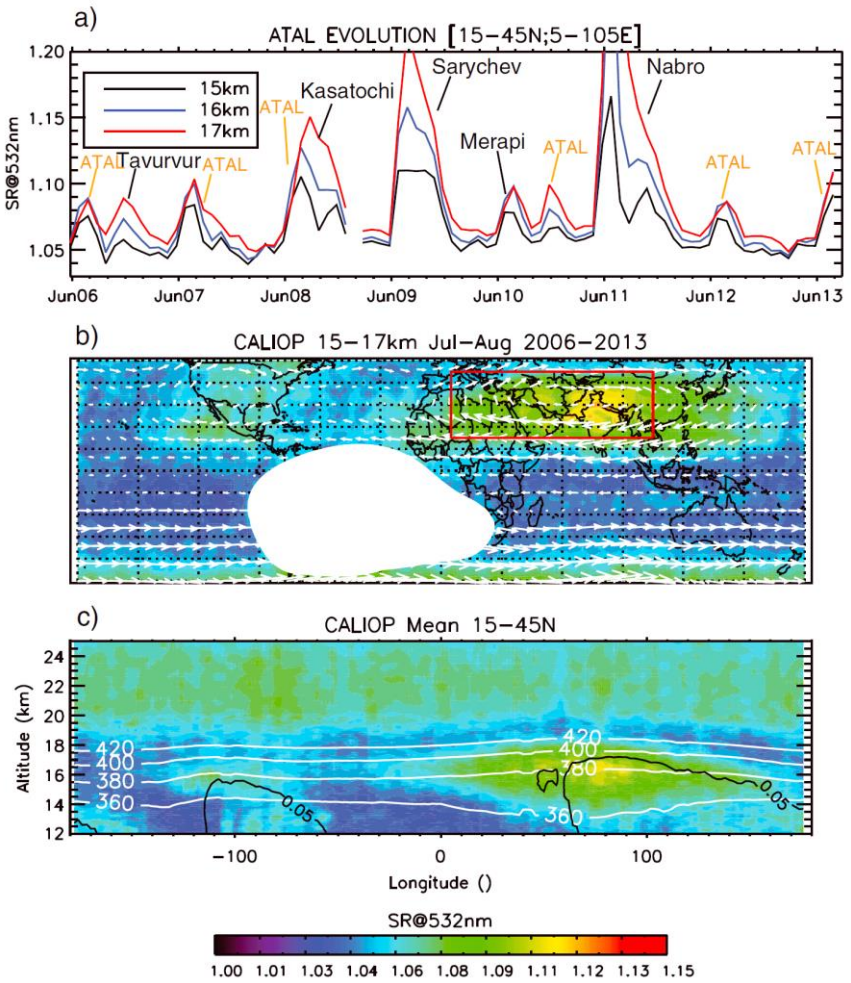
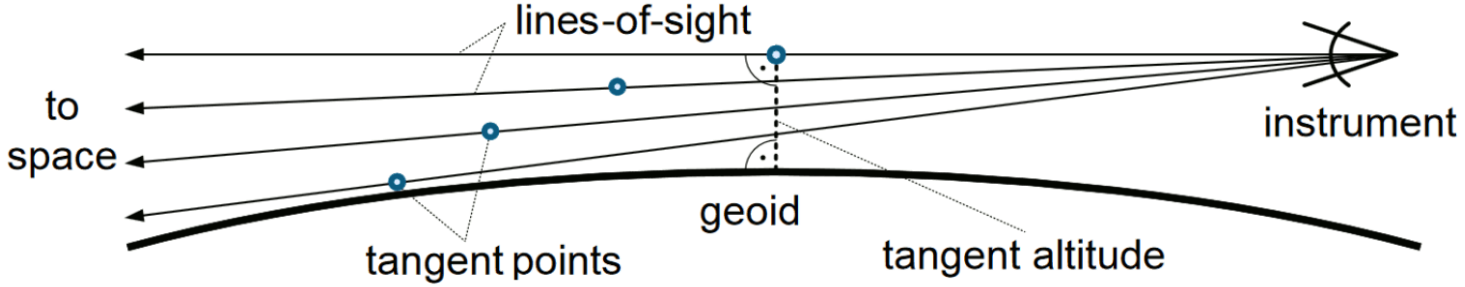
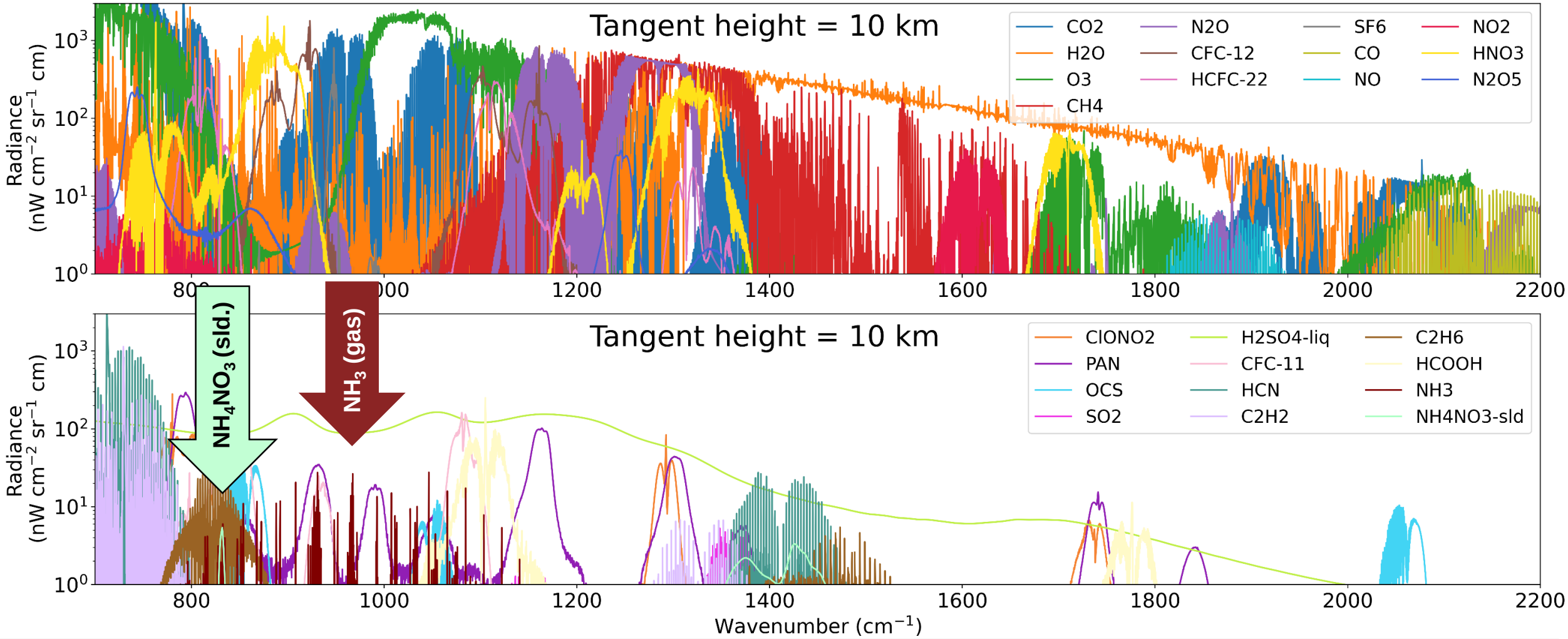


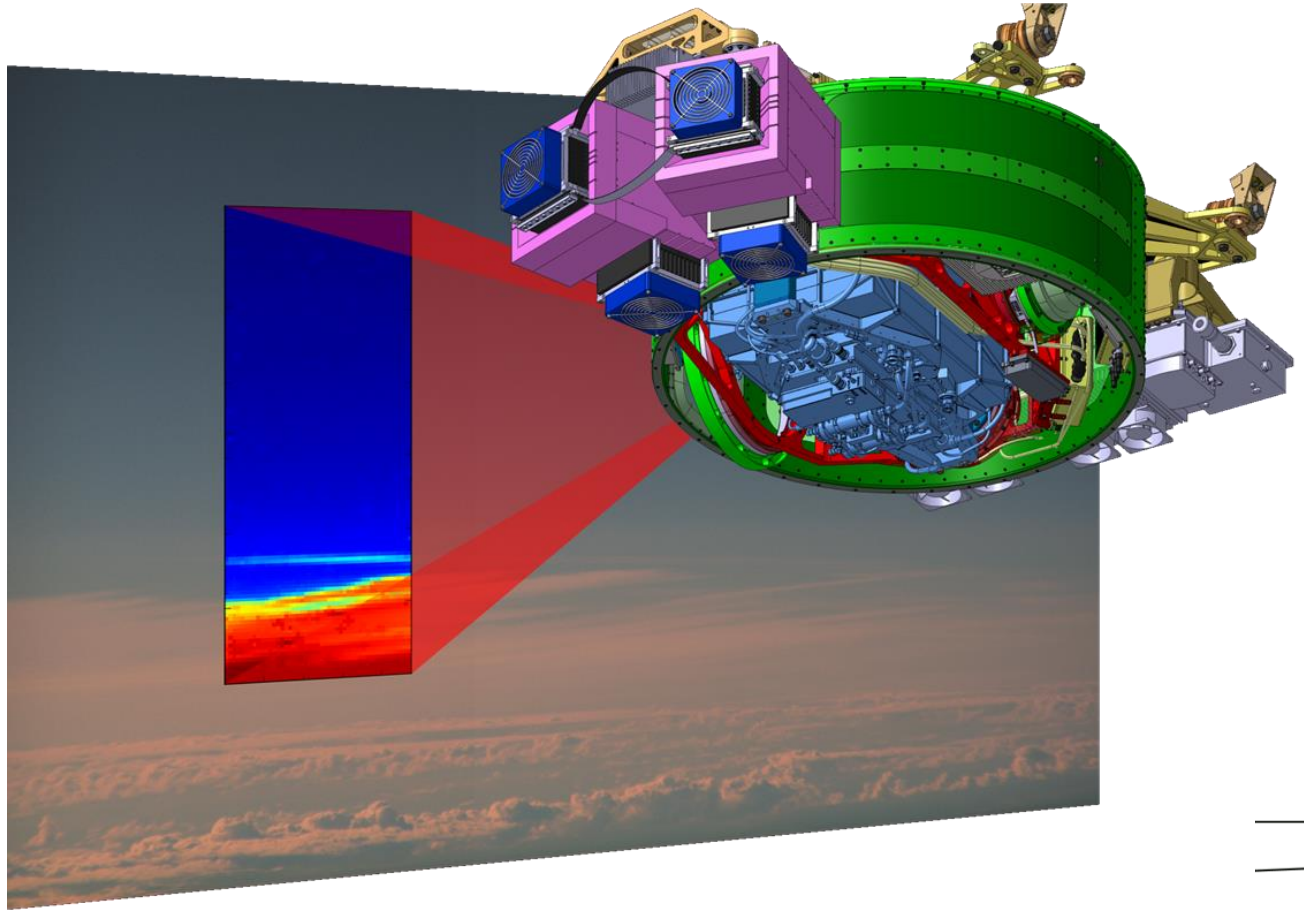
Fig. 2, Vernier et al., JGR, 2014, 10.1002/2014JD022372

The infrared limb spectroscopic measurement concept



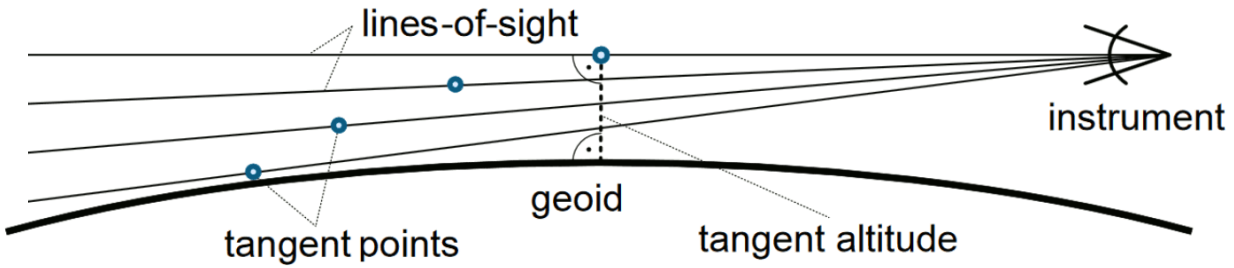


The Gimballed Limb Observer for Radiance Imaging of the Atmosphere (GLORIA)



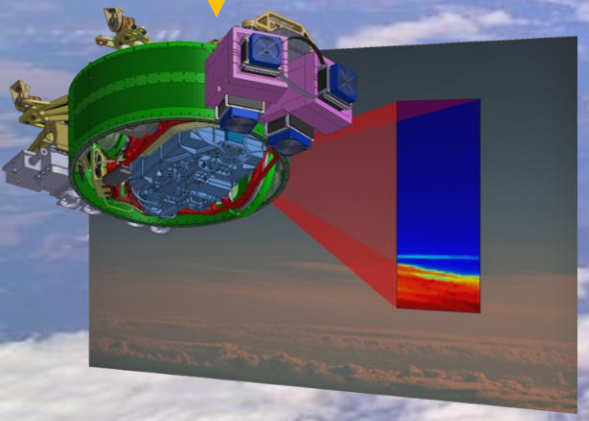
Riese et al. (AMT 2014)
Friedl-Vallon et al. (AMT 2014)

- Cooled Imaging Fourier-Transform Spectrometer (iFTS)
- Instrument consists of
  - Spectrometer
  - Gimballed frame
  - Two external blackbodies
- Unique iFTS for atmospheric limb-sounding



adapted from Ungermann et al. (ACP 2013)

GLORIA aircraft observations (limb-sounding)



3 km

3 km

3 km

Observer

Limb-*images* = data cubes

Spectra

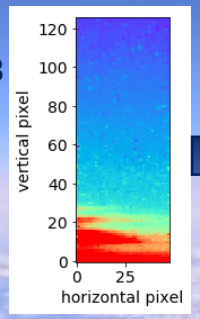
Parameter profiles

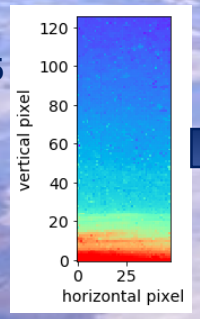
Cross-sections along flight track

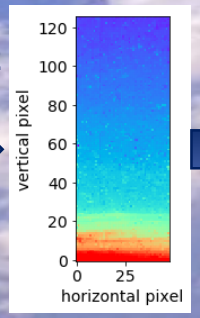
09:00:28

09:00:15

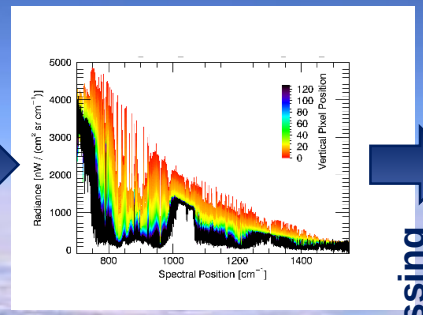
09:00:02

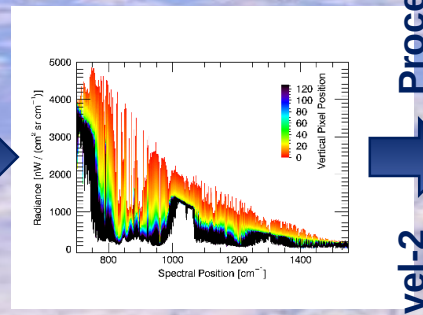


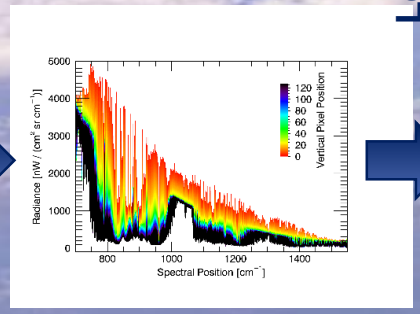




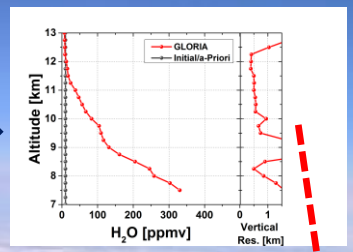
Level-1 Processing

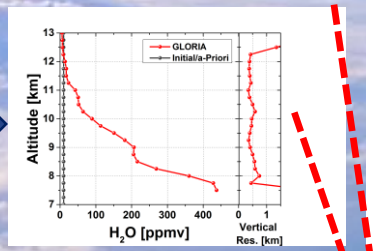


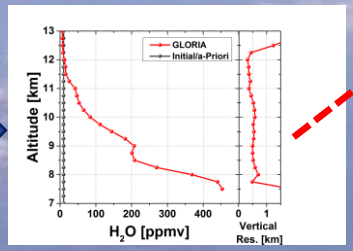


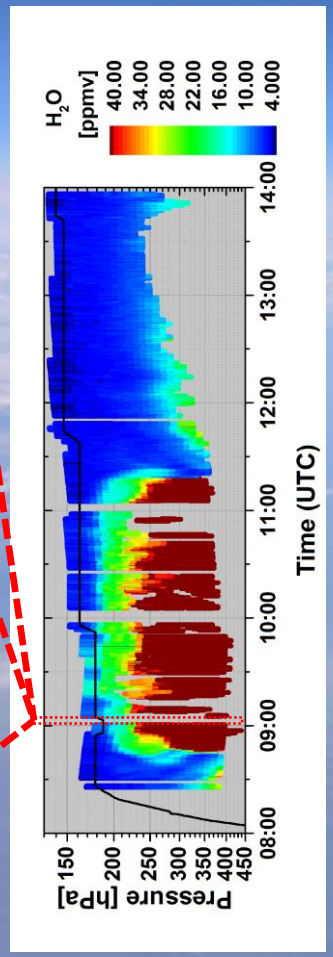


Level-2 Processing









1 image = 128x48 pixels
1 interferogram per pixel

➔128x48 individual spectra binning ➔ **128 spectra**

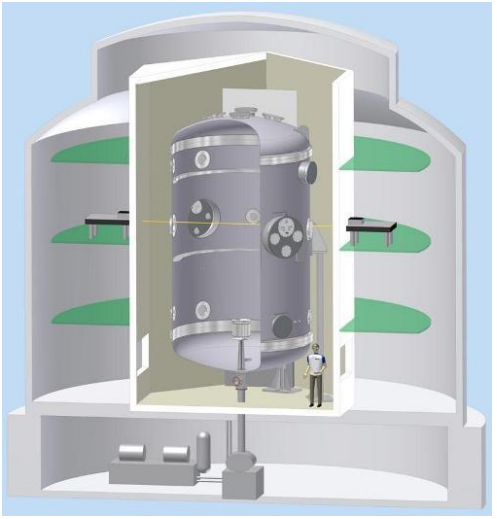
➔**1 profile of T, H_2O, O_3, HNO_3, … per image**

➔**cross-sections of T, H_2O, O_3, HNO_3, …**

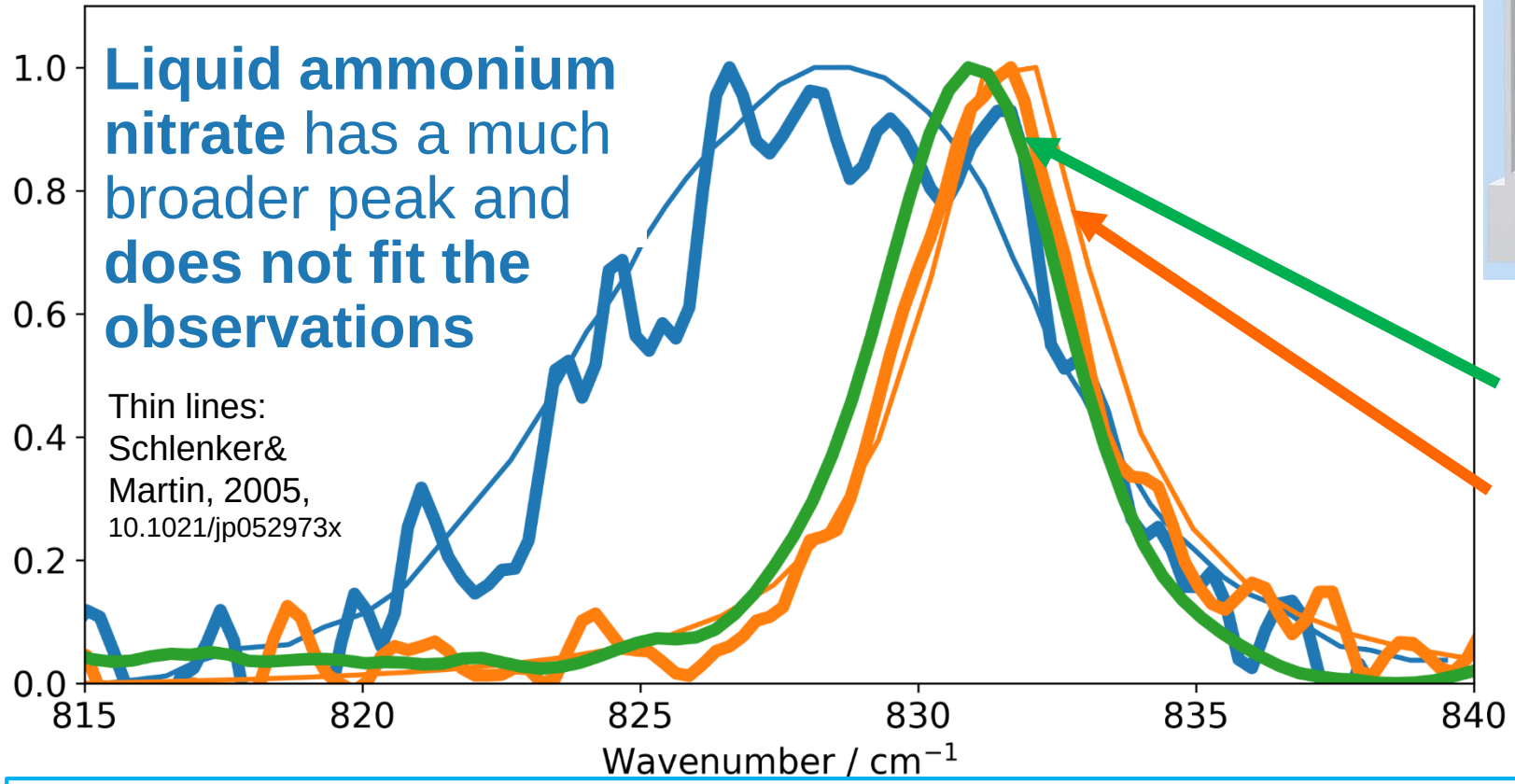
Slide courtesy: W. Woiwode, KIT

Limb-sounding instruments on different platforms analysing the ATAL

Laboratory infrared spectra of the $\nu_2(NO_3^-)$ band of NH_4NO_3 particles compared to the observations



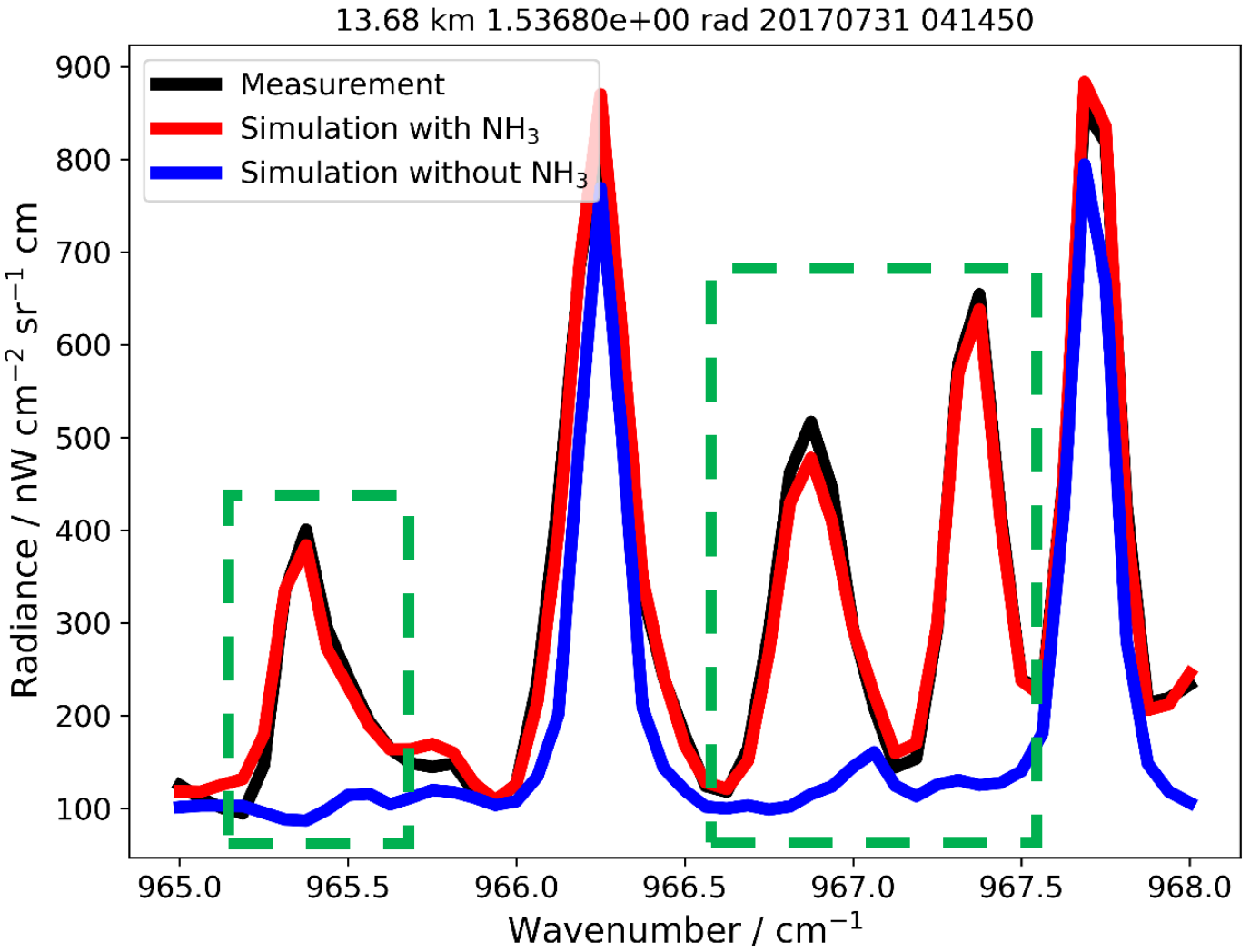
Karlsruhe Institute of Technology



Observed atmospheric peak fits to the signature of **solid ammonium nitrate**

- **A spectral band at 831 cm^{-1} was detected in infrared spectra of satellite (CRISTA, MIPAS) and airborne (GLORIA) instruments only inside the monsoon upper troposphere**
- **Laboratory observations show that the infrared signature is due to solid ammonium nitrate particles**
- **Solid AN particles only form when impurities of ammonium sulfate are present**
- **This allows to derive ammonium nitrate mass concentration profiles from the infrared limb observations**

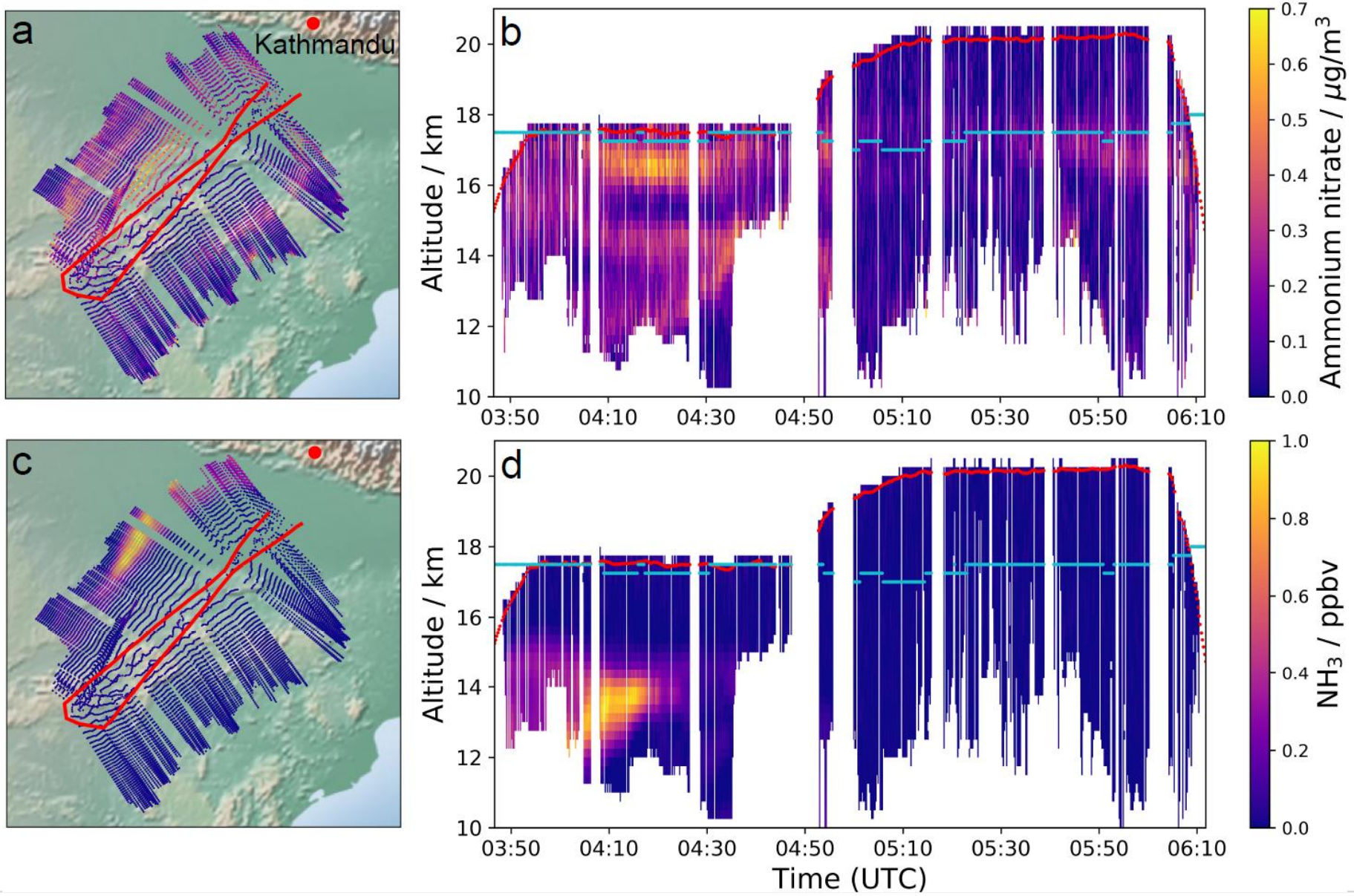
Spectral detection of gas-phase NH_3: a prerequisite for NH_4NO_3 secondary aerosol formation



GLORIA/Geophysica
July 2017

Höpfner et al., 2019

StratoClim flight 31 Jul 2017

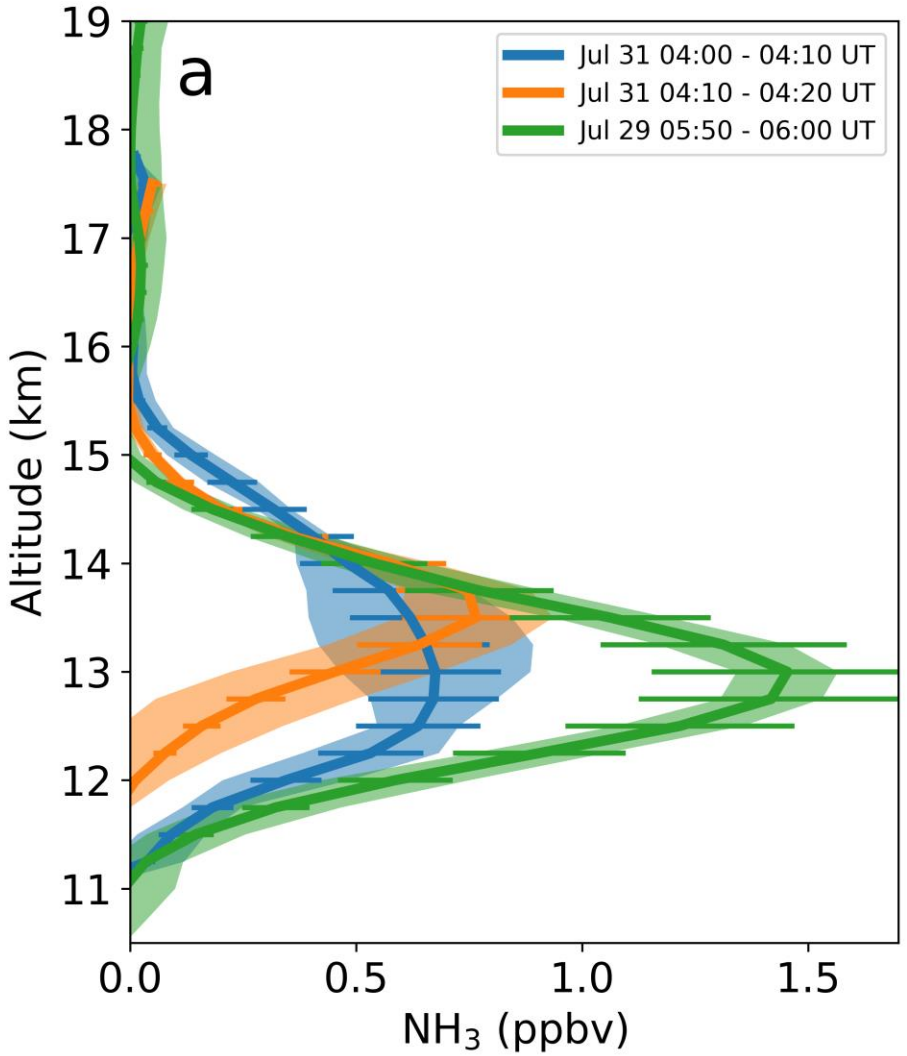


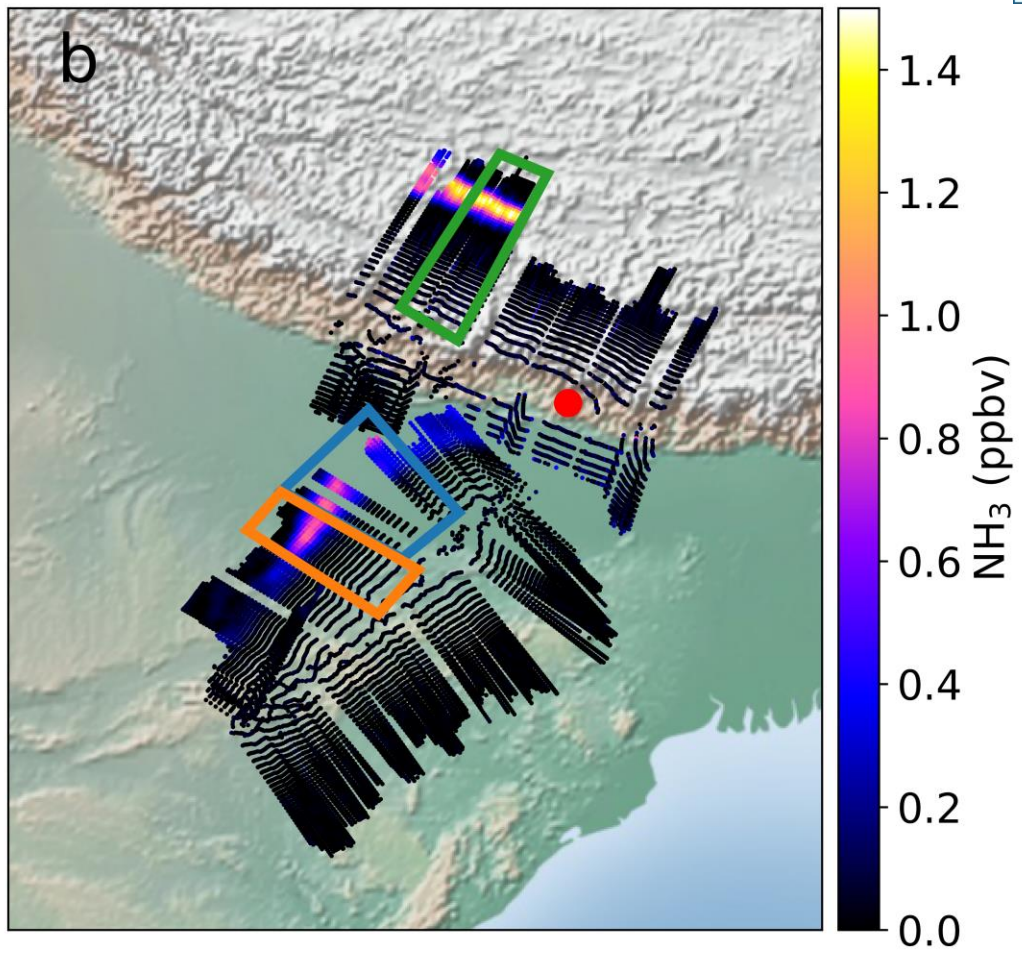
Solid NH_4NO_3 aerosol mass density

NH_3 trace gas mixing ratio

Höpfner et al., 2019

NH_3-plume on 29 and 31 July 2017

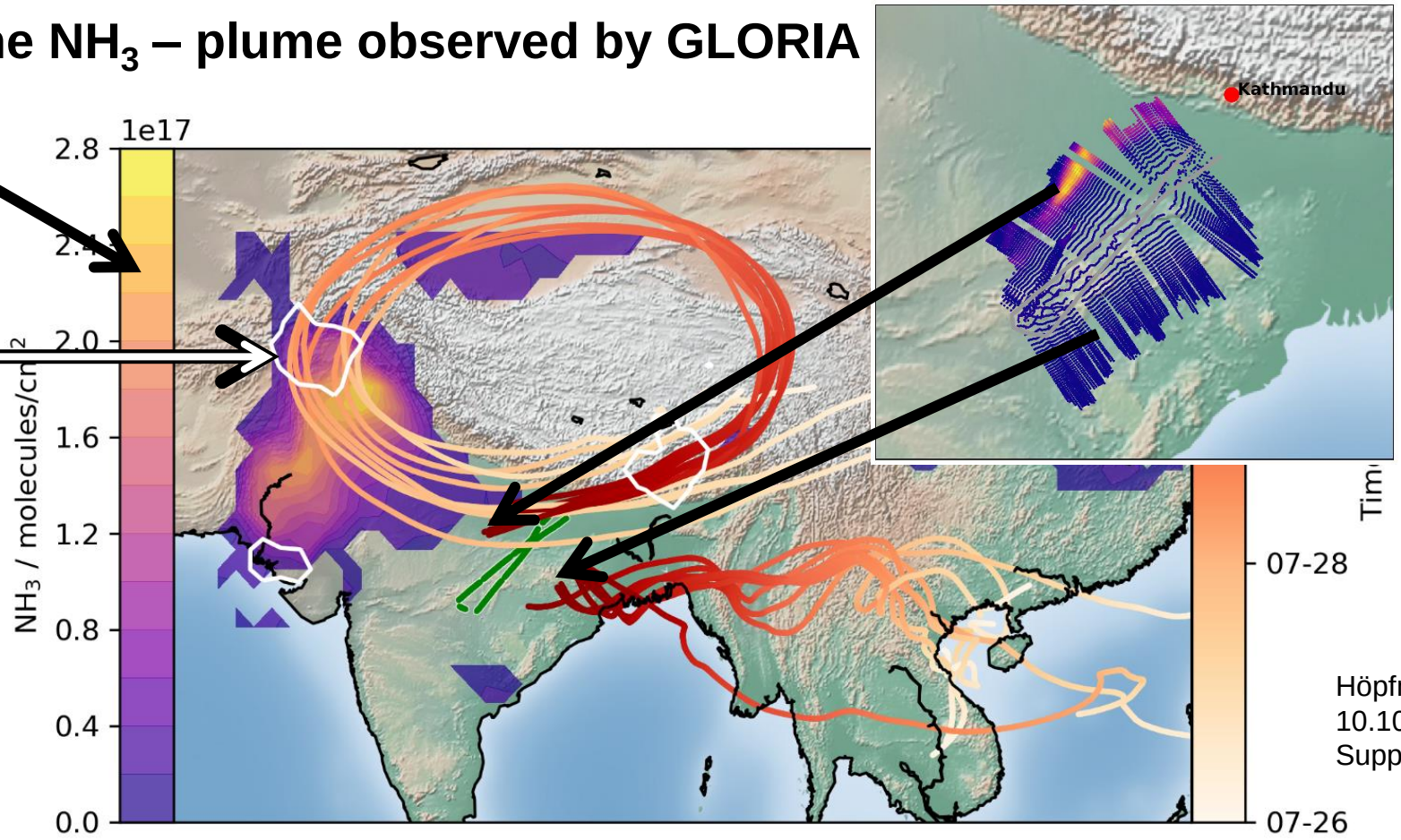




Origin of the NH_3 – plume observed by GLORIA

IASI NH_3 (low-mid troposphere)

Density of convective events along backward trajectories



http://iasi.aeris-data.fr/NH3
The data produced at Université Libre de Bruxelles (ULB) by Simon Whitburn and Martin Van Damme
Van Damme et al., AMT, 2017

Höpfner et al., Nat. Geosci, 2019, 10.1038/s41561-019-0385-8, Suppl. Fig. 10

- **High NH_3 at 14 km altitude traced back to regions with strong convection and with enhanced concentrations of NH_3 in the lower troposphere as detected by the IASI infrared nadir sounder**

Höpfner et al., 2019

Possible importance of ammonia and solid ammonium nitrate in the UTLS

- AIDA-chamber: Solid AN particles are **efficient ice nucleating particles** (Wagner et al., JGR, 2020)

- CERN CLOUD chamber (Wang et al, Nature, 2022):
  - **nitric acid, sulfuric acid and ammonia** form particles **synergistically faster** than from any two of the three components.
  - "..model simulations: ammonia efficiently convected aloft during the Asian monsoon, drives **rapid, multi-acid HNO_3–H_2SO_4–NH_3 nucleation** in the upper troposphere and **produces ice nucleating particles that spread across the mid-latitude Northern Hemisphere**."

- Modelling (Yu et al., GRL, 2022):
  - condensed **nitric acid particles** account for **~20% of the annual mean aerosol mass at the tropical tropopause**, and over 95% in the UTLS at the South Pole in June-July-August.
  - cold ambient conditions in the UTLS of the tropics, ASM and polar regions thermodynamically favor the **condensation of ammonia and nitric acid**
  - widely distributed **nitrate aerosol** in the global UTLS may be **overlooked by climate models**

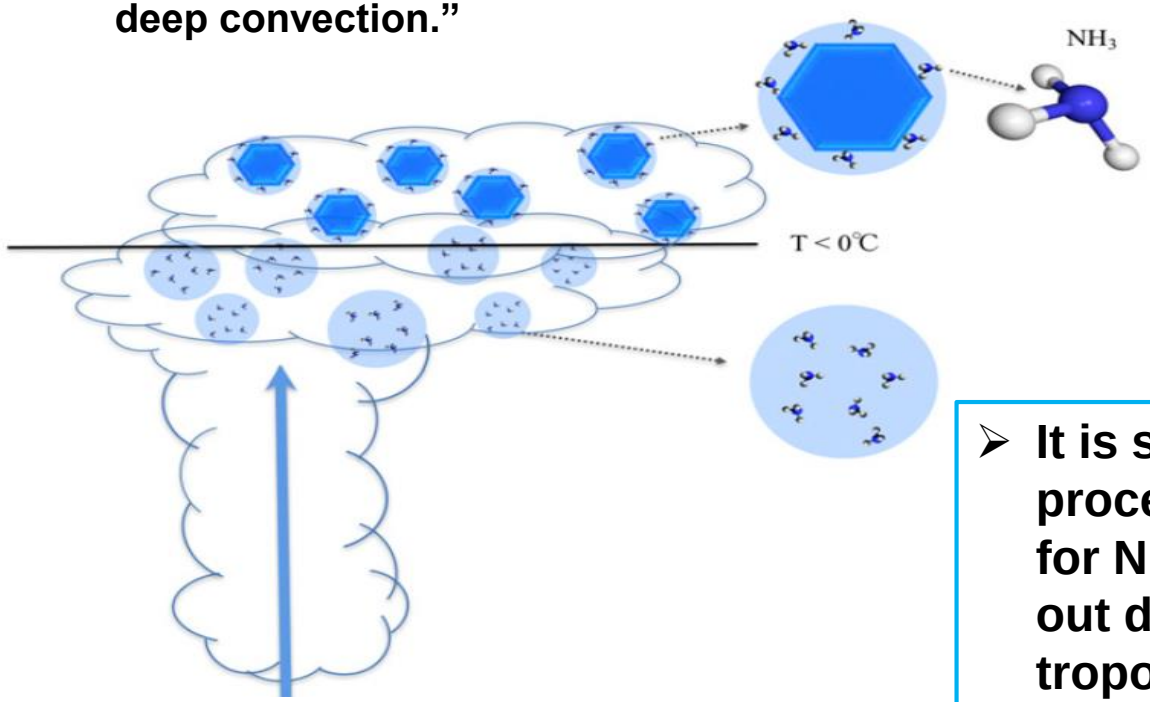
Why is NH_3 not washed out during convection?

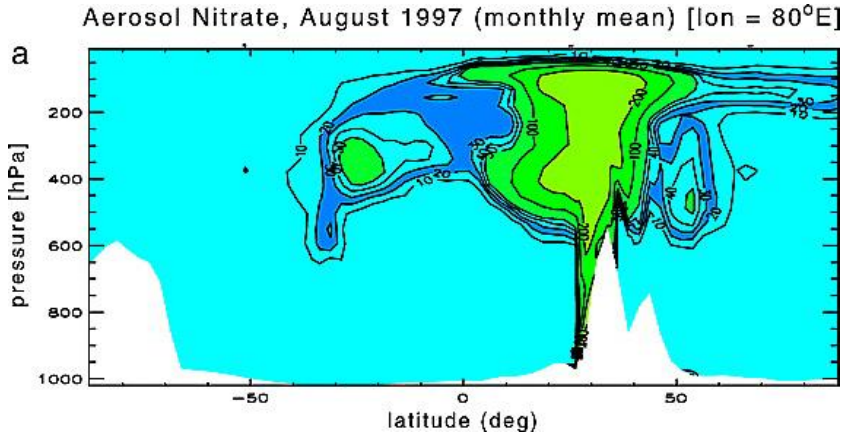
A molecular perspective for global modeling of upper atmospheric NH_3 from freezing clouds

Cui Ge[a,1], Chongqin Zhu[b,1], Joseph S. Francisco[b,2], Xiao Cheng Zeng[b,2], and Jun Wang[a,2]

PNAS, 2018, 10.1073/pnas.1719949115

- **Study trying to explain our MIPAS observations of high concentrations of NH_3 in the upper troposphere**
- **"We show that the NH_3 dissolved in liquid cloud droplets is prone to being released into the UTLS upon freezing during deep convection."**





- **ph – dependence of NH_3 solubility in liquid water: „Convective clouds are hardly acidic so that NH_3 is only partly dissolved and removed by precipitation"**

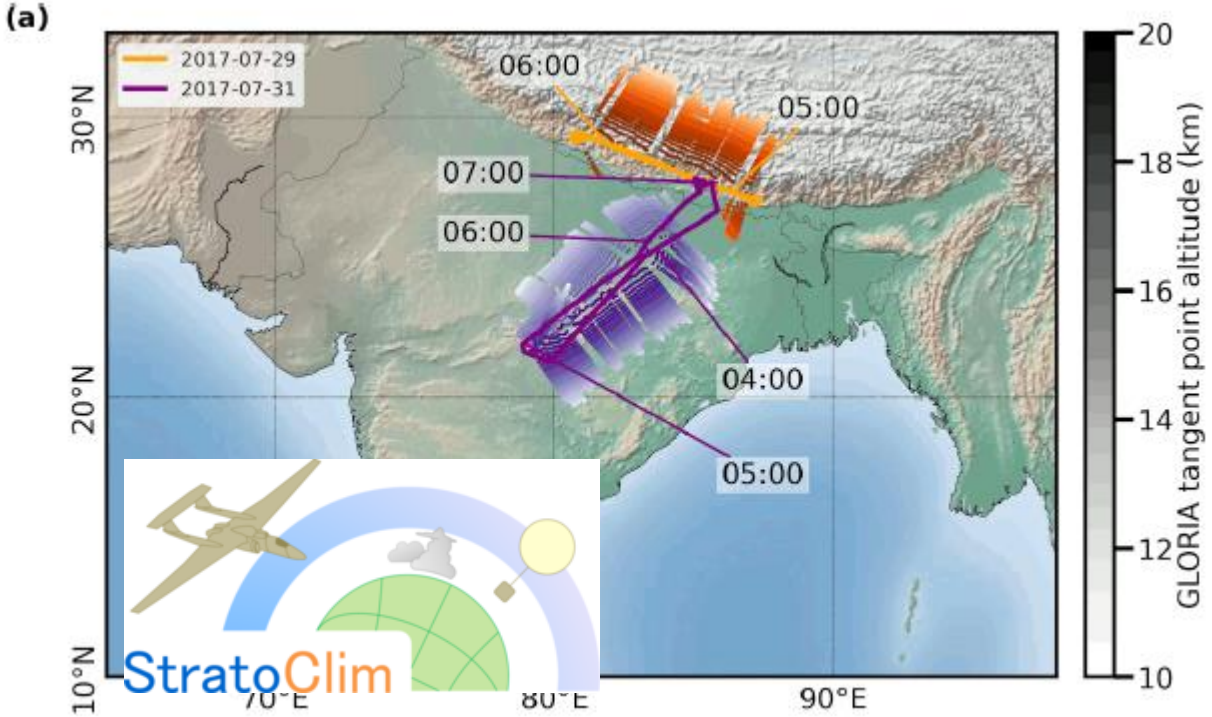
  Metzger et al., JGR, 2002, 10.1029/2001JD001103

➢ **It is still an open question, which of these processes (or a different one) is responsible for NH_3 not being completely washed/rained out during convection but reaching the upper troposphere.**

Ammonia in the UTLS:
GLORIA observations vs. CAMS global model

StratoClim campaign 2017
- Source: agricultural activity
- Upward transport: strong convection

SouthTrac campaign 2019
- Source: biomass burning
- Upward transport: (pyro-?) convection



Johansson, et al., ACP accepted, 2024

CAMS

Atmosphere Monitoring Service

Karlsruhe Institute of Technology

Reanalysis

- Same model setup for all years
- 0.75°x0.75° resolution
- 3h temporal output
- GFAS biomass burning emissions
- PAN distributions reasonably agree with GLORIA measurements

Forecast

- Regular IFS/chemistry upgrades

➔Different versions for 2017 & 2019

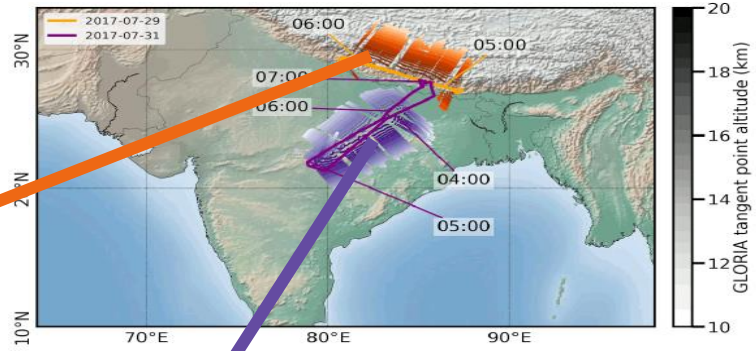
- 0.4°x0.4° resolution
- Initialized 0:00 and 12:00 UTC, output every 3h
- GFAS biomass burning emissions and use of emission heights
- Ammonium and nitrate aerosol species are introduced

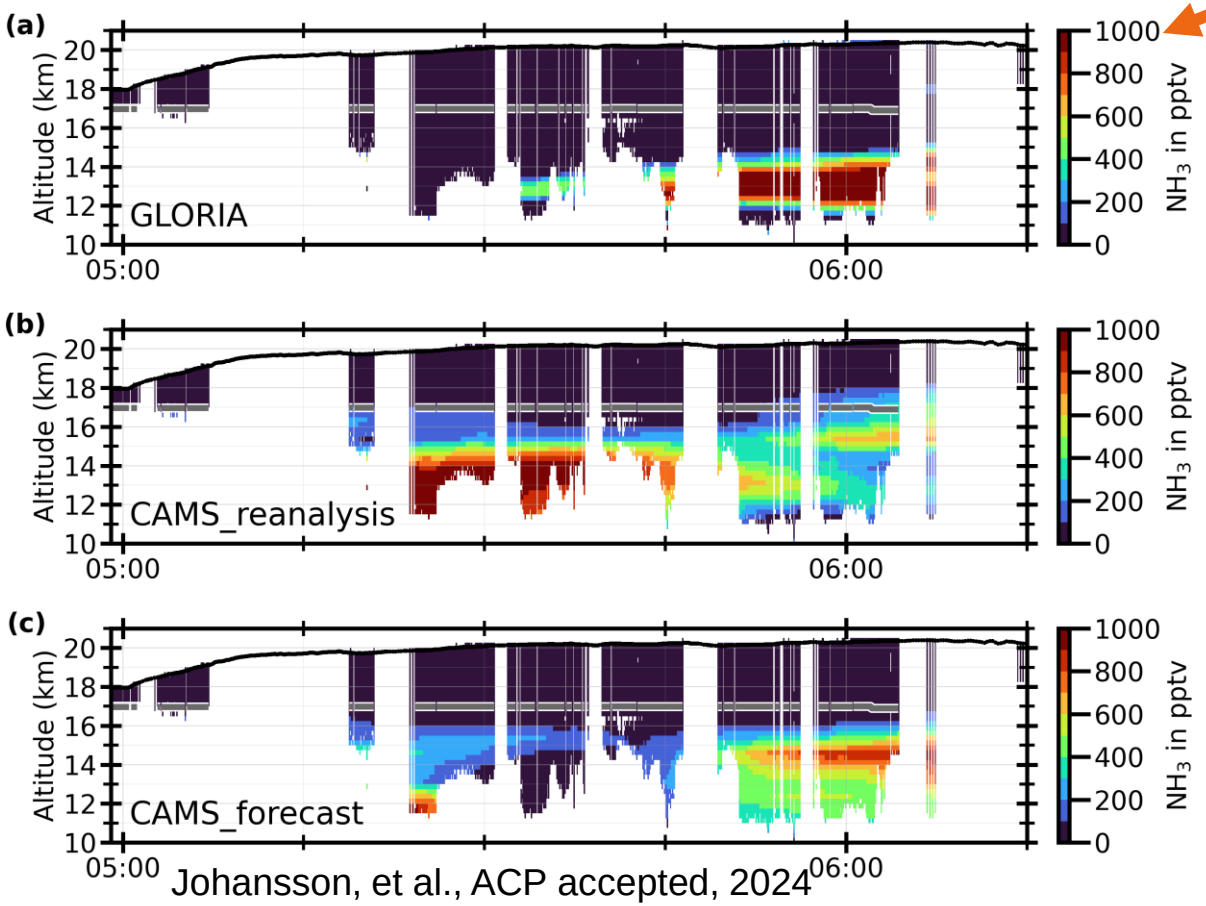
Introduced for 2019 forecasts

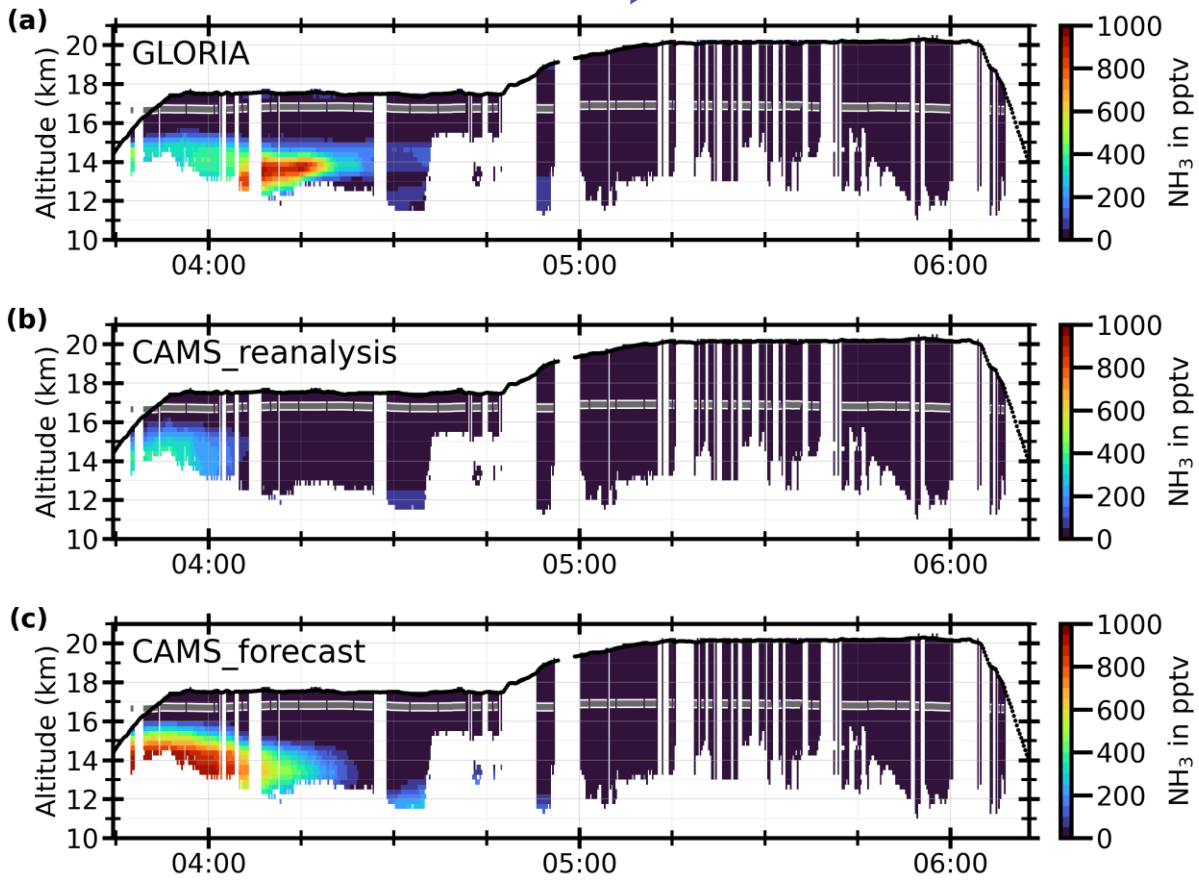
Johansson, et al., ACP accepted, 2024

StratoClim campaign 2017

- CAMS models NH_3 reasonably well compared with GLORIA observations in the monsoon UT
- Better representation in forecast due to updated version and better spatial model resolution (?)

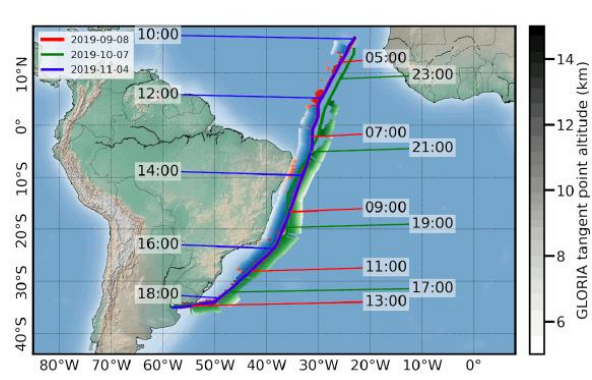






Johansson, et al., ACP accepted, 2024

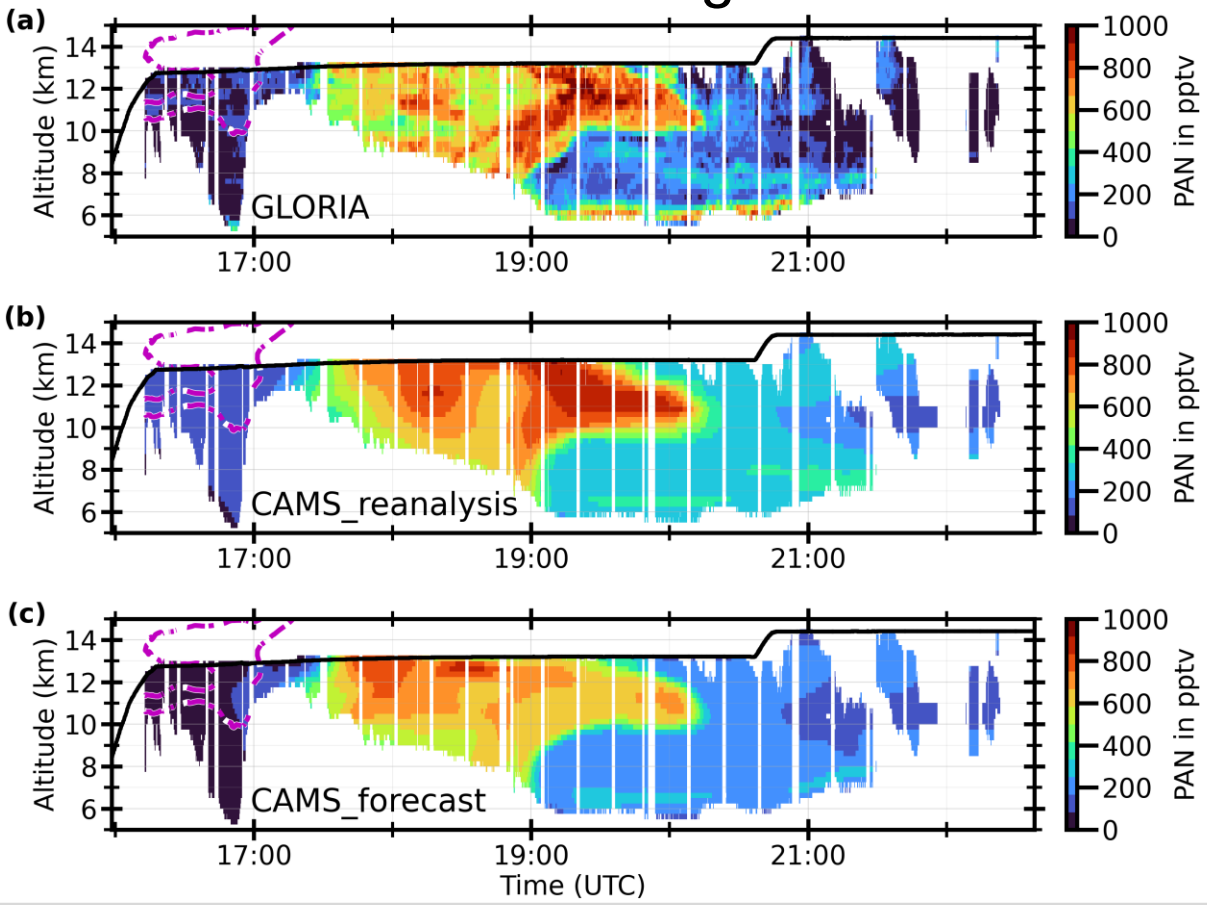
SouthTrac campaign 2019: does biomass-burning contribute to NH_3 in the upper troposphere and is it correctly modelled by CAMS?

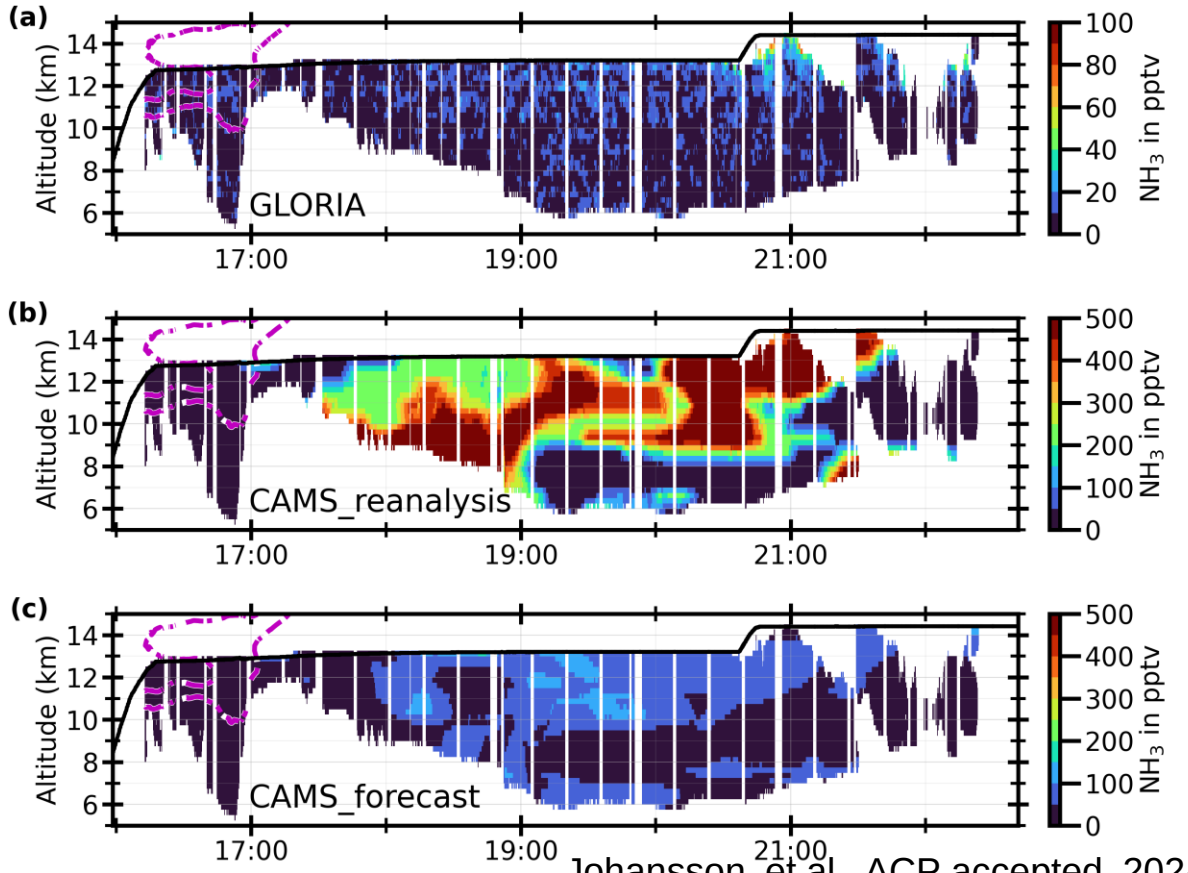


PAN (Peroxyacetyl nitrate): biomass-burning tracer

7 Oct 2019

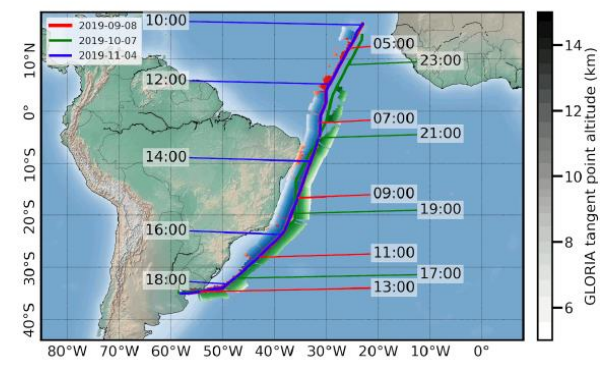
NH_3





Johansson, et al., ACP accepted, 2024

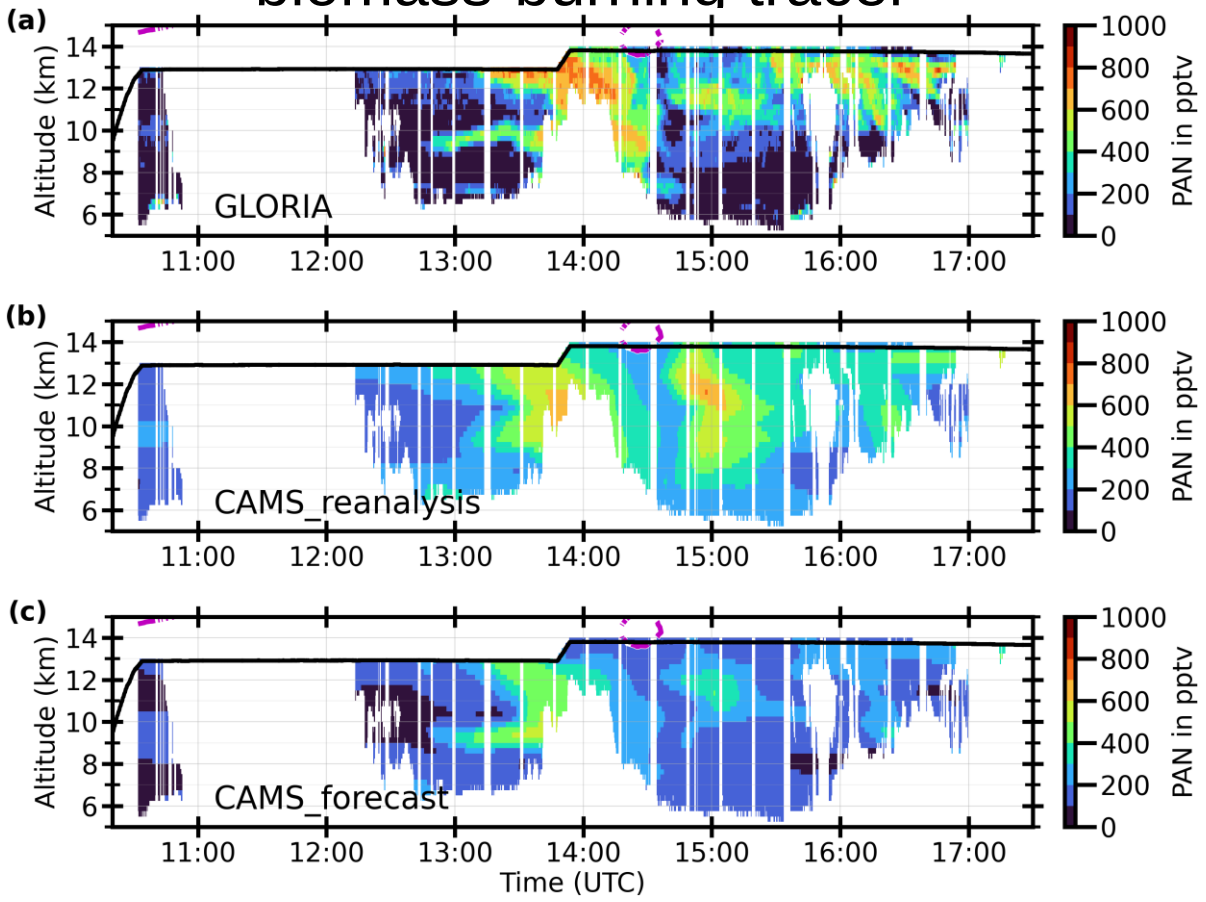
- **GLORIA does not show detectable enhancements of NH_3**
- **CAMS strongly overestimates NH_3 in biomass-burning plumes**

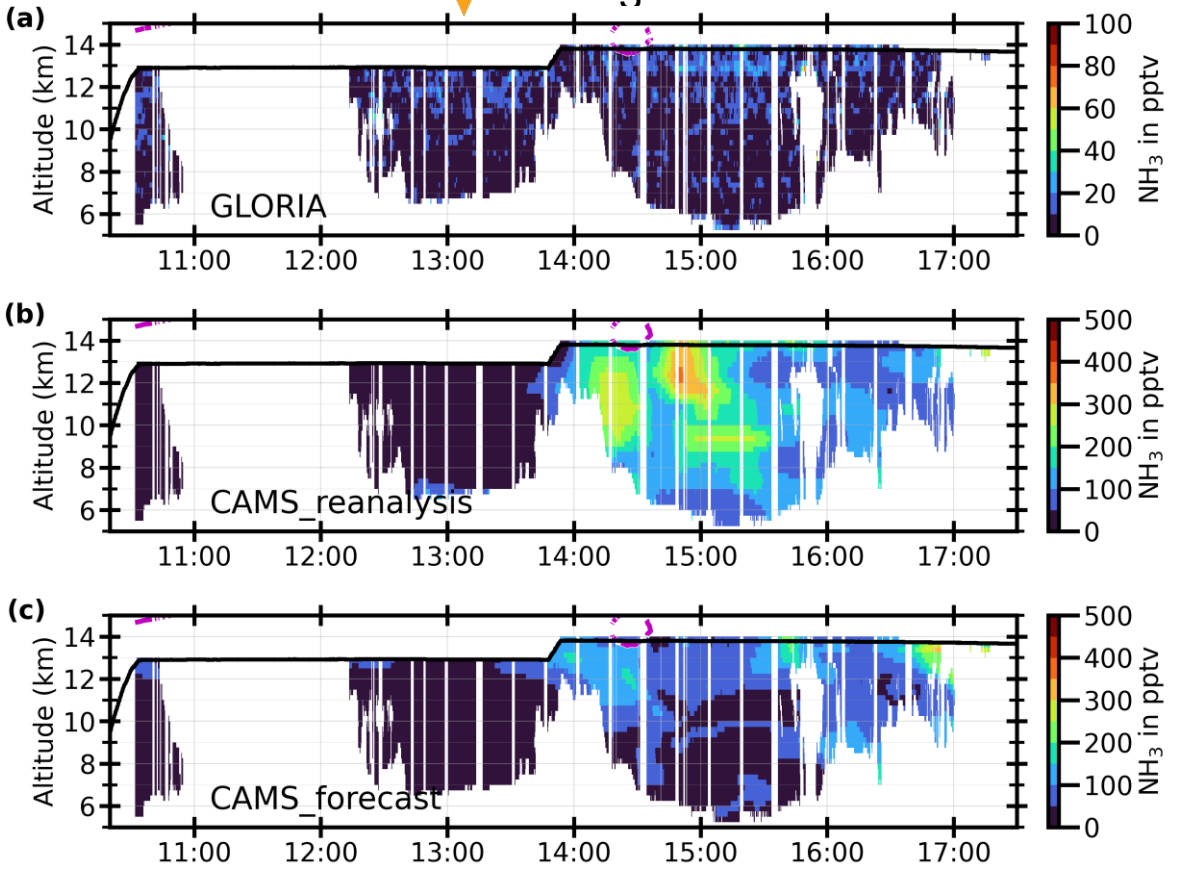


PAN (Peroxyacetyl nitrate): biomass-burning tracer

4 Nov 2019

NH_3





Johansson, et al., ACP accepted, 2024

Possible reasons for NH_3 -overestimation in BB-plumes

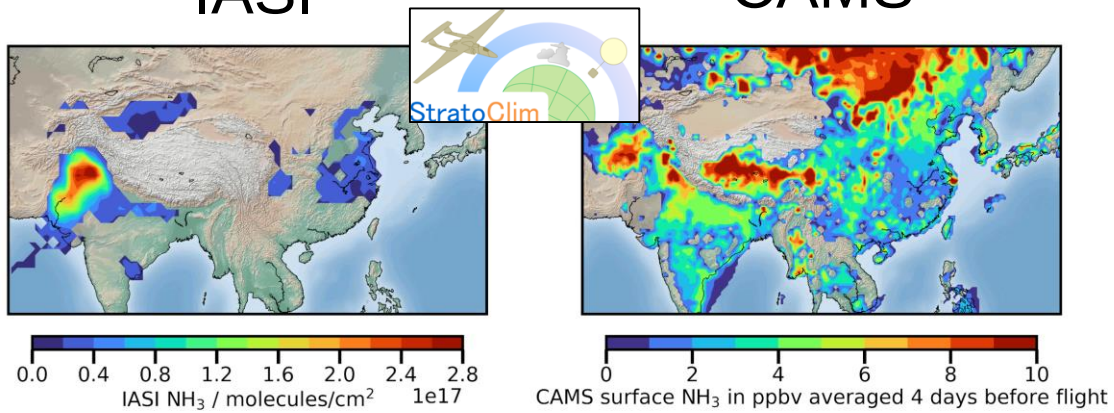
Karlsruhe Institute of Technology

Overestimation of sources?

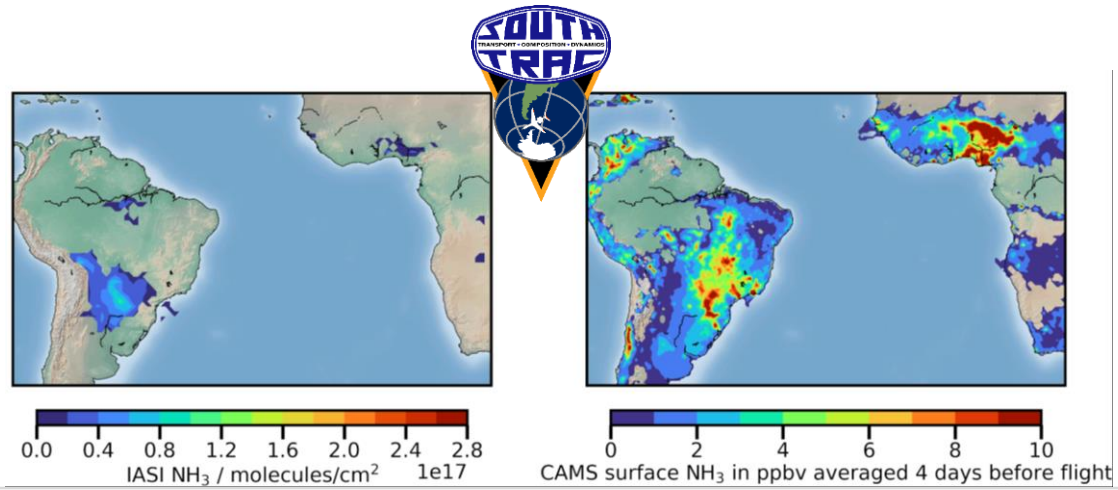
Monsoon

IASI

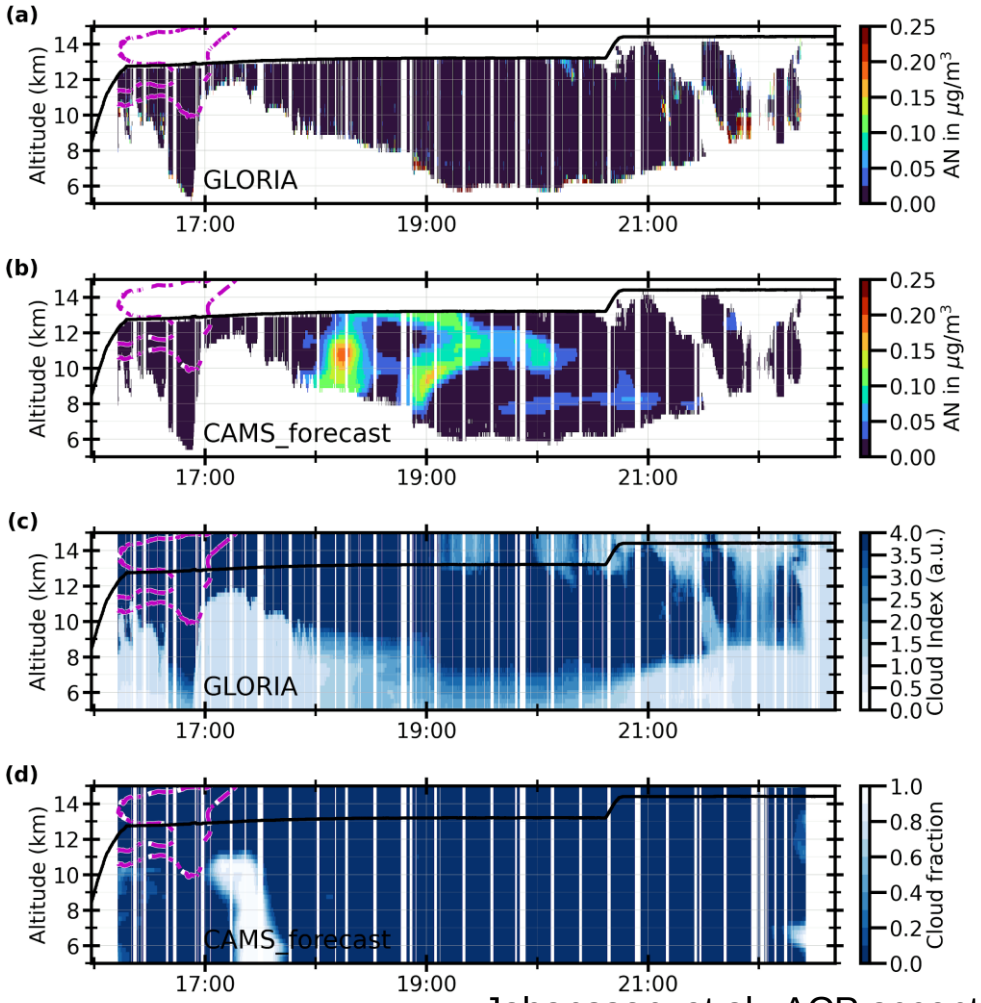
CAMS



Biomass burning



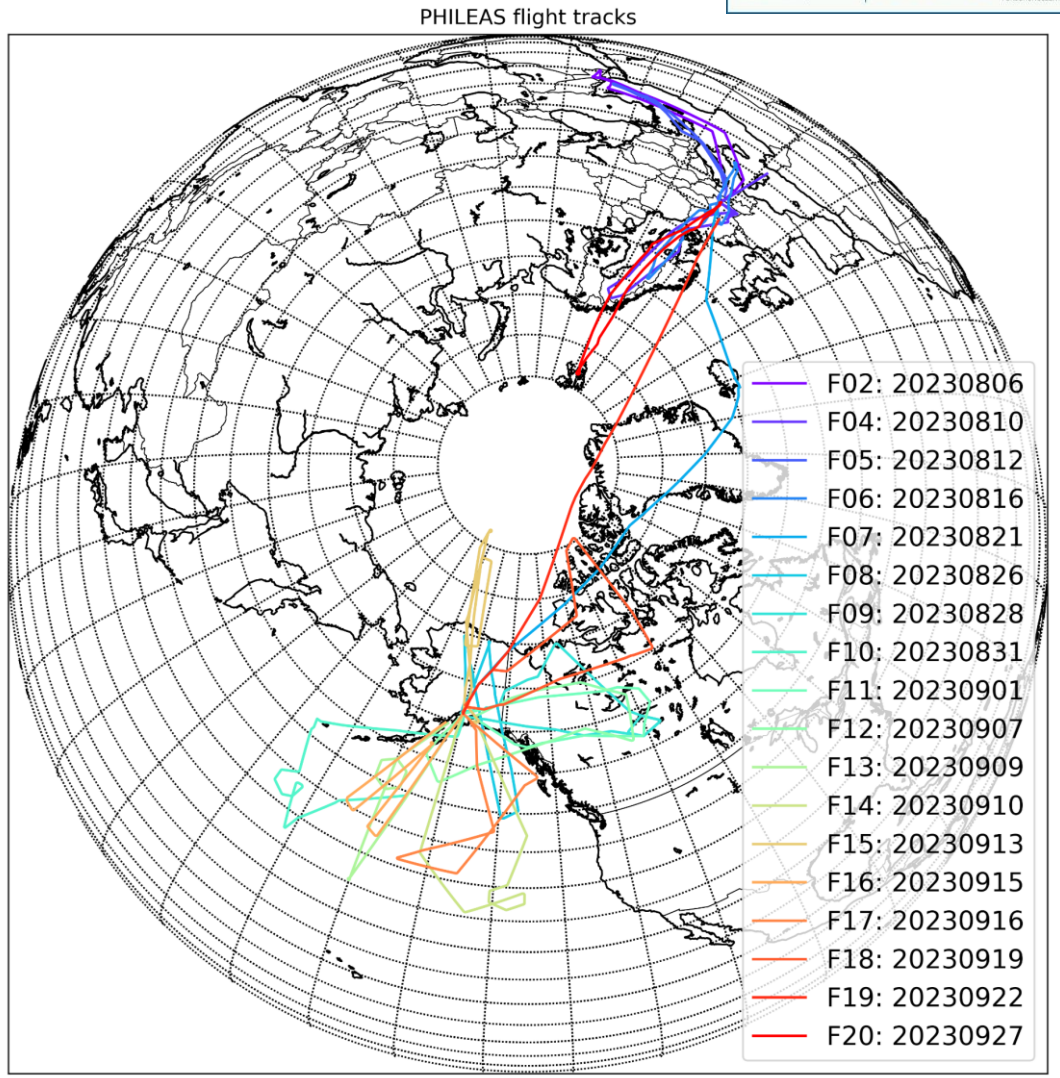
Underestimation of sinks?



Johansson, et al., ACP accepted, 2024

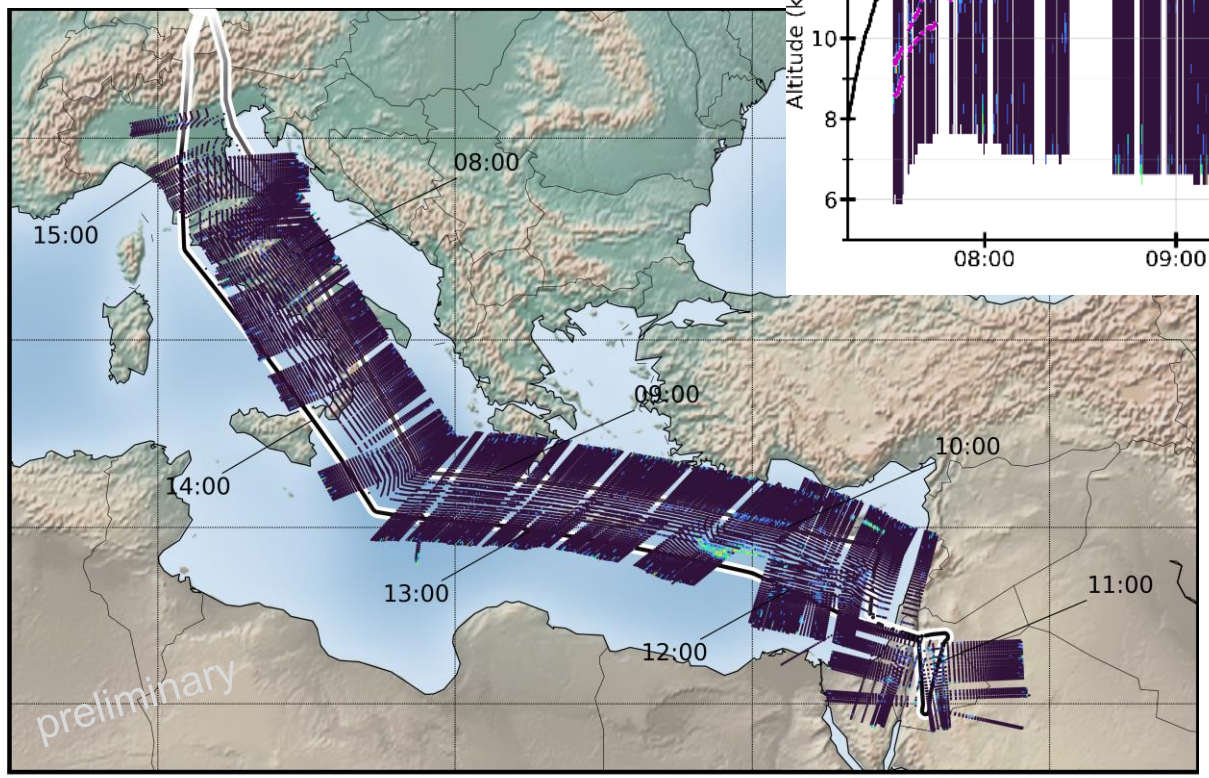
PHILEAS-campaign 2023

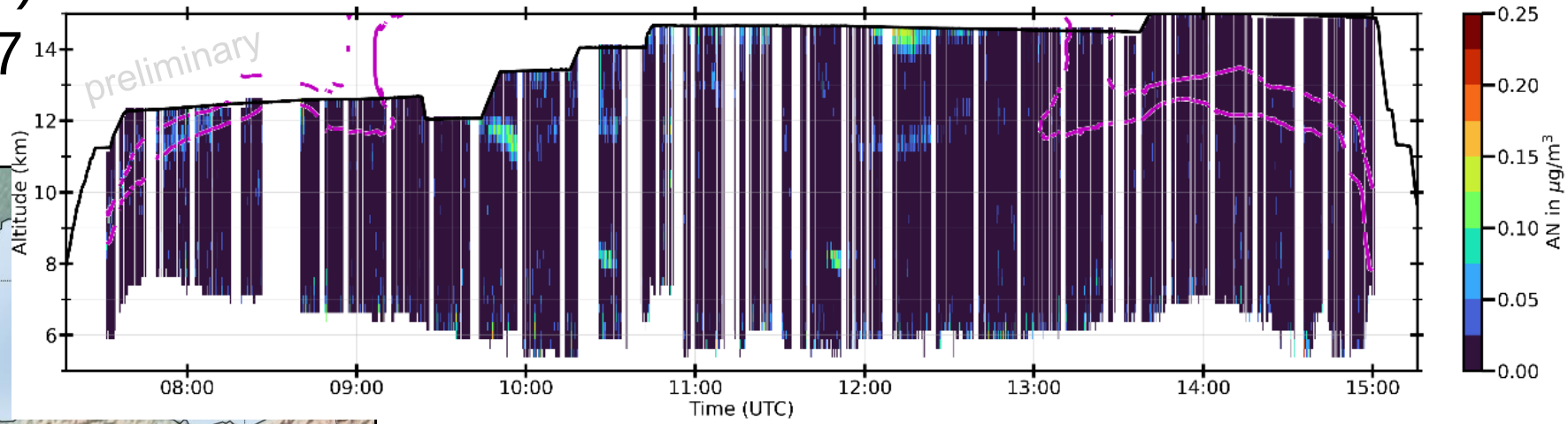
- What are the pathways, time scales and dynamical processes of air mass transport from the Asian summer monsoon into the extratropical UTLS
- How do gas-phase and particulate constituents evolve in large-scale eddies which are shed from the monsoon anticyclone?
- How does eddy shedding from the monsoon impact the extratropical LMS composition in particular the water vapour and radiatively active species?



Flight 6 August 2023

- Ammonium nitrate (AN)
- Observed in AMA 2017

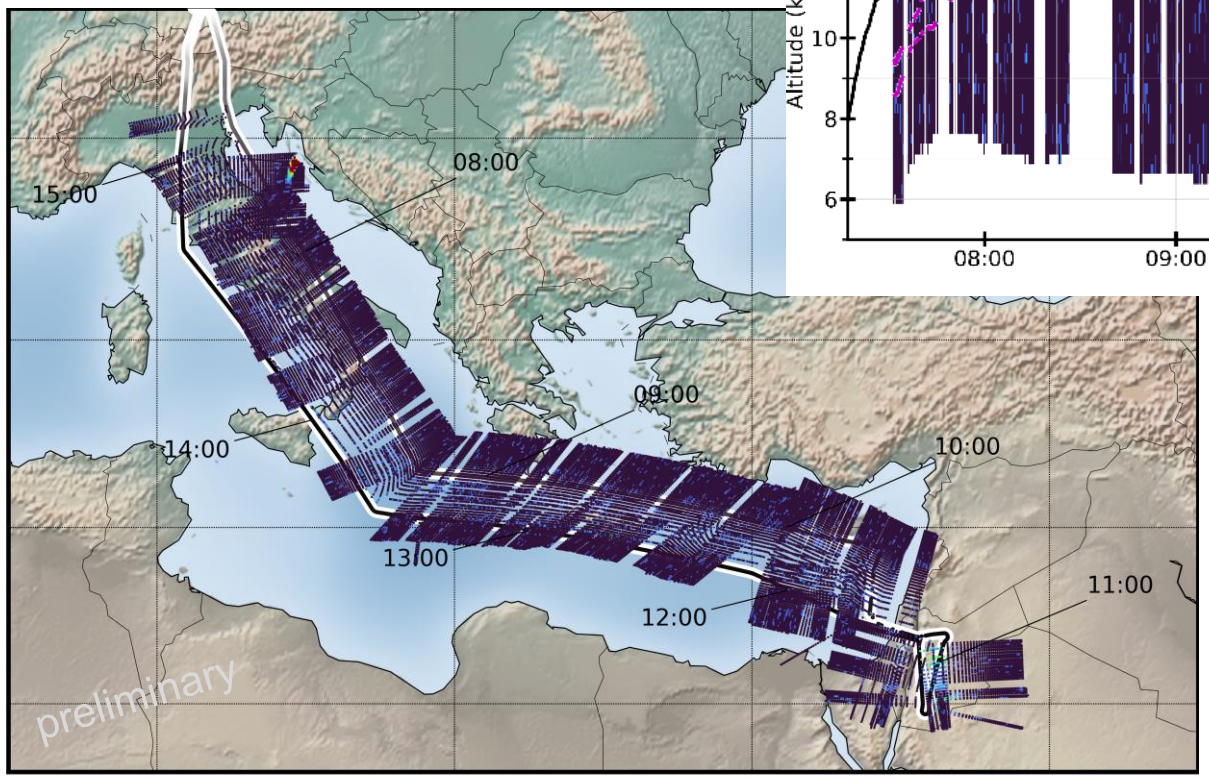


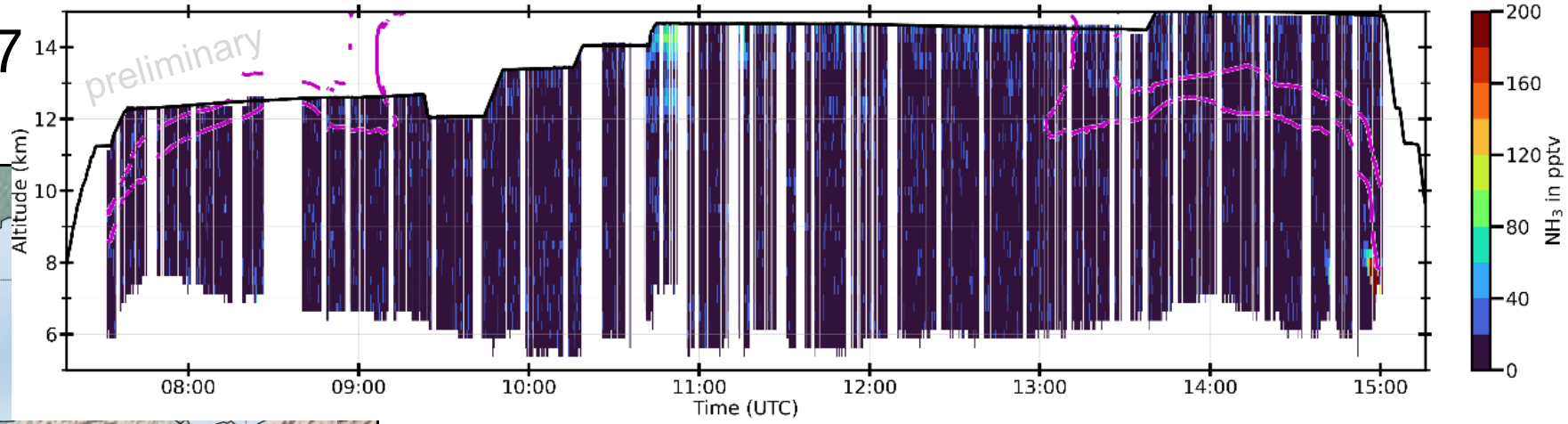


- Filamented structures above eastern Mediterranean
- Same air masses observed twice

Flight 6 August 2023

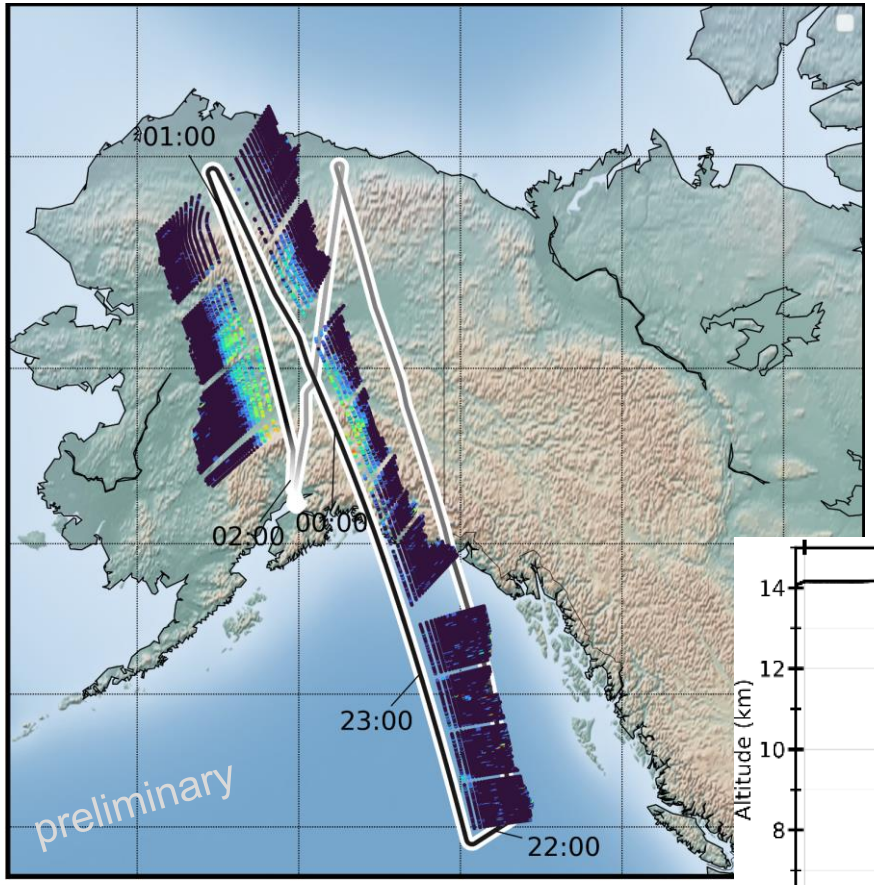
- Ammonia (NH_3)
- Observed in AMA 2017



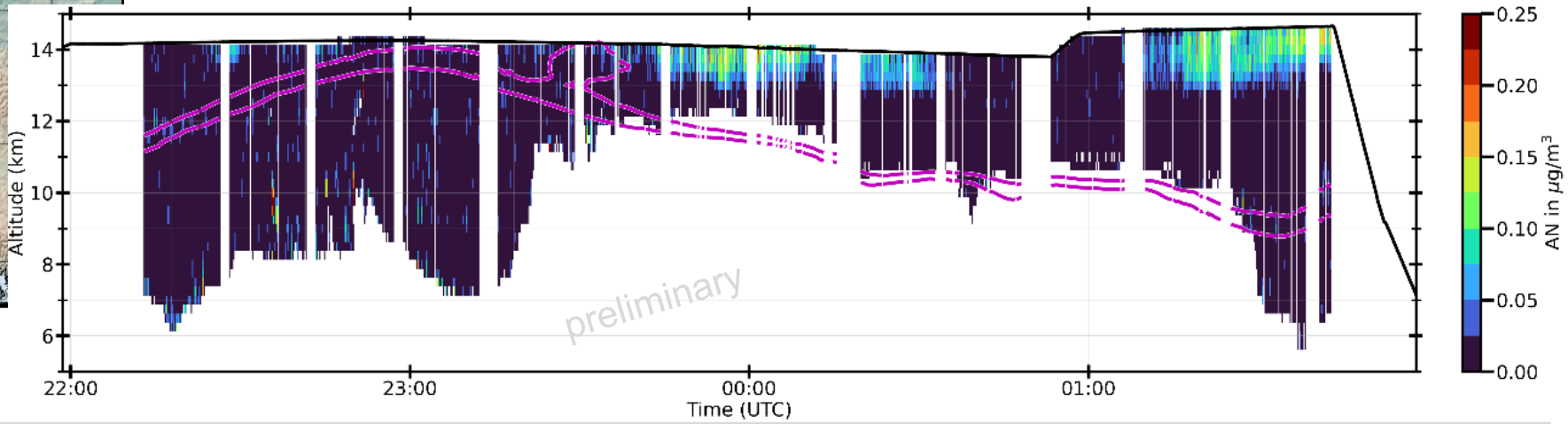


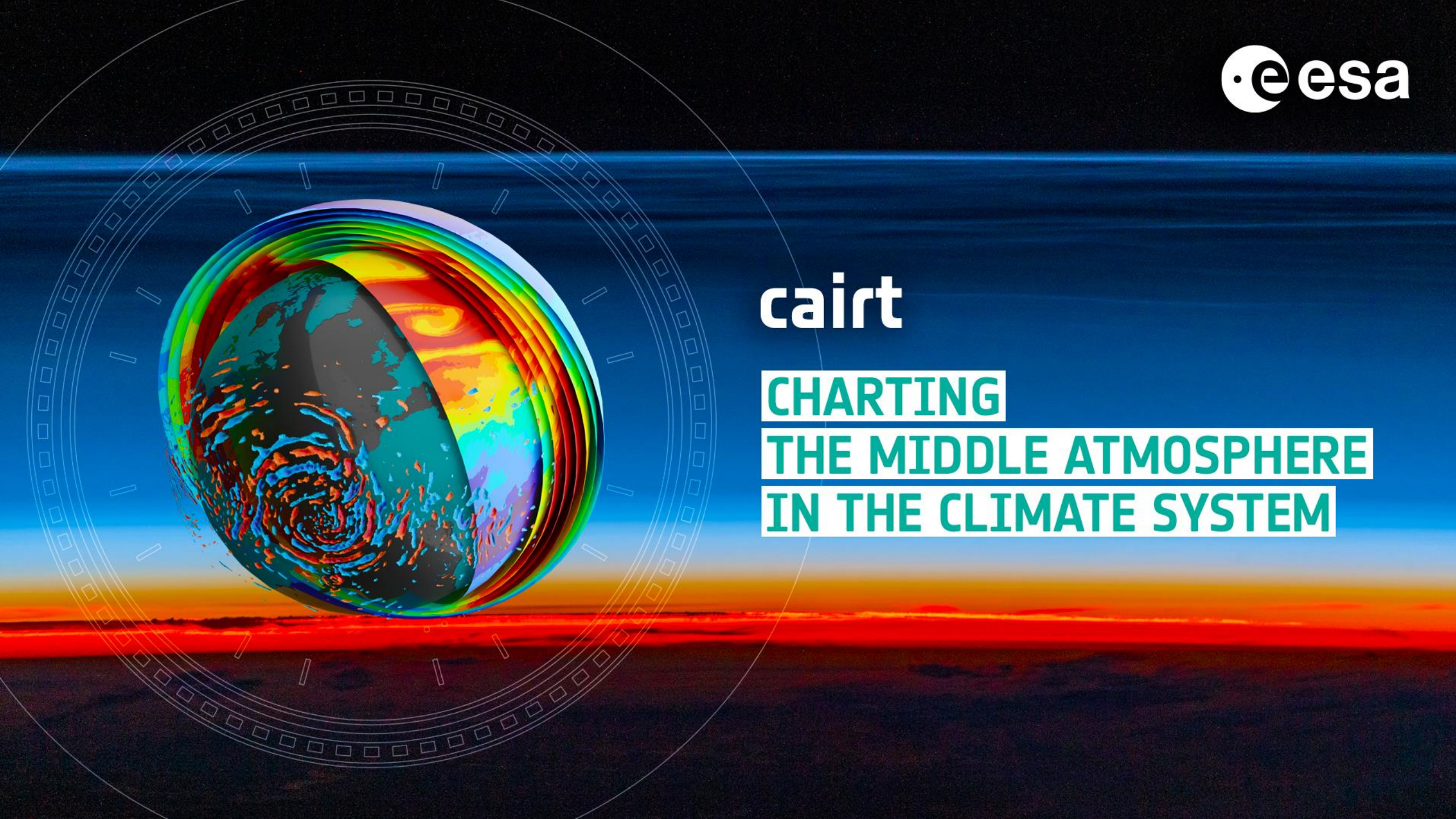
- Tiny enhancement at flight altitude above Israel/Jordan
- Large plume observed above Adriatic Sea (local source?)

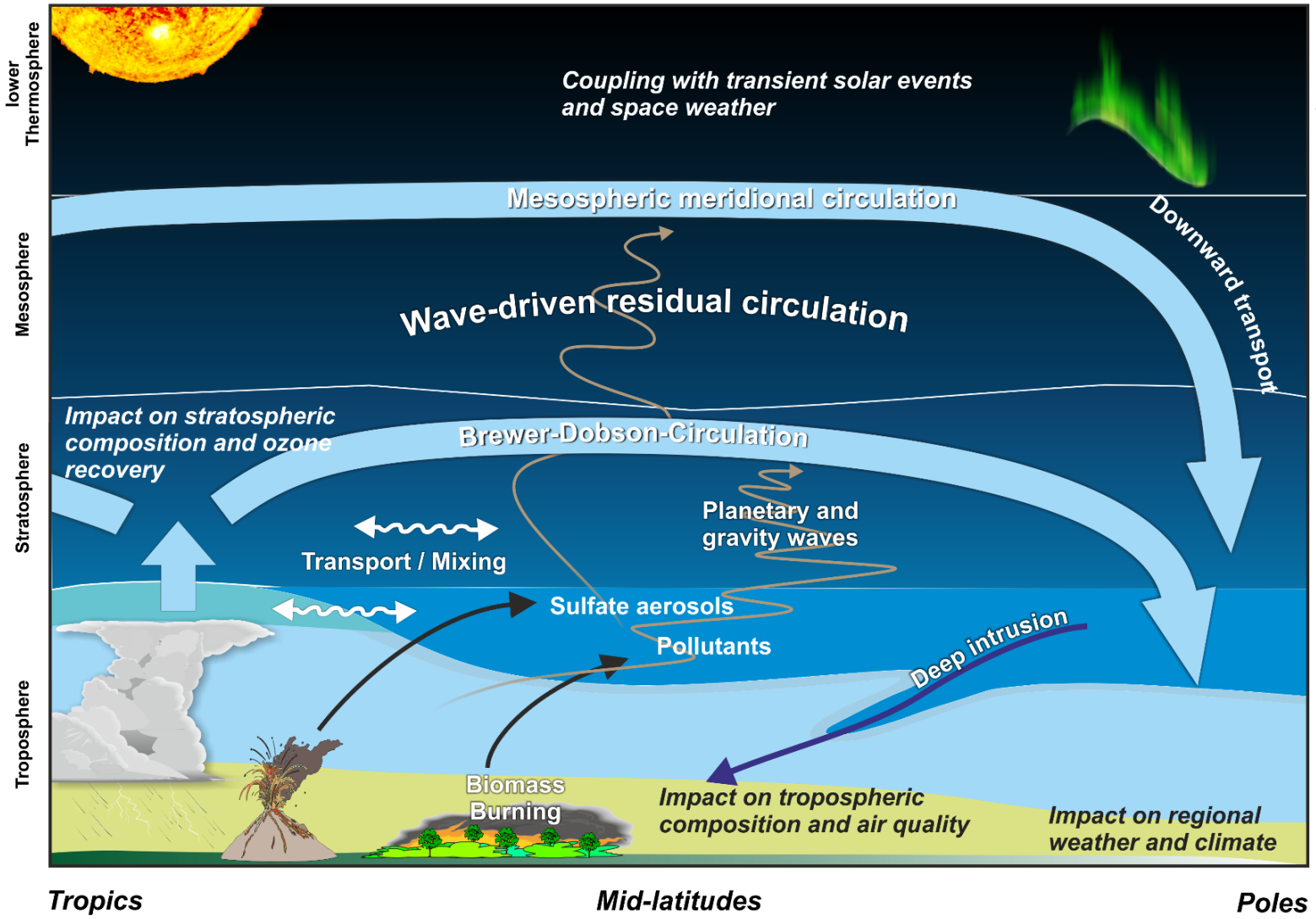
Flight 26 August 2023: first proof of AN in the stratosphere

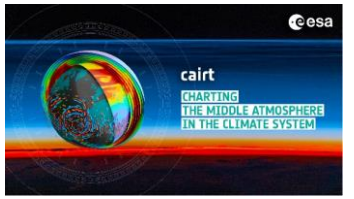


- **Layer of enhanced AN above Alaska in the lower stratosphere**
- Seems to be horizontally and vertically confined
- No indications of NH_3 measured (not shown)




esa
cairt
CHARTING
THE MIDDLE ATMOSPHERE
IN THE CLIMATE SYSTEM





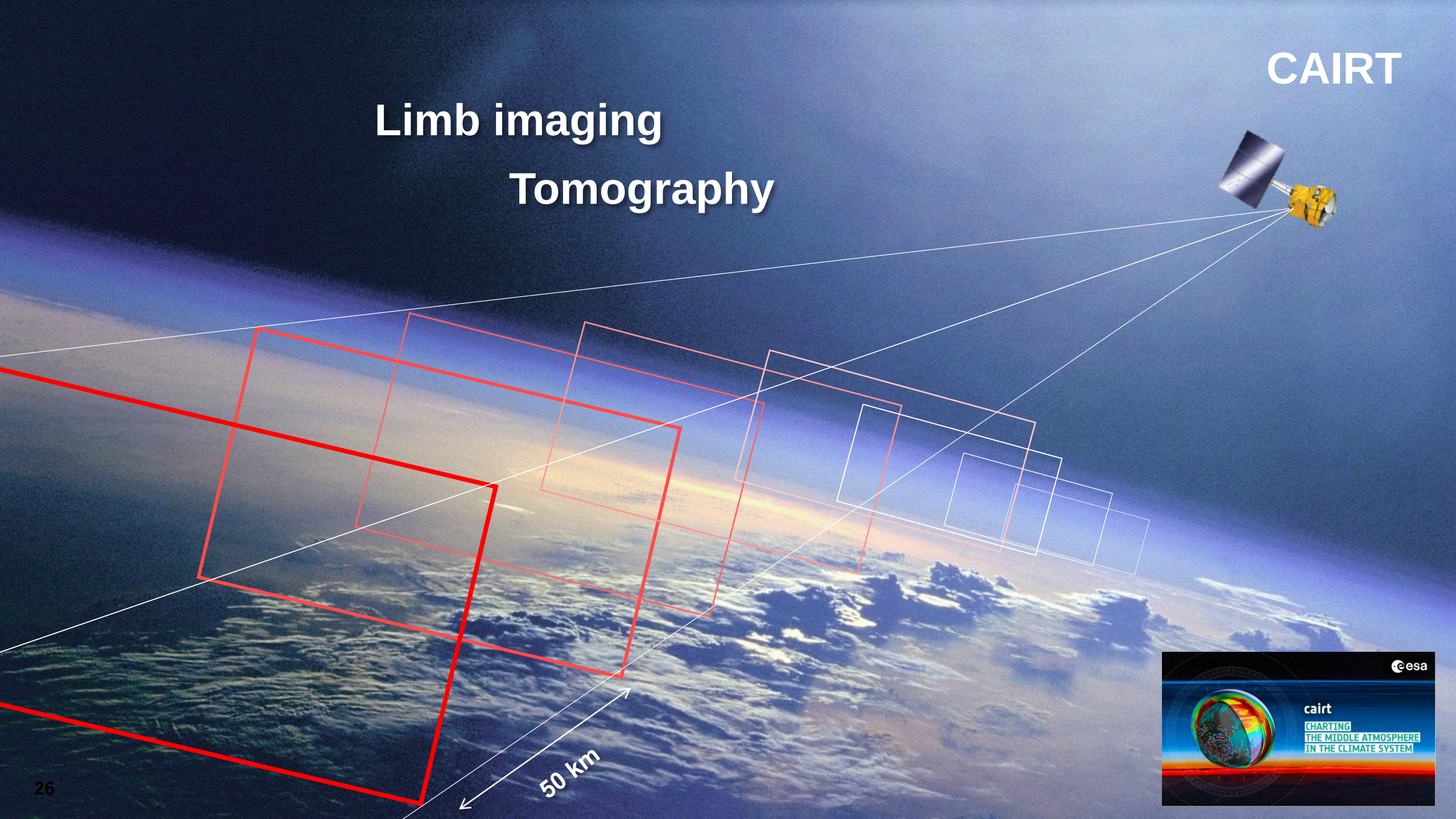
Karlsruhe Institute of Technology

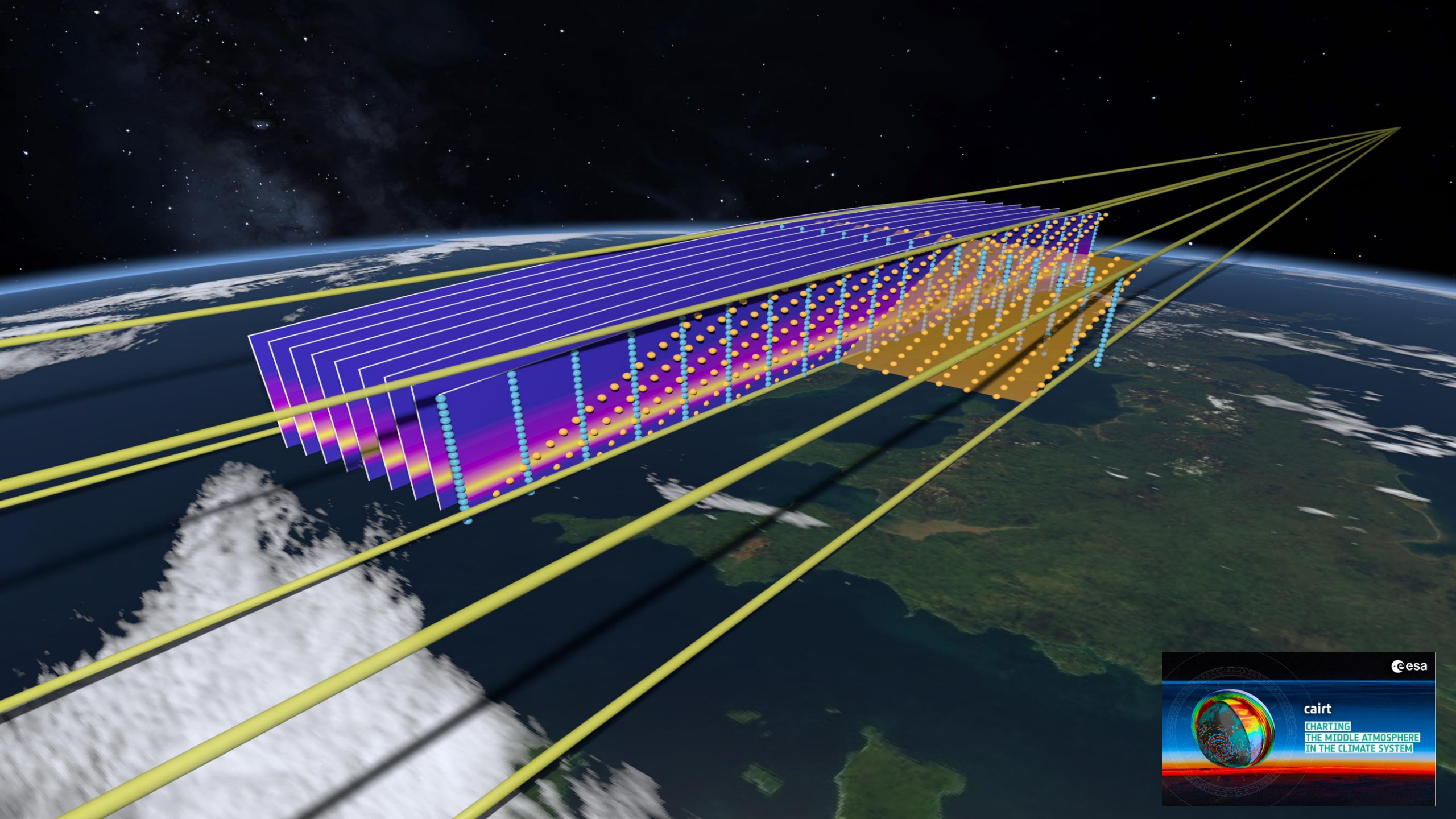
- How is the middle atmosphere circulation changing?
- What is the wave driving of the circulation?
- What is the coupling with the upper atmosphere?
- What is the input by biomass burning, volcanic eruptions, convection, …?
- What is the coupling between composition, circulation and climate?

CAIRT

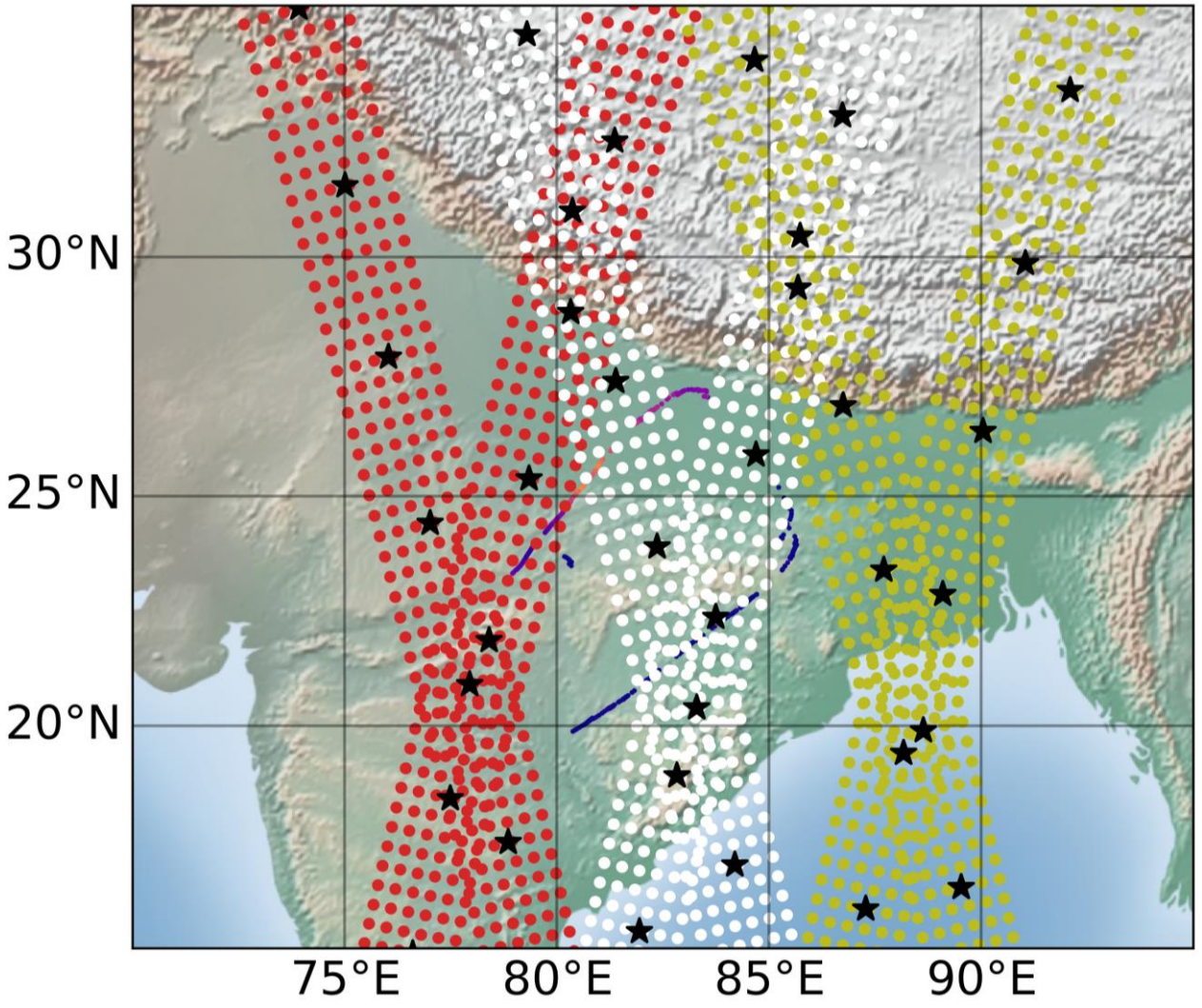
Limb imaging

Tomography



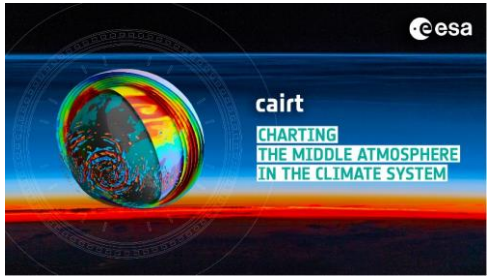

esa
cairt
CHARTING
THE MIDDLE ATMOSPHERE
IN THE CLIMATE SYSTEM

Sampling: CAIRT vs. GLORIA@aircraft vs. MIPAS@Envisat satellite

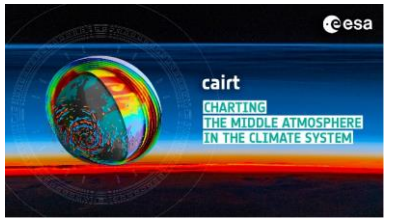


Altitude: 14 km

- GLORIA NH3
- CAIRT day 1
- CAIRT day 2
- CAIRT day 3
- MIPAS day 1-3



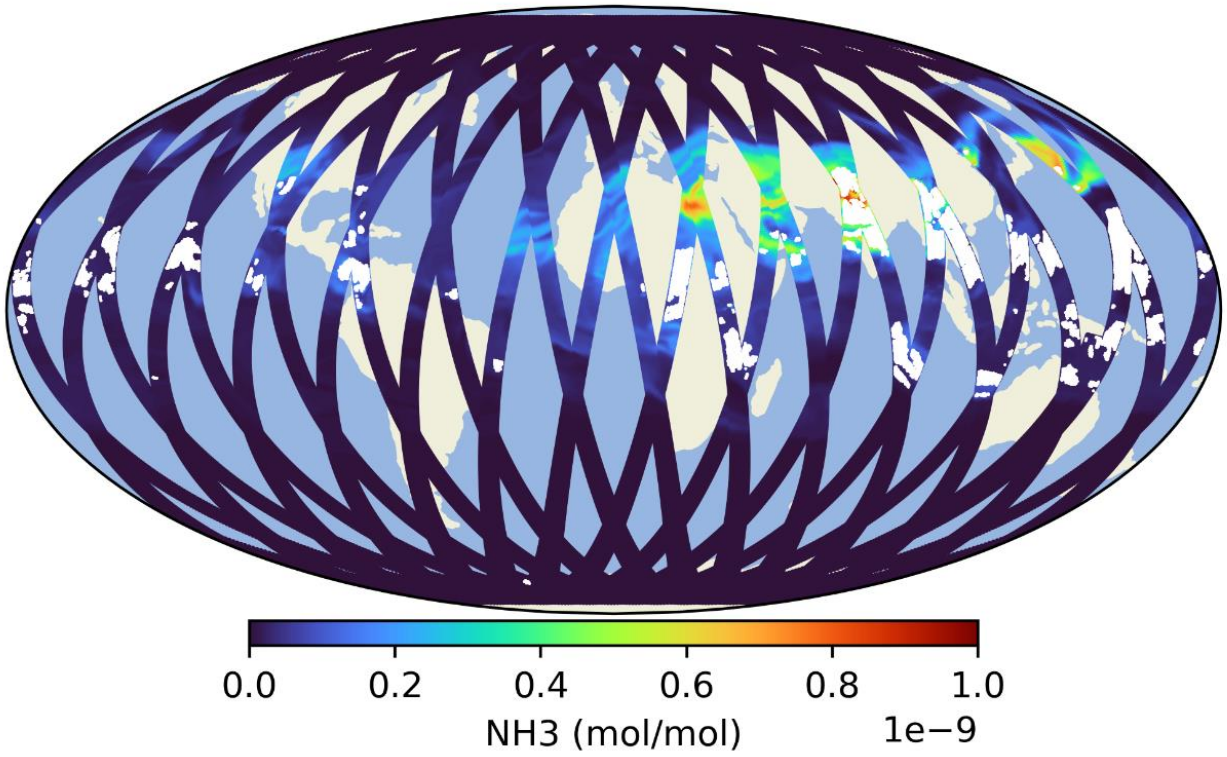
CAIRT: simulated retrieval of NH_3 at 15 km



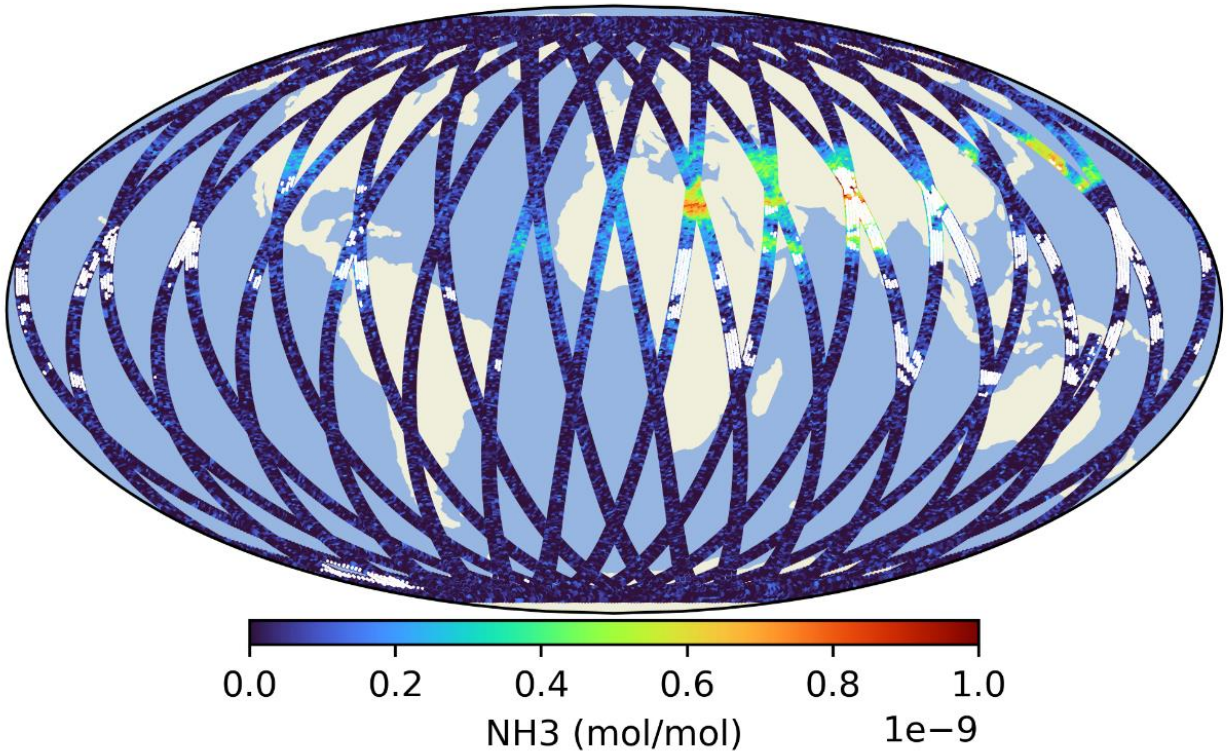
Karlsruhe Institute of Technology

One day of measurements

CAMS sampled at CAIRT grid



CAIRT retrieval incl. random and syst. uncertainty



Summary

- Ammonium nitrate and ammonia: abundant in monsoon upper troposphere

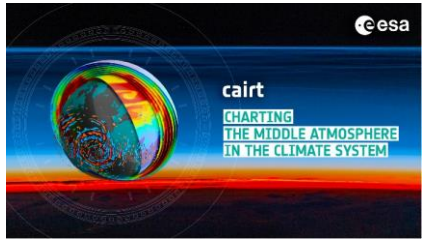
2017

- CAMS simulations agree with GLORIA NH_3 measurements within Asian Monsoon
- CAMS simulations overestimate NH_3 within biomass burning plumes. Reasons: wrong source strength, transport, underestimated sinks ?

2019

- GLORIA observations indicate complex distributions of NO_y constituents (AN, PAN, HNO_3 in monsoon filaments and their vicinity
- Ammonium nitrate detected in filaments in the UT above the Mediterranean
- NH_4NO_3 detected in the lower stratosphere for the first time outside the monsoon region (Alaska)

2023



2032?

- ESA Earth Explorer 11 candidate CAIRT would allow to study an important fraction of NO_y and NH_3 globally, continuously and detailed www.cairt.eu

Thanks

- STIPMEX team for invitation and superb organization
- GLORIA-team (KIT&FZJülich)
- MIPAS-team (KIT)
- AIDA-team (KIT)
- PI’s of the campaigns, aircraft&ground crews
- Mission Advisory Group of CAIRT
- ESA/EU/DFG