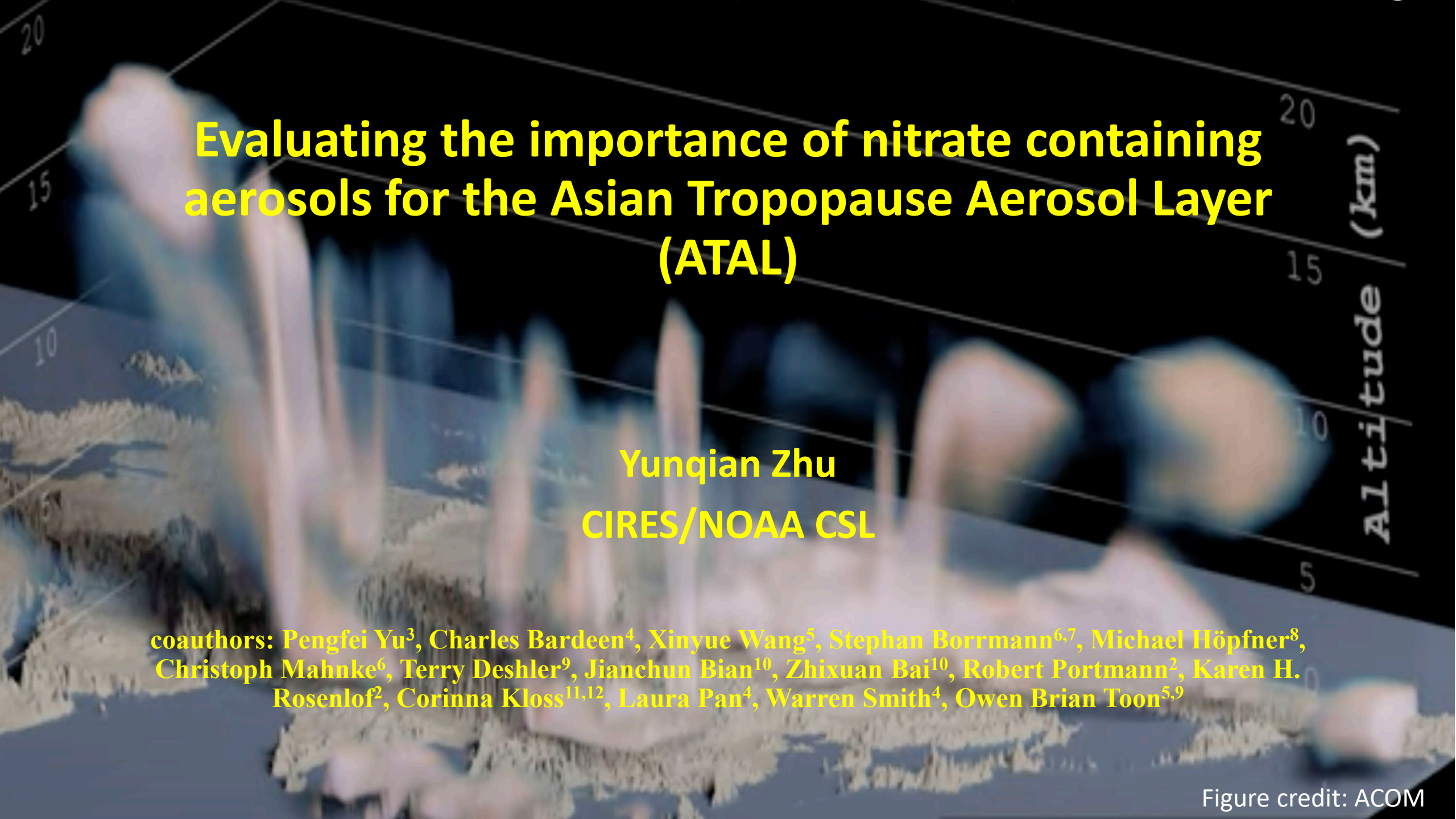


# Evaluating the importance of nitrate containing aerosols for the Asian Tropopause Aerosol Layer (ATAL)

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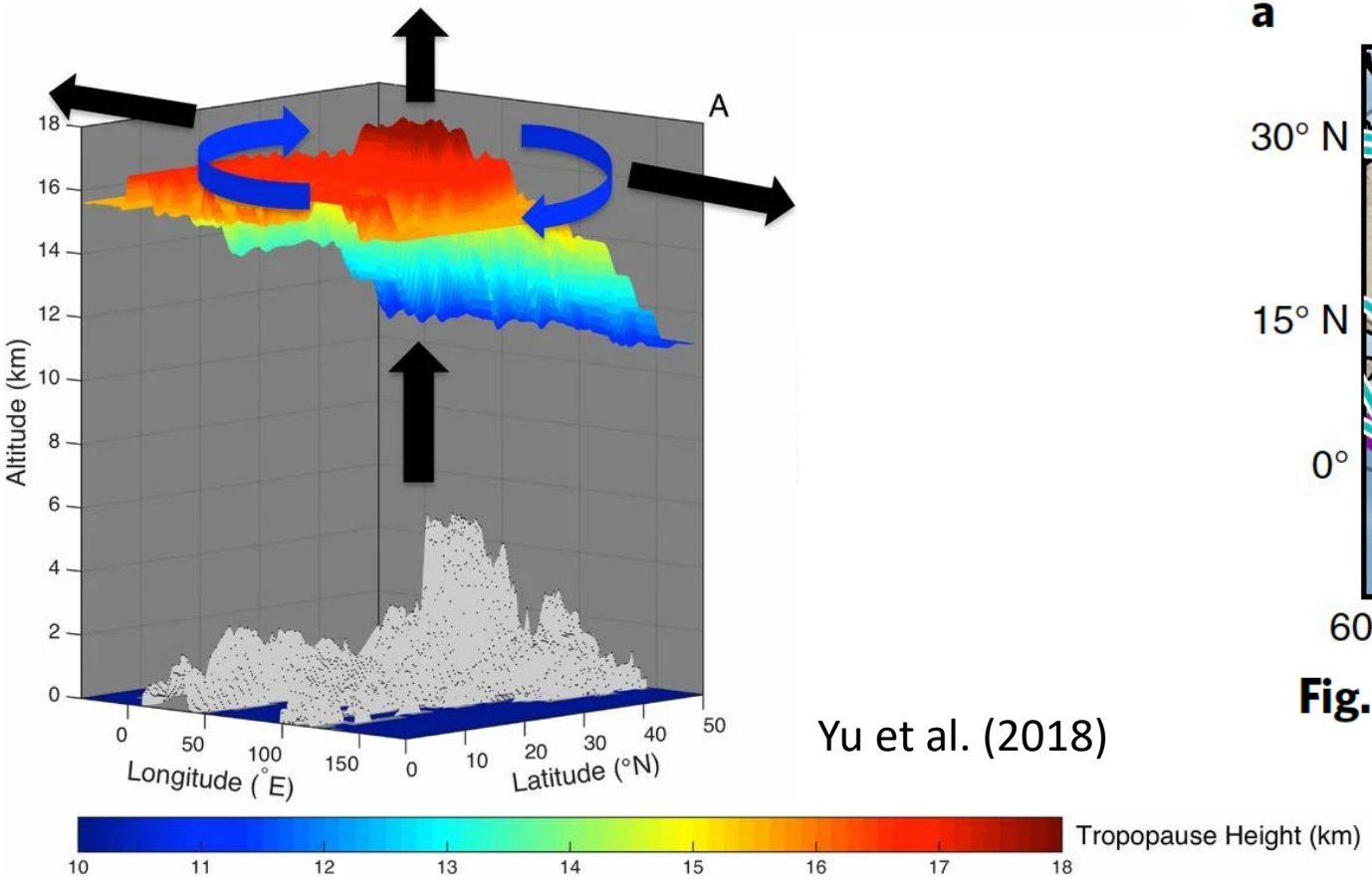
Figure credit: ACOM

# Asian tropopause aerosol layer (ATAL) forms during the ASM season near the tropopause. The composition of ATAL is still under investigation.




Yu et al. (2018)

Hopfner et al. (2019)

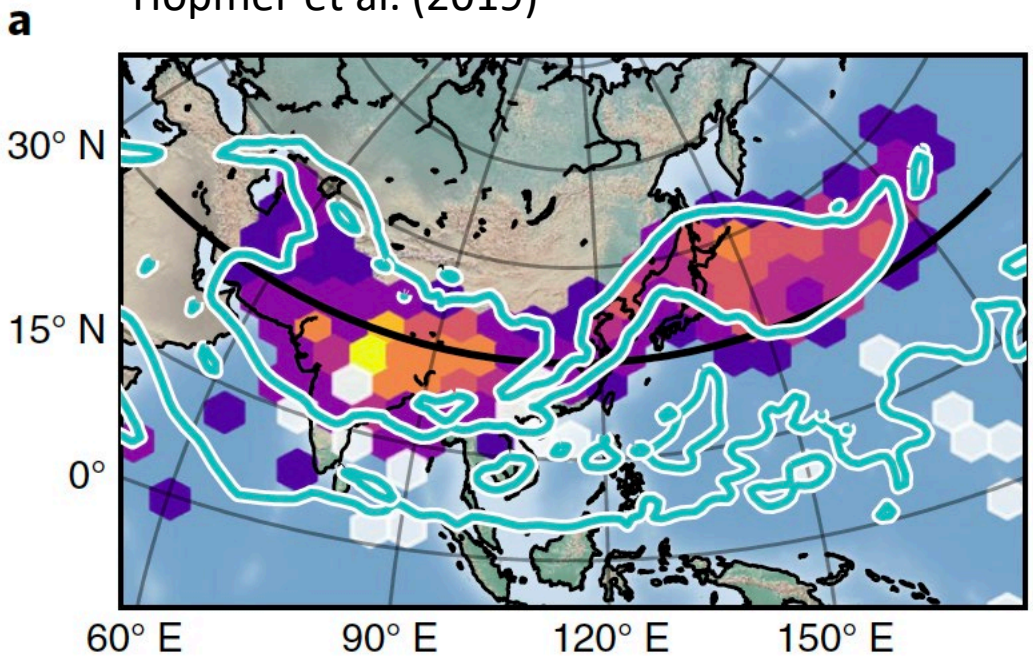



**Fig. 1 | AN observed by CRISTA in the UT in 1997.**

# Model Development History

- Start with Yu et al. 2015, CESM1/CARMA (Which is what we put in the CESM2/CARMA).
  2 groups, pure sulfate and mixed (seasalt, dust, OC, BC, sulfate), bulk SOA

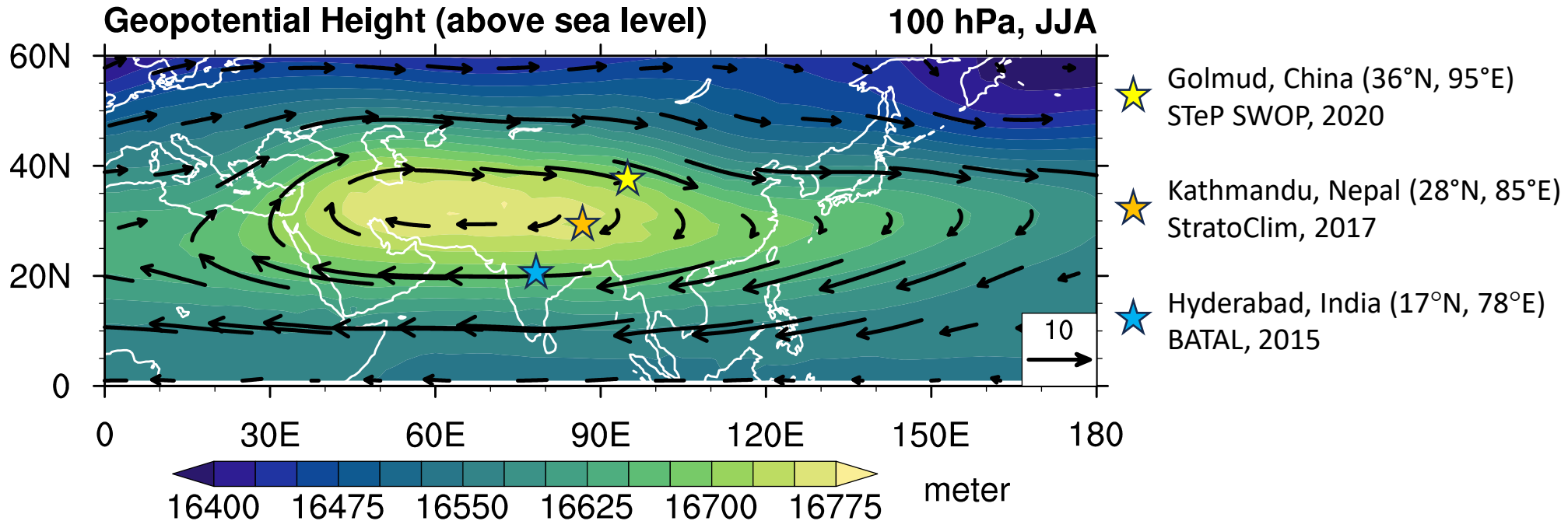
- Yu et al. 2022, add the nitrate in CESM1/CARMA simulating the whole atmosphere. For the nitrate condensation:
  Surface  to 300 hPa: MOSAIC;
  <300 hPa: Lin&Tabazadeh 2001, forming NH3/SO4/NO3 liquid particles.
  2 groups, pure sulfate and mixed (seasalt, dust, OC, BC, sulfate, nitrate, ammonium), bulk SOA

- This work, modifying the nitrate formation when pressure <300 hPa: using Clegg AIM model to estimate the formation of solid ammonium nitrate (AN) and ammonium sulfate (AS). Using Luo1996&Hanson1988 to form liquid $NO_3^-$ particle.
  Reduce the mixed particle minimum radius coverage from 0.05 um to 1 nm.
  Allow more NH3 transport to UTLS; add volcanic injection input files.
  Other parts are the same as Yu 2022.

# AN AS summary

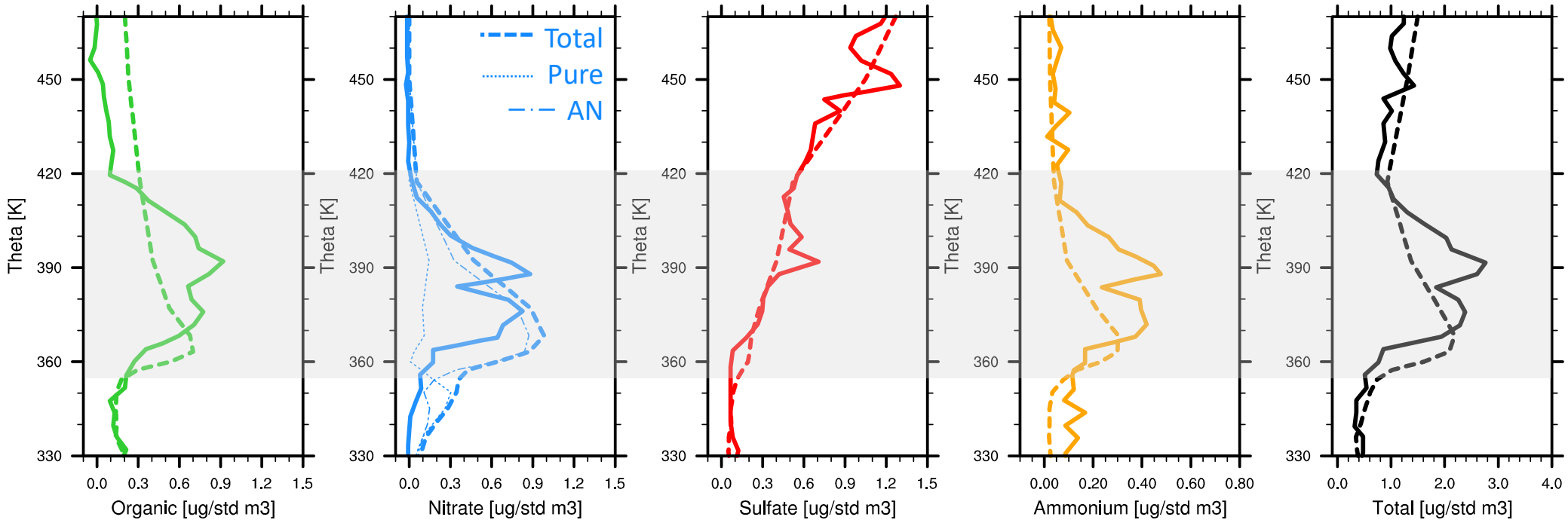
- T>198K, $NH_4^+$ is preferred to form $(NH_4)_3H(SO_4)_2$ (Letovicite) solid first, the rest forms solid $NH_4NO_3$.
- 198K>T>195K, AS (Letovicite) deliquescence, $NH_4^+$ forms $NH_4NO_3$ solid.
- T<195K, AN starts to deliquescence, the $NO_3^-$ growth curve is similar to STS PSCs (Luo et al., 1995).

# We validate the model using three field campaigns and MIPAS satellite.

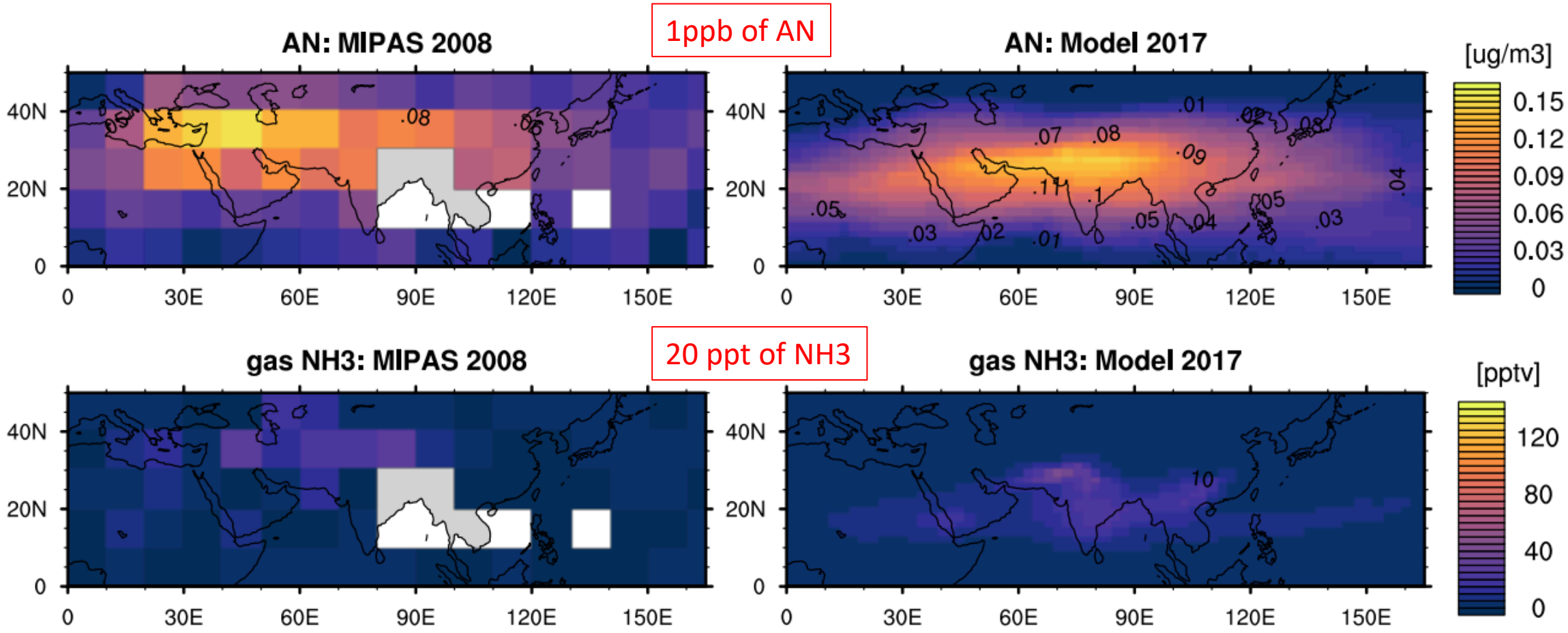


Aerosol composition compared well with ERICA during the StratoClim 2017 Campaign (8 flights), showing organics, nitrate and sulfate near 0.8 ug/m$^3$ at ATAL. The simulated ATAL is slightly lower in altitudes compared with obs.

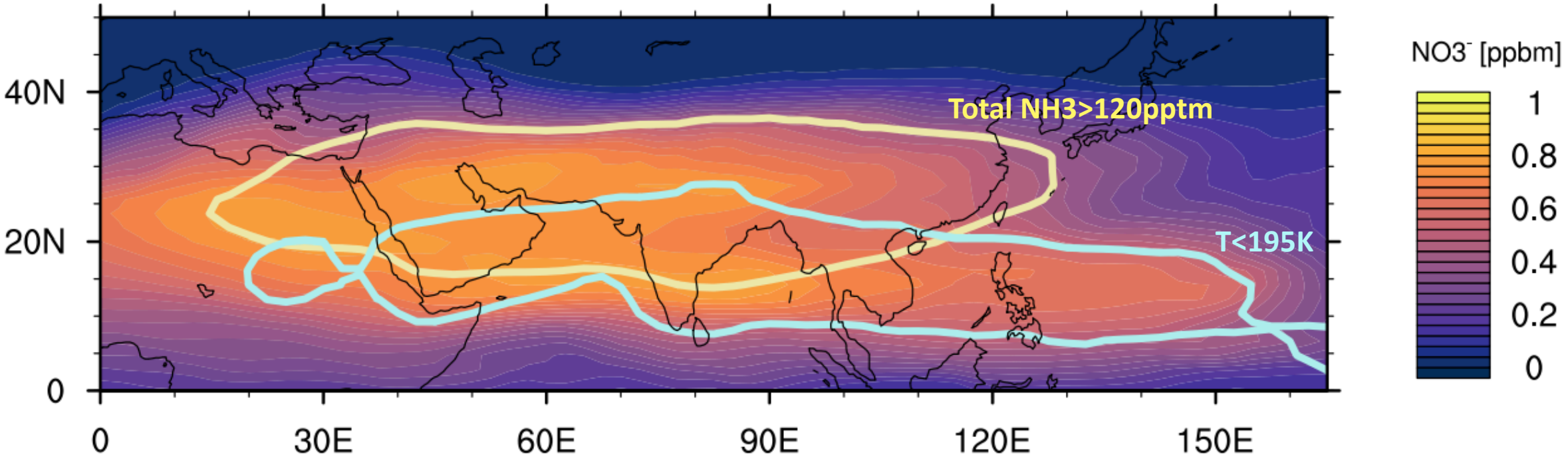
Solid Lines: ERICA from Appel et al. (2022); dash lines: Model; is the approximate ATAL region.



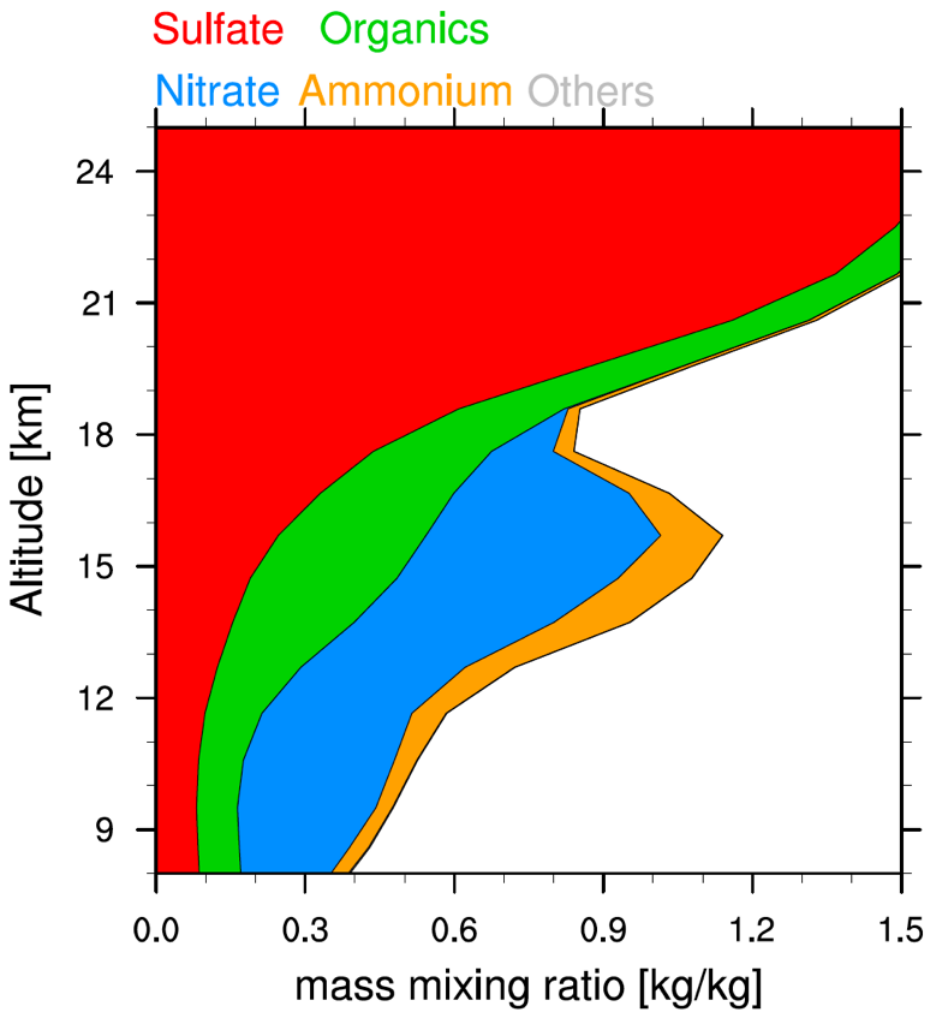
# ATAL composition: Ammonium Nitrate and NH3 gas abundant is consistent with MIPAS observation.



# Simulated $NO_3^-$ peaks due to different reasons at 100 hPa.



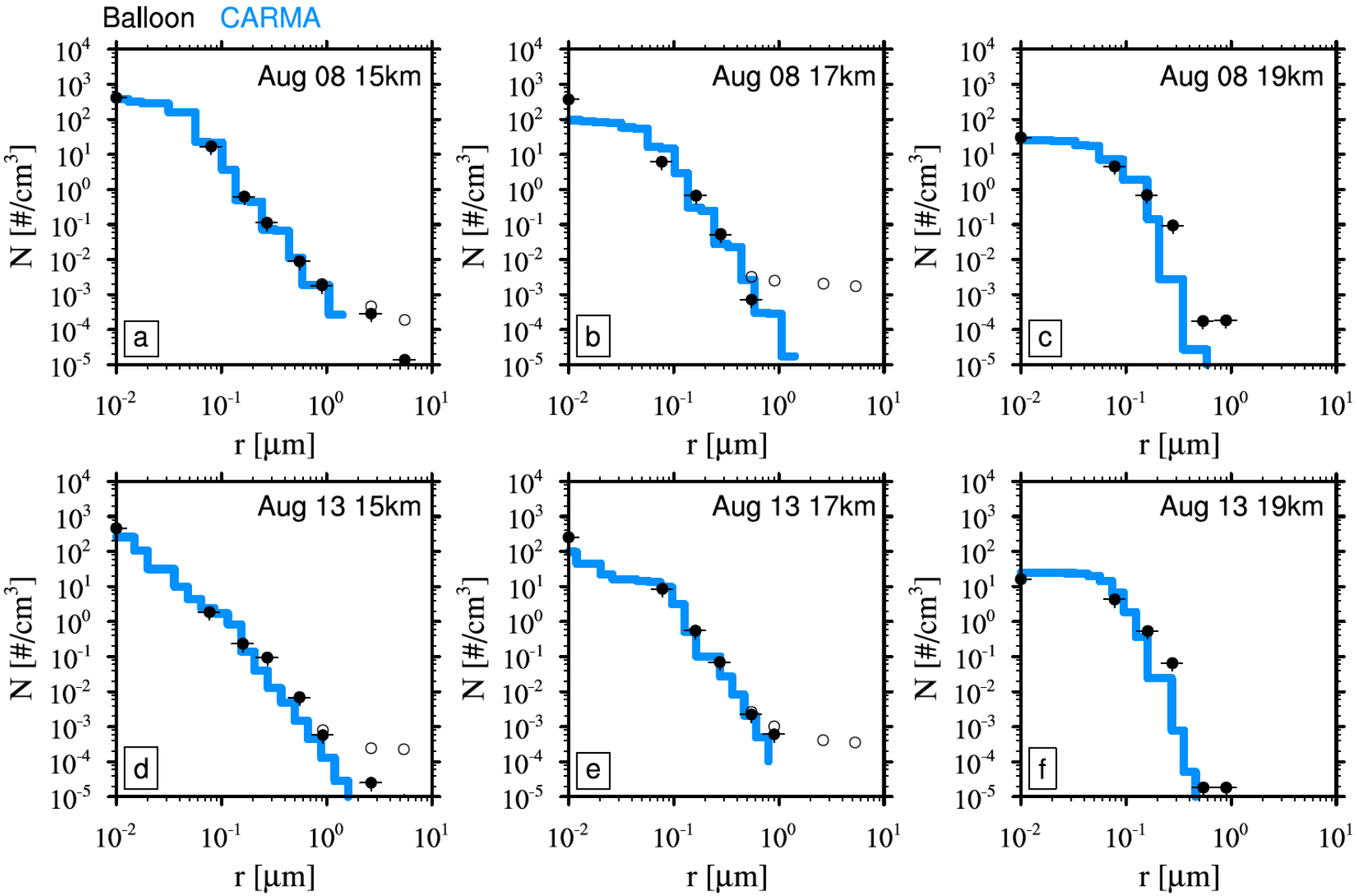
# Secondary aerosols (Sulfate, Organics and Ammonium nitrate) are major contributors to ATAL.



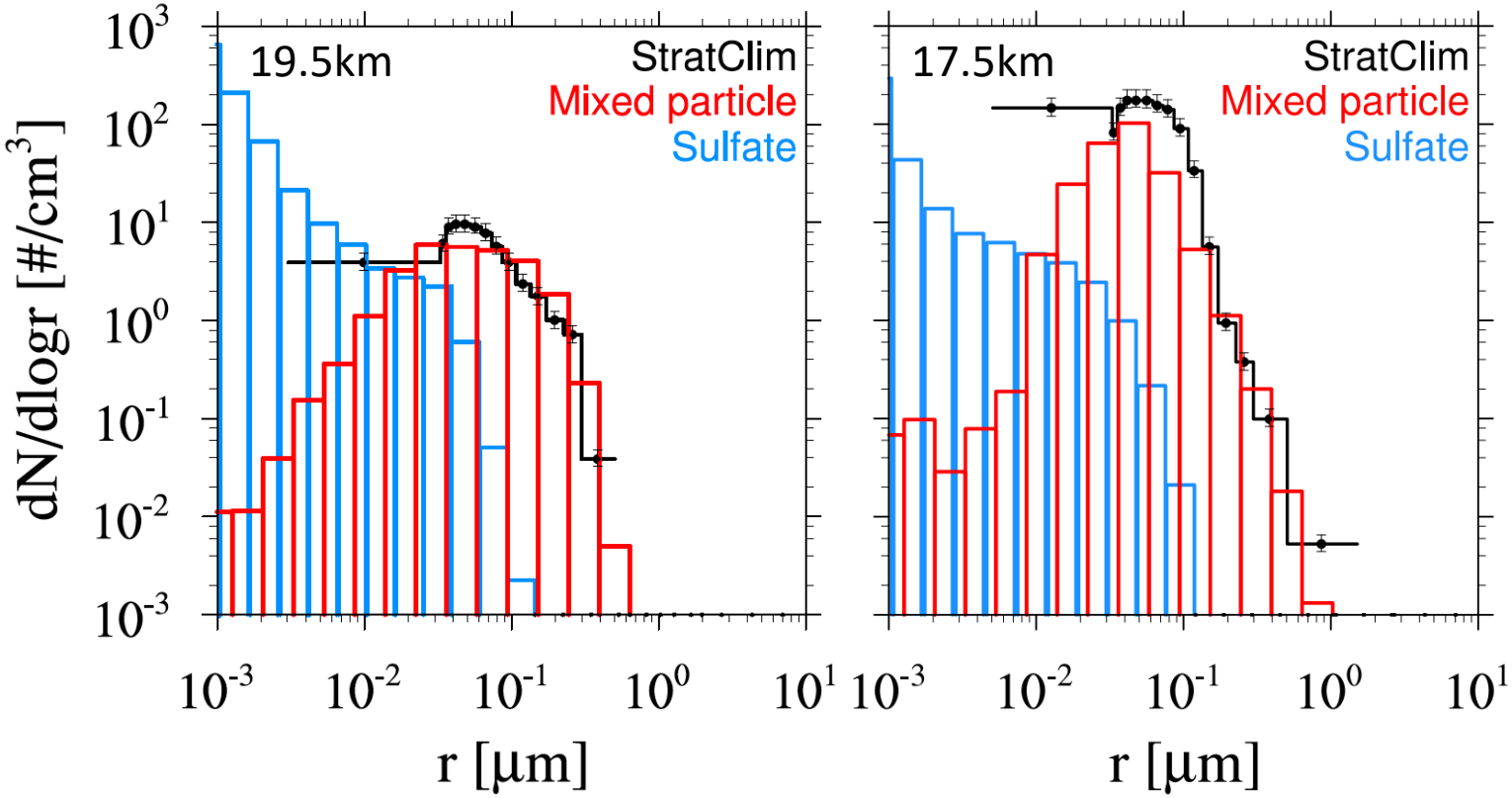

**Nitrate aerosol at ASM UTLS:**

1. **Ammonium nitrate forms in the UTLS (major)**
2. **$HNO_3$ condenses due to cold temperatures (moderate)**
3. **Ammonium nitrate is transported to the UTLS from the surface (minor)**

# Modeled size distribution in 2015 is consistent with BATAL balloon observations.

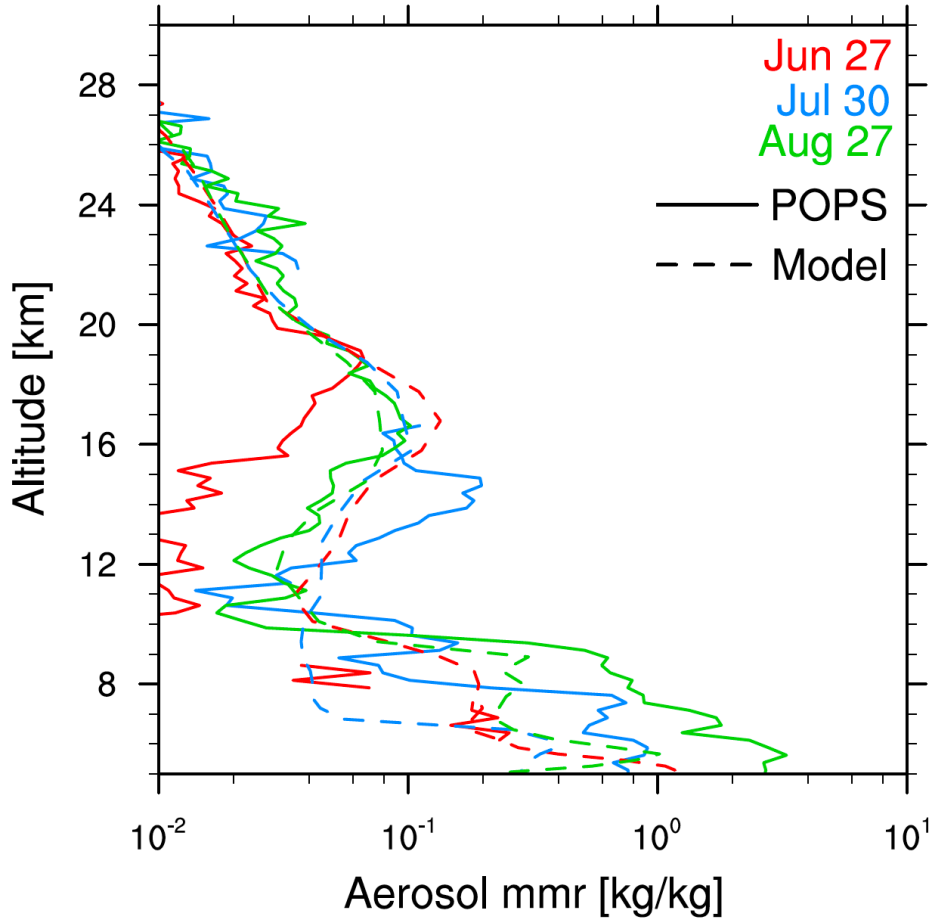


# Adding nitrate growth is important to reproduce the size distribution during the StratoClim campaign.

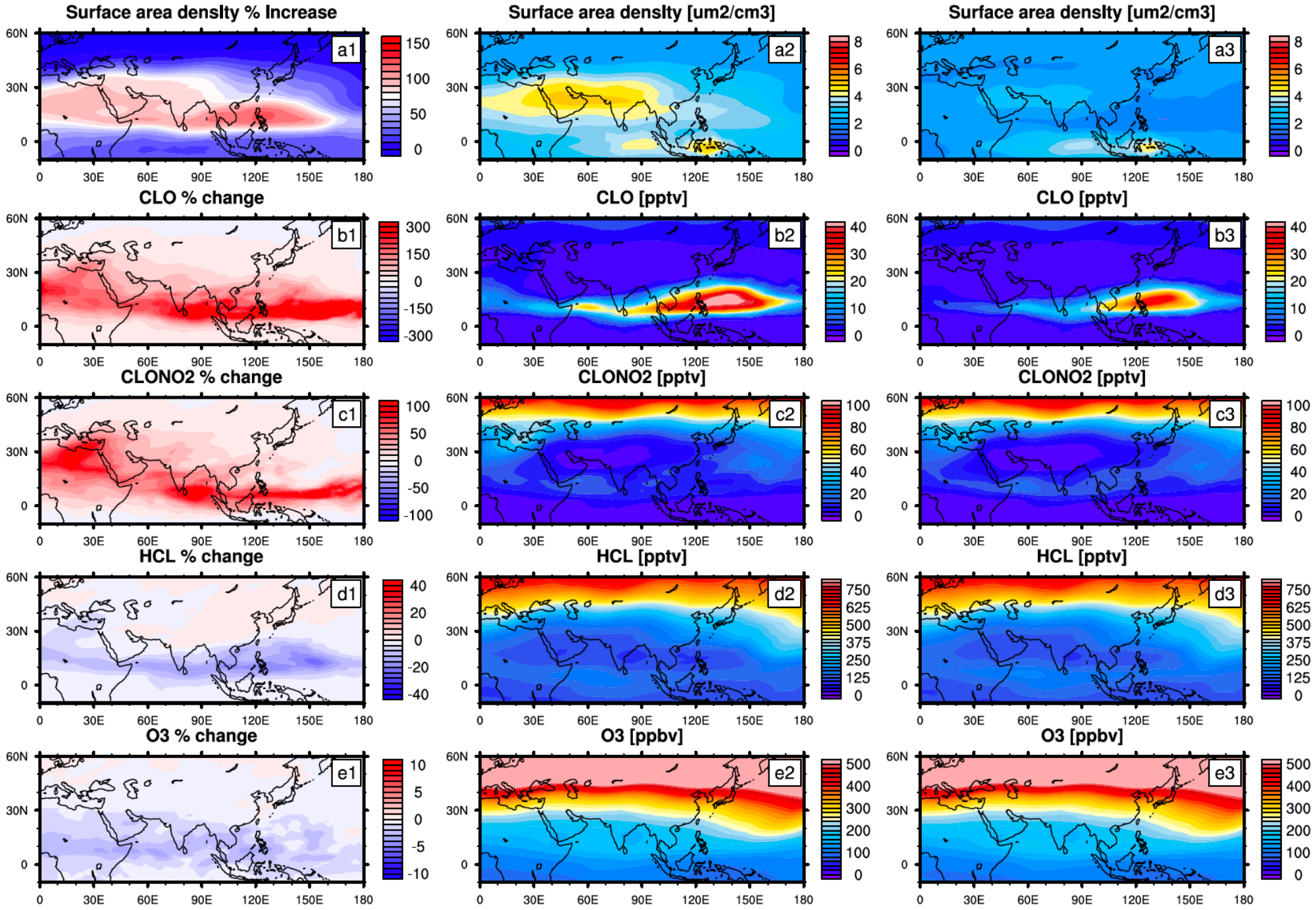



StratoClim Obs (Mahnke et al., 2020)

During the Golmud 2020 campaign, particle mixing ratio is consistent at ATAL region(15-20 km), although the POPS observation shows variations of ATAL altitudes in these three days while the model has ATAL at a consistent altitude.



Including nitrate containing aerosol in the model increases the surface area density at UTLS and changes the partitioning of chlorine.



# Conclusion

- Nitrate aerosol in the stratosphere mainly consists of ammonium nitrate formed locally by ammonium neutralization and nitric acid formed by vapor condensation at the cold tropopause.

- For the ASM region, nitrate, organics, and sulfate are the three major contributors to the aerosol composition by mass, while the contributions of dust and sea salt are minimal.

- Including nitrate in the model doubles the surface area density in the UTLS in the region 0-45˚N, 0-180˚E, changing the chlorine partitioning in the region.