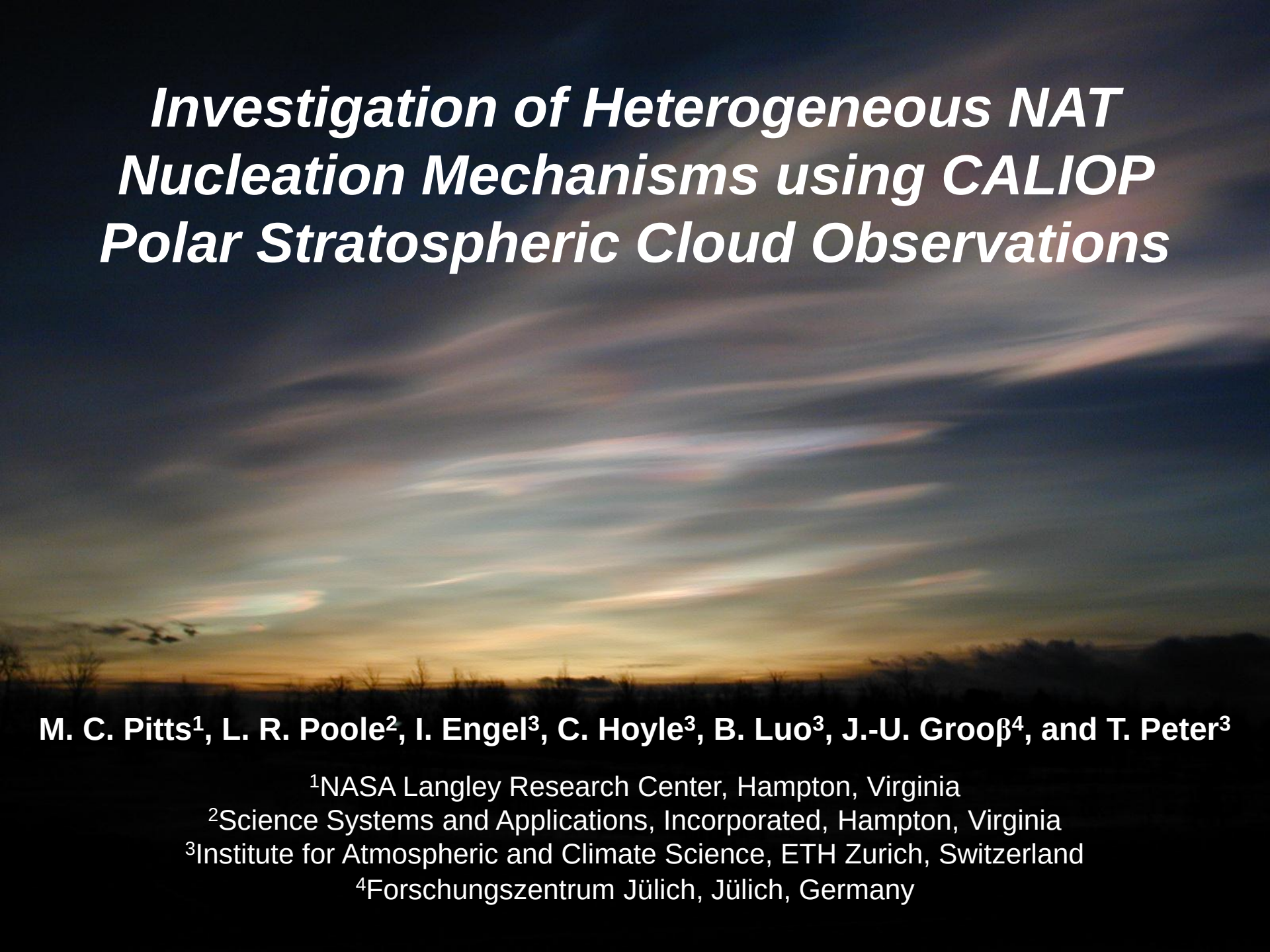

Investigation of Heterogeneous NAT Nucleation Mechanisms using CALIOP Polar Stratospheric Cloud Observations

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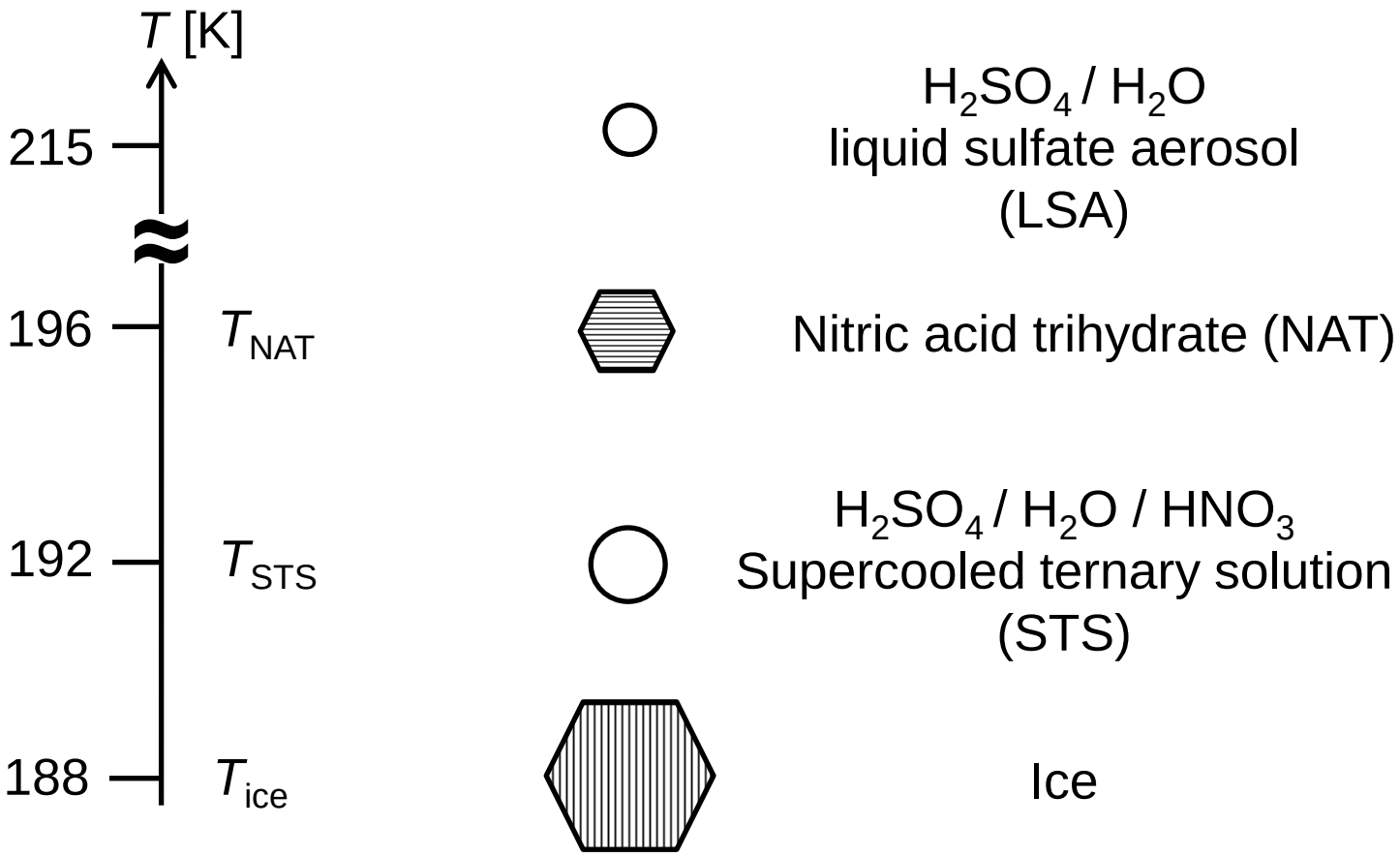
Why are we interested in polar stratospheric clouds (PSCs)?

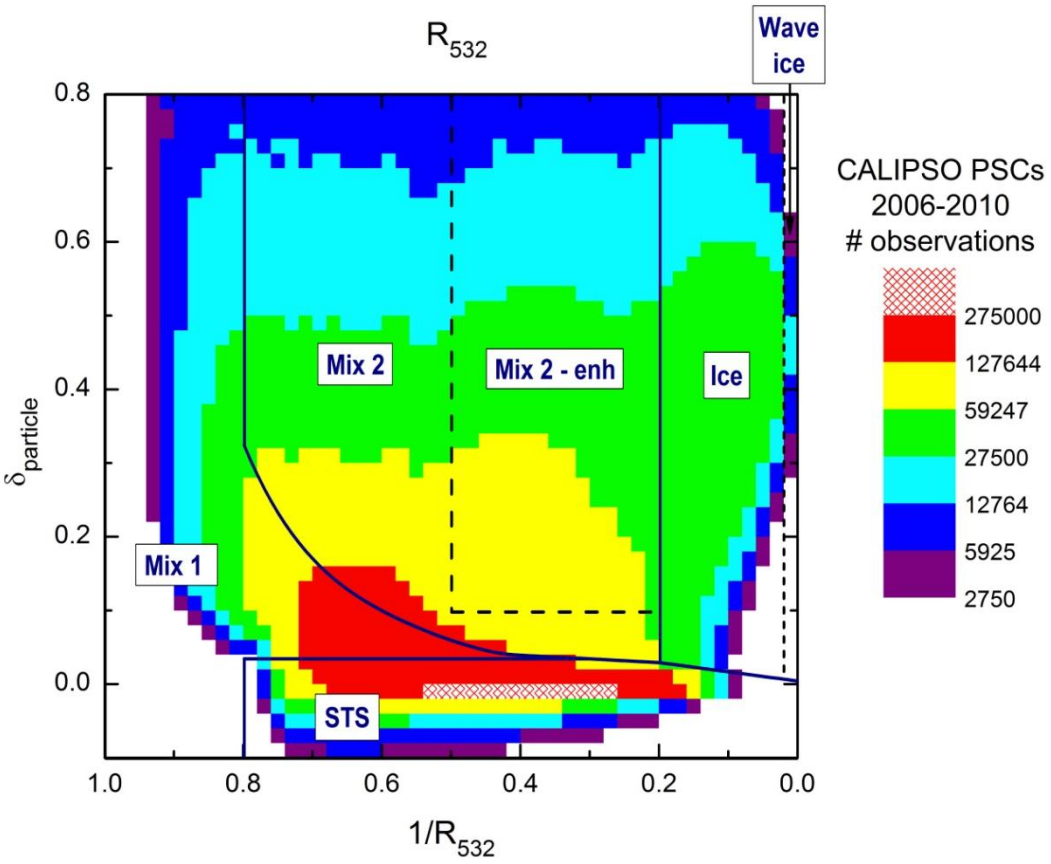
- They play major roles in polar O_3 depletion.
  a) Sites for heterogeneous reactions that activate chlorine; efficiency = f(particle composition, surface area).
  b) Large nitric acid trihydrate (NAT) PSC particles sediment and denitrify the stratosphere, prolonging O_3 depletion cycle.
- Our lack of understanding of solid particle formation (especially NAT) limits our ability to represent PSCs in global models and accurately predict O_3 recovery.
- The CALIOP dataset provides many opportunities for detailed studies of PSC formation mechanisms.

PSC Thermodynamics

50 hPa, 5 ppmv H_2O, 10 ppbv HNO_3



CALIOP PSC Composition Classes



Composite 2-D histogram of all CALIOP PSC observations from 2006-2010 in the aerosol depolarization ratio ($\delta_{particle}$) vs. inverse scattering ratio ($1/R_{532}$) coordinate system (Pitts et al., 2011)

STS: Supercooled ternary solution droplets (~ 10 particles per cm^3)

Mix 1: Liquid + low number density NAT (< 10^{-3} particles/cm^3)

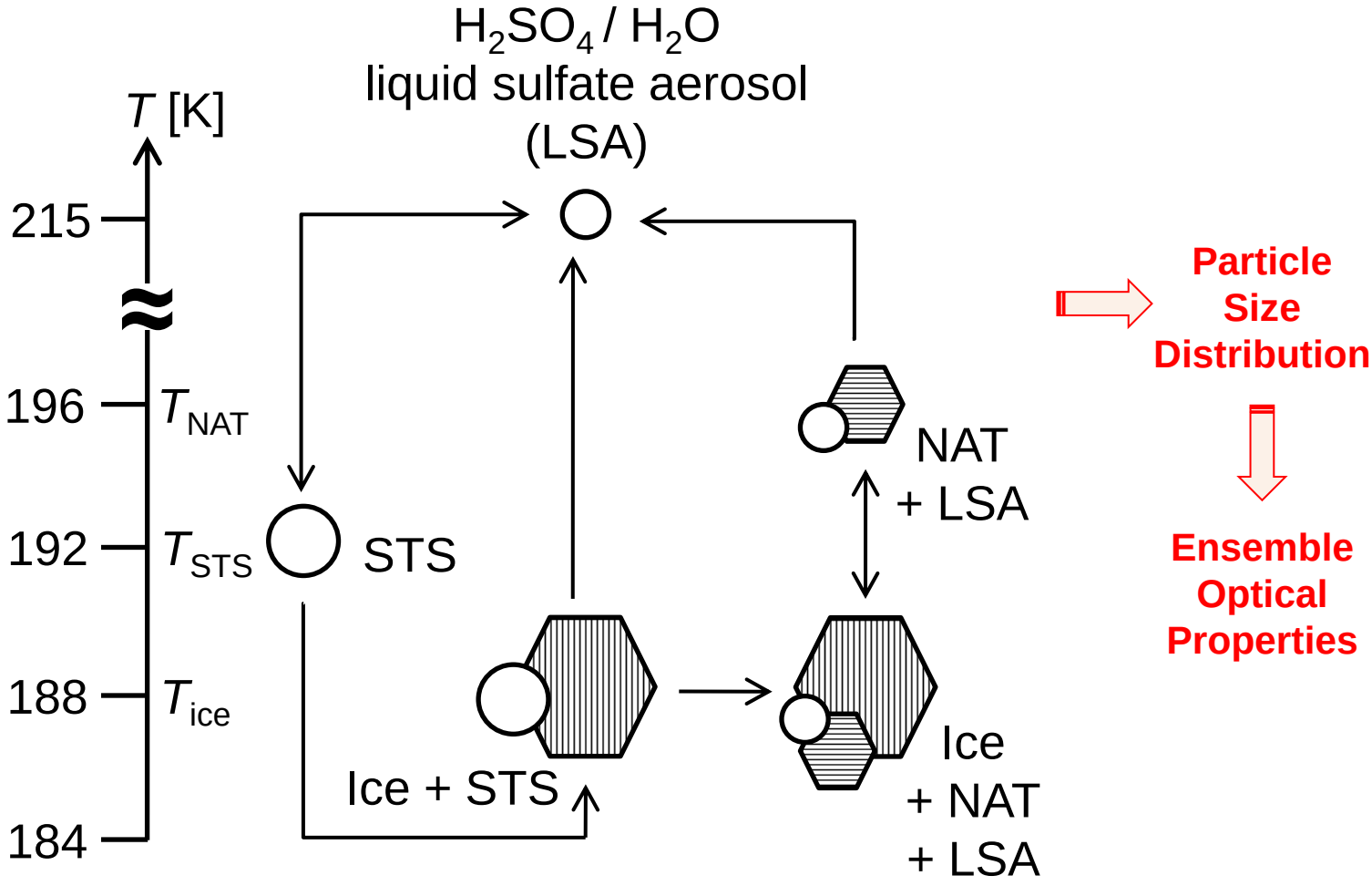
Mix 2: Liquid + intermediate number density NAT (10^{-3} – 10^{-1} particles/cm^3)

Mix 2-enh: Liquid + high number density NAT (~10^{-1} particles/cm^3)

Ice: Water ice (10^{-3} – 10^{-1} particles/cm^3)

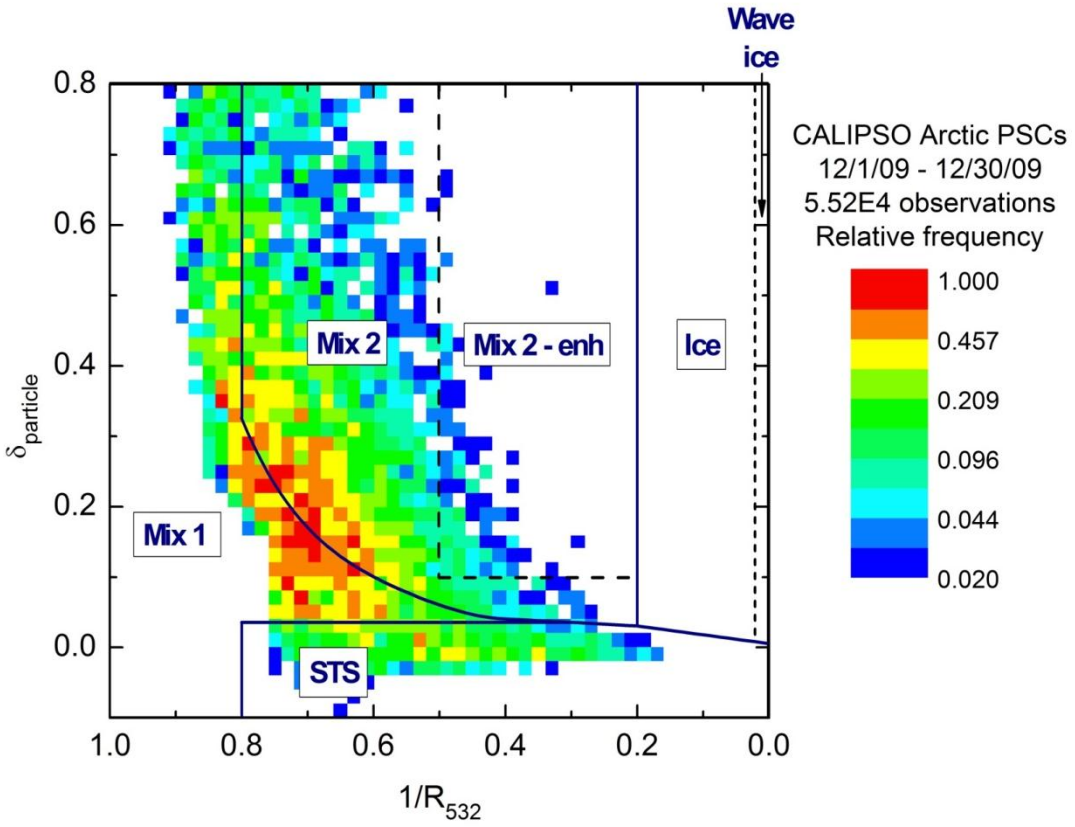
Wave Ice: Intense mountain-wave induced ice clouds (~10 particles/cm^3; 1-1.5 μm radius)

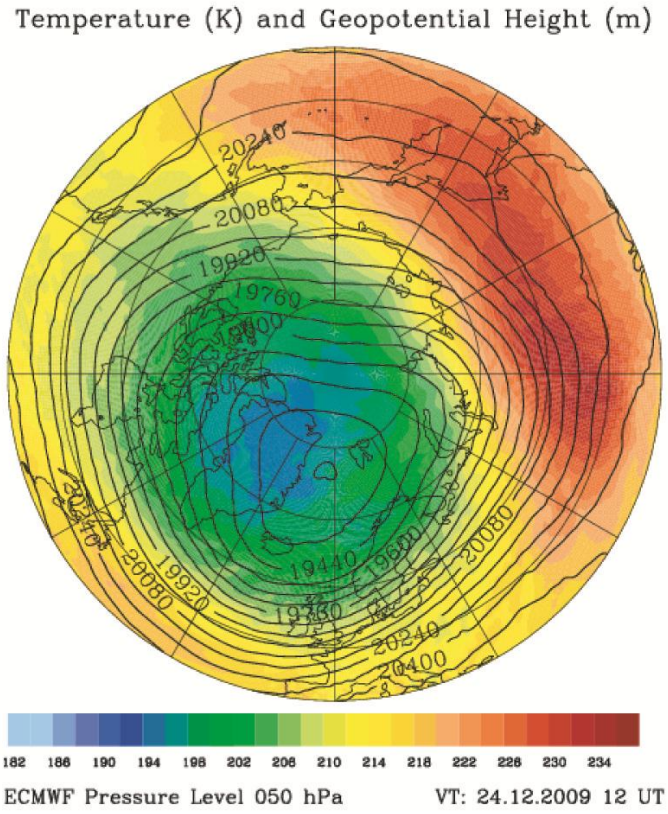
PSC Microphysical Modeling: Conventional Wisdom



CALIPSO Arctic PSC Observations 15-30 December 2009

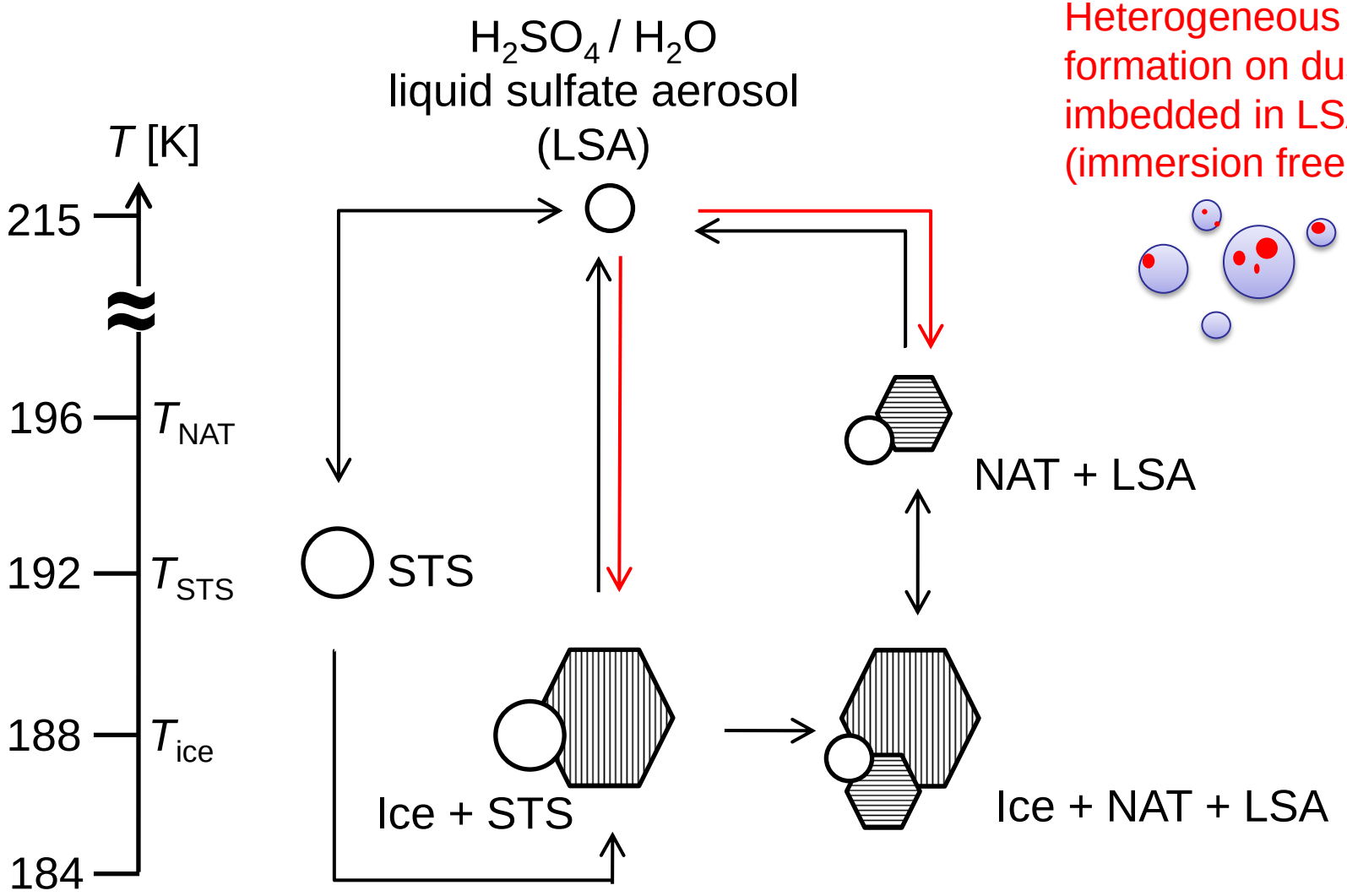
→ NAT observed before ice was present



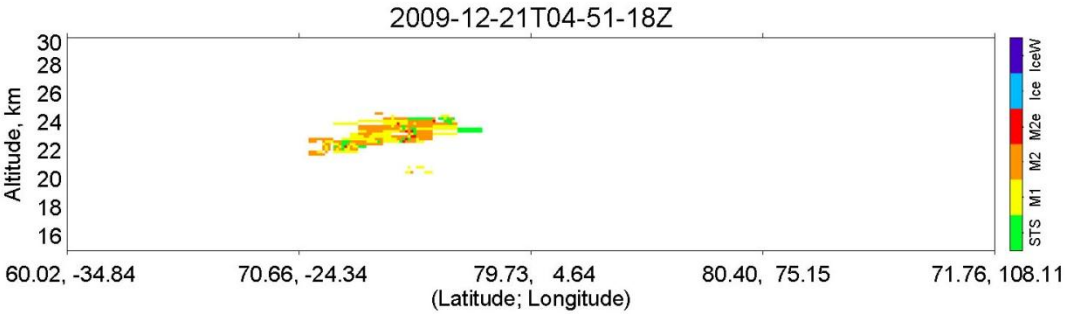


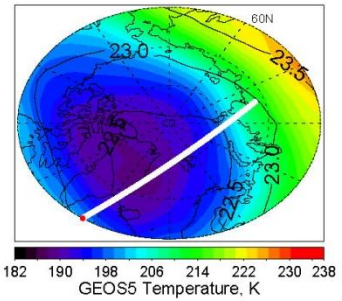
From Pitts et al. (2011)

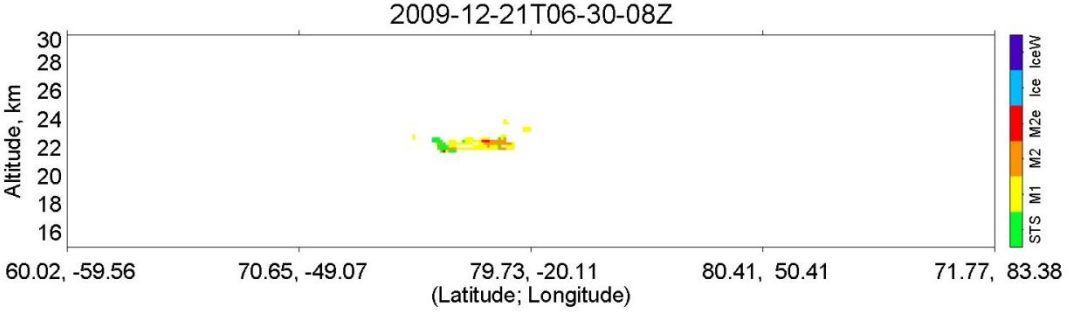
Modified ETH PSC Microphysical Model

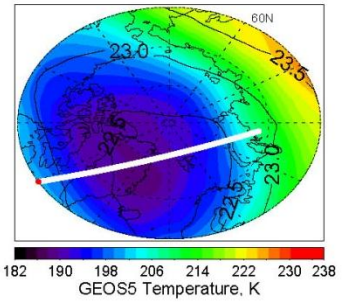


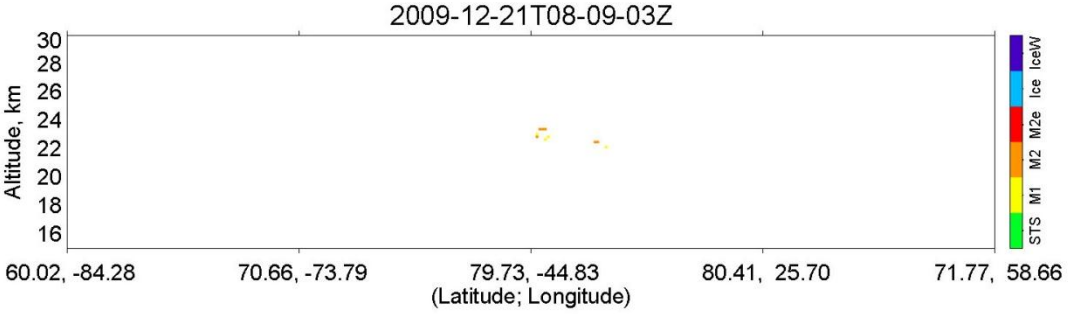
CALIPSO PSC Composition Examples: 21 December 2009

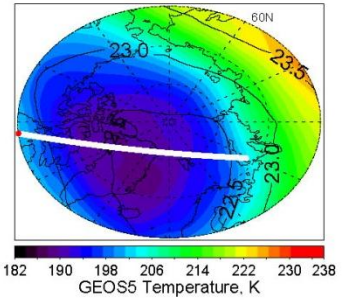




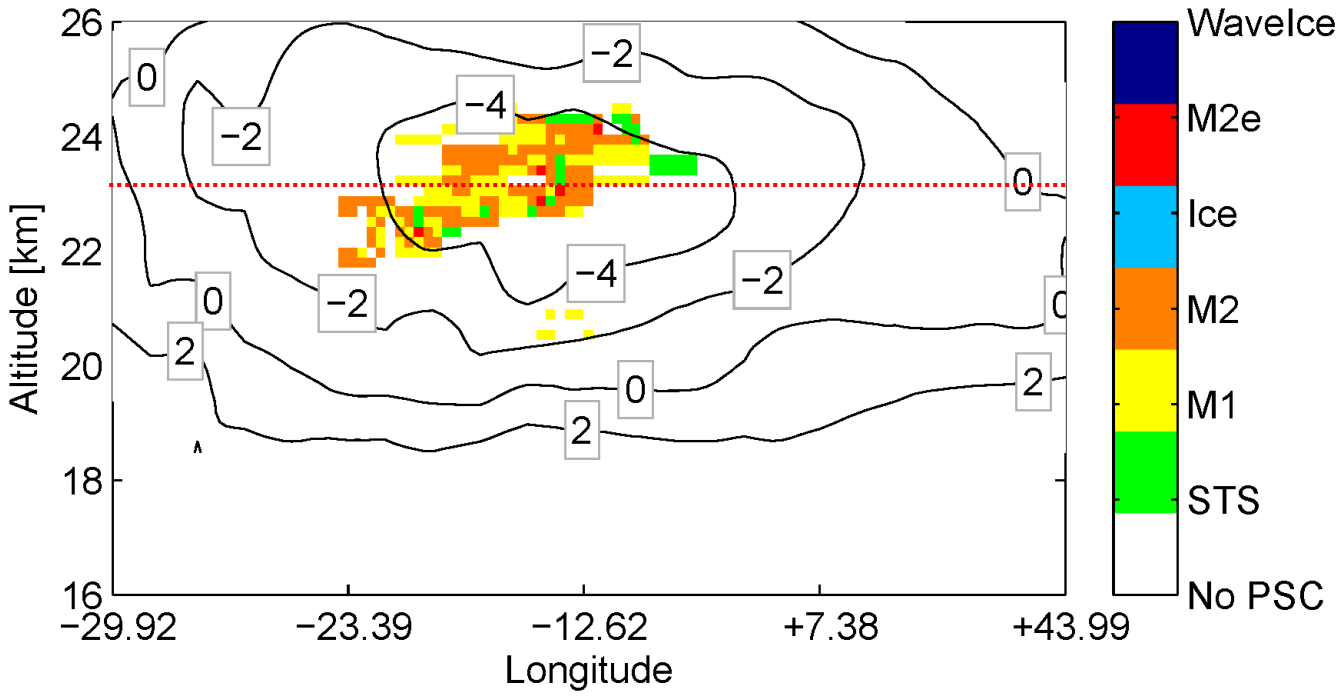


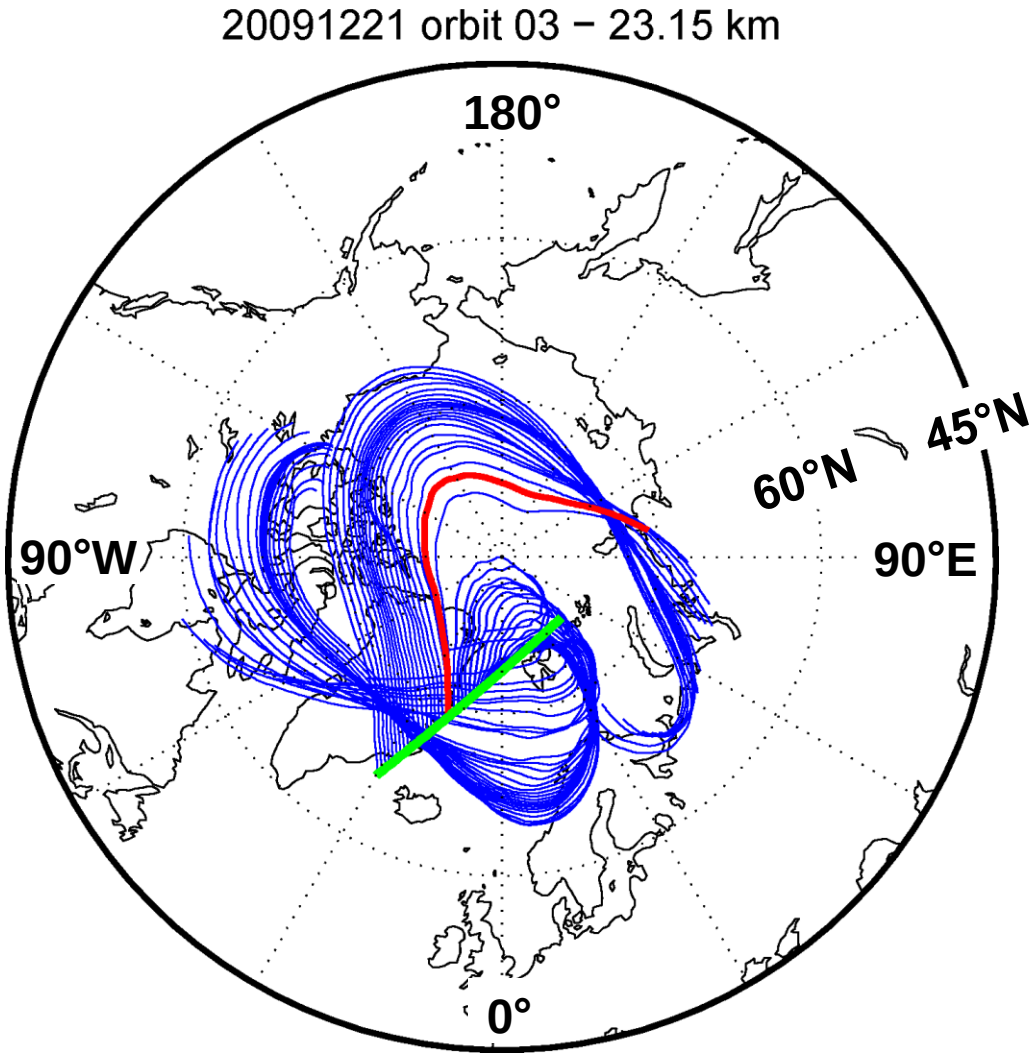




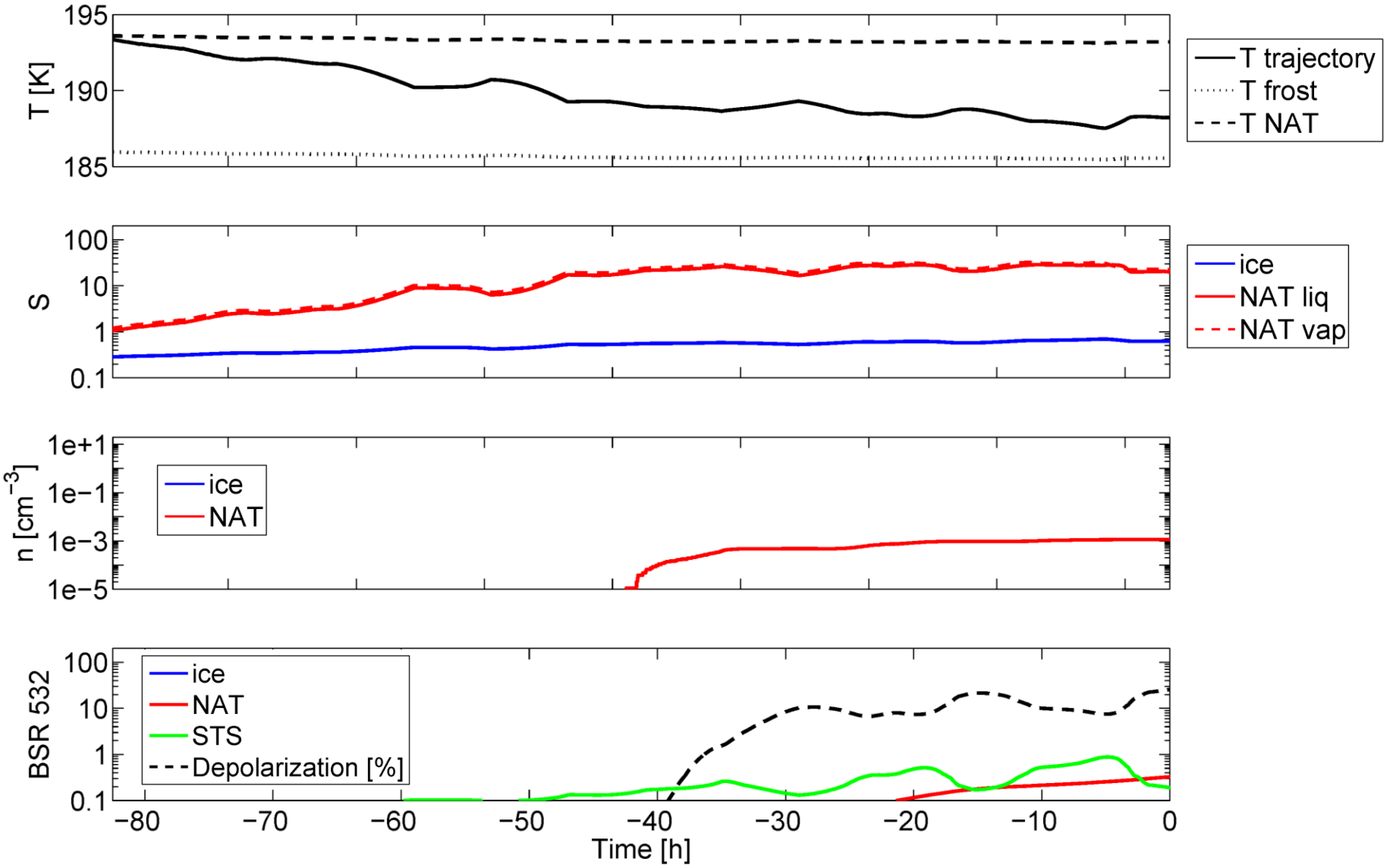


21 December 2009
CALIPSO composition classification and ECMWF $T-T_{NAT}$

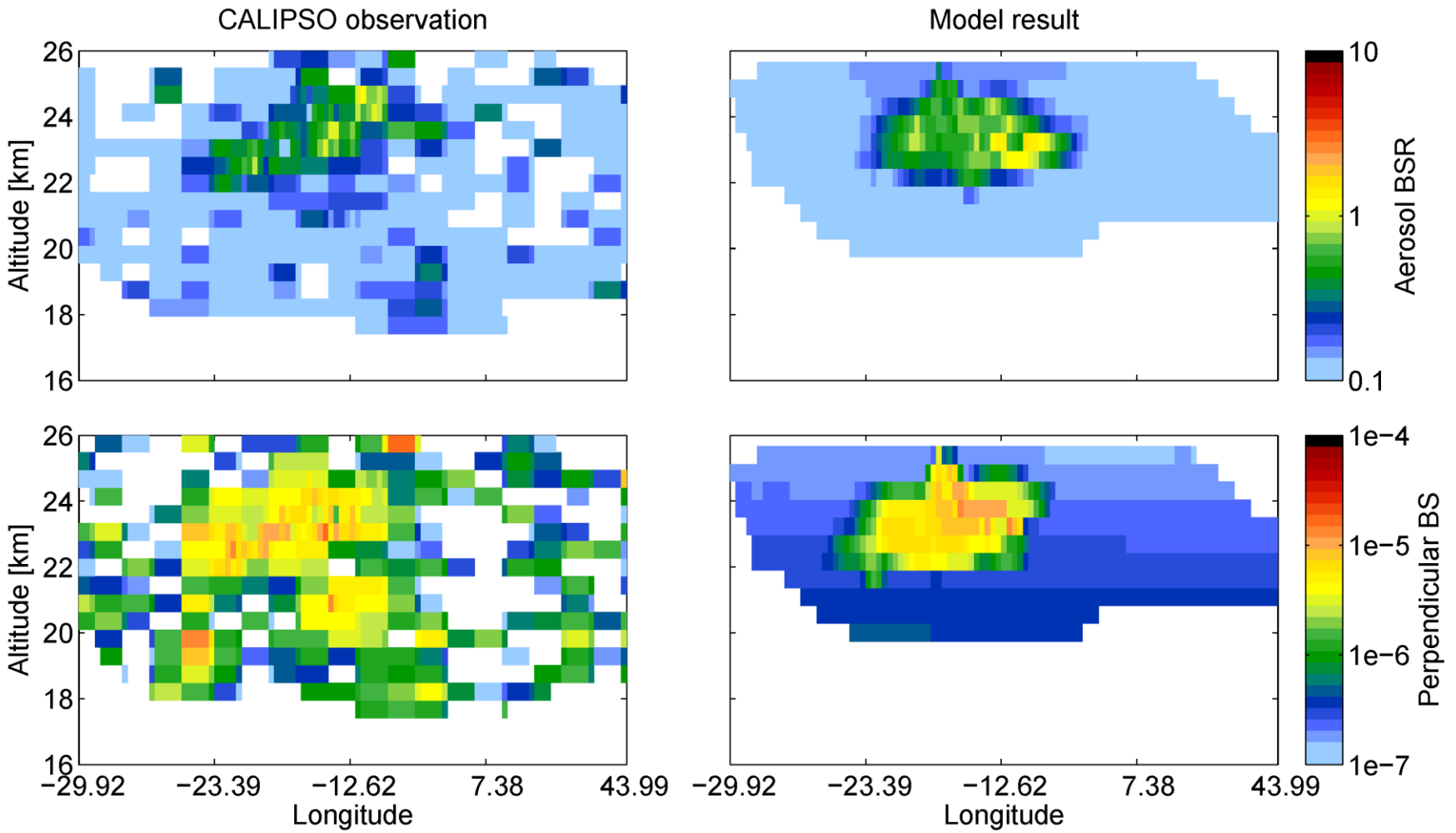



20091221 orbit 03 − 23.15 km
180°
45°N
60°N
90°W
90°E
0°

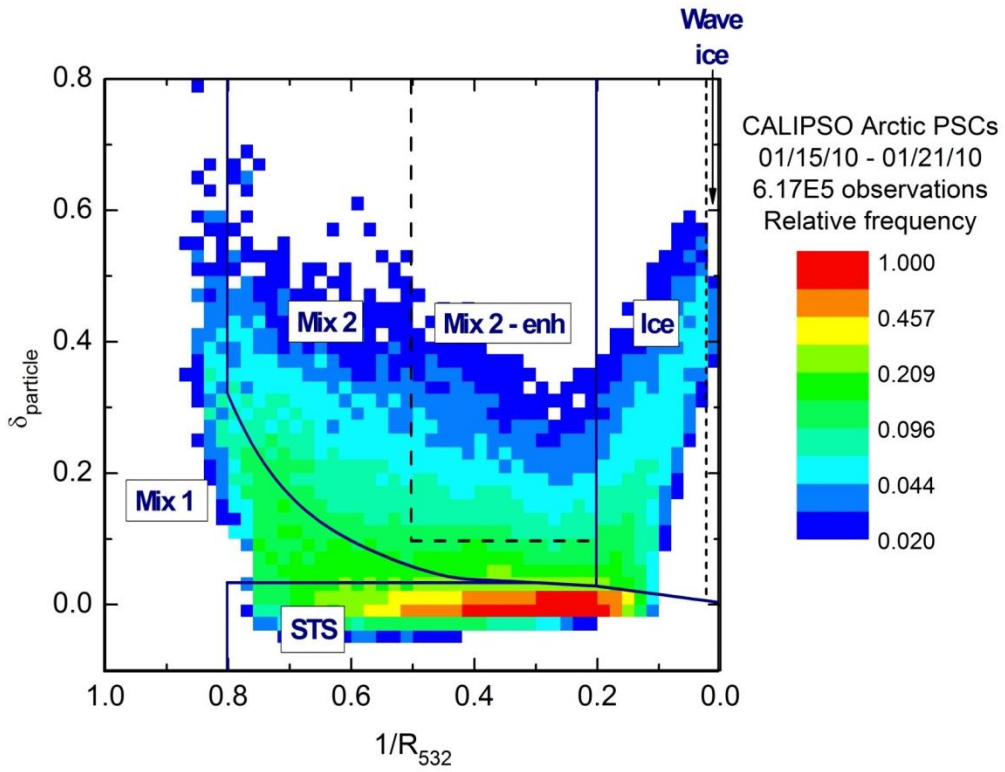
Sample Model Results

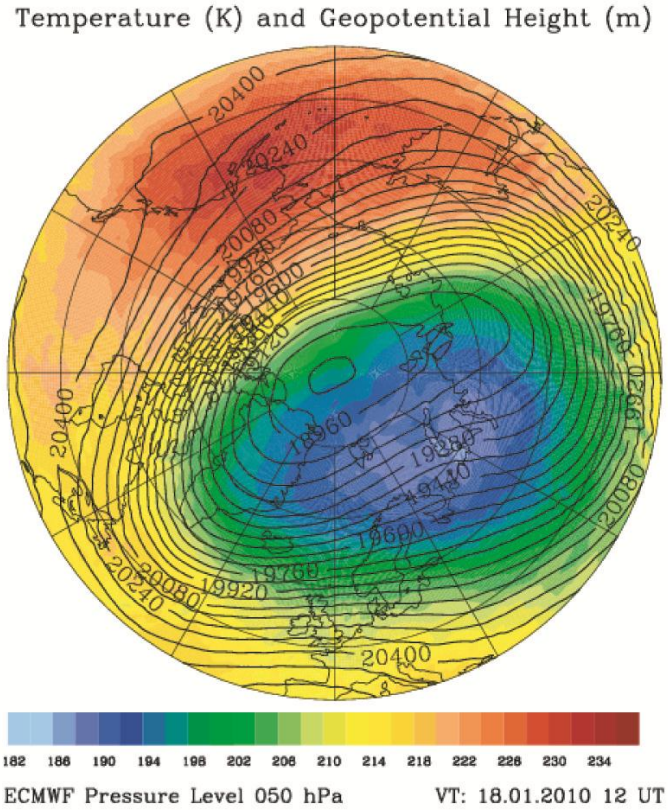


21 December 2009
CALIPSO observations ↔ model result



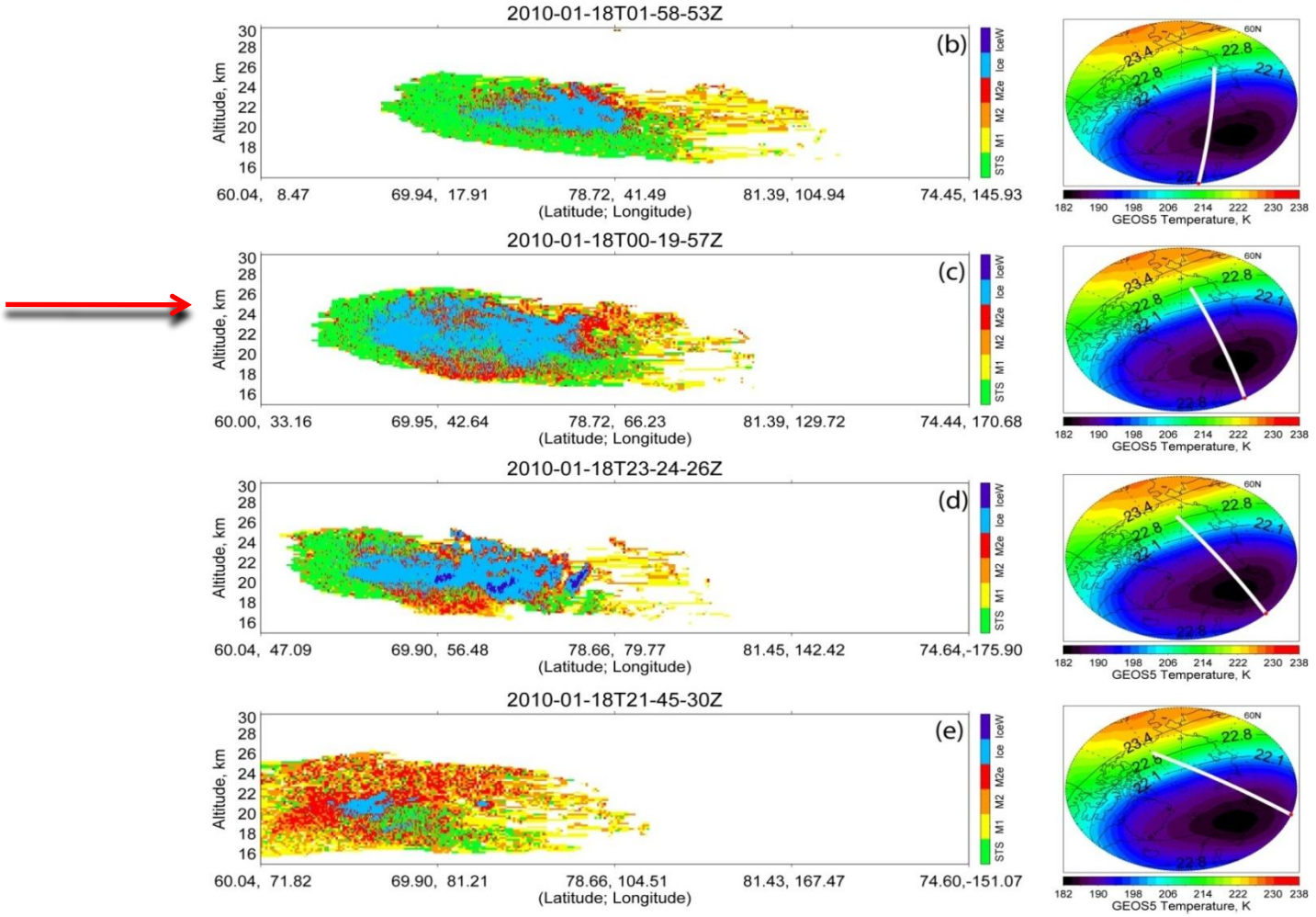
CALIPSO Arctic PSC Observations
15-21 January 2010



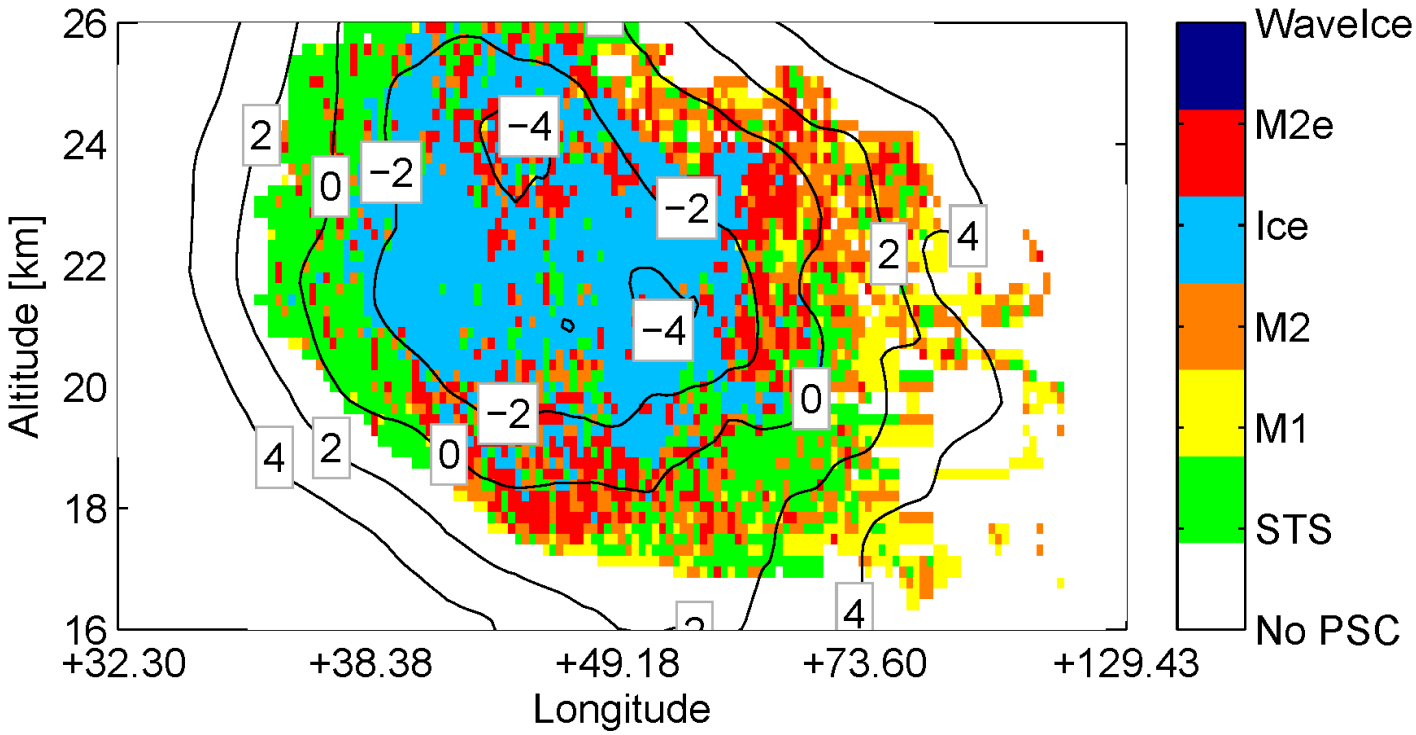


- ***Extensive synoptic-scale ice PSCs***
- ***Smaller fraction of Mix 2 and Mix 2-enh PSCs***
- ***Nucleation of synoptic-scale ice PSCs on pre-existing NAT and dust imbedded in LSA***

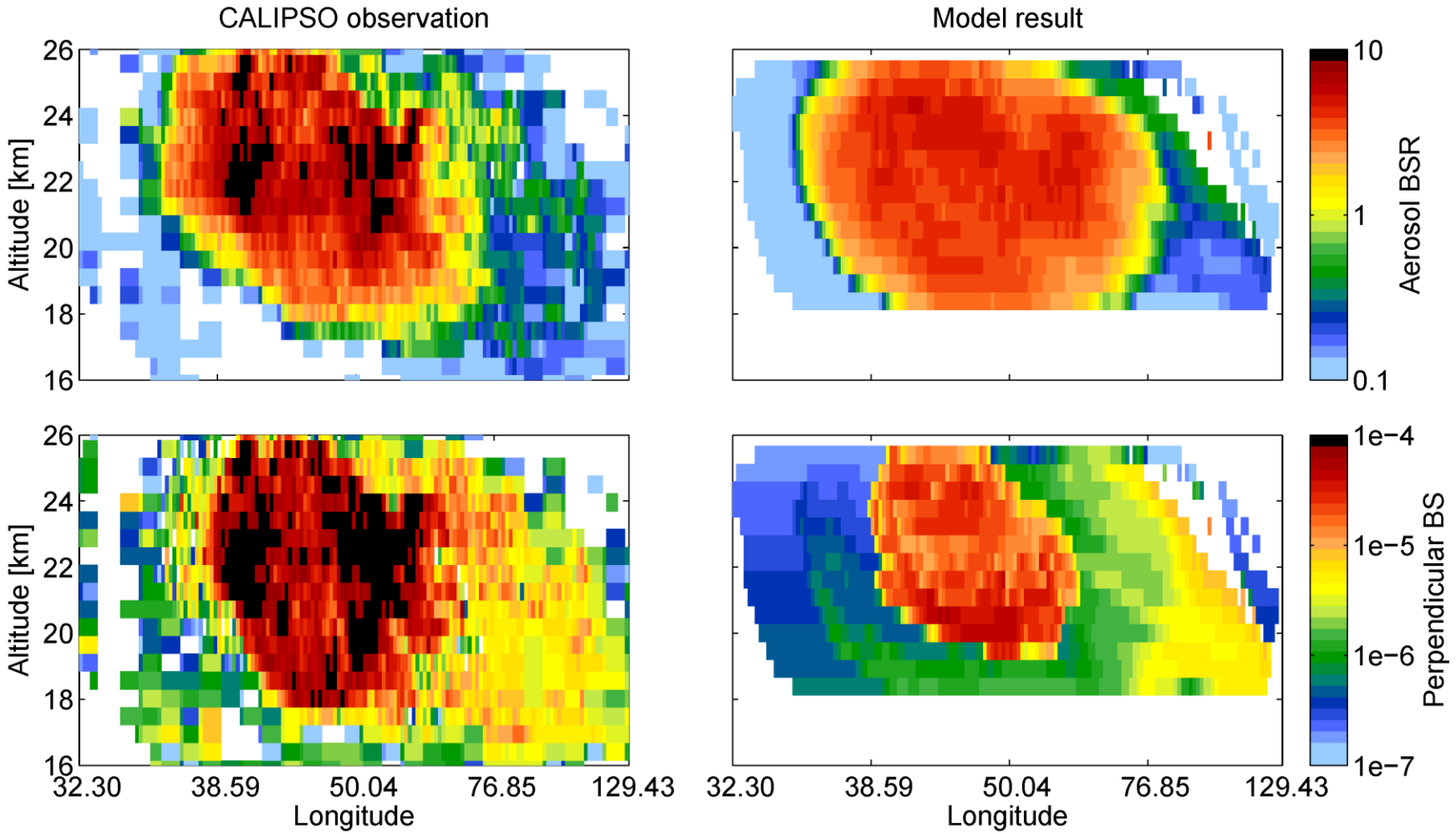
CALIPSO PSC Composition Examples: 18 January 2010



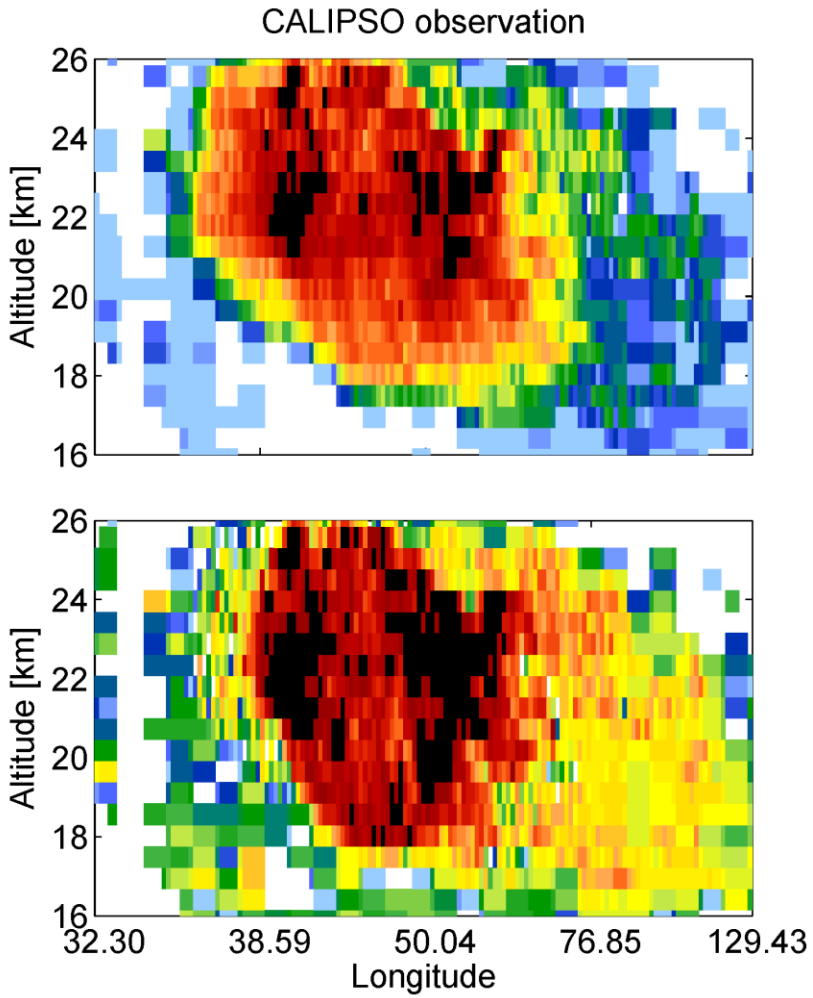
18 January 2010
CALIPSO composition classification and ECMWF T-T_{ice}



18 January 2010
CALIPSO observations ↔ model result



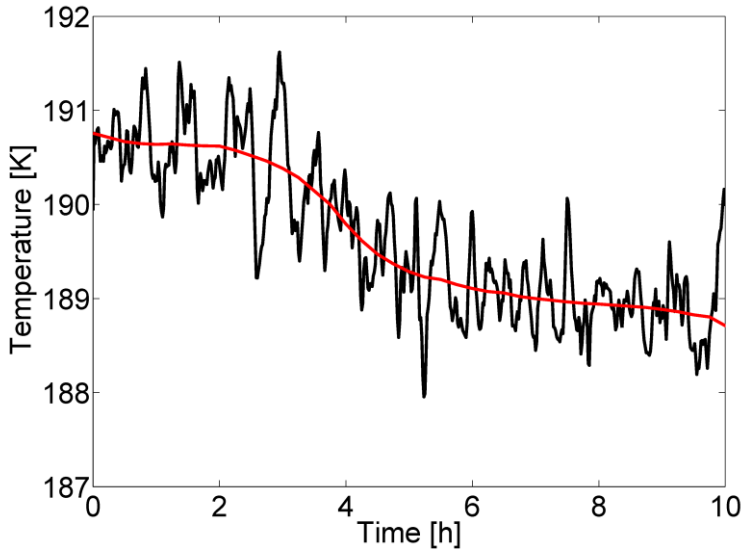
18 January 2010
CALIPSO observations ↔ model result



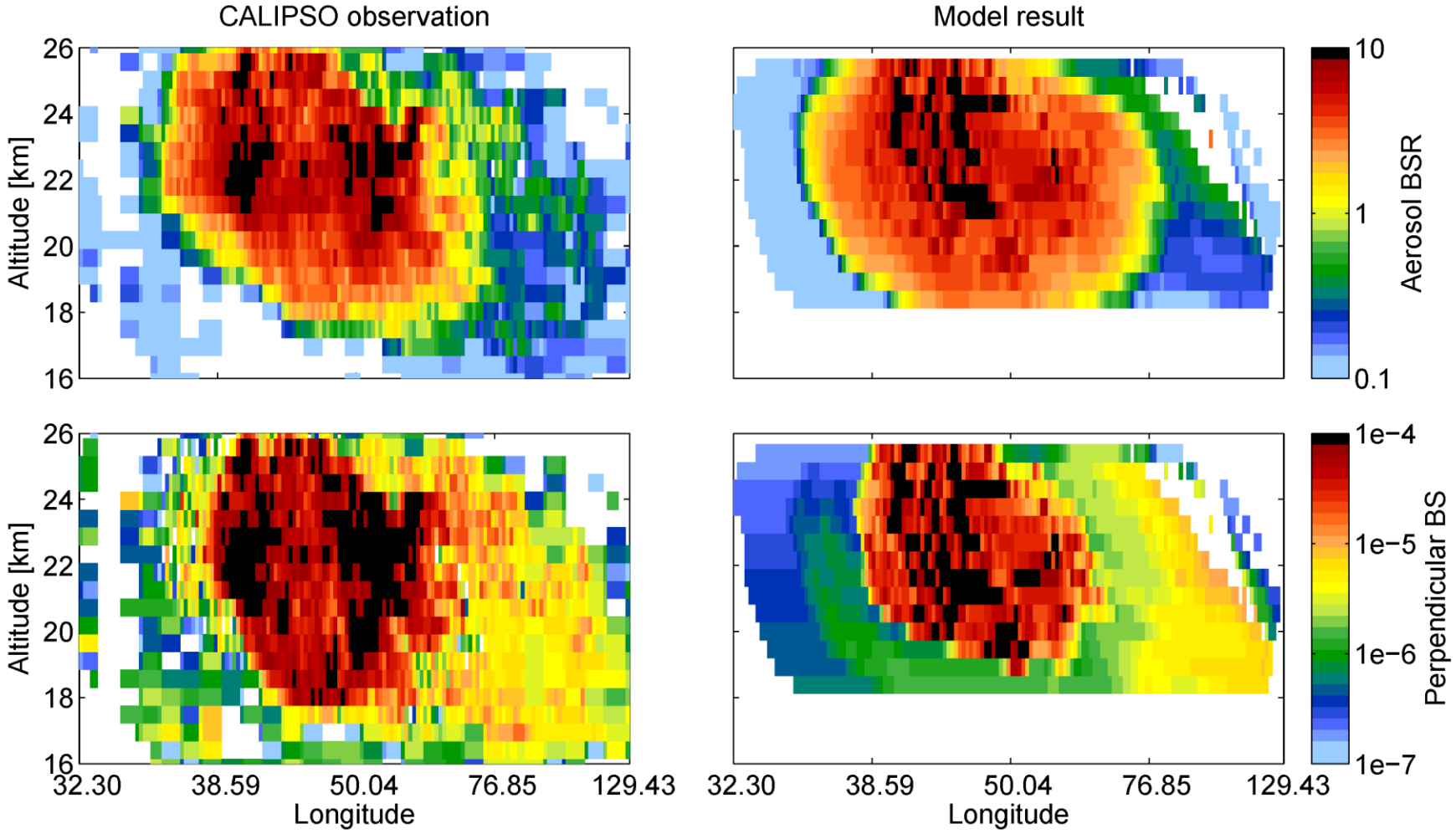
Improve model result:

ECMWF might not provide sufficient cooling rates

→ Add small scale temperature fluctuations to ECMWF trajectories (Hoyle et al., 2005)



18 January 2010 CALIPSO observations ↔ model result incl. small scale temperature fluctuations



Conclusions

Heterogeneous nucleation of …

- NAT on pre-existing ice cannot explain CALIPSO observations in December 2009
- NAT on dust could explain CALIPSO observations in December 2009
- Ice (synoptic scale) on dust and NAT could explain CALIPSO observations in January 2010
  - Small scale temperature fluctuations appear necessary to capture small scale variability in data.