What controls the Low Ice Number Concentration in the Upper Tropical Troposphere?

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Motivation

- High super saturation and low number of ice crystals (<100#/L) were frequently found near the tropical tropopause layer (e.g. Krämer et al. 2009, Jensen et al. 2013)
- Low ice number densities are inconsistent with models of cirrus cloud formation involving homogeneous freezing of liquid aerosols.
- Possible explanations of the low ice number
  1. Suppression of homogeneous freezing by glassy organic aerosols/solid ammonium sulfate [Murray et al., 2010;Jensen et al., 2010]
  2. Pre-existing ice [Kuebbeler et al., 2014, Shi et al. 2014]

  3. Dynamic equilibrium [Barahona and Nenes, 2011]
  4. Special dynamic condition: slow large-scale ascending + fast gravity wave [Spichtinger and Krämer, 2012]

The last two proposals does not require the suppression of homogeneous freezing!

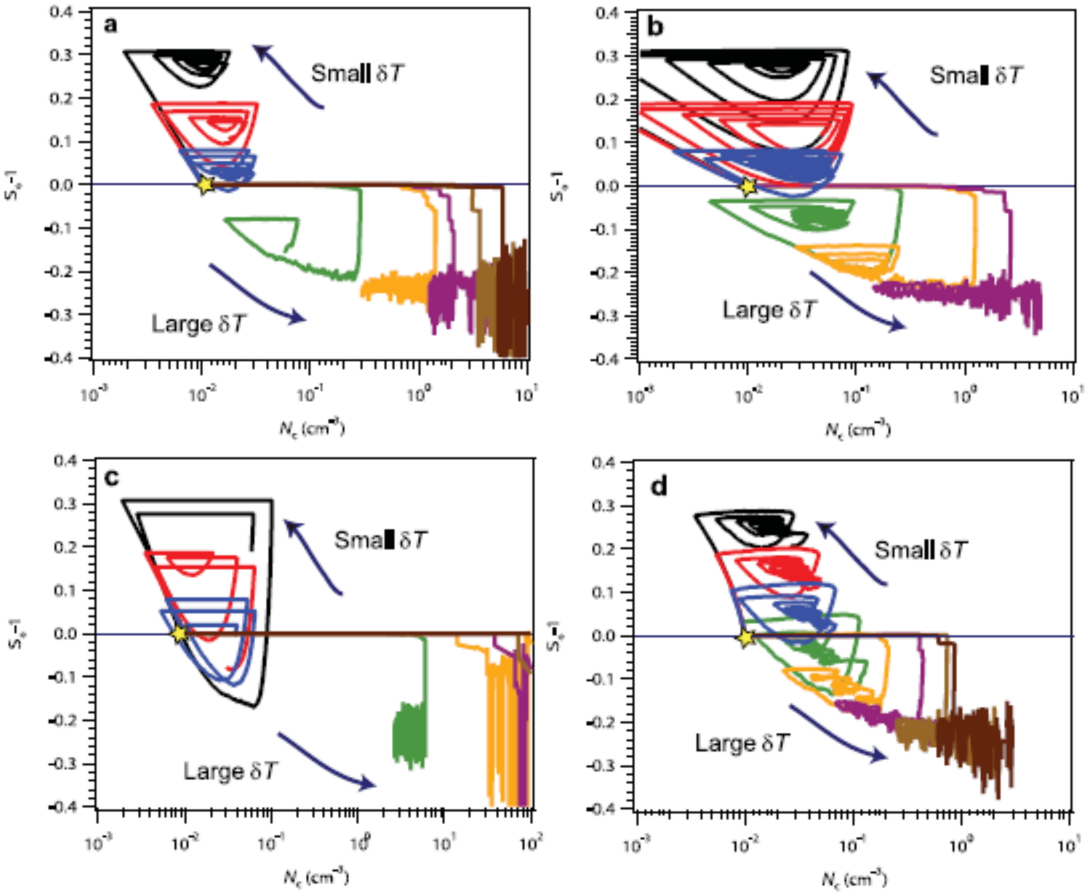


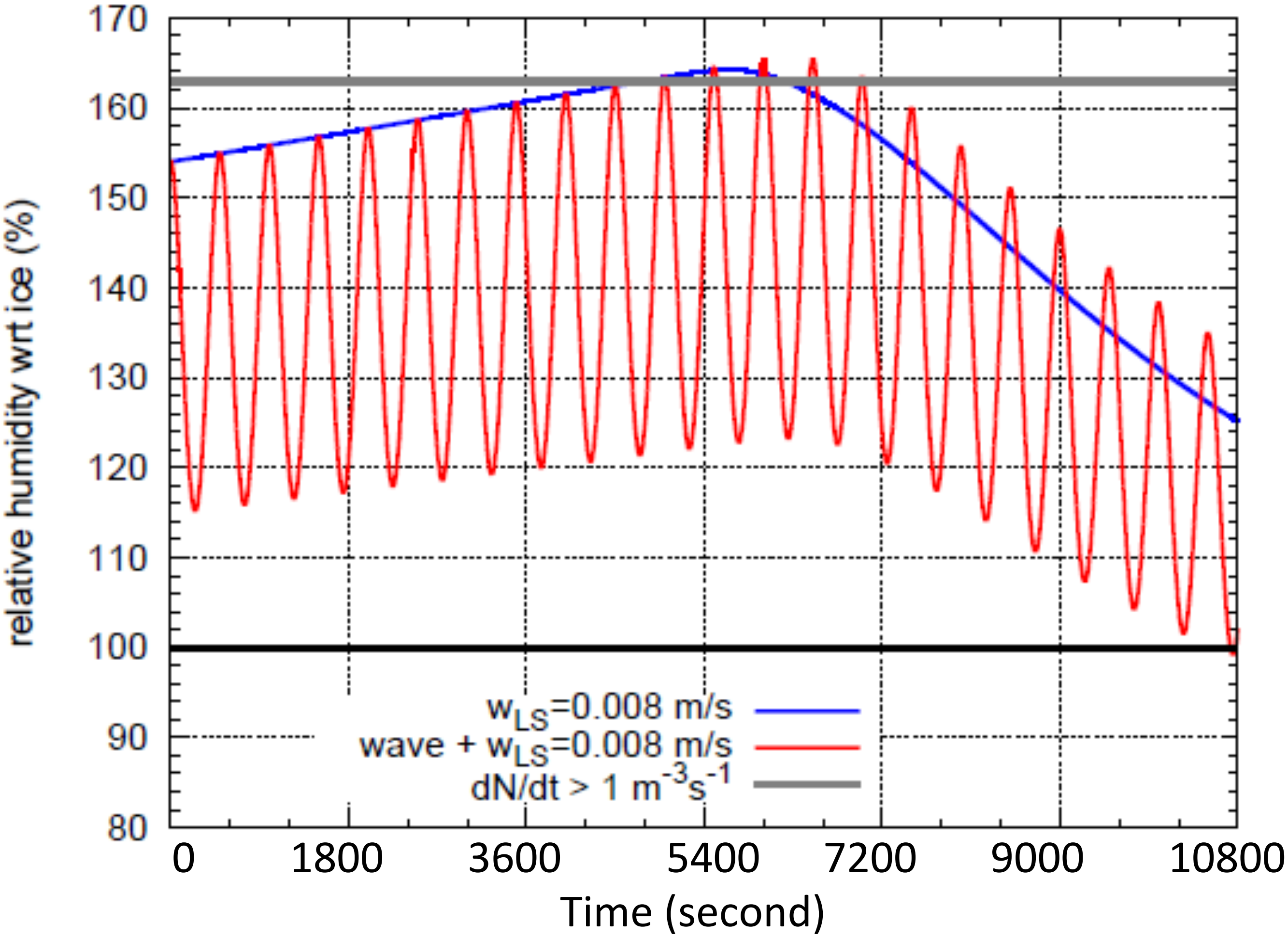
Fig. 6. Sensitivity of N_c and S_o evolution to cloud formation conditions for different values of δT (color scheme same as in Fig. 4); **(a)** same conditions as in Fig. 4, **(b)** cloud thickness, $H = 100\,\mathrm{m}$ (increased ice crystal removal rate), **(c)** deposition coefficient equal to 0.006 (Magee et al., 2006) (slow water vapor transfer), and **(d)** initial temperature 225 K and cloud lifting at 5 cm s^{-1}. The yellow star in each panel indicates initial conditions. The arrows indicate the temporal progression along each trajectory. The integration time was 40 h cases, except in **(d)** were it was 15 h.

"Dynamic equilibrium"
from [Barahona and Nenes, 2011]

Special dynamic condition:

slow large-scale ascending + fast gravity wave

Time evolution of RHi from Spichtinger and Krämer [2012]



Blue lines indicate constant updraft

Red lines denote superposition of large-scale updraft and a short wave.

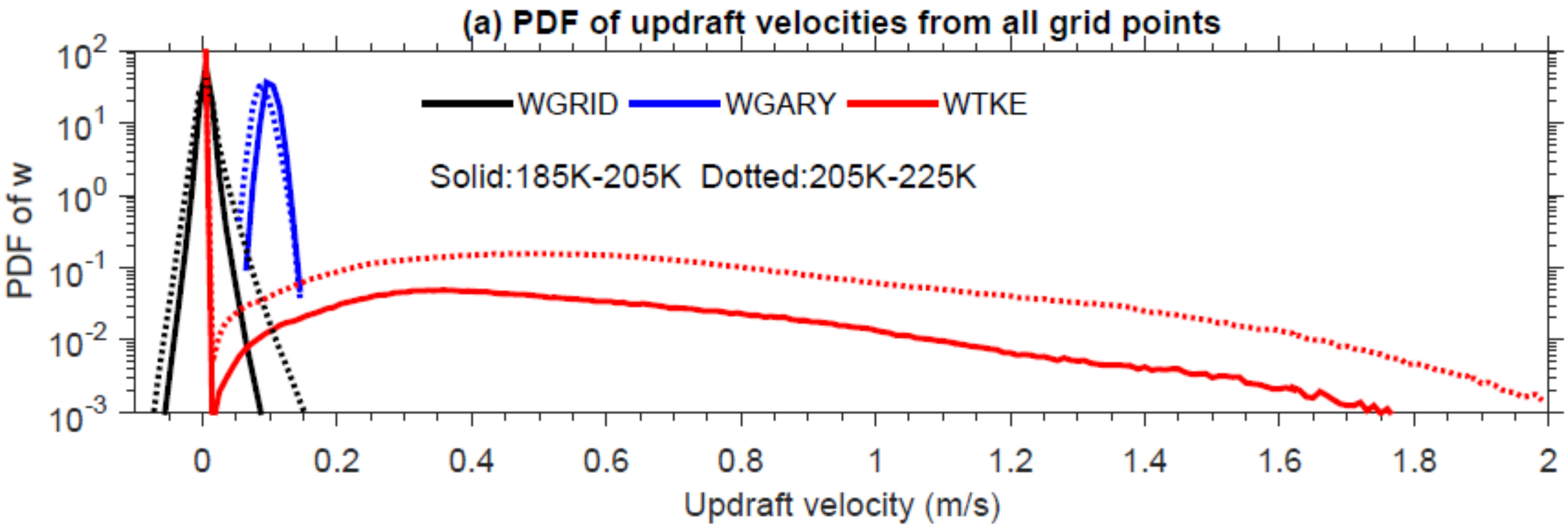
Our goal

Use a coupled CAM5/IMPACT model to study the effect from following factors on the ice number concentration in the upper troposphere.

1. Glassy organic aerosol IN (here SOA)
2. Different updraft velocities/cooling rates
3. Pre-existing ice effect
4. Different accommodation coefficients of water vapor to ice (0.1 vs. 1.0)

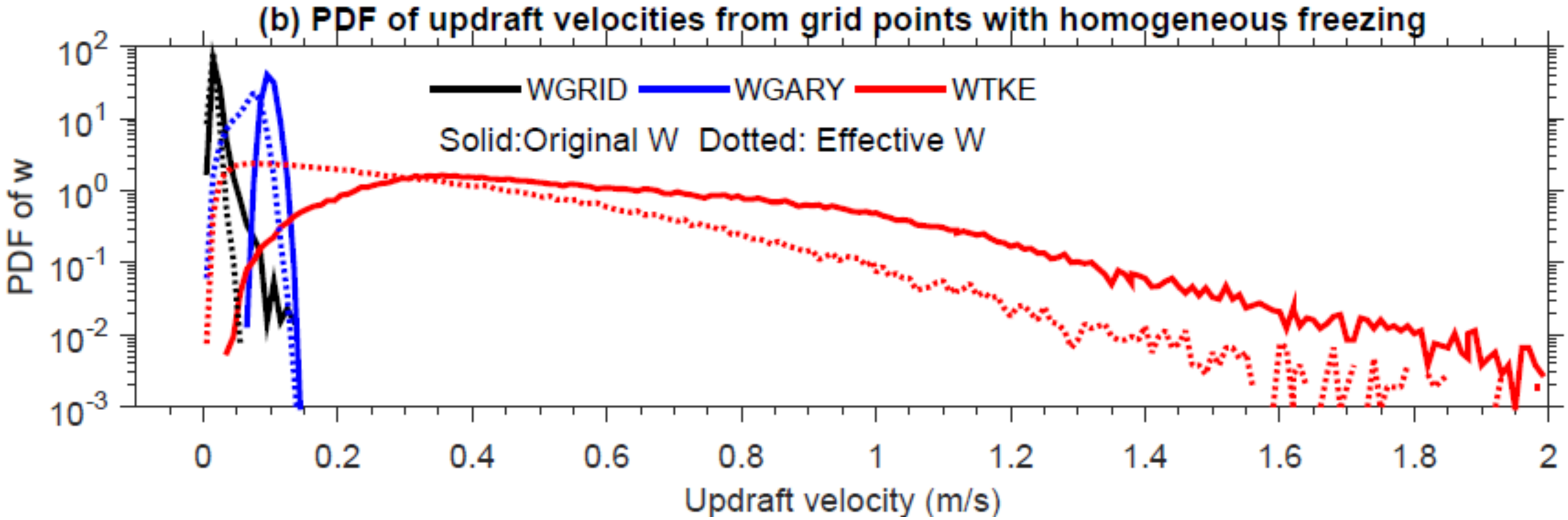
Choices of updraft velocities

1. **WGrid:** Large-scale grid resolved W
2. **WGary:** Updraft velocity based on observed temperature fluctuations (Gary 2006, 2008)
3. **WTKE:** $w_{sub} = \sqrt{\frac{2}{3}TKE}$, where TKE is the modeled subgrid turbulence kinetic energy



Pre-existing ice effect

- The updraft velocity acts to increase relative humidity by cooling the air parcel through adiabatic expansion
- The pre-existing ice particles act to decrease the relative humidity by consuming the excessive water vapor above the ice supersaturation.
- So mathematically, one could combine the two effects together. This is equivalent to have a reduced updraft velocity. This reduced updraft velocity is termed the effective updraft velocity.



Model introduction - 1

- We used the coupled **CAM5/IMPACT** model. The **IMPACT** module simulates a total of **50** aerosol types and/or size bins and ~**200** gaseous species:
  - 3 sizes representing the number and mass of pure sulfate aerosols (i.e. nucleation, Aitken and accumulation modes),
  - 3 types of fossil/bio-fuel soot that depend on its hygroscopicity or the amount of sulfate on the soot
  - 1 biomass soot mode
  - 4 dust sizes
  - 4 sea salt sizes
  - **35 Secondary Organic Aerosols (SOA)**
- Ice nucleation parameterization Baharona and Nenes [2008,2009]

Model introduction – 2
SOA formation

1. **Gas phase:** SOA formed from gas-particle partitioning of semi-volatile organic compounds together with aerosol phase reactions. For example,

$$Toluene \xrightarrow{OH} CRES \xrightarrow{OH} POXY \xrightarrow{\mathrm{NO2}} NITP \xrightarrow{NO3} R4N2\ldots$$

$$\mathrm{NITP_{gas}} \xleftrightarrow{\text{partitioning}} \mathrm{ev\_NITP_{aerosol}} \xrightarrow{\text{1 day}} \mathrm{ne\_NITP_{aerosol}}$$

2. **In Aerosol Water:** SOA formed from the reactive uptake of glyoxal, methylglyoxal, and epoxide onto sulfate aerosol.

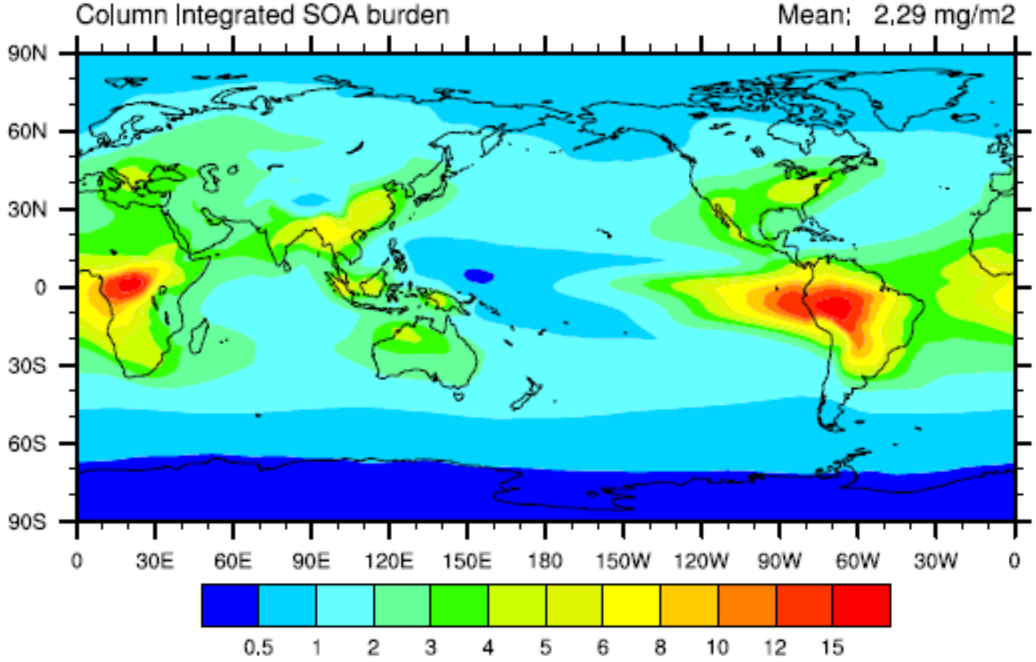
$$\frac{dC_{aq}}{dt} = \frac{1}{4} \cdot \gamma \cdot A \cdot < \nu > \cdot C_g$$

3. **In Cloud Water:** SOA formed from the aqueous phase reactions of glyoxal and methylglyoxal in the cloud water.

$$\frac{dC_{aq}}{dt} = R_{aq} + \frac{k_t}{RT} \cdot P_g - \frac{k_t}{HRT} Q \cdot C_{aq}$$

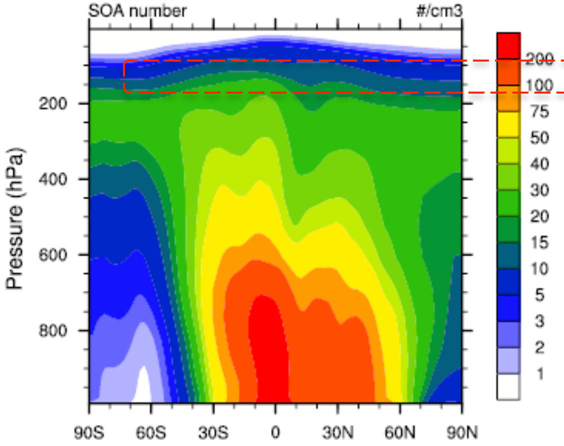
SOA burden and number

Column integrated burden

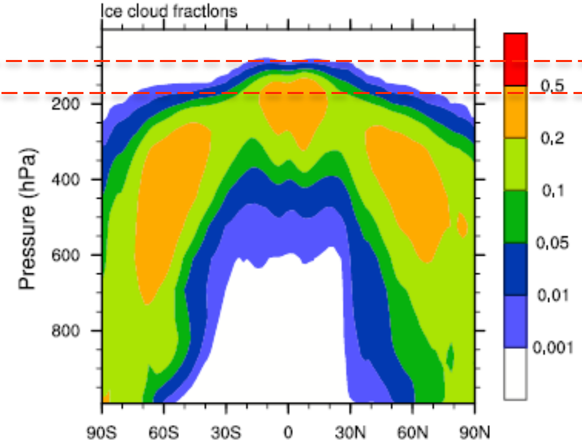


- Global is burden is ~1.16Tg
- Zonal mean: ~1-40 #/cm3 near tropopause

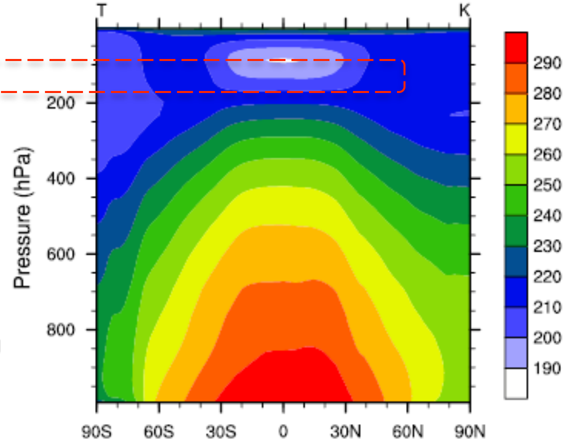
Zonal mean SOA #



Ice cloud fractions

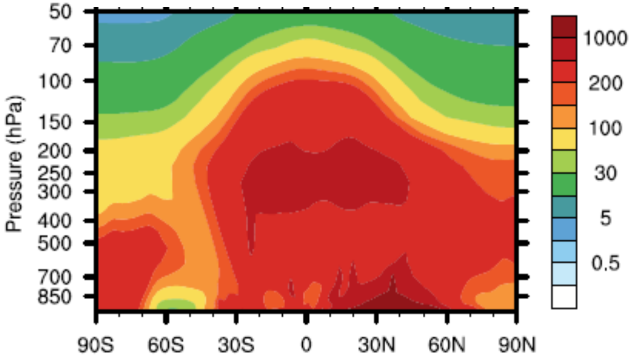


Zonal mean T

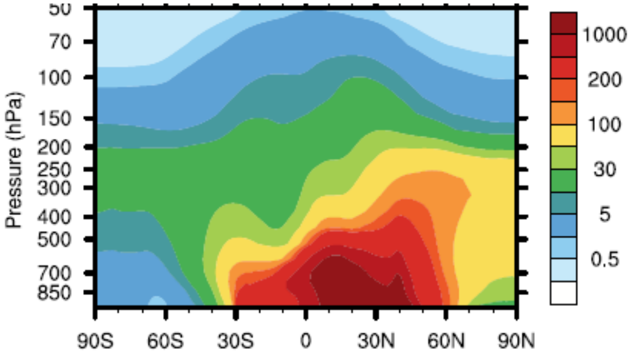


Solution droplet number and Heterogeneous IN number

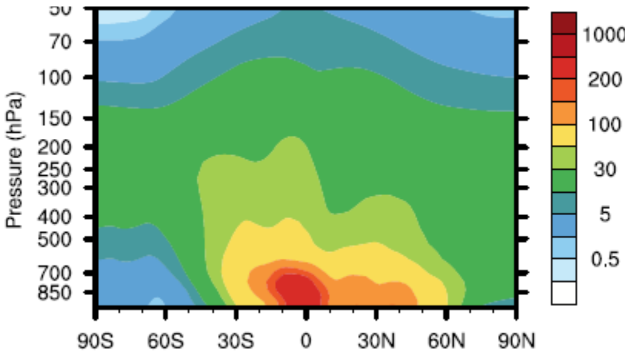
Aitken+Accum sulfate #/cm3



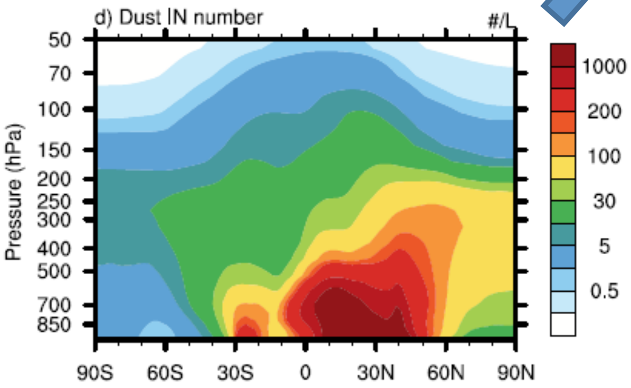
Total background IN #/L



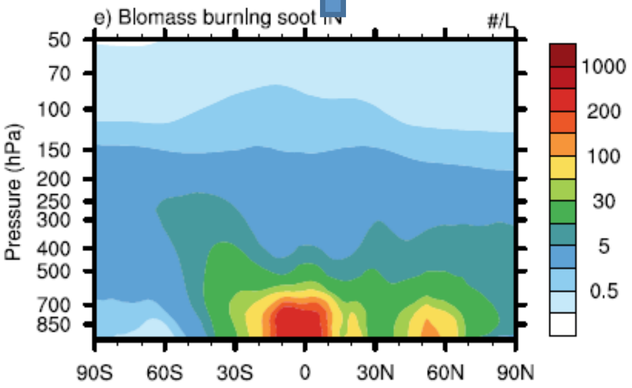
0.1% of SOA acting as glassy IN #/L



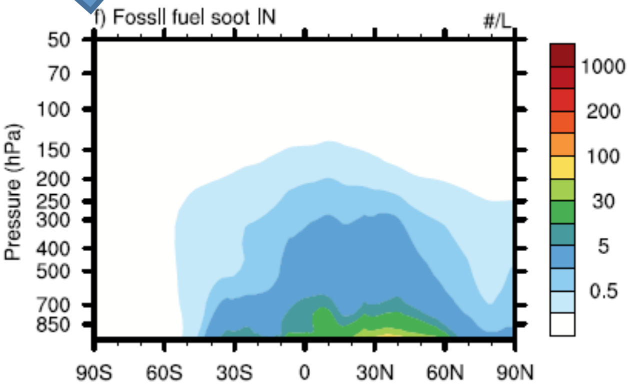
SOA # concentration is at least 1 order larger than POM near TTL !!



10% of total dust number

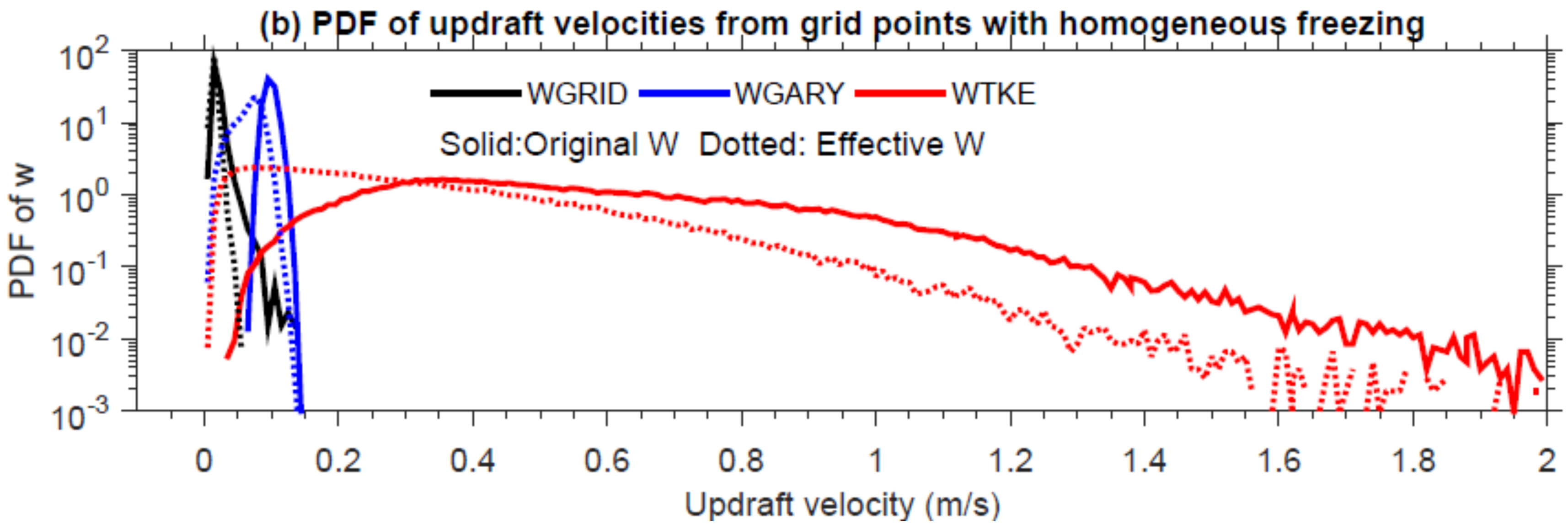


0.1% of biomass burning soot

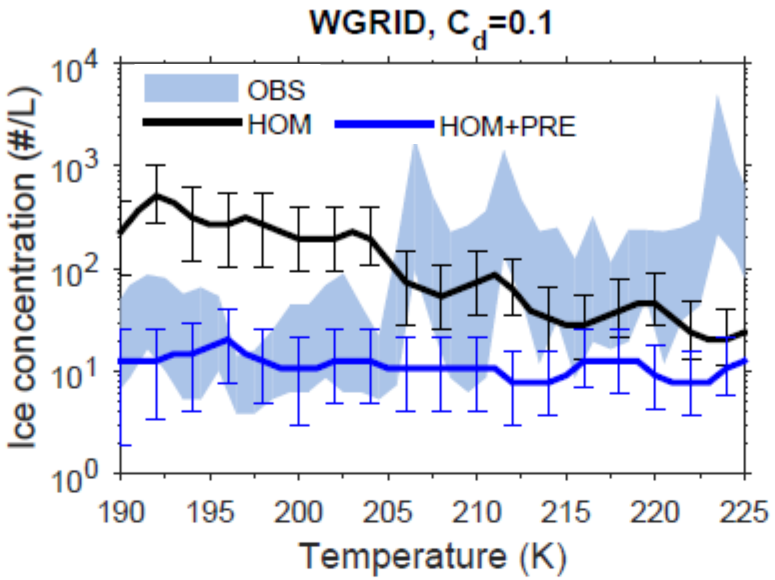


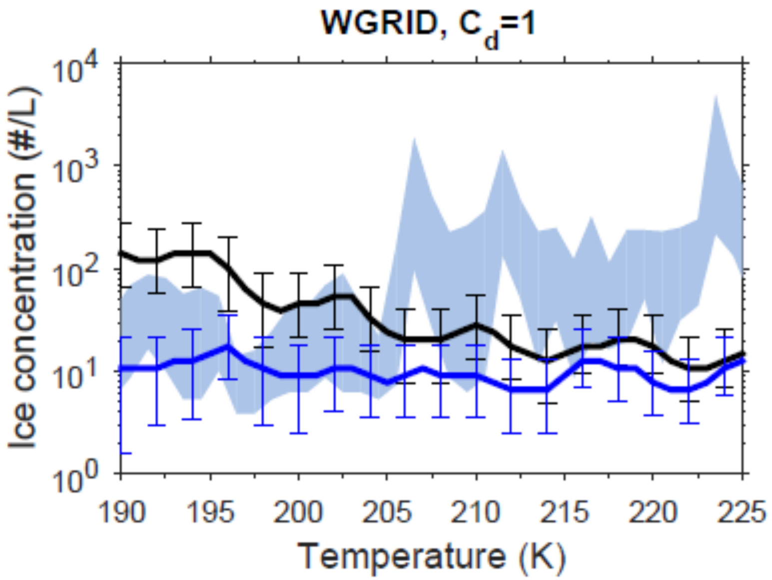
0.1% of hydrophilic fossil fuel soot
0.05% of hydrophobic fossil fuel soot

In-cloud ice number vs. T from WGRID



In-cloud ice number vs. T from WGRID

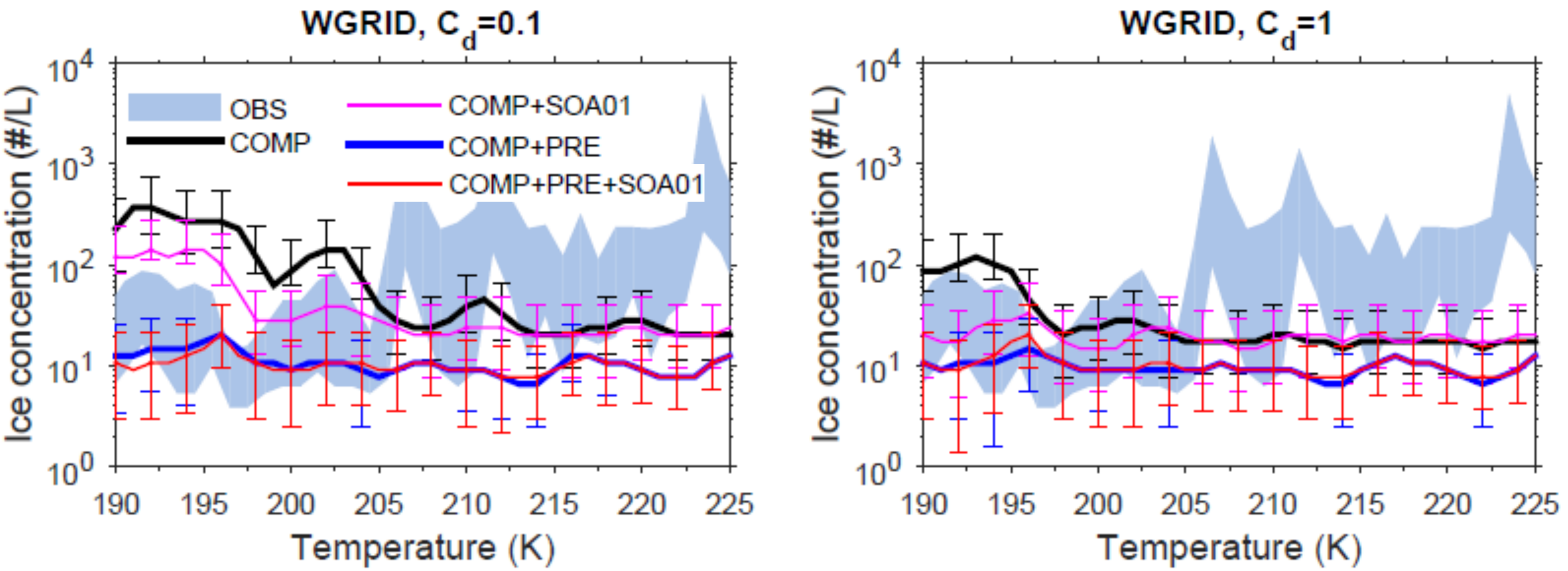




- **OBS: Shade** shows the 25%-75% percentiles of observed in-cloud ice number concentration from Krämer et al. (2009)
- **HOM :** only homogeneous freezing occurs in ice nucleation, no heterogeneous nucleation.
- **PRE:** pre-existing ice effect is considered.
- **Solid curves** show the 50% percentiles of simulated in-cloud ice number concentration.
- **Error bars** show 25%-75% percentiles of simulated in-cloud ice number concentration.
- **Cd:** Accommodation coefficient of water vapor to ice

Homogeneous freezing plus pre-existing ice effect can explain the low ice # at T<205K!

In-cloud ice number vs. T from WGRID



- **OBS: Shade** shows the 25%-75% percentiles of observed in-cloud ice number concentration from Krämer et al. (2009)
- **COMP:** competition between homogeneous freezing and heterogeneous nucleation in ice nucleation.
- **SOA01:** 0.1% of total SOA act as glassy IN.
- **PRE:** pre-existing ice effect is considered.
- **Solid curves** show the 50% percentiles of simulated in-cloud ice number concentration.
- **Error bars** show 25%-75% percentiles of simulated in-cloud ice number concentration.
- **Cd:** Accommodation coefficient of water vapor to ice

Pre-existing ice effect can explain the low ice # at T<205K!

WGRID, Cd=0.1, at ~100 hPa

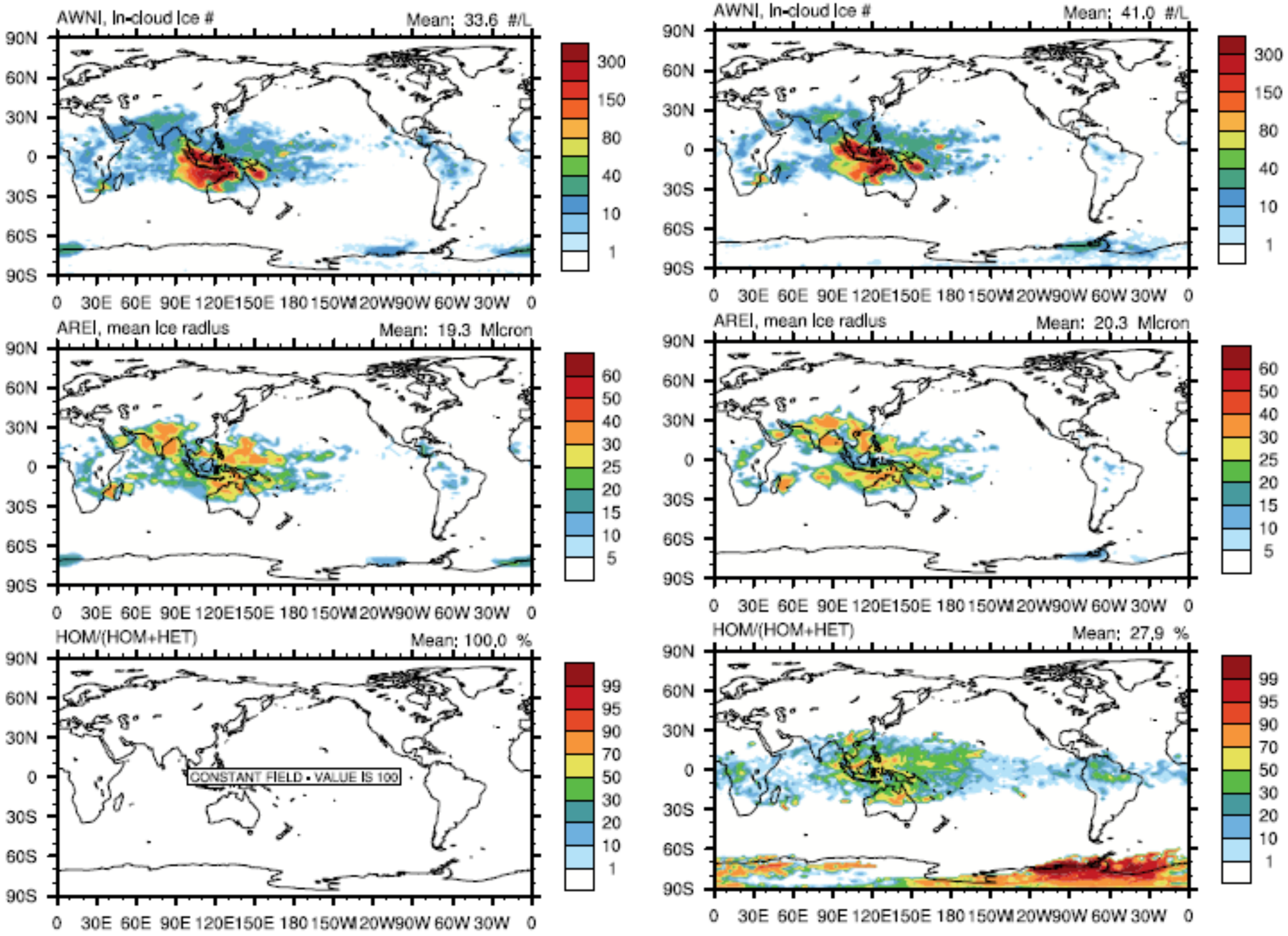
HOM+PRE

COMP+PRE

In-cloud ice #

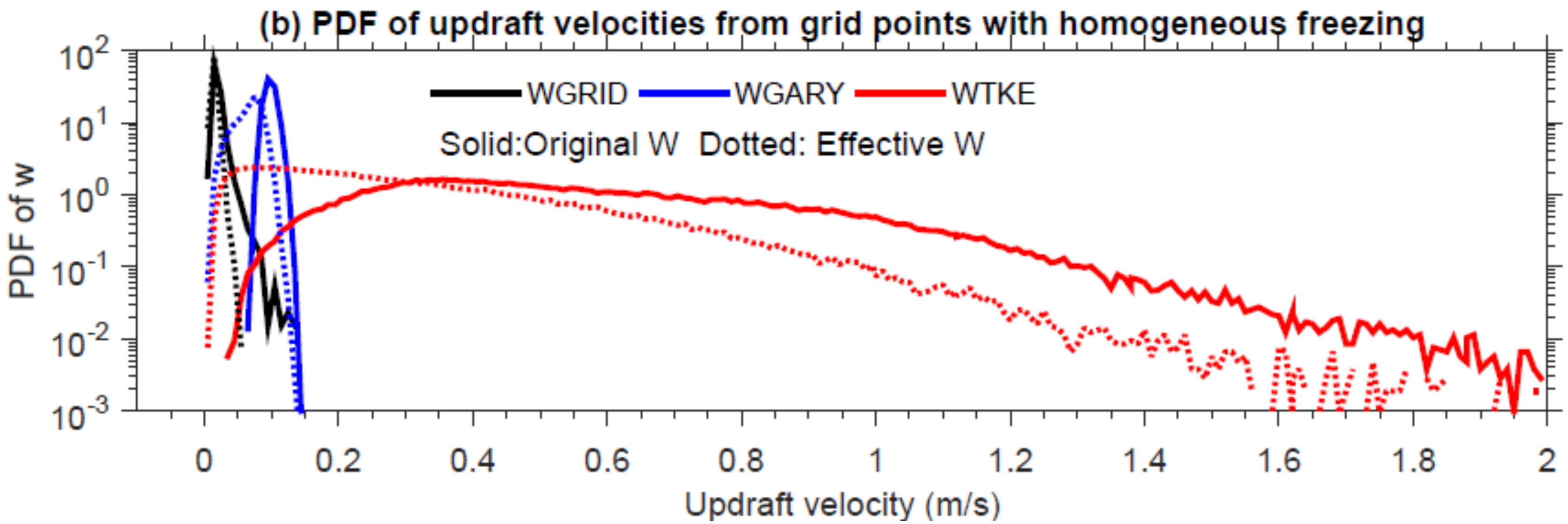
Ice radius

Fraction of ice from homo. freezing,

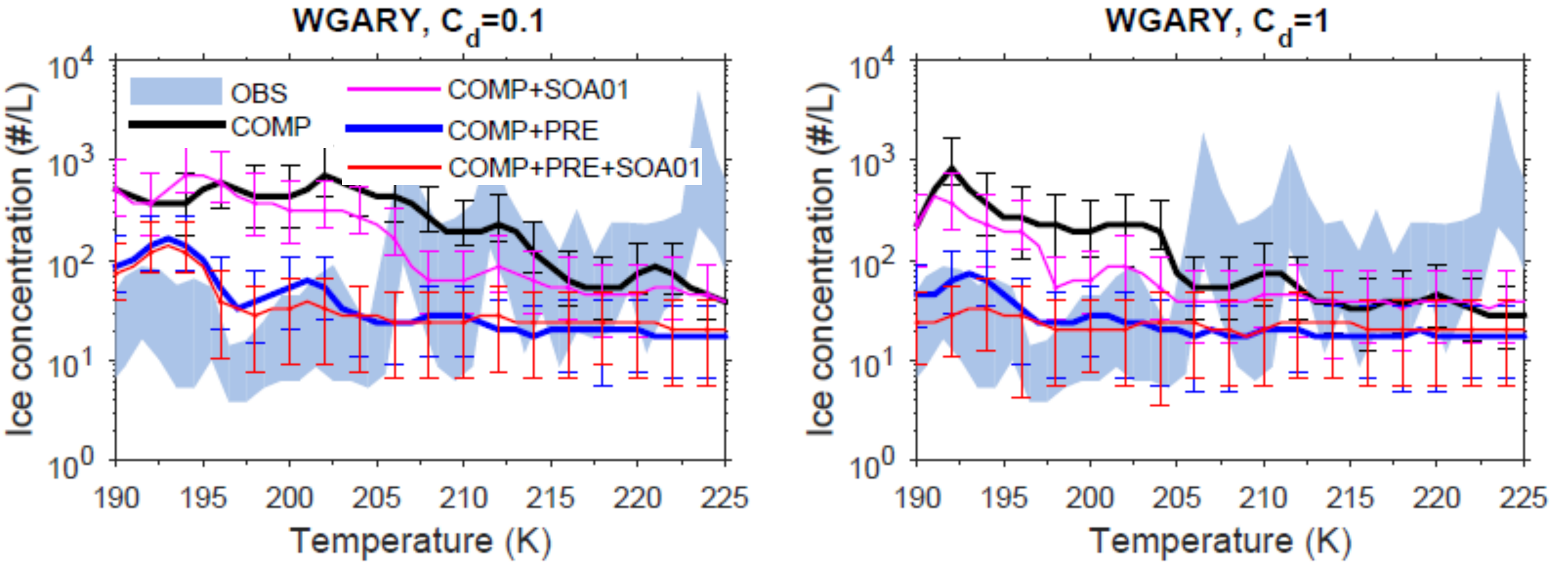


Whether the ice formed from homogeneous or heterogeneous freezing does not matter.

In-cloud ice number vs. T from WGARY



In-cloud ice number vs. T from WGARY



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- **Solid curves** show the 50% percentiles of simulated in-cloud ice number concentration.
- **Error bars** show 25%-75% percentiles of simulated in-cloud ice number concentration.
- **Cd:** Accommodation coefficient of water vapor to ice

Pre-existing ice effect + SOA IN + larger Cd can explain the low ice # at T<205K!

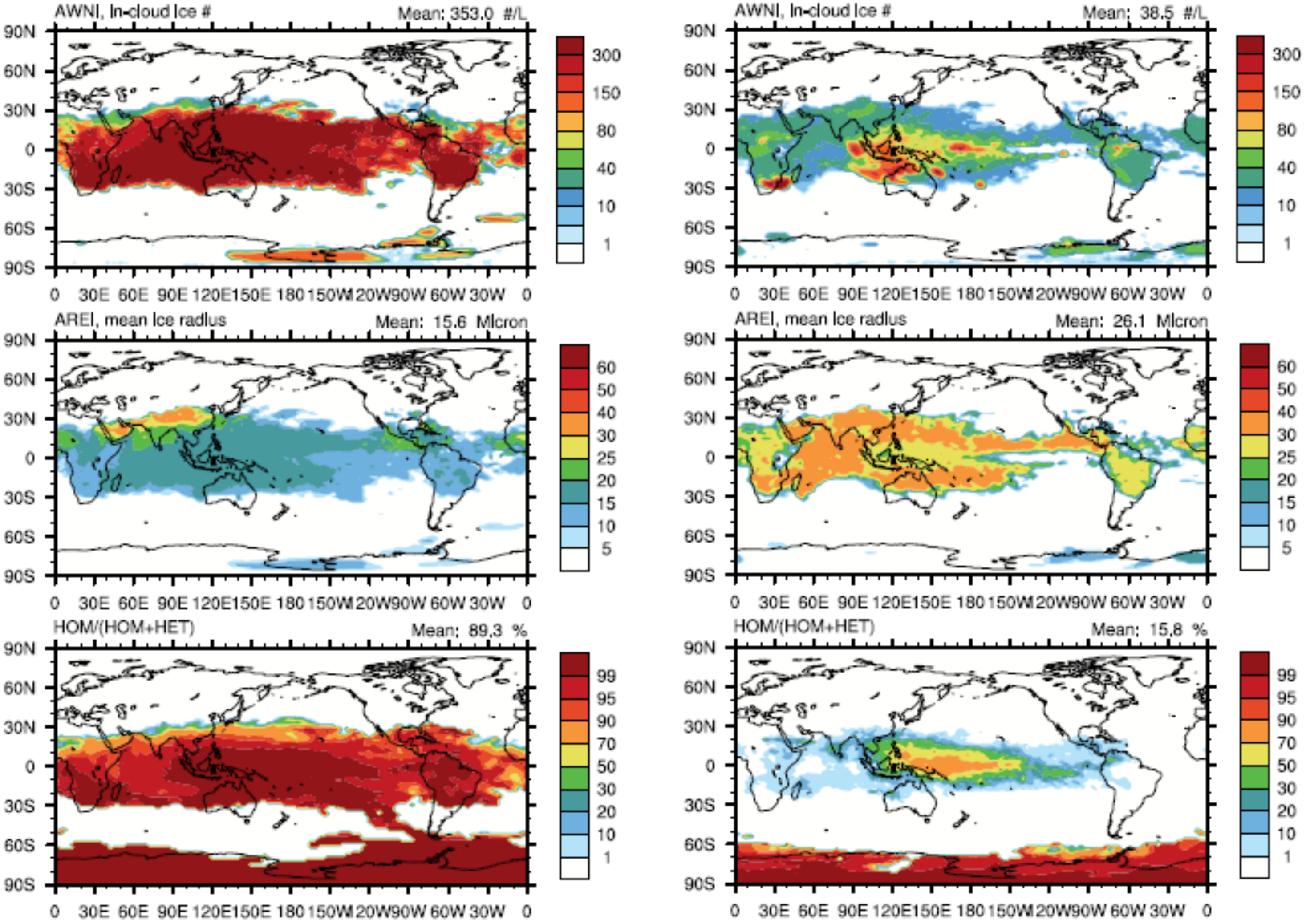
WGARY, Cd=1, at ~120 hPa

COMP — COMP+PRE+SOA01

In-cloud ice #

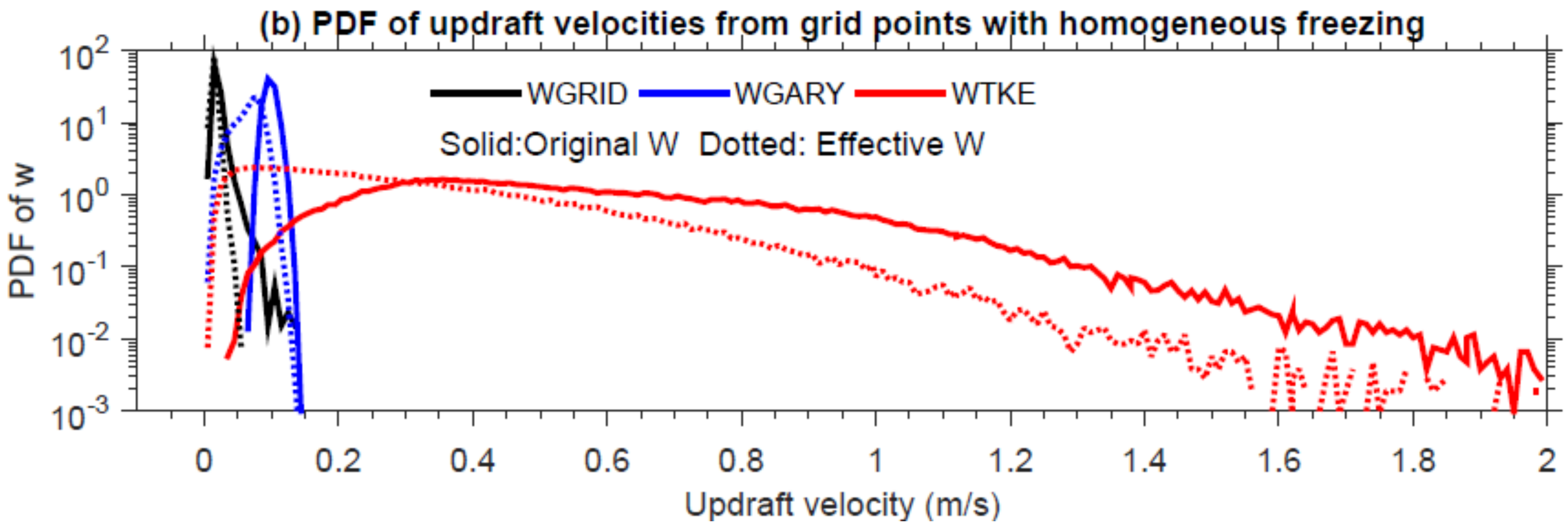
Ice radius

Fraction of ice from homo. freezing

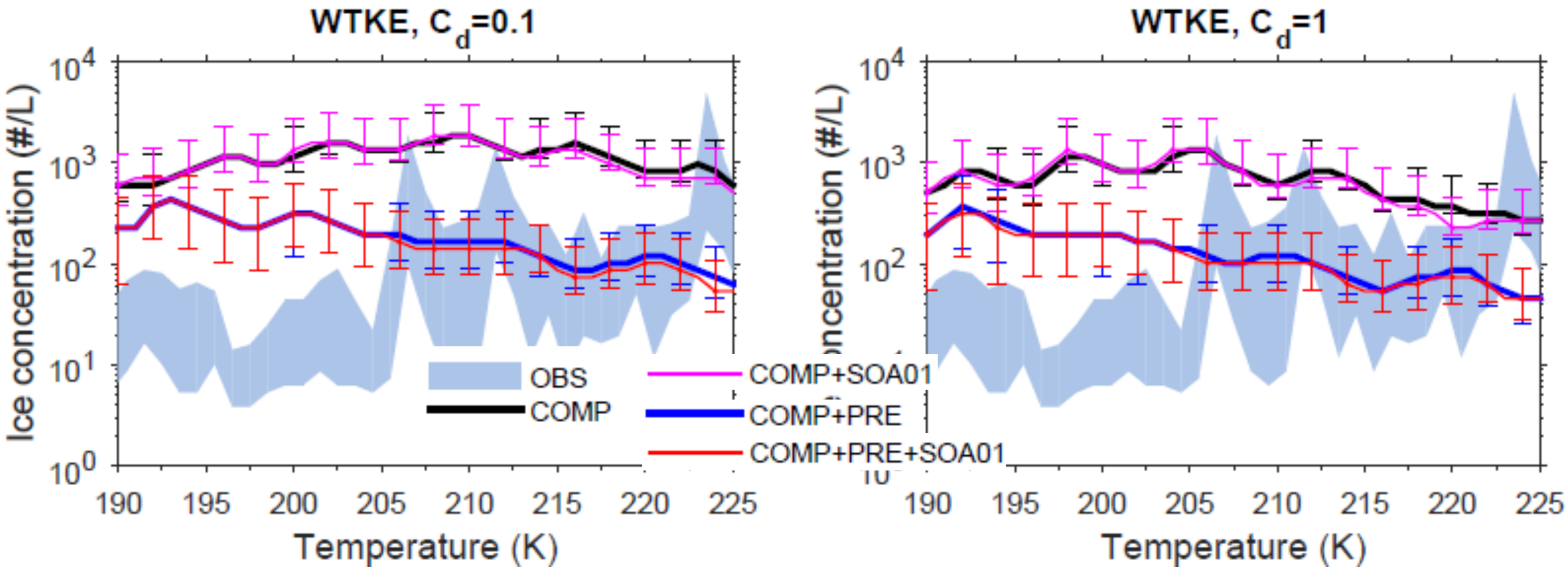


Ice number is 2 orders smaller, radius is doubled, homo. fraction from 90% to 16%.

In-cloud ice number vs. T from WTKE



In-cloud ice number vs. T from WTKE



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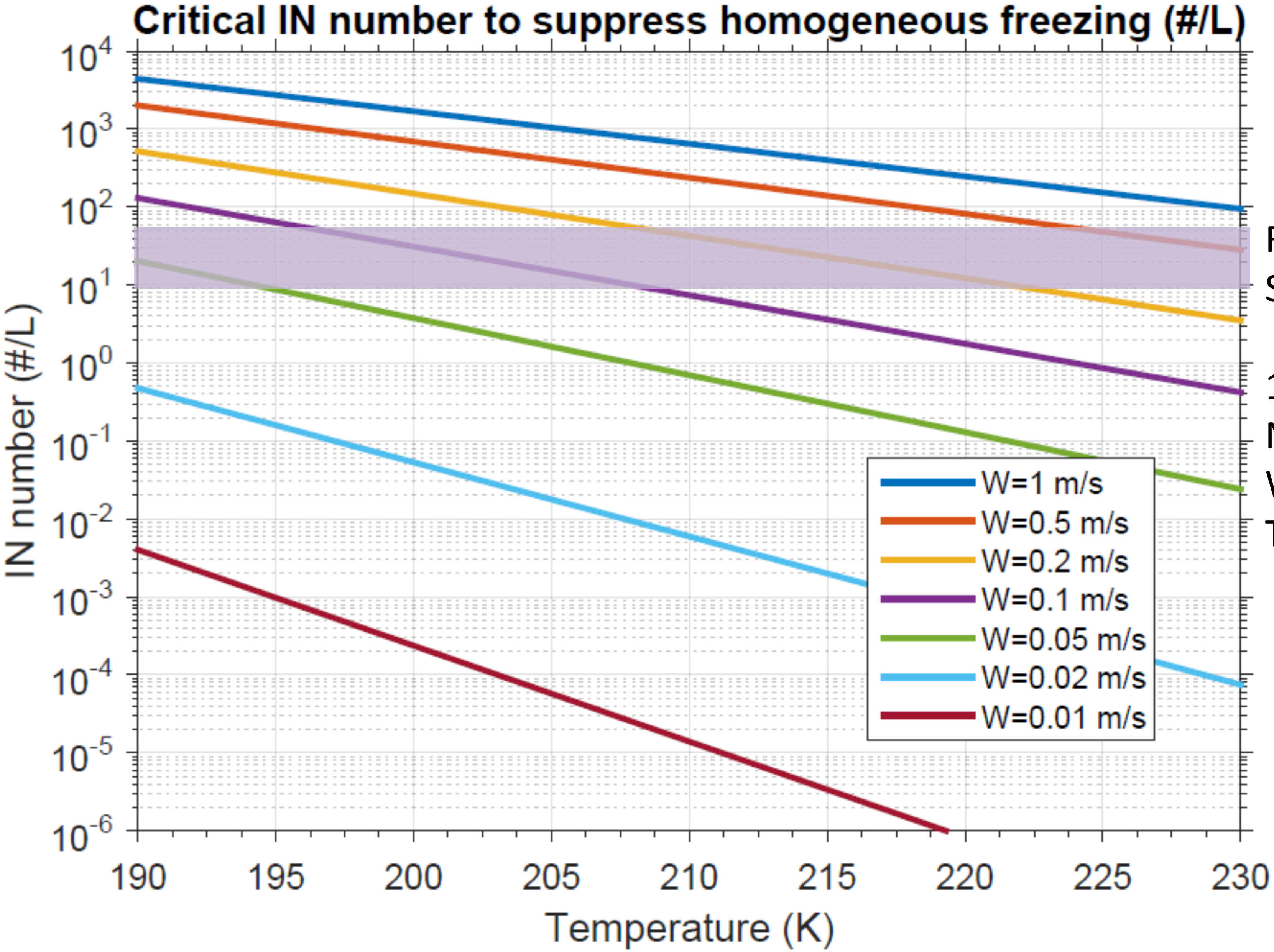
- **Cd:** Accommodation coefficient of water vapor to ice

Pre-existing ice effect has bigger effect while SOA IN has much smaller effect.



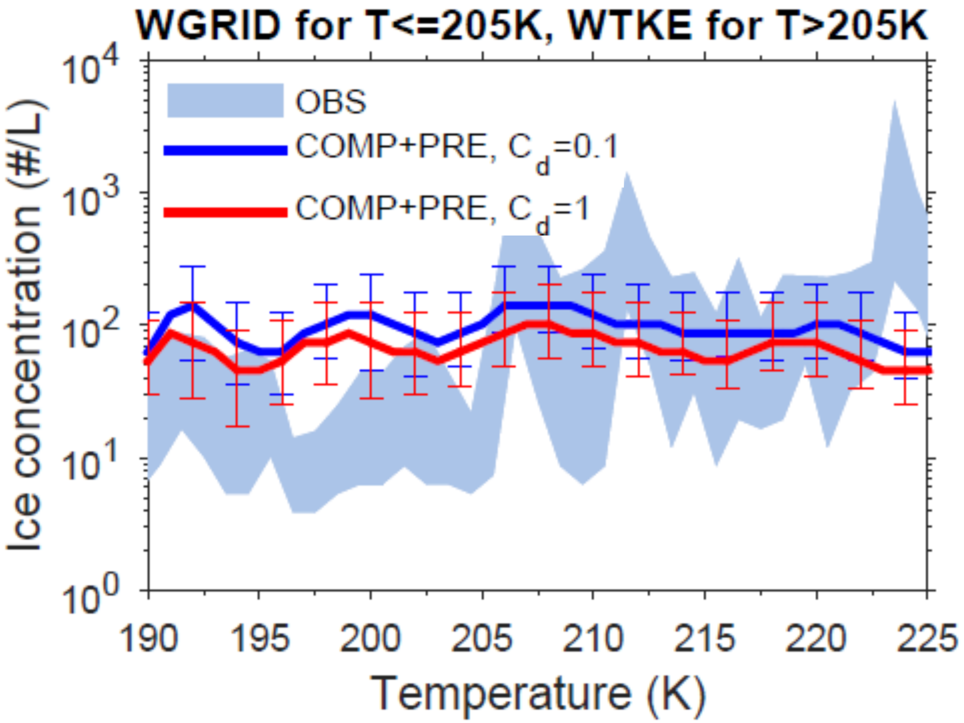
Range of
SOA IN #

100#/L is
Needed for
W>0.2m/s
T<205K

Results are based on the parcel model results from Liu and Penner (2005).
Baharona and Nenes [2009] scheme require 1 order larger IN number.s

Hybrid updraft velocities

- Use WGRID in cold cirrus clouds (T<=205K)
- Use WTKE in warm cirrus clouds (T>205K)
- **Reasoning:**
  - Inertial gravity waves have higher frequencies in cold cirrus clouds?
  - CAM may overestimate TKE in cold cirrus near TTL?

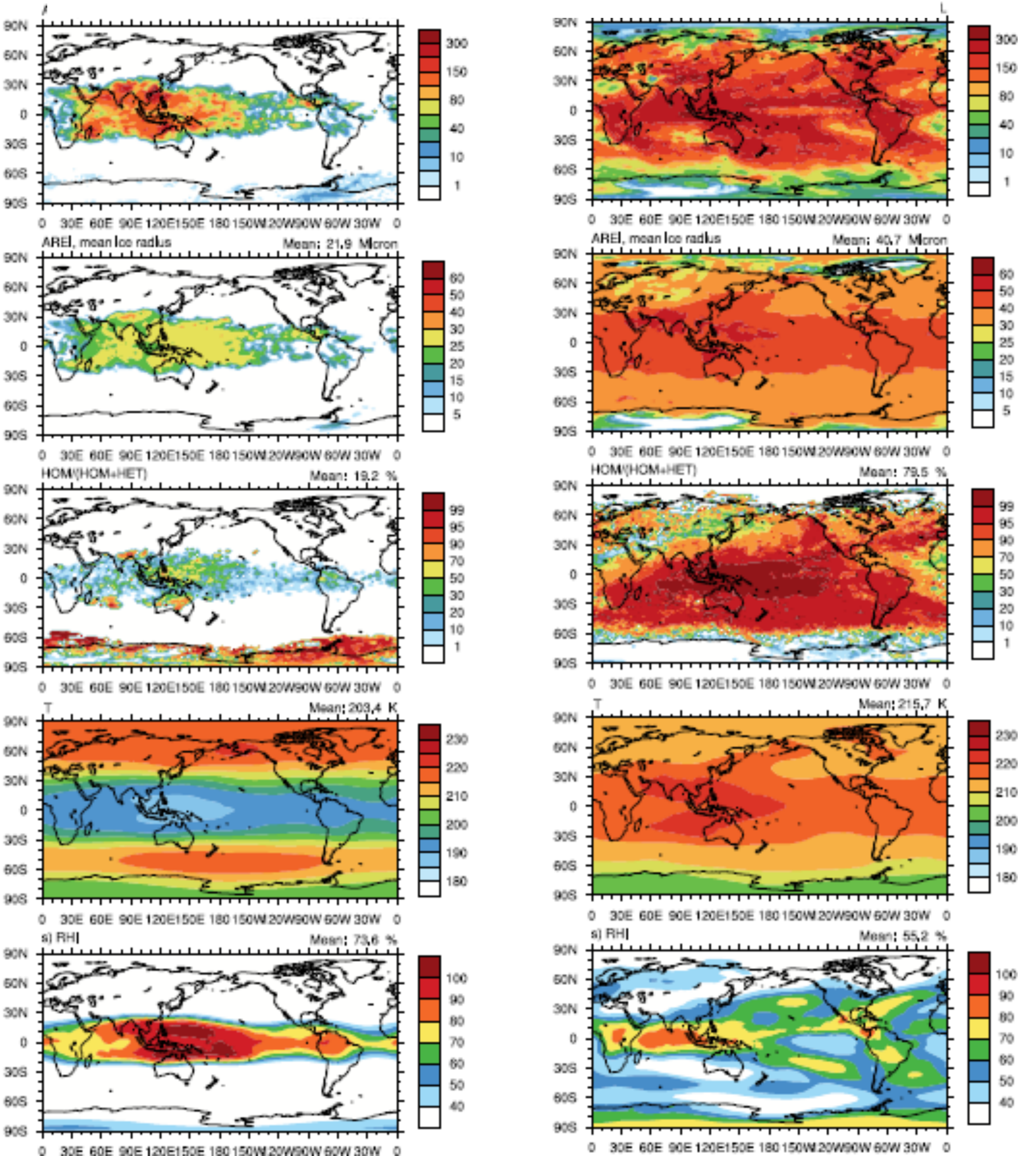


Ice number for T<205K is higher than that from WGRID cases. Vertical transport of ice from lower altitudes?

Hybrid updraft velocities case: WGRID for T<=205K and WTKE for T>205K, Cd=0.1,COMP+PRE

At ~100 hPa

At ~200 hPa



In-cloud ice #

Ice radius

Fraction of
ice from homo.
Freezing

T

RHI

Conclusion

- **SOA IN, pre-existing ice effect, and bigger accommodation coefficient of water vapor to ice** can all contribute to reduce the ice number concentration.
- **SOA IN** becomes less important when pre-existing ice effect is considered. This is because the pre-existing ice can have bigger size and larger number than SOA IN.
- **Updraft velocity** plays the most important role in determining the ice number:
  - **WGRID:** pre-existing ice effect alone is able to explain the low ice number for T<205K.
  - **WGARY:** pre-existing ice effect+SOA IN+larger Cd are needed to explain the low ice number for T<205K.
  - **WTKE:** No combination can explain the low ice number for T<205K. But ice number compares best with observation for T>205K when pre-existing ice effect is considered.
- **Hybrid use** of WGRID in cold cirrus clouds (T<205K) and WTKE in warm cirrus clouds (T>205K) produces the best ice number concentration.
  - Vertical transport of ice from warm cirrus clouds to cold cirrus clouds may play a role in determining the ice number in cold cirrus clouds.

THANK YOU!

WGRID
Cd=0.1
at ~100 hPa

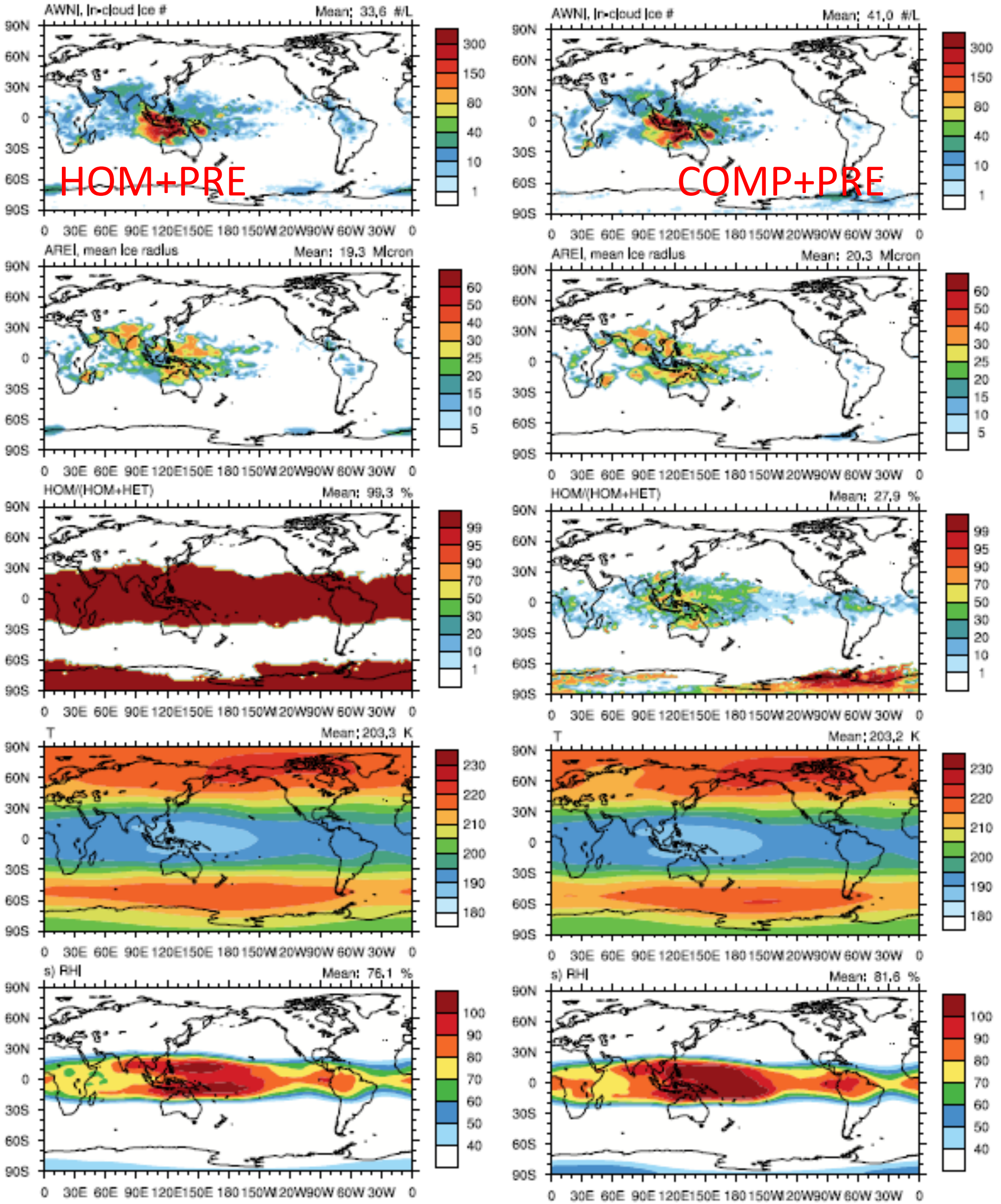
In-cloud ice #

Ice radius

Fraction of ice from homo. Freezing

T

RHI

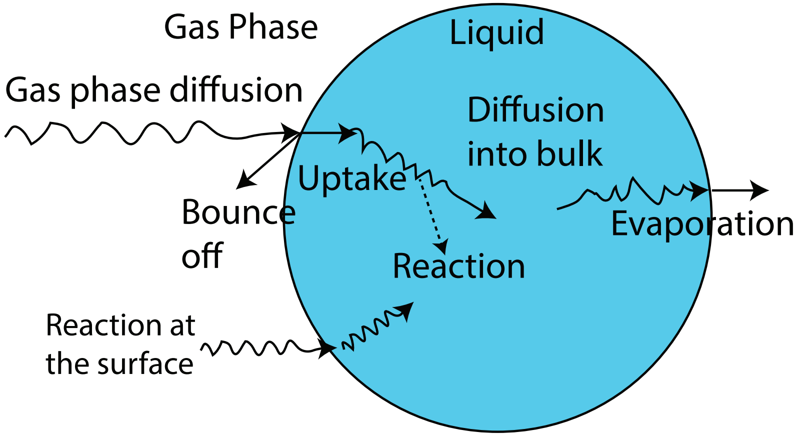


Global atmospheric chemical transport model (IMPACT)

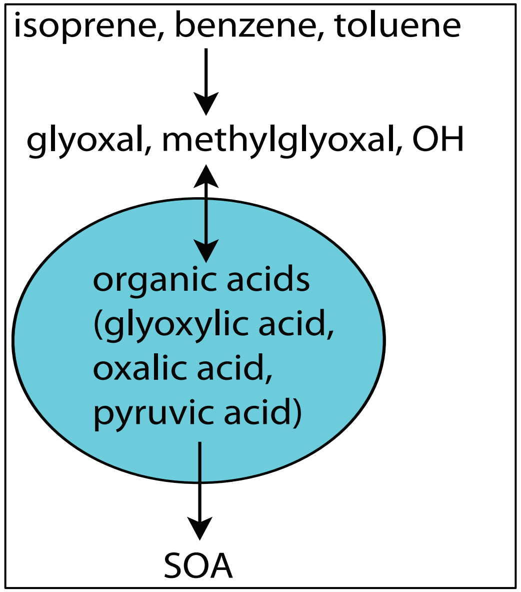
- Gas phase chemistry
  - Basic photochemistry of O_3, OH, NO_x and VOCs (Ito et al., 2007).
  - Epoxide formation from isoprene (Paulot et al., 2009).
  - HO_x regeneration through isoprene oxidation proposed by Peeters et al. (2009) but with a recycled rate reduced by a factor of 10.
- SOA formation mechanism
  - SOA formed from gas-particle partitioning
    - Select species that form SOA based on criteria from *Griffin et al*. (2002).
    - Species vapor pressures estimated as in *Myrdal and Yalkowsky* (1997) with modifications from *Camredon and Aumont* (2006).

SOA formation mechanism

SOA formed from the cloud processing of gloyxal and methylglyoxal



Schematic diagram of uptake and reaction of gases in liquid. Based on Finlayson-Pitts and Pitts, (2000)


isoprene, benzene, toluene

glyoxal, methylglyoxal, OH

organic acids (glyoxylic acid, oxalic acid, pyruvic acid)

SOA

SOA formed from the reactive uptake of glyoxal, methylglyoxal and epoxide onto sulfate aerosol

$$\frac{dC_{SOA}}{dt} = \frac{1}{4} \cdot \gamma \cdot A \cdot < v > \cdot C_{gas}$$

Y: reactive uptake parameter.
A: surface area of aqueous sulfate aerosols
Major products: oligomers and organosulfate