



# Aerosol modeling at ECPL



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# Global Cloud Condensation Nuclei (CCN) simulations: Robustness and implication for droplet formation

Atmos. Chem. Phys. Discuss., <https://doi.org/10.5194/acp-2018-1340>

Manuscript under review for journal Atmos. Chem. Phys.

Discussion started: 18 January 2019

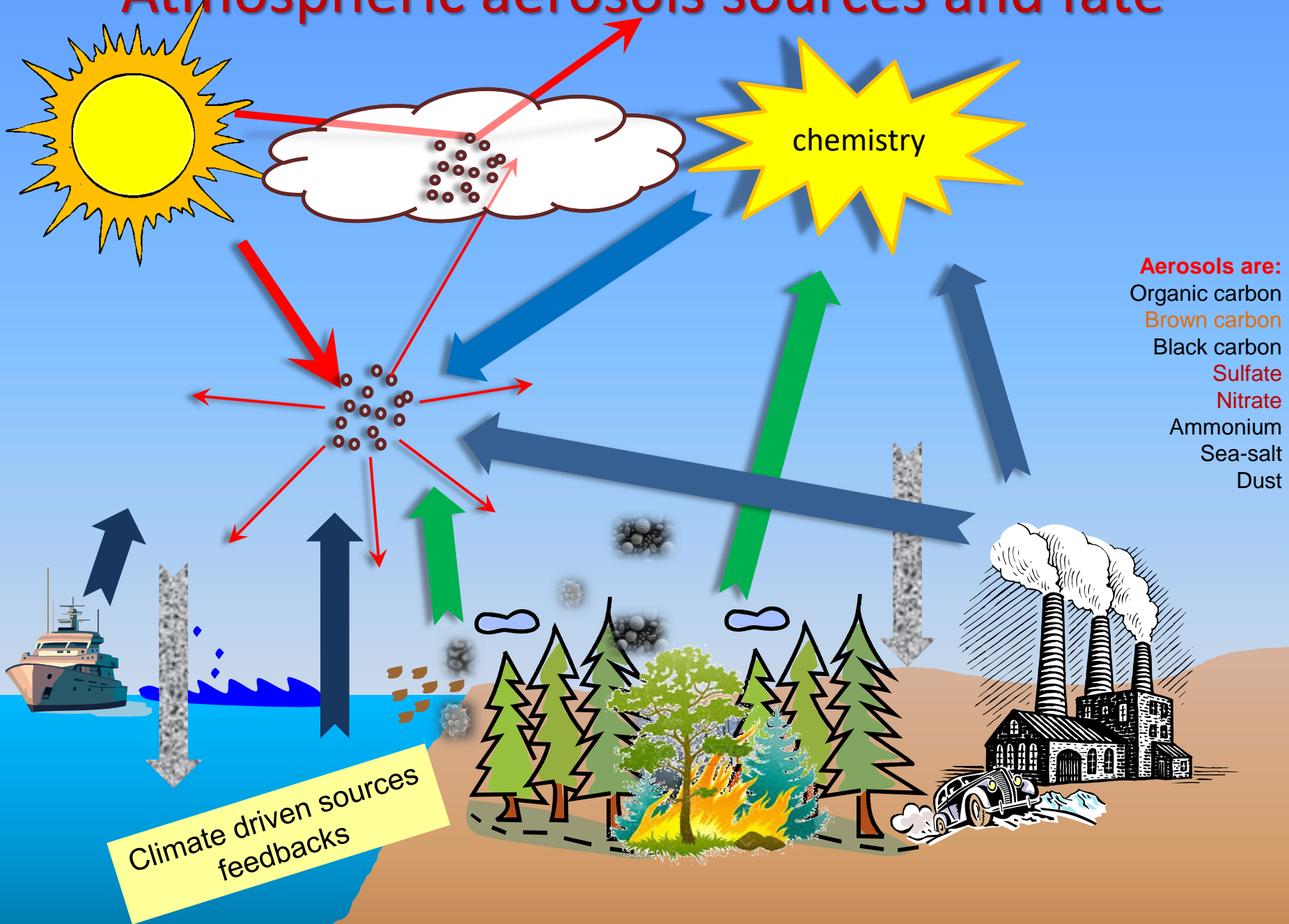
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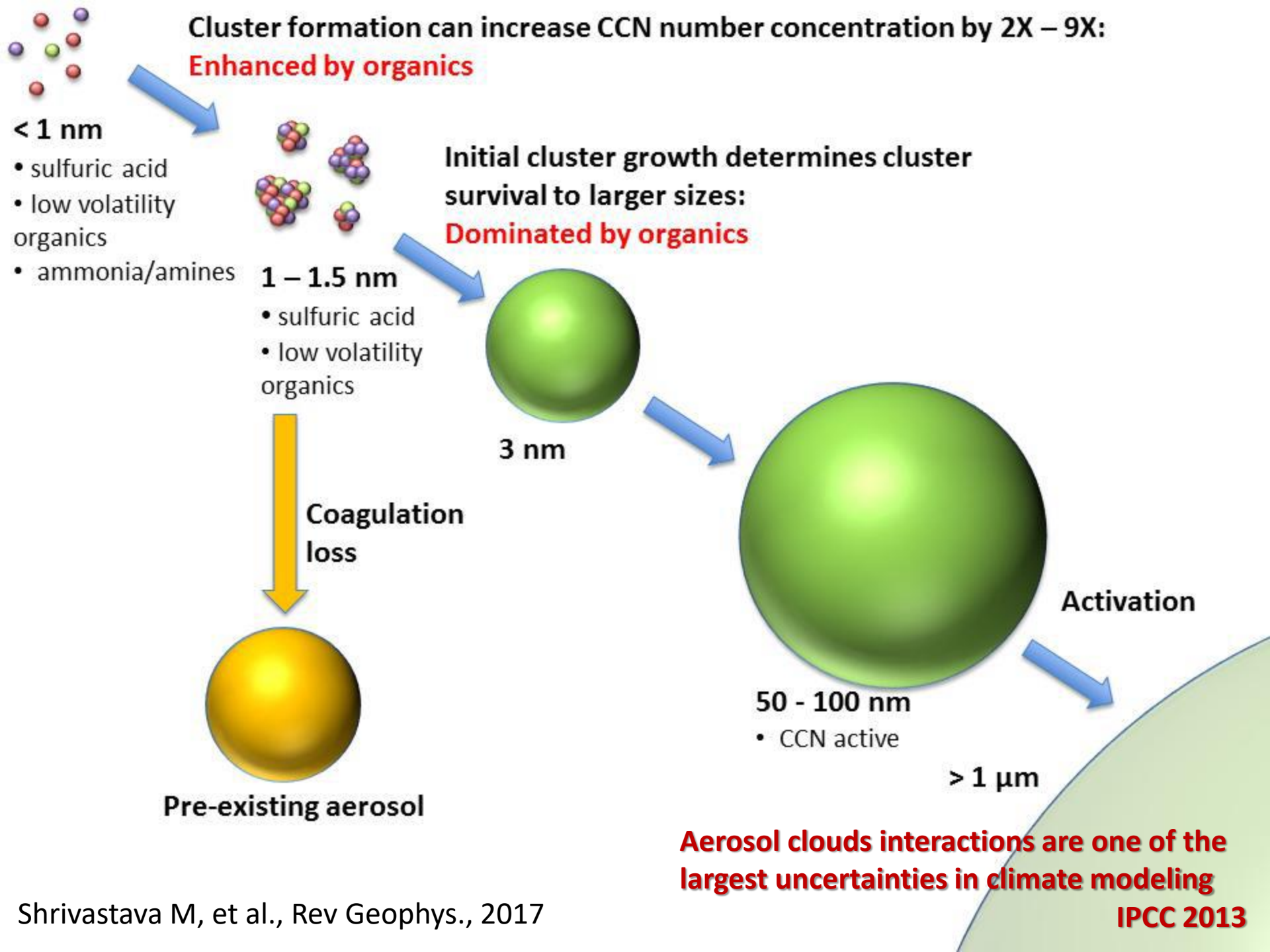


## **Evaluation of global simulations of aerosol particle number and cloud condensation nuclei, and implications for cloud droplet formation**

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# Atmospheric aerosols sources and fate





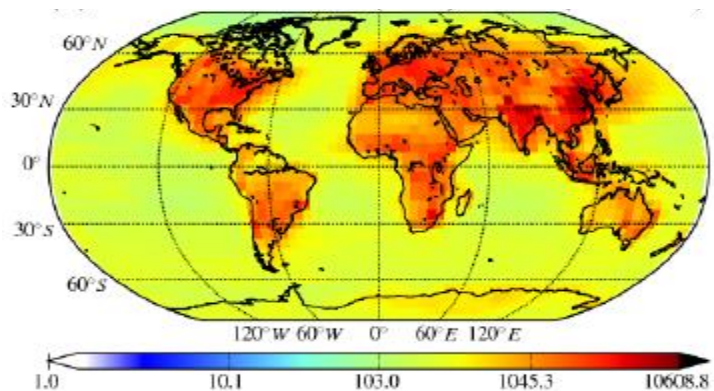
## aim of the study

Evaluate general circulation & global chemistry-transport models for their ability to simulate

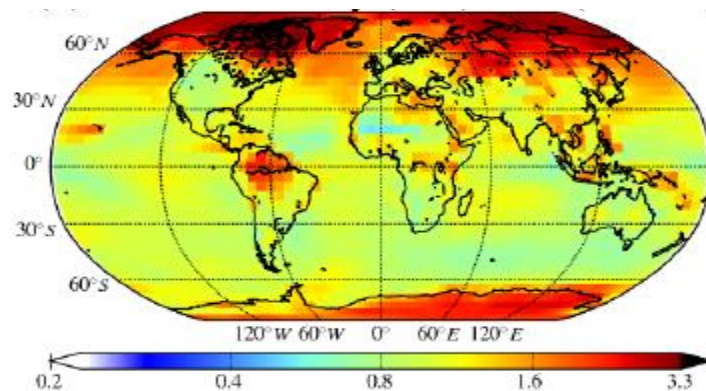
- ✓ Aerosol number concentrations ( $N_a$ )
  - ✓ Cloud condensation nuclei (CCN)
  - ✓ Cloud droplets number concentration (CDNC)
  - ✓ the long-term seasonal variability & the short-term dynamical behavior of aerosol particles and CCN
- 
- 15 global models (GCMs & CTMs) for the years 2010-2015
  - 8 European observatories (from ACTRIS) and 1 site in Japan



Annual Multi-model median

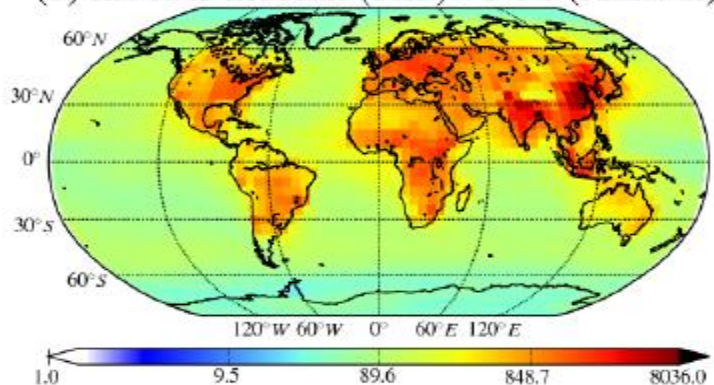


Model diversity (stdev/mean)

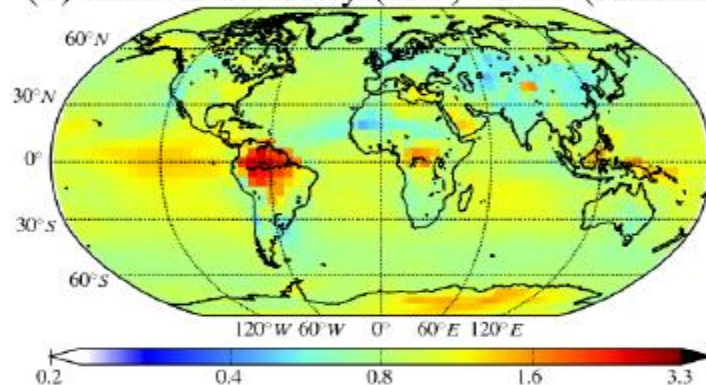


**N3**

(c) models median (5x5) - N50 (Annual)

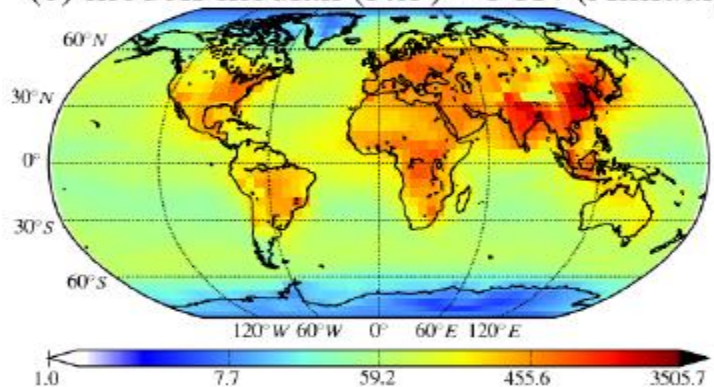


(d) models diversity (5x5) - N50 (Annual)

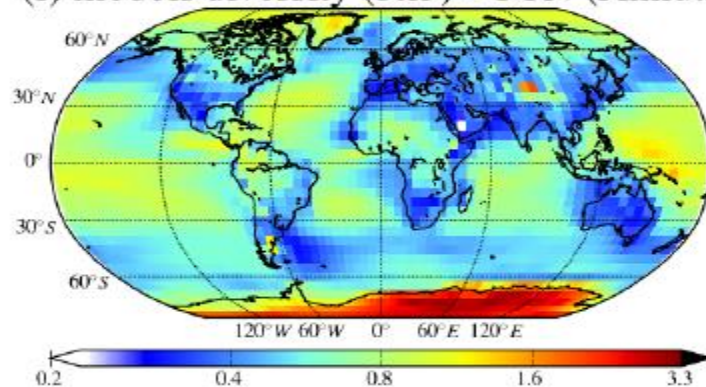


**N50**

(e) models median (5x5) - CCN (Annual)



(f) models diversity (5x5) - CCN (Annual)



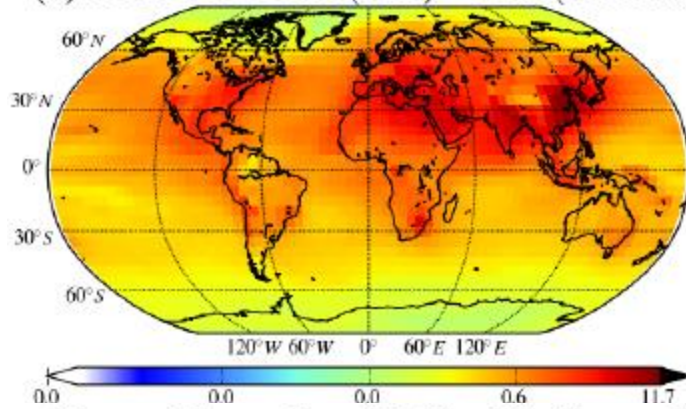
**CCN**

multi-model results at surface

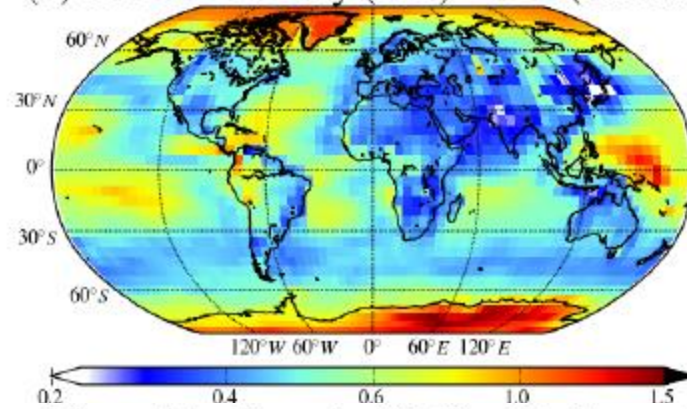


# PM<sub>1</sub> multi-model median chemical composition- surface

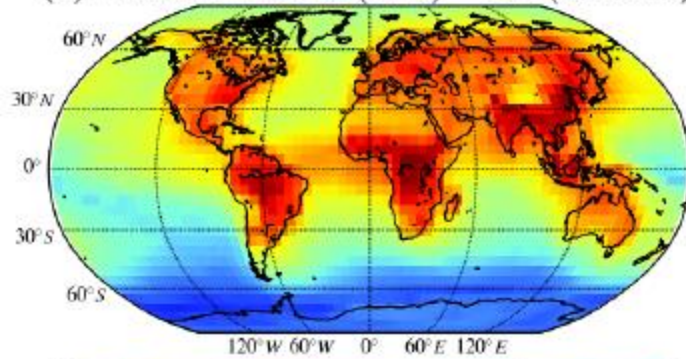
(a) models median (5x5) - SO<sub>4</sub> (Annual)



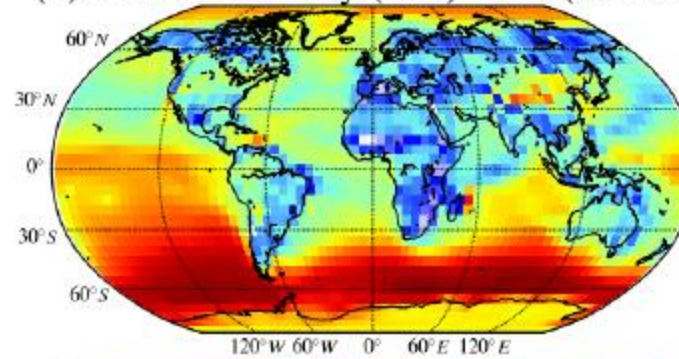
(b) models diversity (5x5) - SO<sub>4</sub> (Annual)



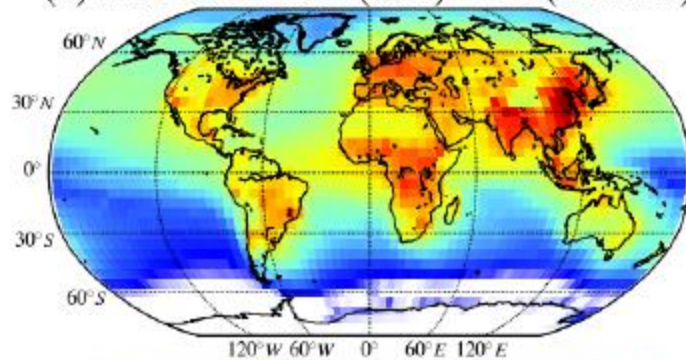
(c) models median (5x5) - OA (Annual)



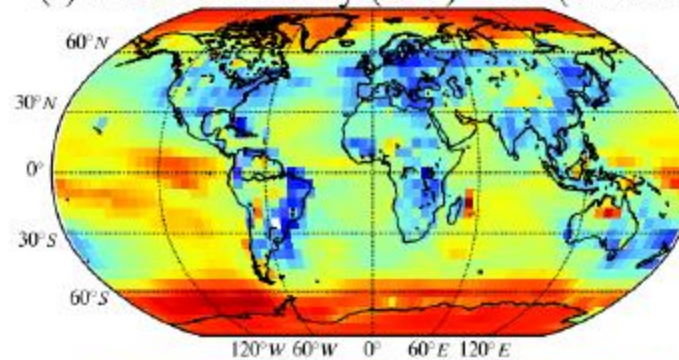
(d) models diversity (5x5) - OA (Annual)



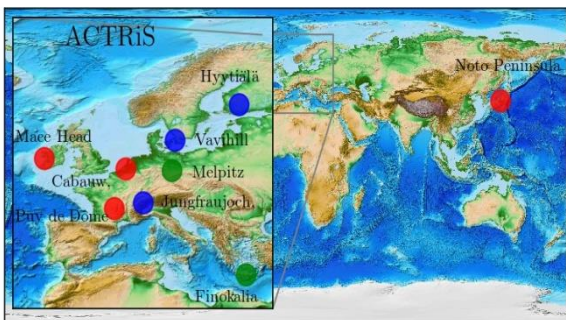
(e) models median (5x5) - BC (Annual)



(f) models diversity (5x5) - BC (Annual)



# BACCHUS model intercomparison 15 models vs AMS- organics



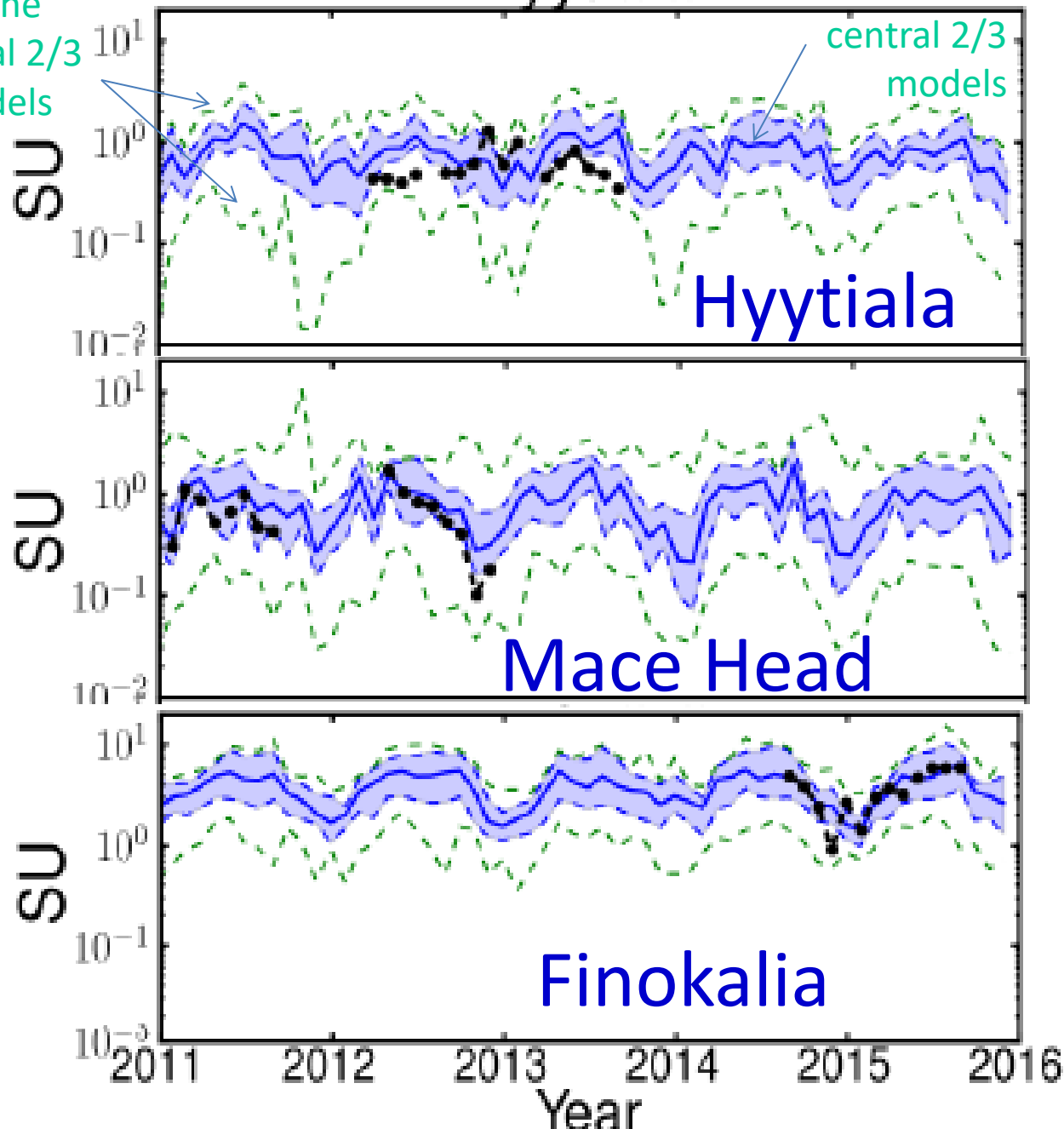
Data (black dots) from ACTRIS - Schmale et al. Scientific Reports, 2017

MMM – blue line  
min, max model green dashed

Min max of the central 2/3 models

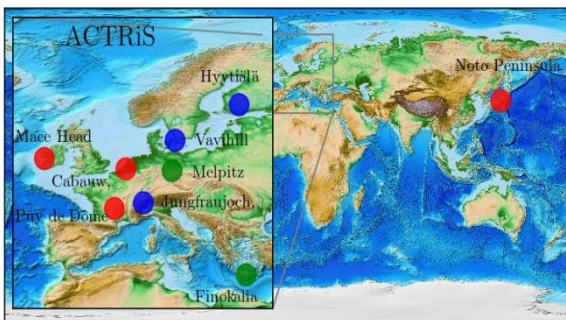
## Sulfate

Geometric mean of central 2/3 models



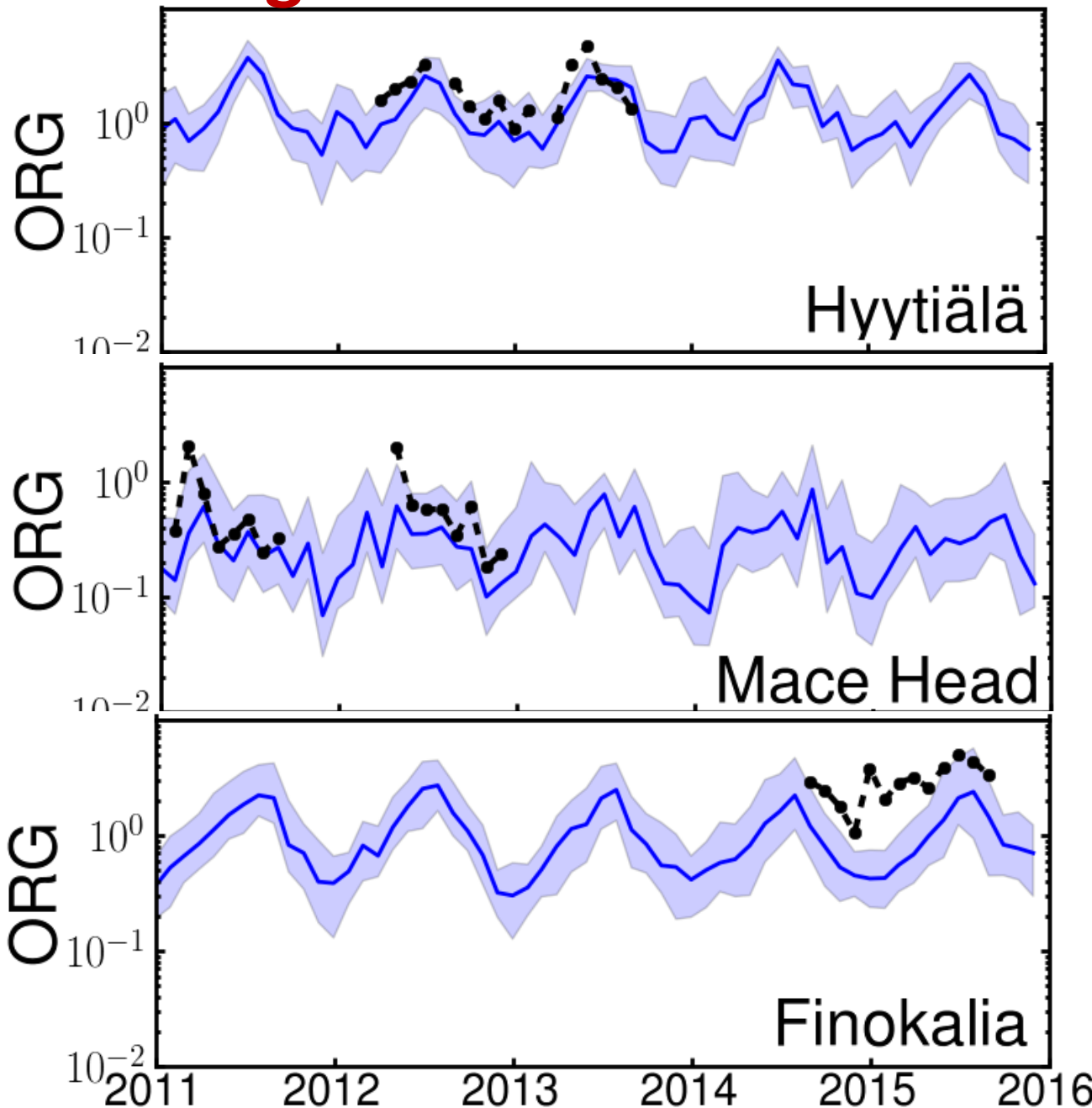


# BACCHUS model intercomparison 15 models vs AMS- organics

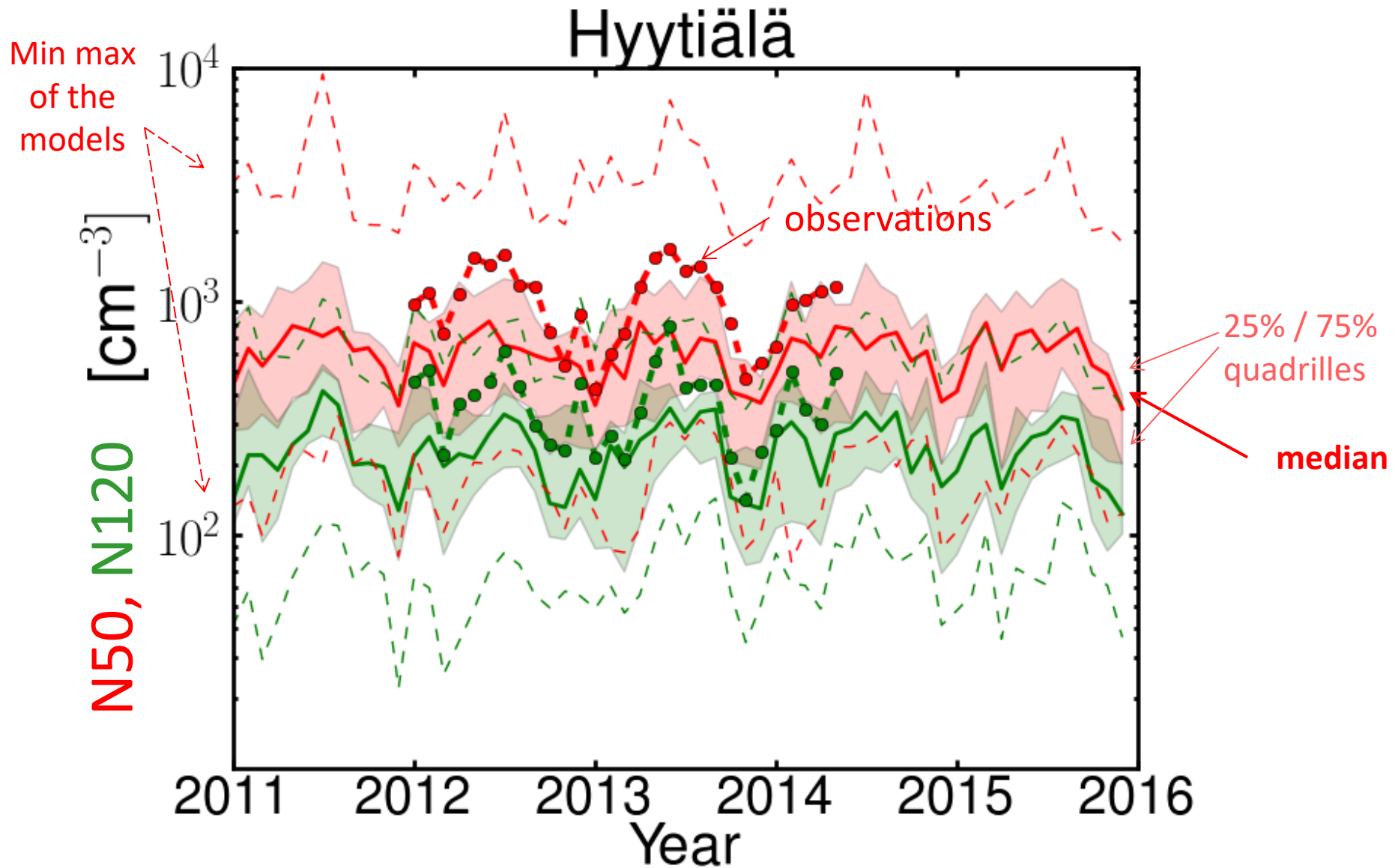


Data from ACTRIS -  
Schmale et al. Scientific  
Reports, 2017

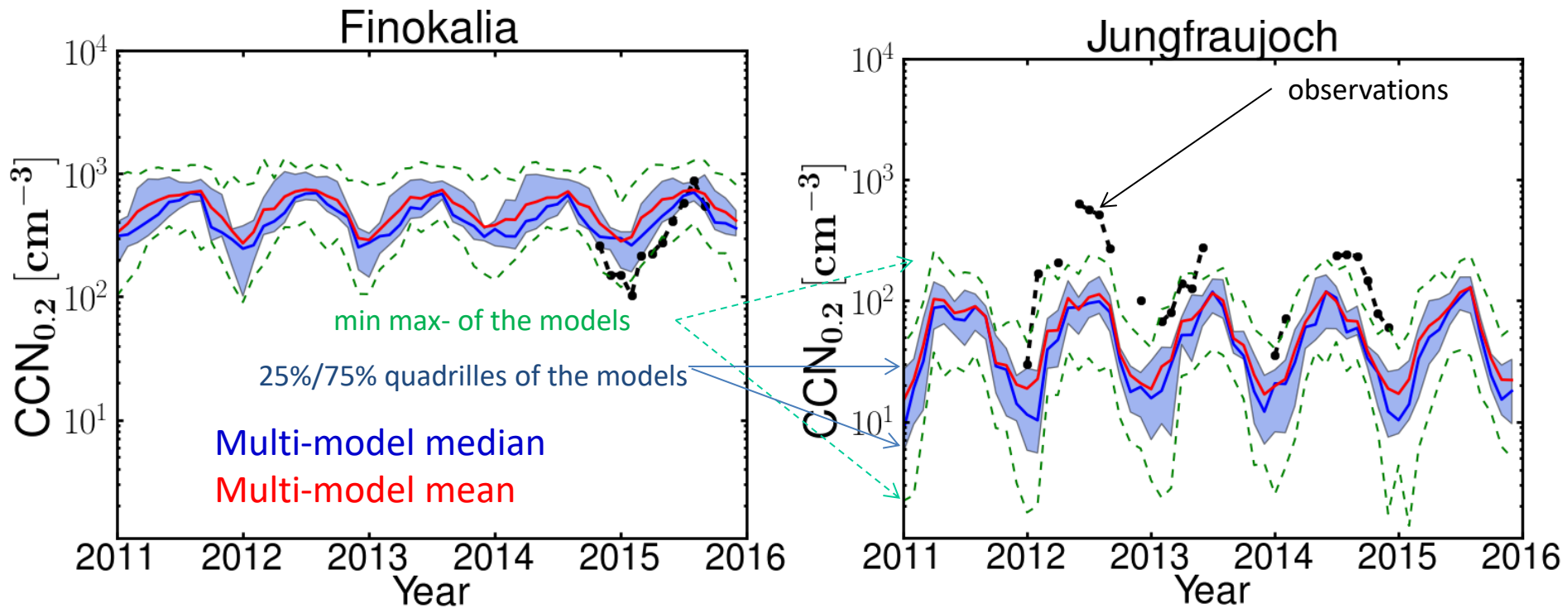
## Organics



# Number concentration of aerosols



# CCN at 0.2% supersaturation

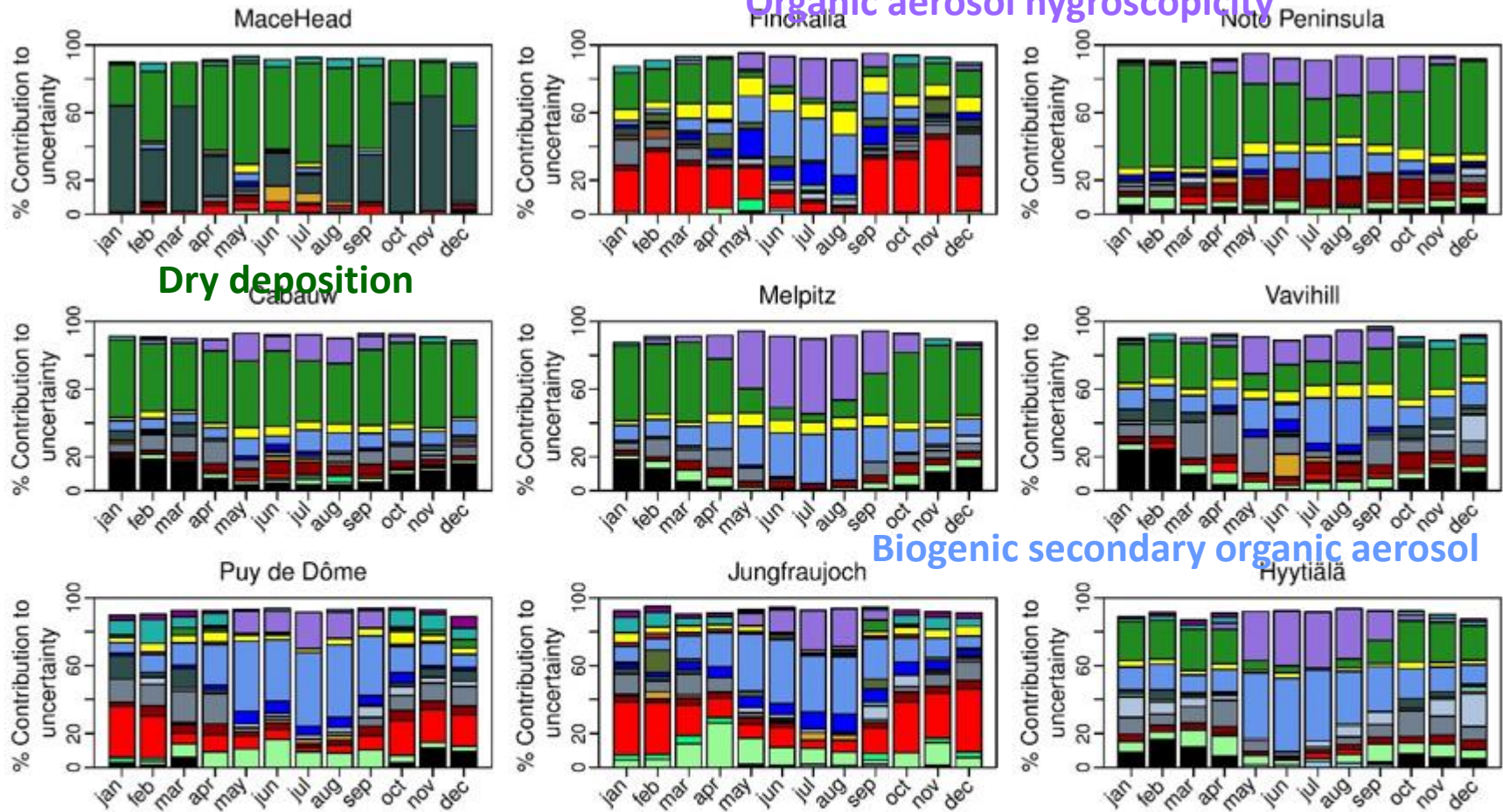


Overall NMB -37%



# Major contributors to model uncertainty – perturbed parameter ensemble

Organic aerosol hygroscopicity



Dry deposition

Biogenic secondary organic aerosol

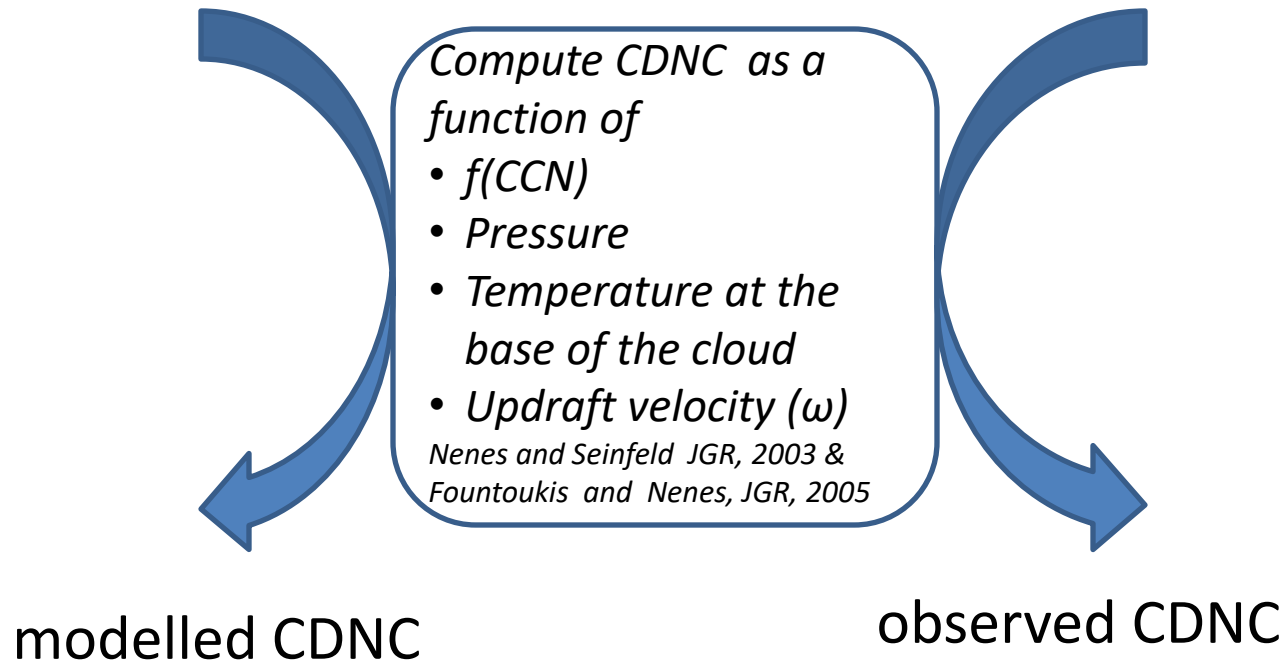


HadGEM3-UKCA  
Yoshioka et al., in  
prep.

# How the CCN uncertainty reflects in CDNC (cloud droplet number concentration) calculations?

modelled CCN spectra  
 $CCN=f(ss)$

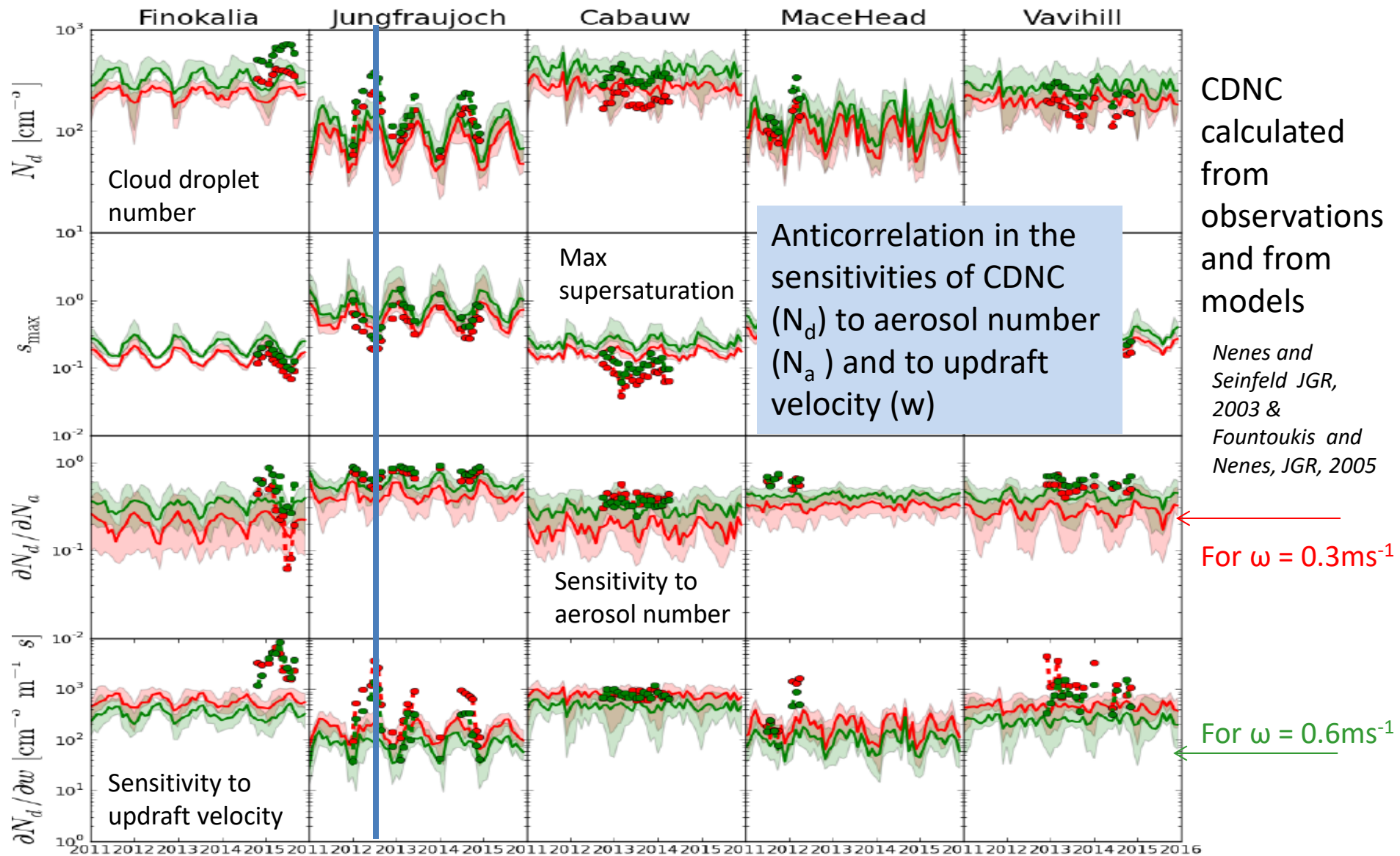
observed CCN spectra  
 $CCN=f(ss)$



$\omega = 0.3\text{ms}^{-1}$  typical for stratiform clouds

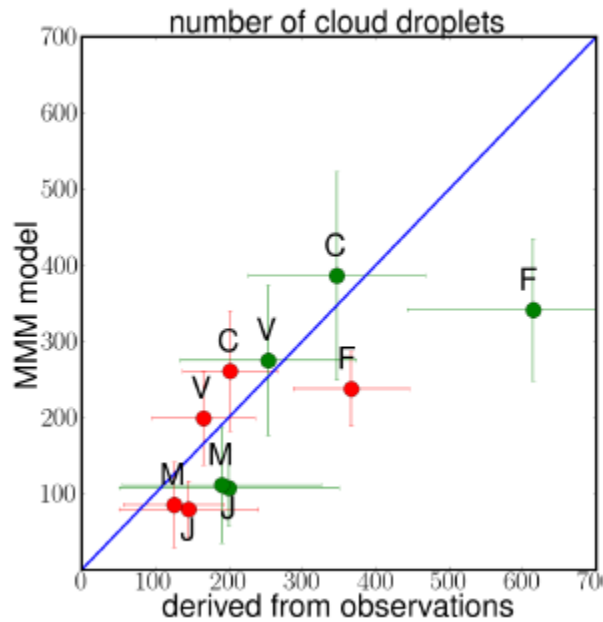
$\omega = 0.6\text{ms}^{-1}$  typical for cumulus clouds

# Cloud droplet number and its sensitivity to aerosol number and updraft velocity



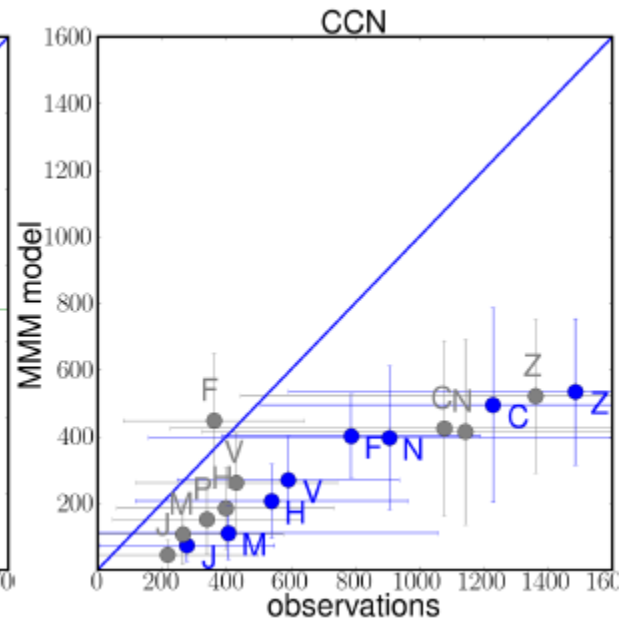


# Cloud droplet number and its sensitivity to aerosol number and updraft velocity



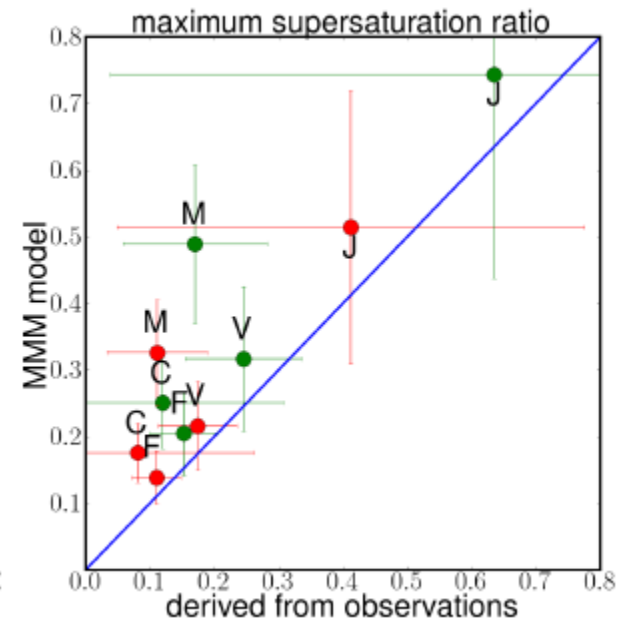
updraft velocity  $0.6 \text{ m s}^{-1}$

updraft velocity  $0.3 \text{ m s}^{-1}$



CCN at supersaturation 0.2%

CCN at max measured supersaturation (0.8%-1.0%)



updraft velocity  $0.6 \text{ m s}^{-1}$

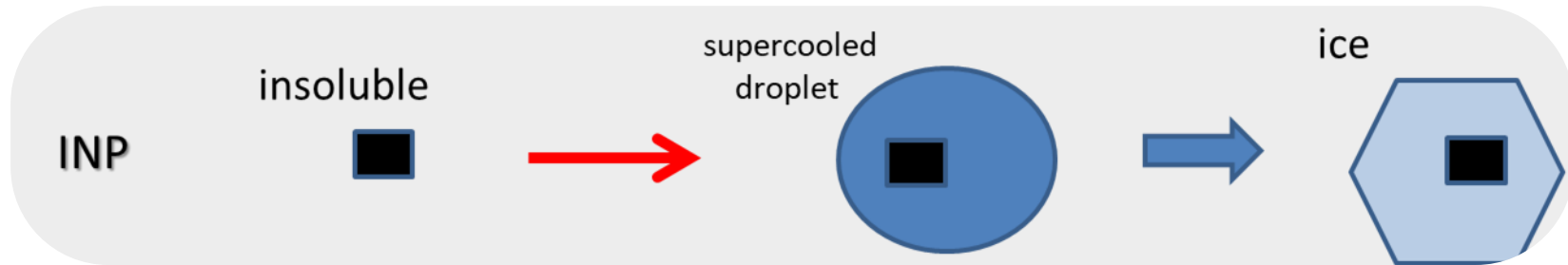
updraft velocity  $0.3 \text{ m s}^{-1}$

*The number of CCN at a prescribed supersaturation cannot be used as indicator of CDNC, as supersaturation is dynamically determined and can vary considerably for a given site*

# Summary

- First comparisons of model results with experimentally derived CDNC.
- **The spread of models for CDNC is smaller than the spread for  $N_a$  and for CCN**
- The sensitivities of CDNC to  $N_a$  and to updraft velocity,  $\omega$ , are negatively correlated. **The variability in  $N_a$  and  $\omega$ , is controlling that of CDNC**
- The models underestimate
  - i) N50, N120, CCN
  - ii) Organic aerosol mass in PM1
- **OA is important contributor: ? to CCN**  
**? to summer time uncertainty in CCN**
- More N3 particles in the models with higher diversity between models over the NH continents than CCN indicating differences in the **size distribution of the primary emissions and/or in the NPF and growth.**

# Ice Nuclei simulations Chatziparaschos et al 2018



Experimental Parameterizations of ice active surface density ( $N_s$ ) are used for the simulations:

INPs :

- Marine OA (ocean biota)



➤ Wilson et al., 2015 (Numbers/TOC)

- Dust (feldspar)



➤ Atkinson et al., 2013 , Niemand et al., 2012,  
Boose et al., 2016 ( $/m^2$ )

- Pollen

- Soot

- Fungal

- Bacteria

(functional groups via hydrogen bonds with -OH, -NH<sub>2</sub>)

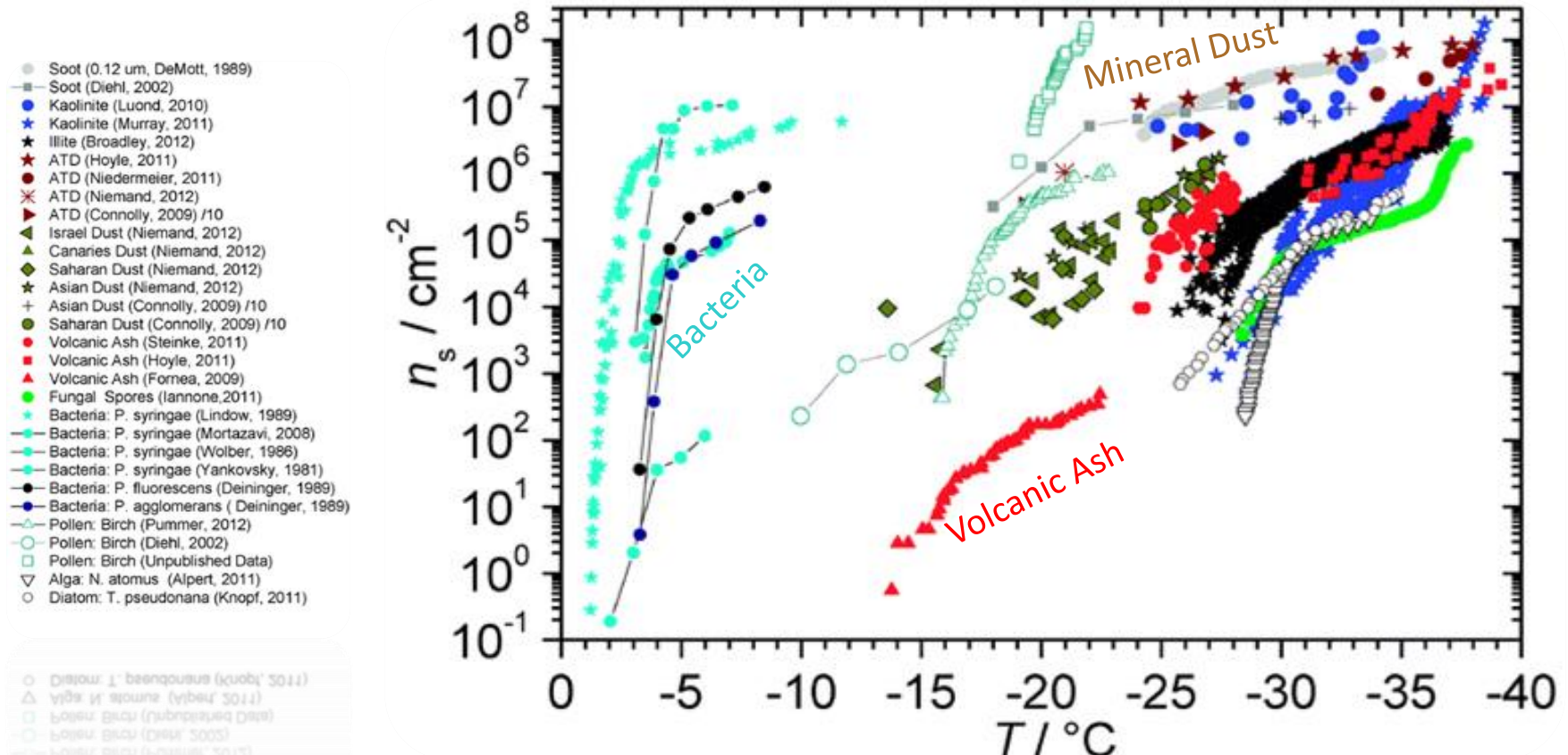
➤ McCluskey et al., 2018 ( Terrestrial) ( $/m^2$ )



# Singular description of Active sites

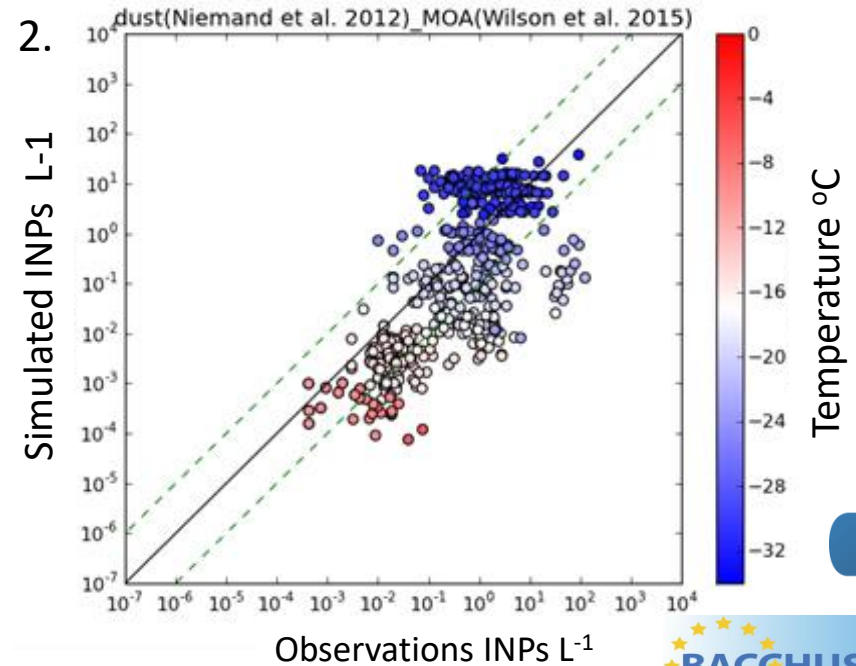
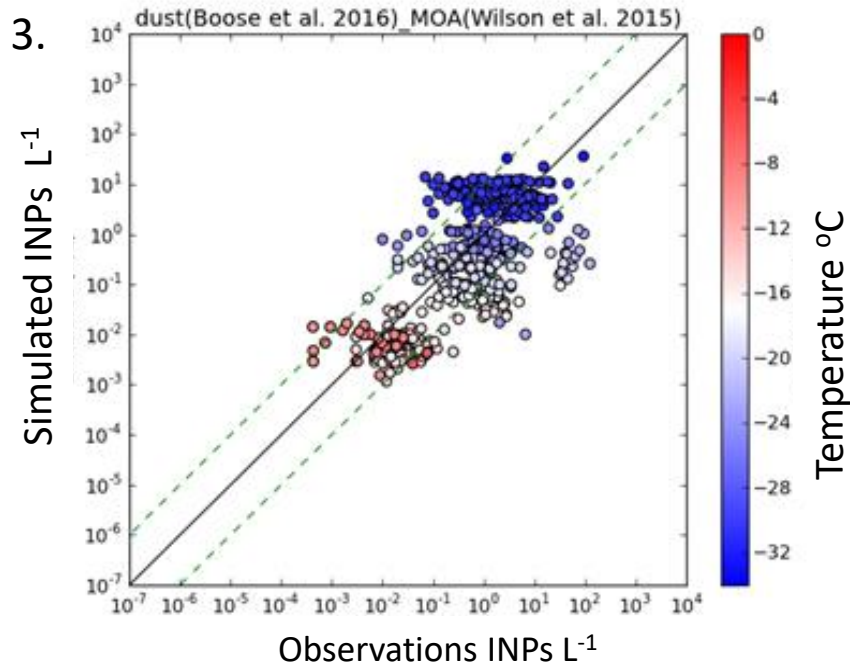
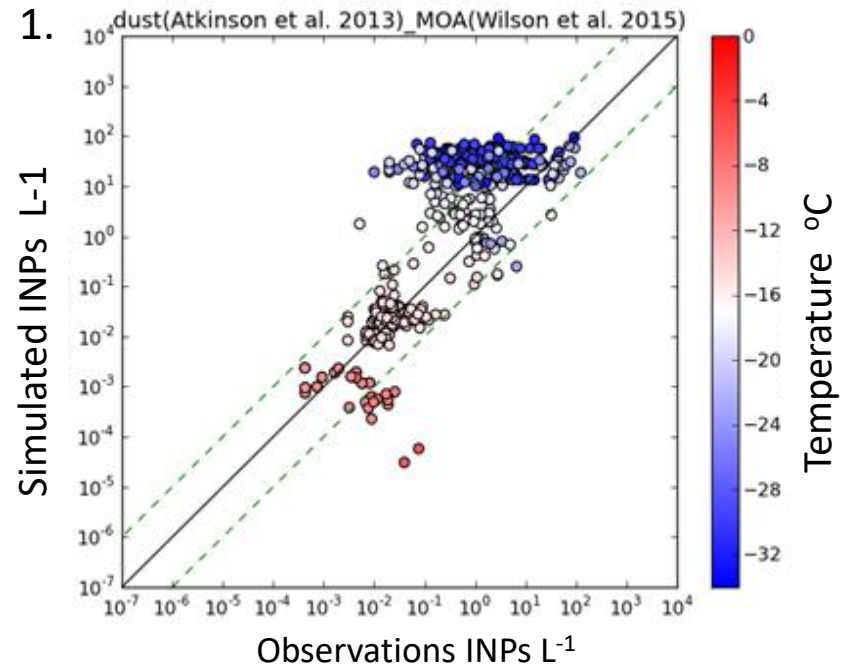
- describes ice active variability of different particles
- ice nucleation based on Temperature  $n_s(T)$
- **experimentally derived**
- $T_c$  is the critical temperature below which the multiple active sites present on an IN surface activate to form ice

**Active sites** : surface has steps and cavities as a result of mechanical fracture of natural weathering, which have functional groups Hydroxyl–OH and act as hydrophilic sites (Freedman et al., 2015)



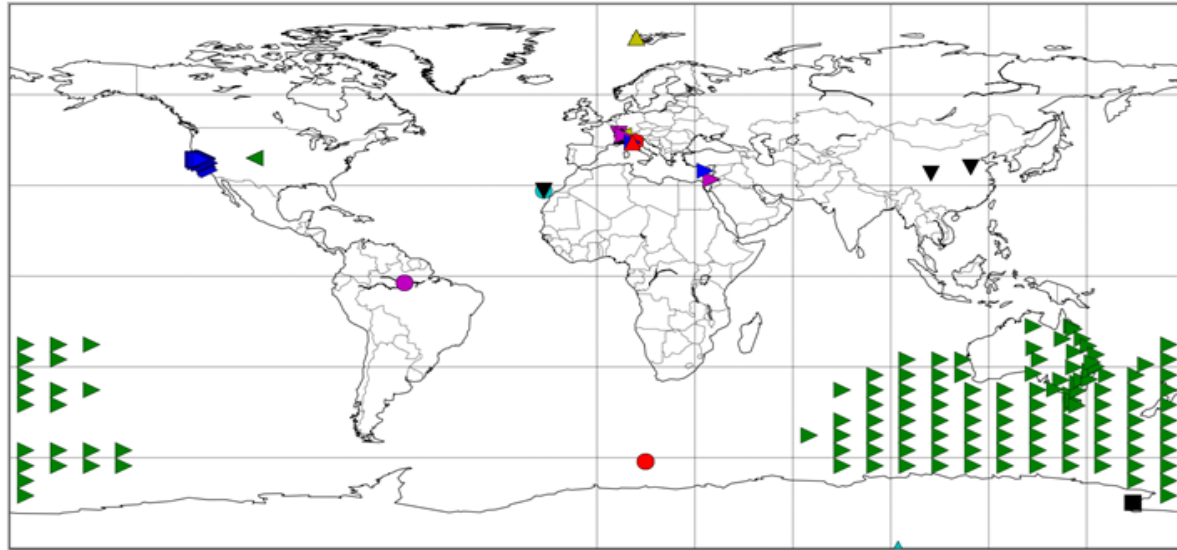
# Testing parameterizations for dust INPs

INPs category	Equation	Reference
Marine OA	$n_m = e^{11.2186 - (0.4459 * T_c)}$	Wilson et al., 2015
Dust-Feldspar	$n_s = e^{(-1.038 * T_k + 275.26)}$	Atkinson et al., 2013
	$n_s = e^{(-0.517 * (T_k - 273.15) + 8.934)}$	Niemand et al., 2012
	$n_s = e^{(-0.33 * (T_k - 273.15) + 15.64)}$	Boose et al., 2016



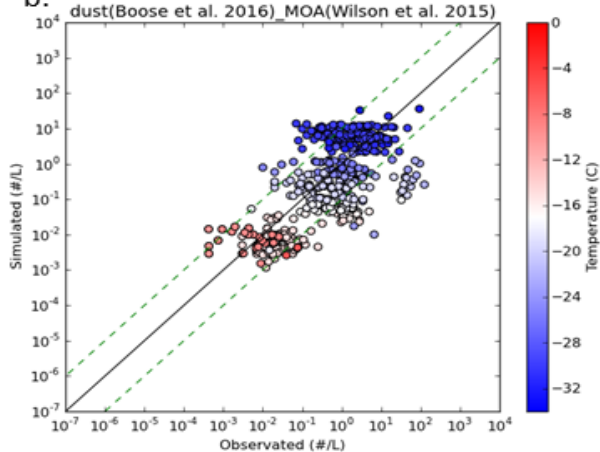
# Accounting only for marine organics and dust INPs : missing sources

a.

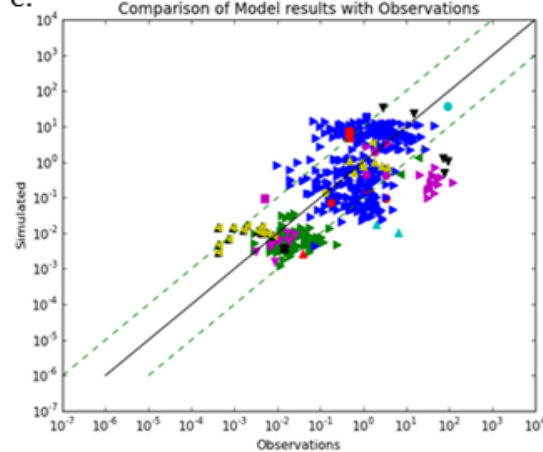


- bigg73\_1969\_1972.xls
- Abbatt\_2013\_Ucluelet.
- BACCHUS\_FRIDGE\_MARTIN
- conen\_Chauumont\_Jun\_20
- conen\_JFJ\_2012\_jun.xl
- conen\_JFJ\_3580m\_JUN\_2
- DeMott\_Calwater\_2011.
- ETHCALIMA2013\_Aug.xls
- ETHCALIMA2014\_Aug.xls
- ETHCLACE2012\_Jan.xls
- ETHCLACE2013Feb.xls
- ETHCLACE2014\_Jan.xls
- ISAC\_CNR\_2012\_Antarct
- ISAC\_CNR\_May\_2014\_San
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- KAD\_SouthPole\_2009.xl
- San\_PietroCap2014\_Feb
- Yin\_China\_2012.xls
- BACCHUS\_FRIDGE\_AMAZON
- BACCHUS\_FRIDGE\_SVALBA
- CSU\_CFDC\_archive\_BEAC
- ISAC\_CNR\_May\_2014\_MtC
- ISAC\_CNR\_Oct\_2015\_MtC
- ISAC\_CNR\_August\_2015\_
- BACCHUS\_CHARMEX\_CYPRU

b.

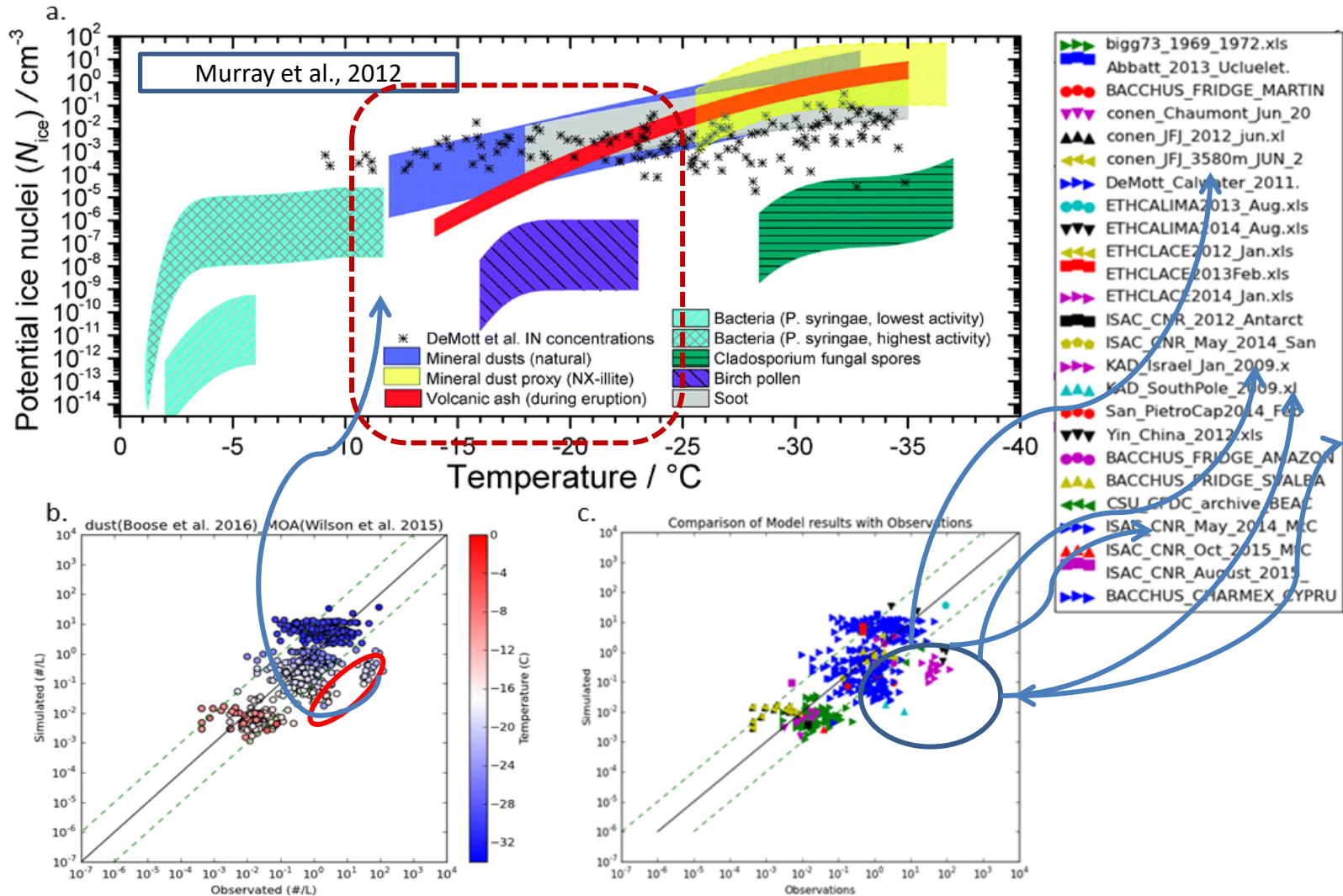


c.



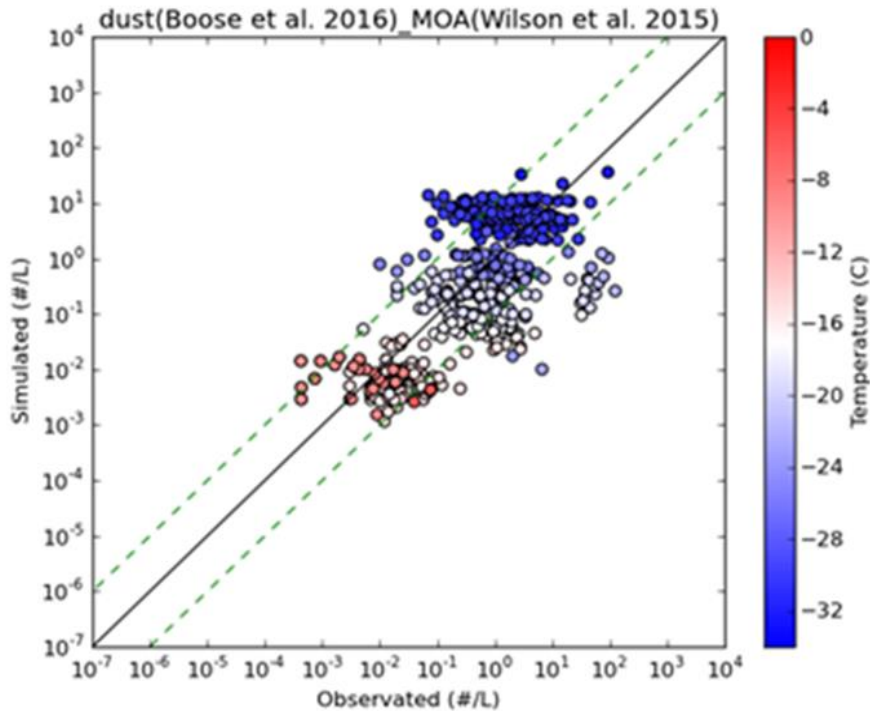


# Accounting only for marine organics and dust INPs : missing sources

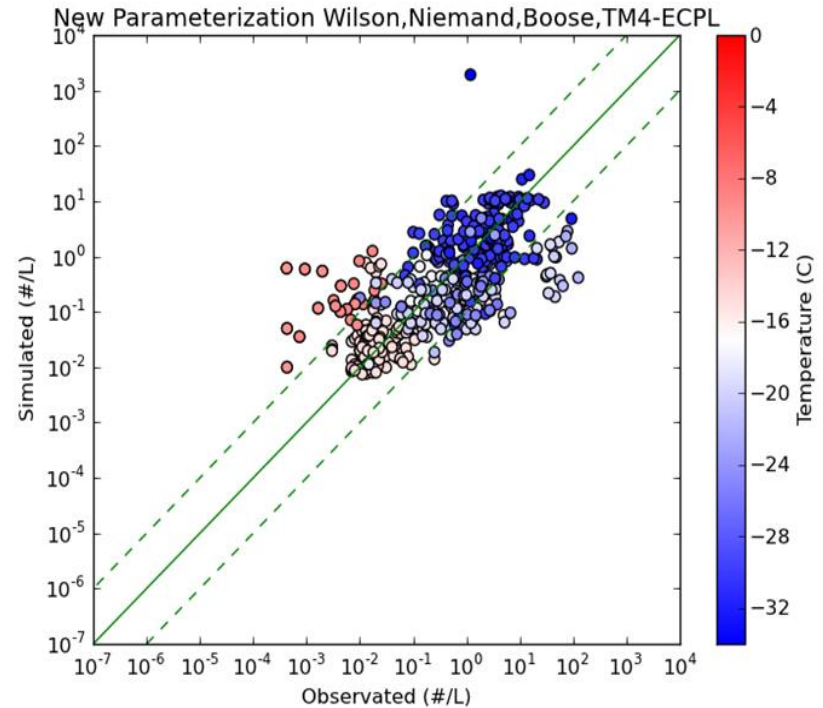


# Improvements: adding terrestrial bioaerosol

Dust feldspar & marine aerosol



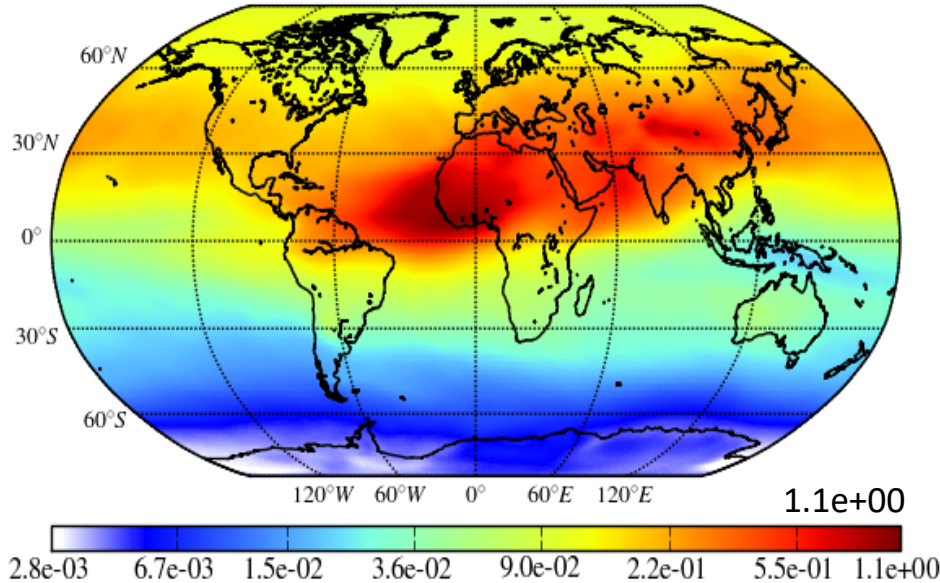
Adding also terrestrial bioaerosol



Using McCuskey et al., 2018  
based on number of insoluble  
bacteria, pollen, fungi

# Annual Mean Distribution of INPs

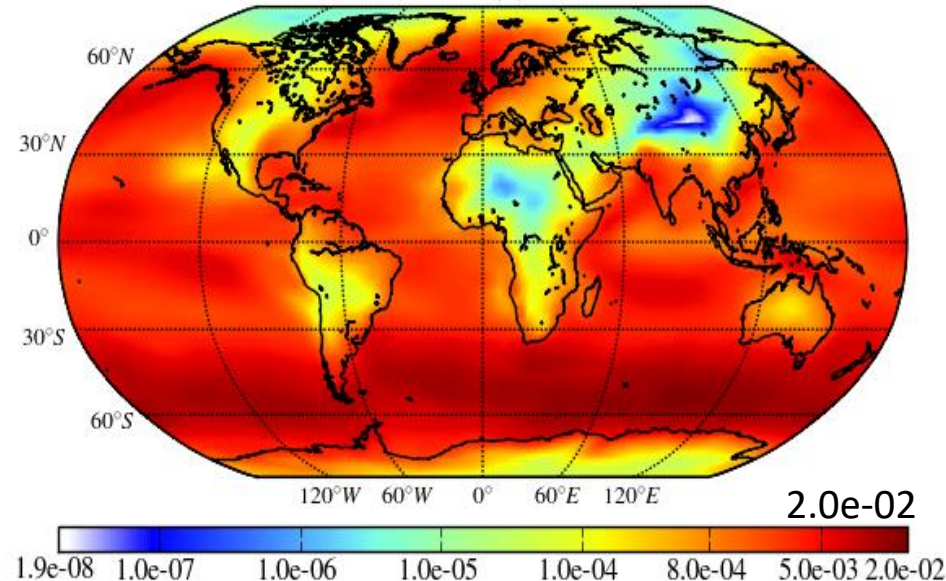
$\text{INPs}_{dust} [L^{-1}]$   $T_{-20(C)}$  600 hPa Annual



## INPs from mineral Dust:

- Dominate across Northern Hemisphere, North Africa (Sahara), Asia (Gobi)
- More abundant in number than marine INPs
- Transported to mid and high-altitudes
- Affect regions far from their emission point

$\text{INPs}_{marine} [L^{-1}]$   $T_{-20(C)}$  850 hPa Annual



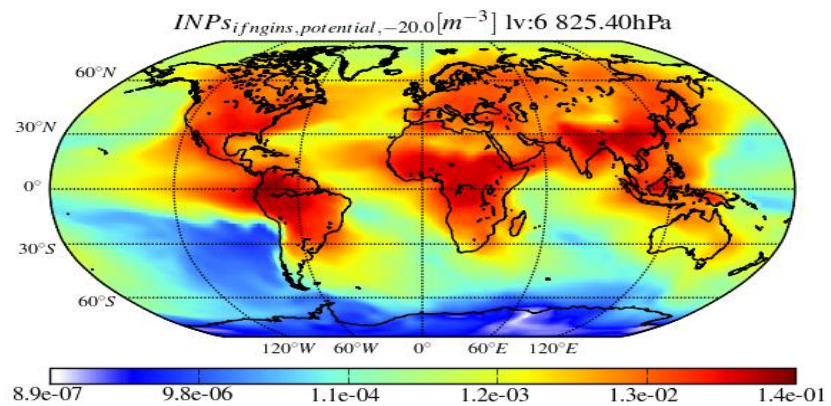
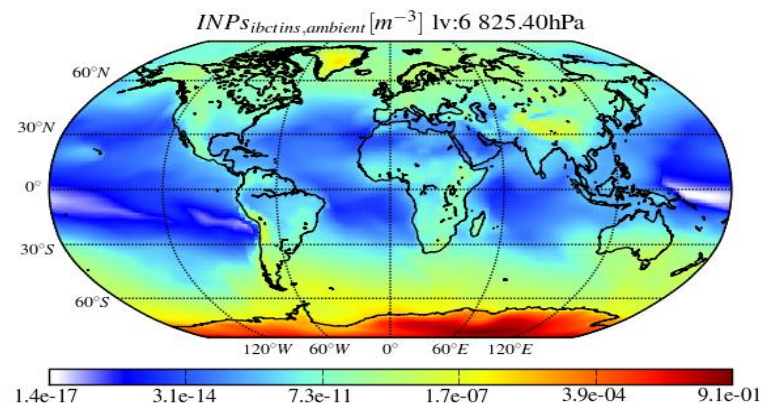
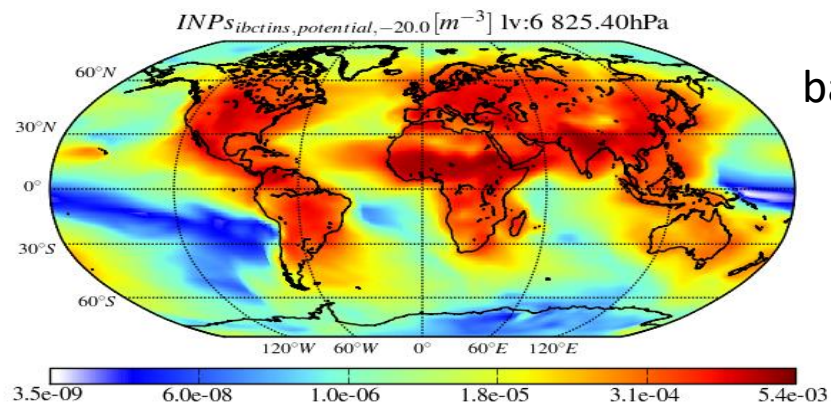
## INPs from Marine Organic:

- Dominate across Southern Hemisphere, (remote oceans)
- Several spots → oceanic biota activity
- Generally lower number concentration than dust INPs
- Affect regions close to their emissions
- Important in coastal regions → (North East Coast of America -North Atlantic) → comparable concentrations with dust INPs

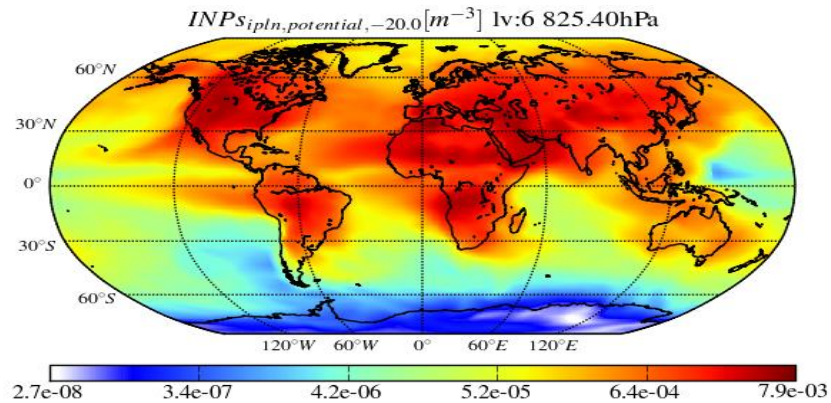
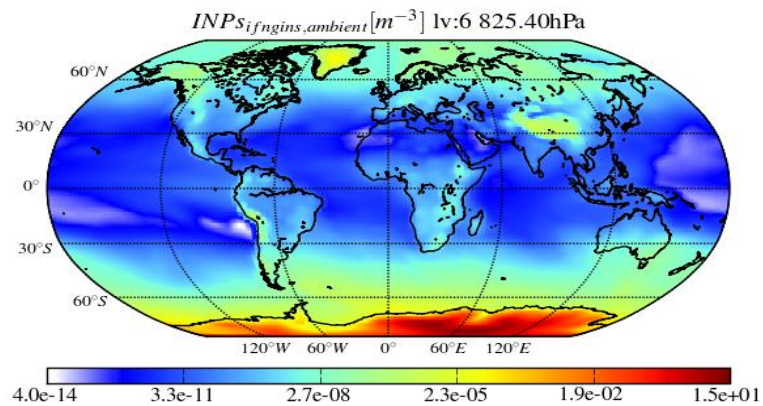
Simulation year 2008



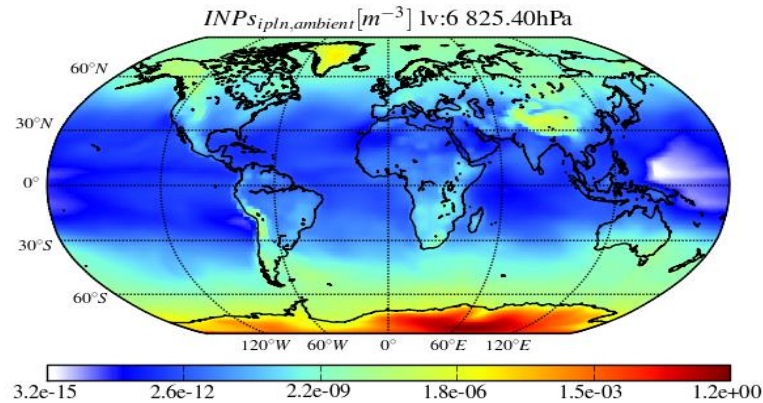
at -20C ← [INP] → at ambient temperature



fungi



pollen





# Perspectives of INP/CCN modeling

- further implemented parameterizations for INPs missing sources improve discrepancies between model and measurements
- the simulated number concentrations of CCN and of INPs are compared and their significance for precipitation rates, cloud lifetime and cloud coverage and albedo will be investigated.

