



# Towards an improved representation of IN particle sources in models: adding the missing pieces

July 27, 2020

**Susannah M. Burrows**  
1<sup>st</sup> virtual INP colloquium



PNNL is operated by Battelle for the U.S. Department of Energy





## Office of Science Laboratories

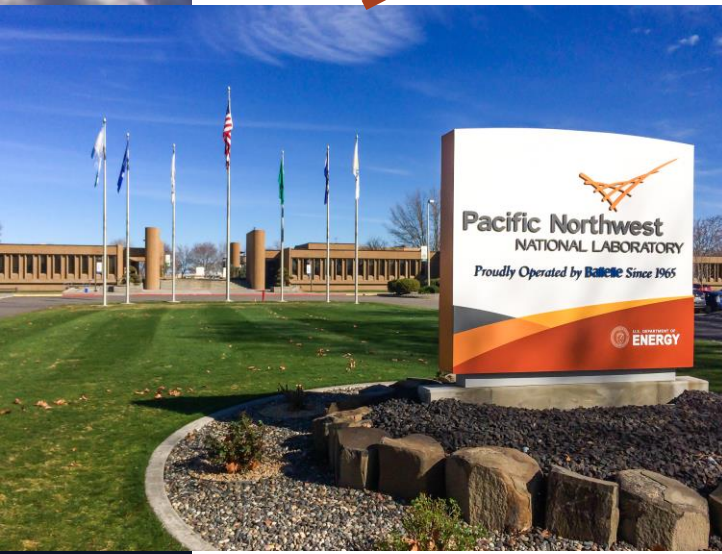
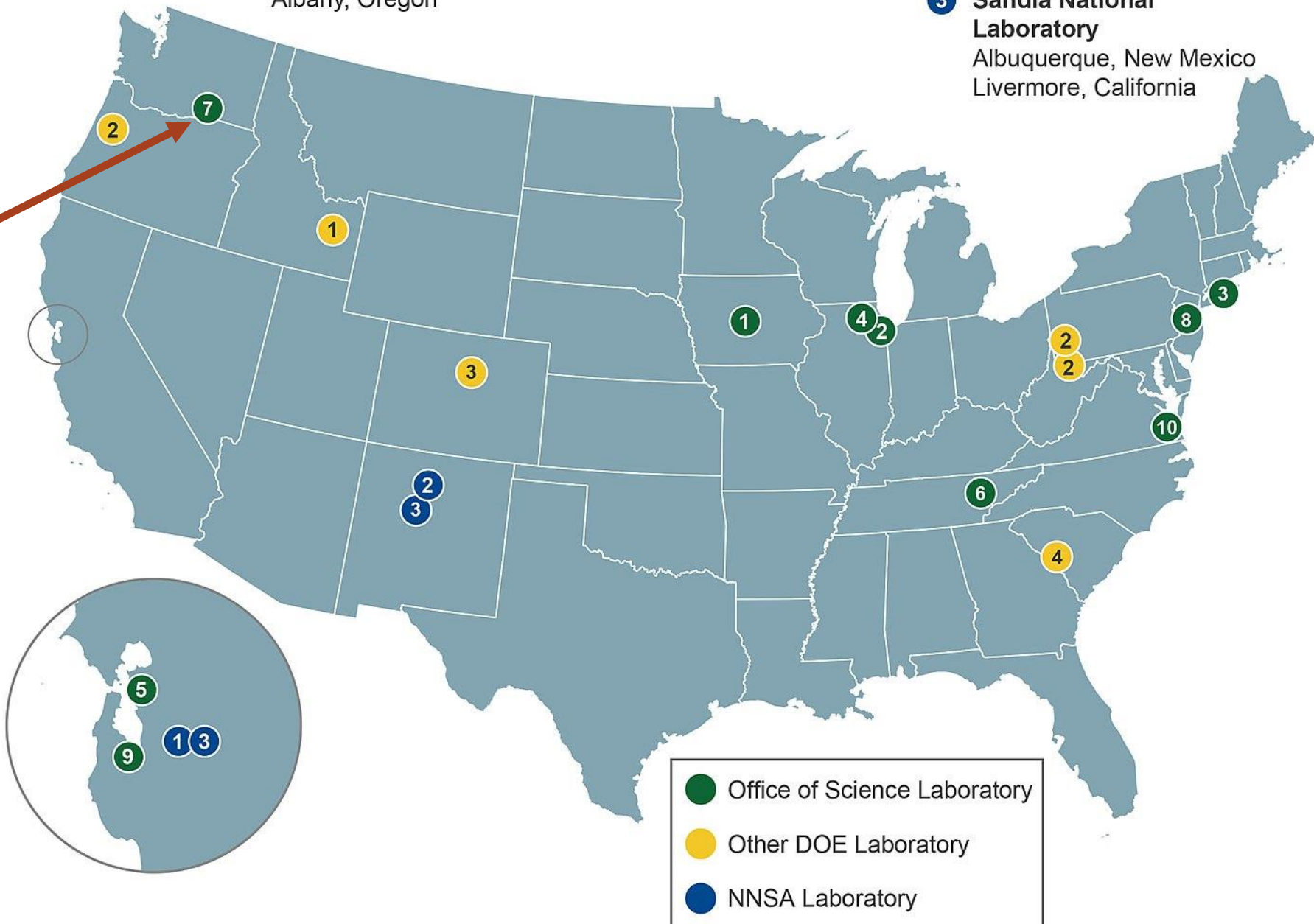
- 1 Ames Laboratory  
Ames, Iowa
- 2 Argonne National Laboratory  
Argonne, Illinois
- 3 Brookhaven National Laboratory  
Upton, New York
- 4 Fermi National Accelerator Laboratory  
Batavia, Illinois
- 5 Lawrence Berkeley National Laboratory  
Berkeley, California
- 6 Oak Ridge National Laboratory  
Oak Ridge, Tennessee
- 7 Pacific Northwest National Laboratory  
Richland, Washington
- 8 Princeton Plasma Physics Laboratory  
Princeton, New Jersey
- 9 SLAC National Accelerator Laboratory  
Menlo Park, California
- 10 Thomas Jefferson National Accelerator Facility  
Newport News, Virginia

## Other DOE Laboratories

- 1 Idaho National Laboratory  
Idaho Falls, Idaho
- 2 National Energy Technology Laboratory  
Morgantown, West Virginia  
Pittsburgh, Pennsylvania  
Albany, Oregon
- 3 National Renewable Energy Laboratory  
Golden, Colorado
- 4 Savannah River National Laboratory  
Aiken, South Carolina

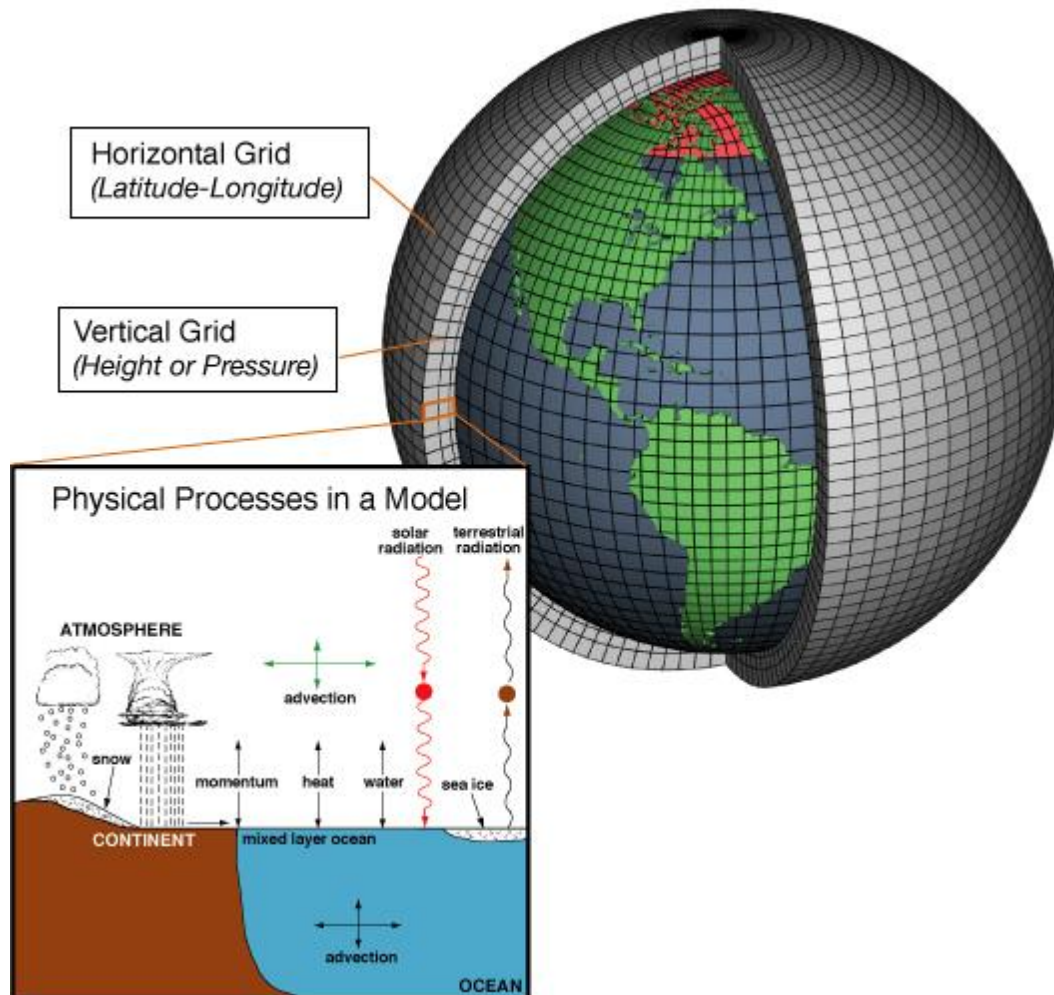
## NNSA Laboratories

- 1 Lawrence Livermore National Laboratory  
Livermore, California
- 2 Los Alamos National Laboratory  
Los Alamos, New Mexico
- 3 Sandia National Laboratory  
Albuquerque, New Mexico  
Livermore, California



- Office of Science Laboratory
- Other DOE Laboratory
- NNSA Laboratory



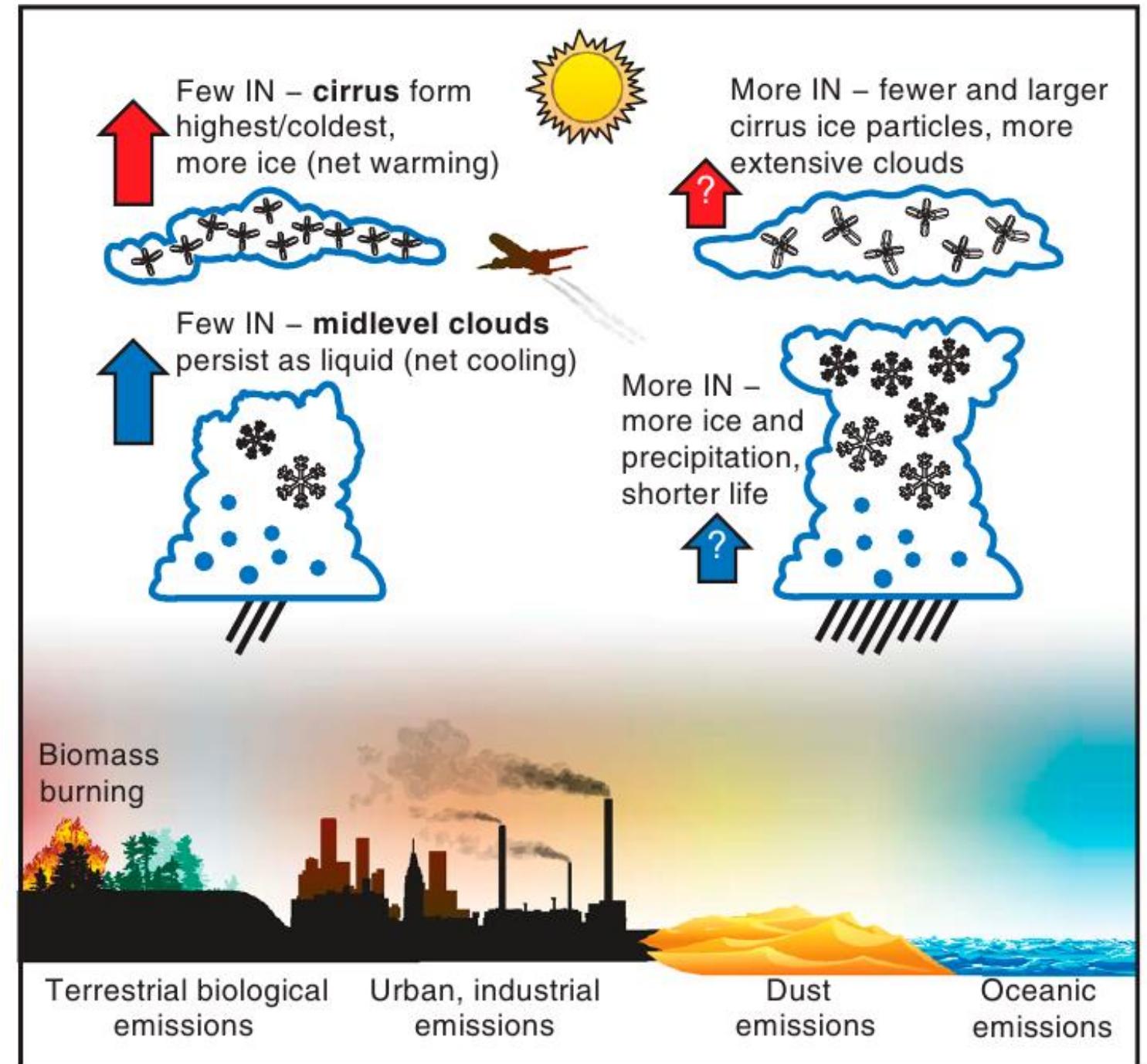


Earth system  
dynamics & local  
process  
representations



# Climate effects and sources of atmospheric ice nuclei

- Freezing by ice-nucleating particles affects:
  - Cloud phase
  - Ice crystal number concentration
  - Formation of precipitation
  - Convective invigoration(?)
  - Lifetime/persistence of clouds (e.g. anvils)
  - Cloud radiative effects



# Most precipitation is formed in clouds containing ice

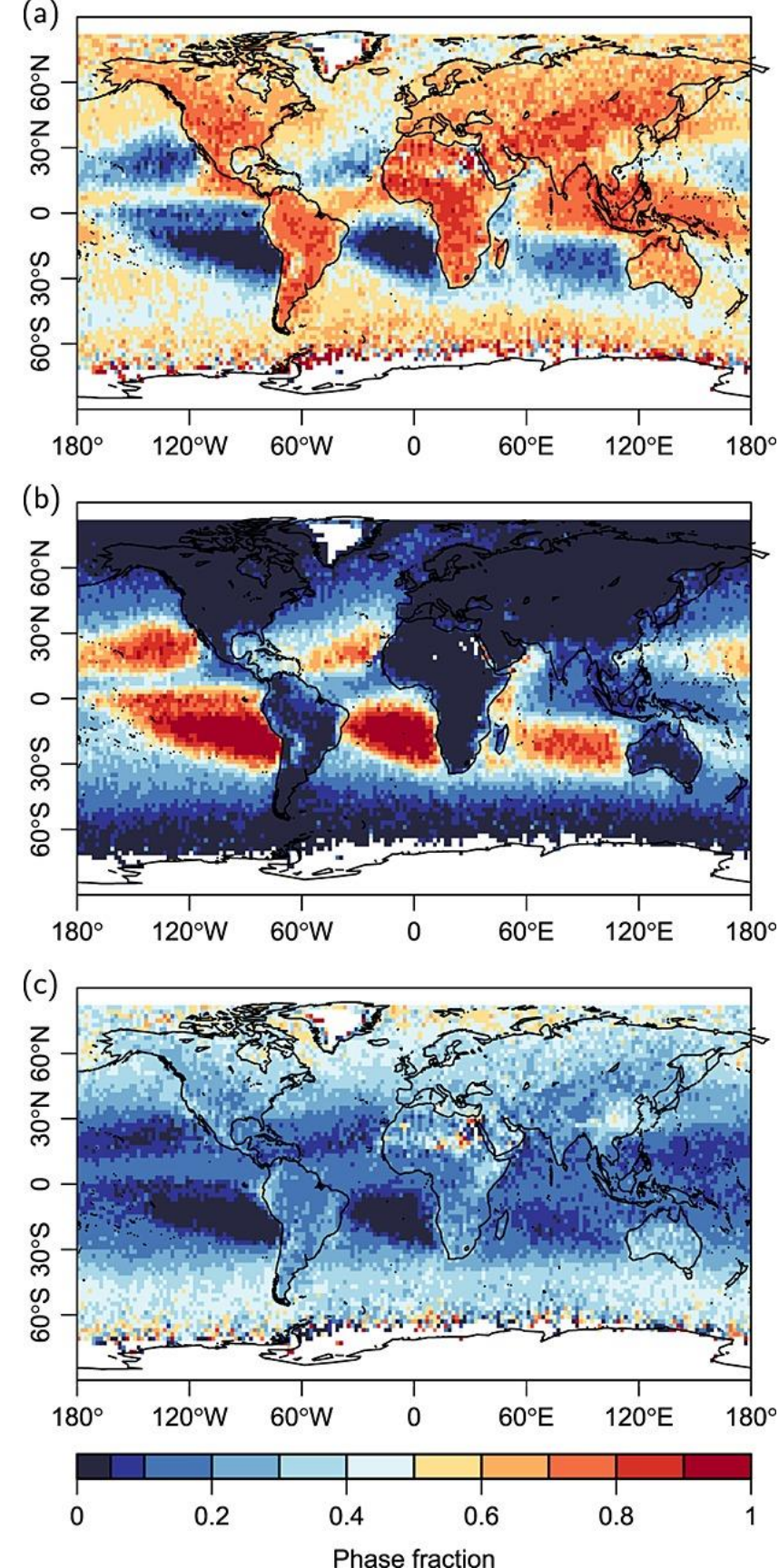
Fraction of raining clouds that are

- (a) ice-phase,
- (b) liquid-phase, and
- (c) mixed-phase

...at cloud **top**.

...averaged over 5 years of collocated  
CloudSat–CALIPSO data (2006-2011)

Mülmenstädt, et al., 2015



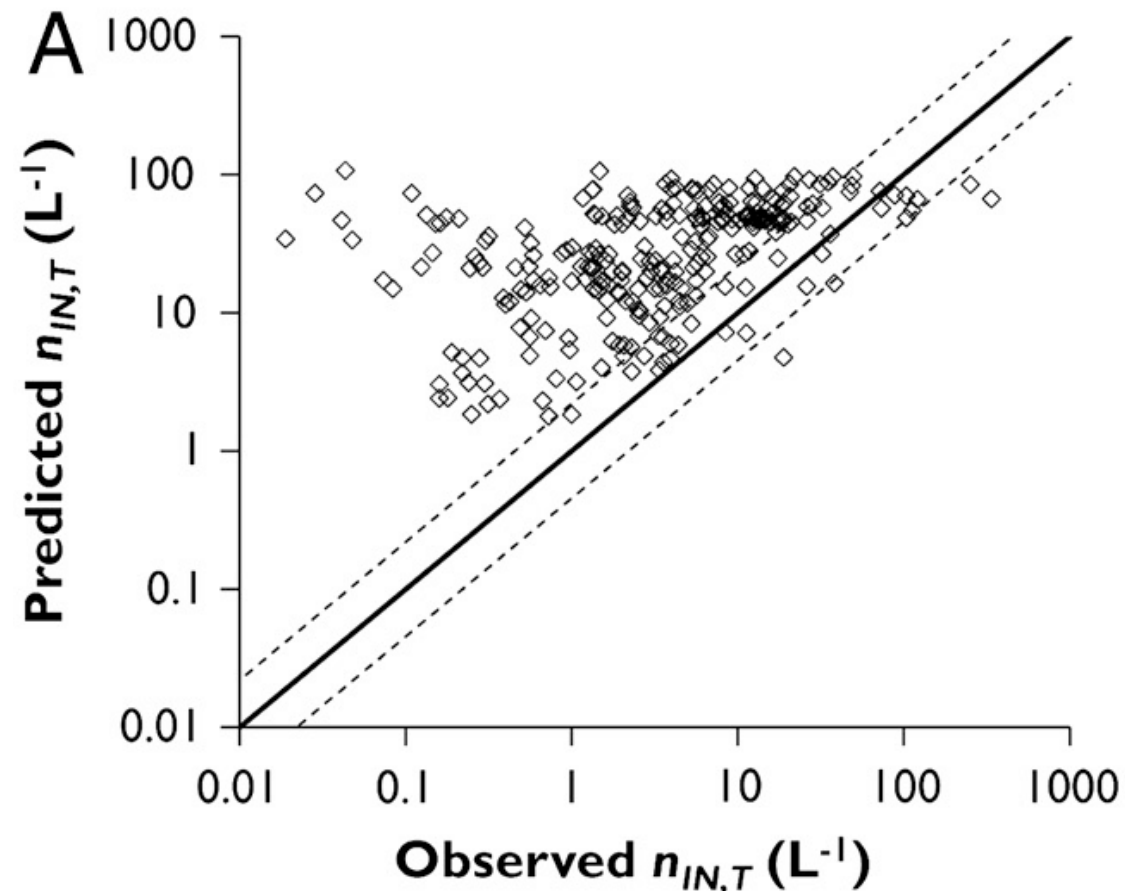


# Aerosols are responsible for much of the variance in INP concentrations

- INP concentrations vary by several orders of magnitude, and much of this variance can be explained by variations in aerosol
- Very few atmospheric particles are effective INP:  $\sim 1$  per  $10^5 - 10^6$
- Understanding which particles contribute to INP is important to better prediction of their numbers, temporal and spatial variability, and potential response to global change

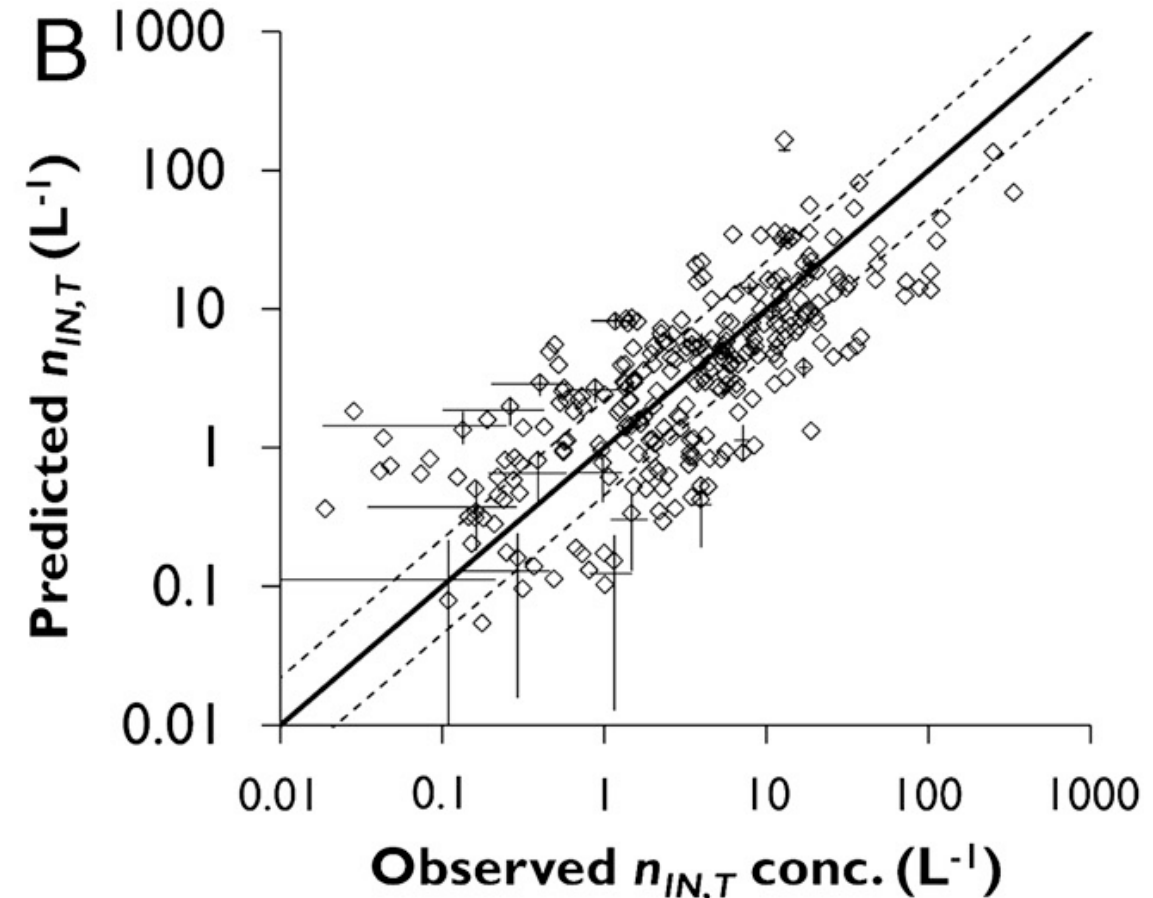
Meyers et al. 1992

$$N_{id} = N_0[(S_i - 1)(S_0 - 1)^{-1}]^b \exp(aT_{\text{sup}}).$$



DeMott et al., 2010, PNAS

$$n_{IN,T_k} = a(273.16 - T_k)^b (n_{\text{aer},0.5})^{(c(273.16 - T_k) + d)},$$



# A measurements-to-models approach to advance understanding of ice-nucleating particles

Aim: Improve **predictive understanding** of INP variability:

- We **understand** the particle sources and atmospheric processes that control INP variability at different places and times.
- We can use this understanding to **skillfully predict** INP concentrations in models.

Laboratory experiments probe processes, identify key drivers, and constrain parameter values. Models quantify atmospheric relevance / importance of proposed processes.

**Atmospheric & Process Models**

Models identify where and when observations will be most useful in testing hypotheses and constraining models. Challenging models with observations identifies limitations and knowledge gaps.

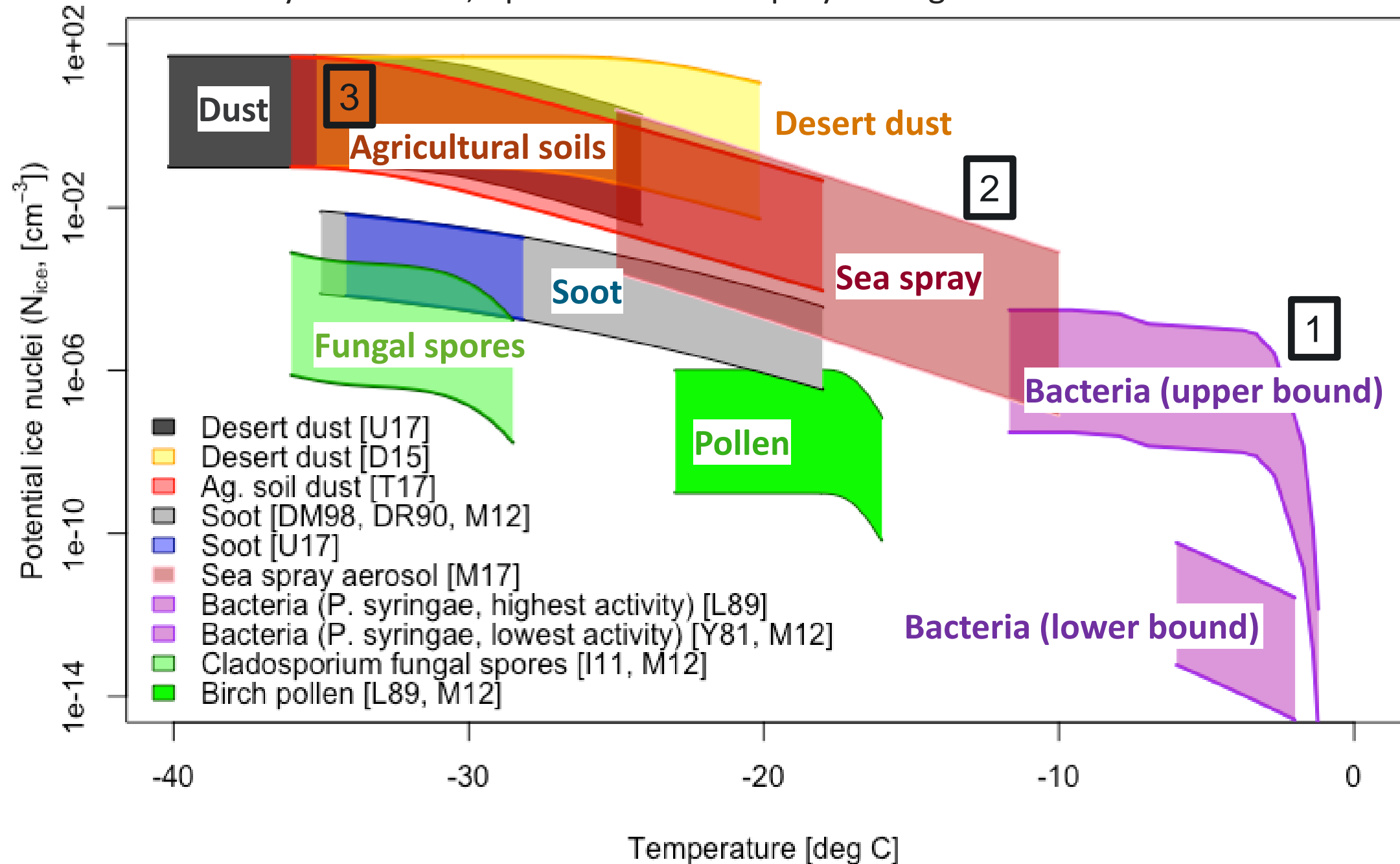
**Laboratory Experiments**

**Remote Sensing & Field Studies**

Laboratory analyses facilitate interpretation of field observations. New hypotheses to explain field observations can be tested in the laboratory.

# Which particles drive immersion-mode INP number for mixed-phase clouds?

Based on Murray et al. 2012; updated with sea spray and agricultural soil sources



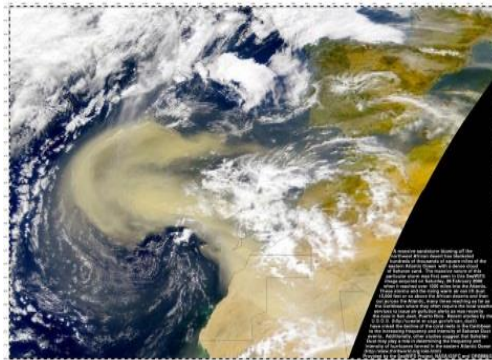


# Missing piece 1: Airborne bacteria (??)

Certain bacteria (e.g. *Pseudomonas syringae*) are known to be effective ice nucleators.

Goal: to place upper/lower bounds on the potential of bacteria-bearing particles to contribute to atmospheric INPs.

## Real-world mechanisms of bacteria emissions



Lofted with dust and soil



Shaking leaves

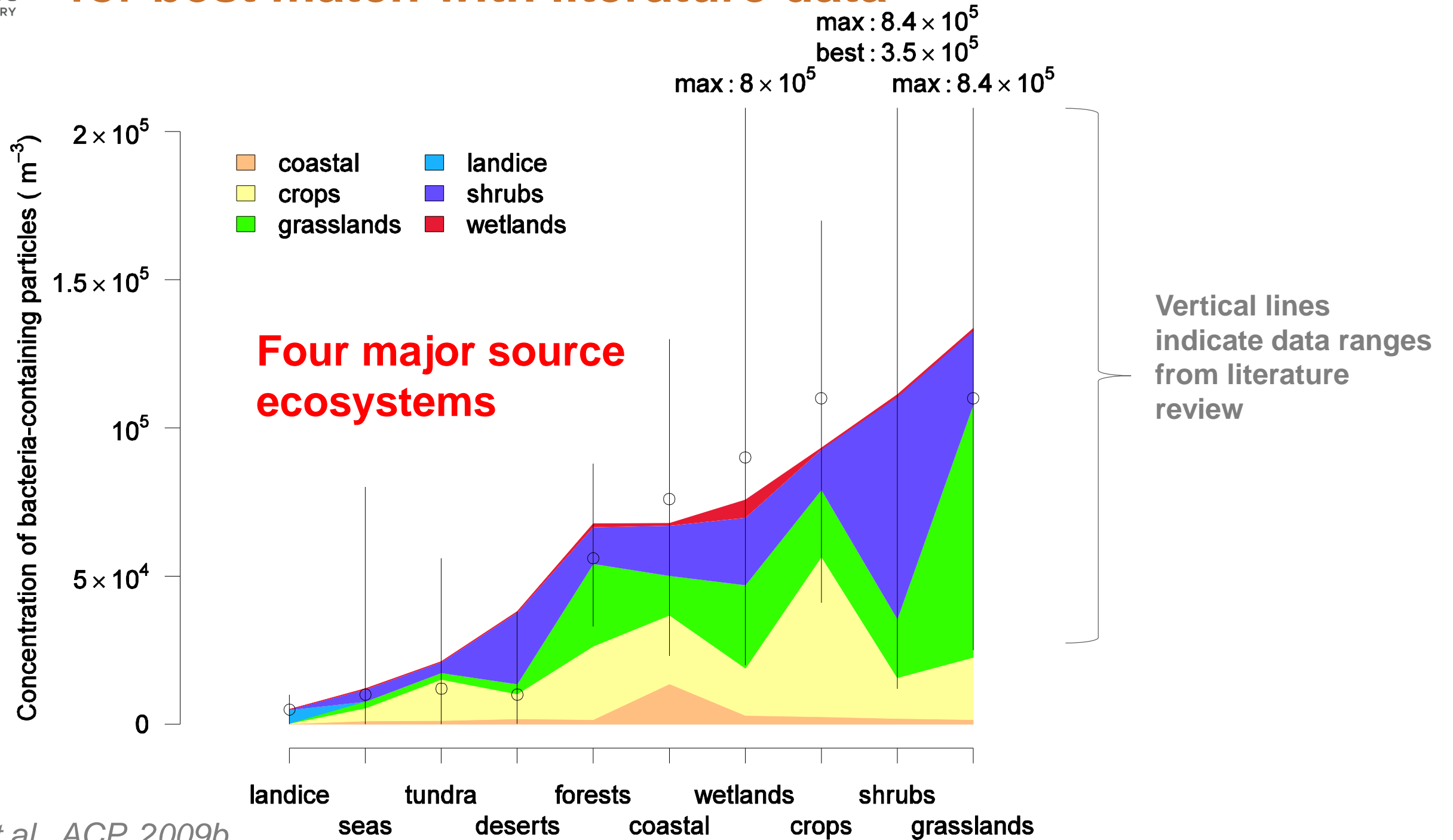


Sea spray





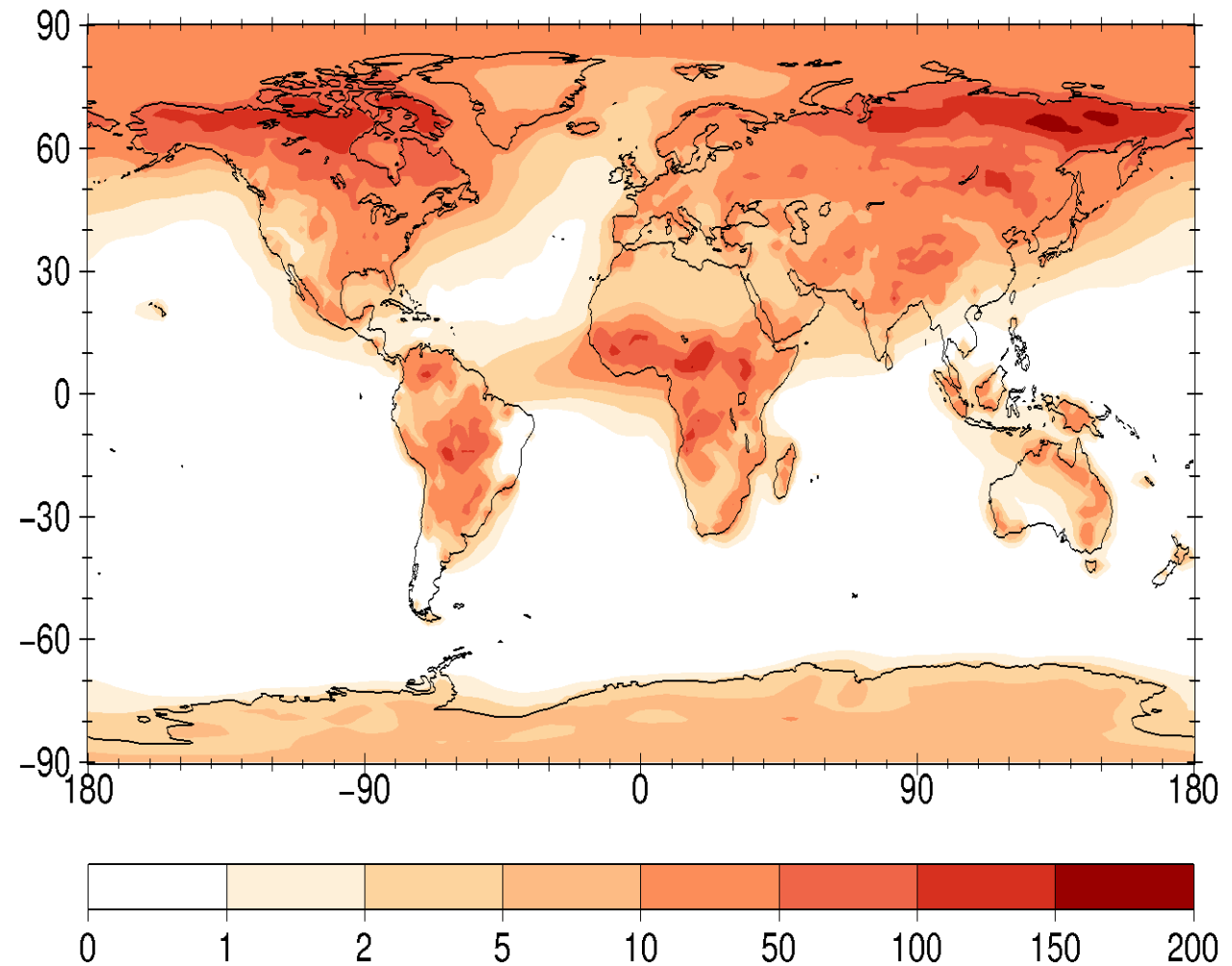
# Simulated bacteria-containing particle concentrations in near-surface air by ecosystem, after optimizing fluxes for best match with literature data



# Global estimates of bacteria emissions

	Ensemble mean	Ensemble range (5%-95%)
Mean emissions from land (particles $m^{-2} s^{-1}$ )	250	140 - 380
Mean emissions from seas (particles $m^{-2} s^{-1}$ )	0	0 - 226
Global emissions (particles per year)	$1.4 \times 10^{23}$	$(7.6 - 35) \times 10^{23}$
Global emissions (Gg per year)	740	400 - 1800

**Modeled concentration of bacteria-containing particles ( $10^6$  per  $m^{-3}$ )**





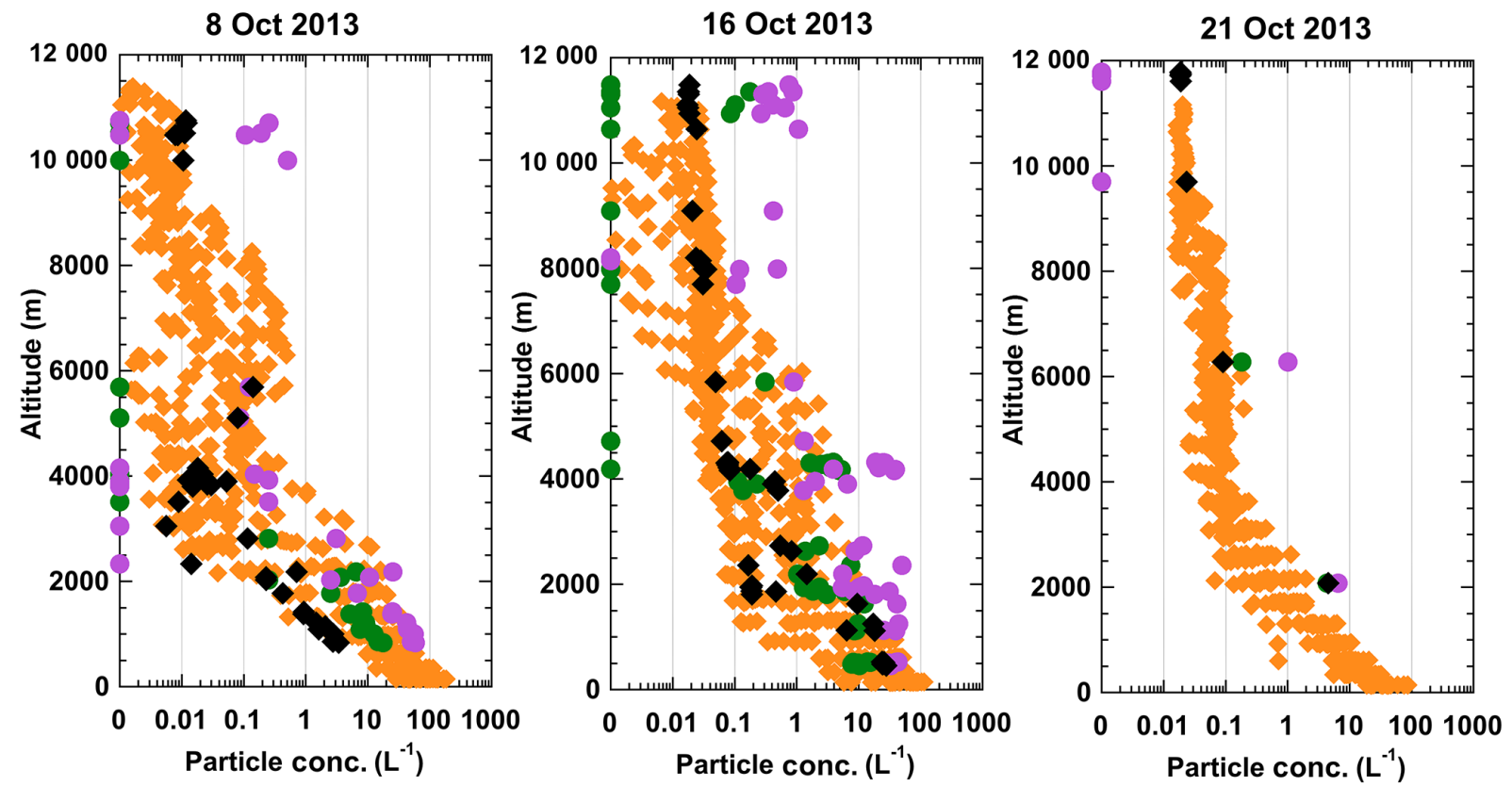
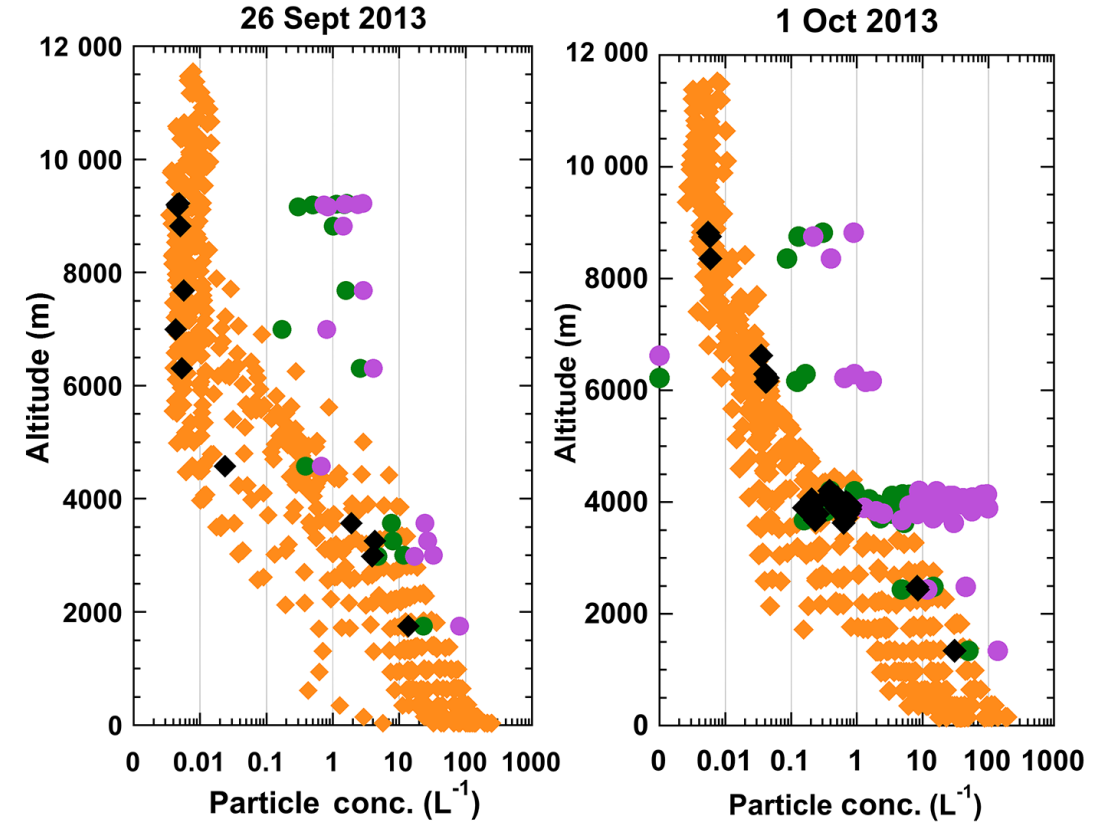
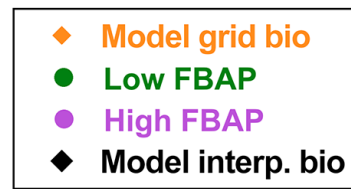
# Comparison of models with airborne measurements of biological particles

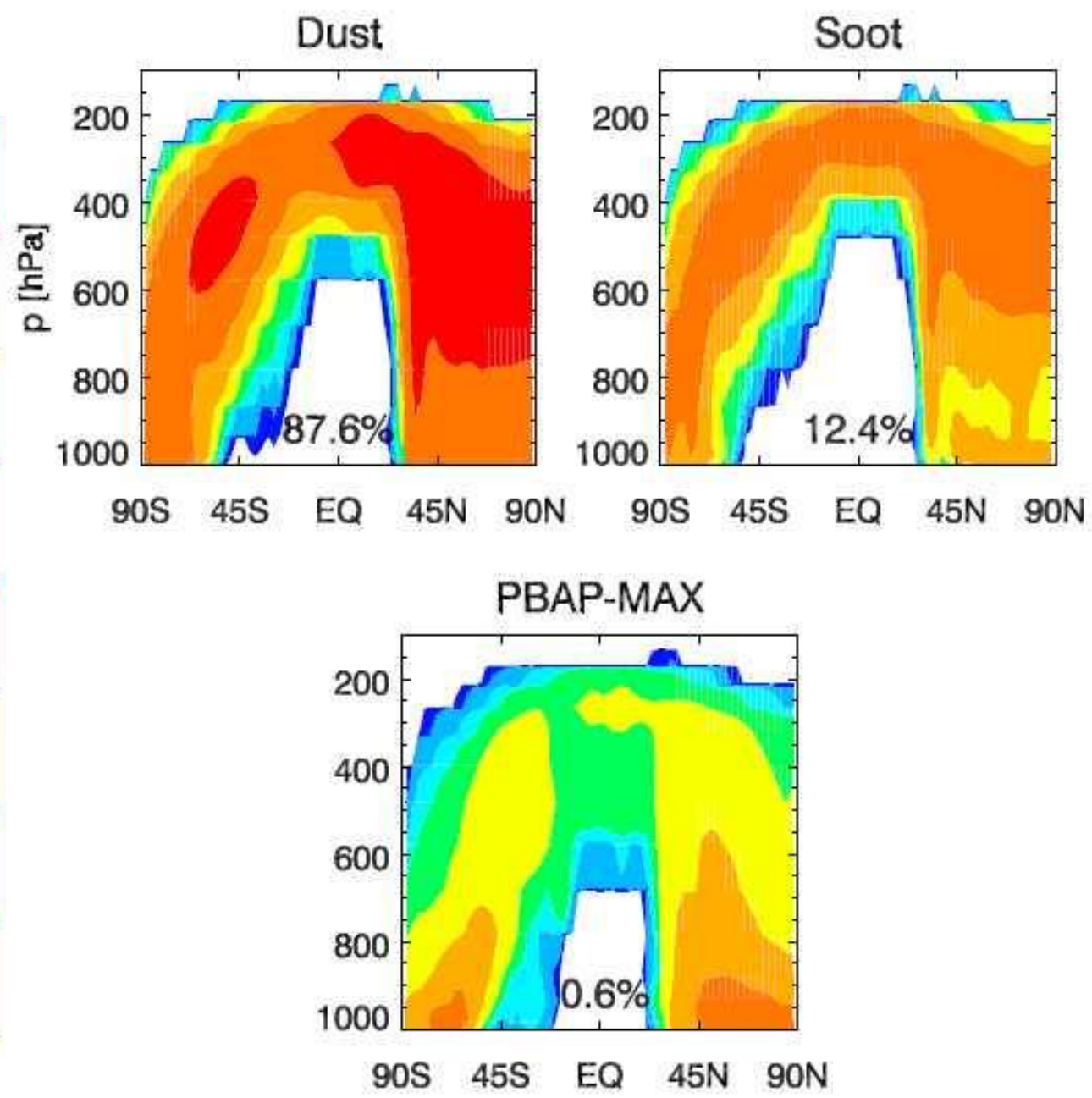
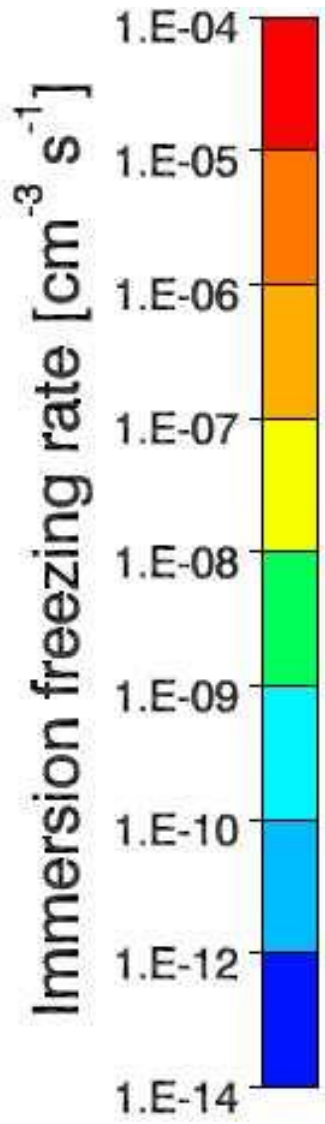
FBAP =  
“Fluorescent Biological  
Aerosol Particles”

Real-time measurement  
of particle fluorescence  
and aerodynamic size by  
WIBS instrument.

Over US Western Plains  
in autumn.

*Twohy et al., 2016, ACP*



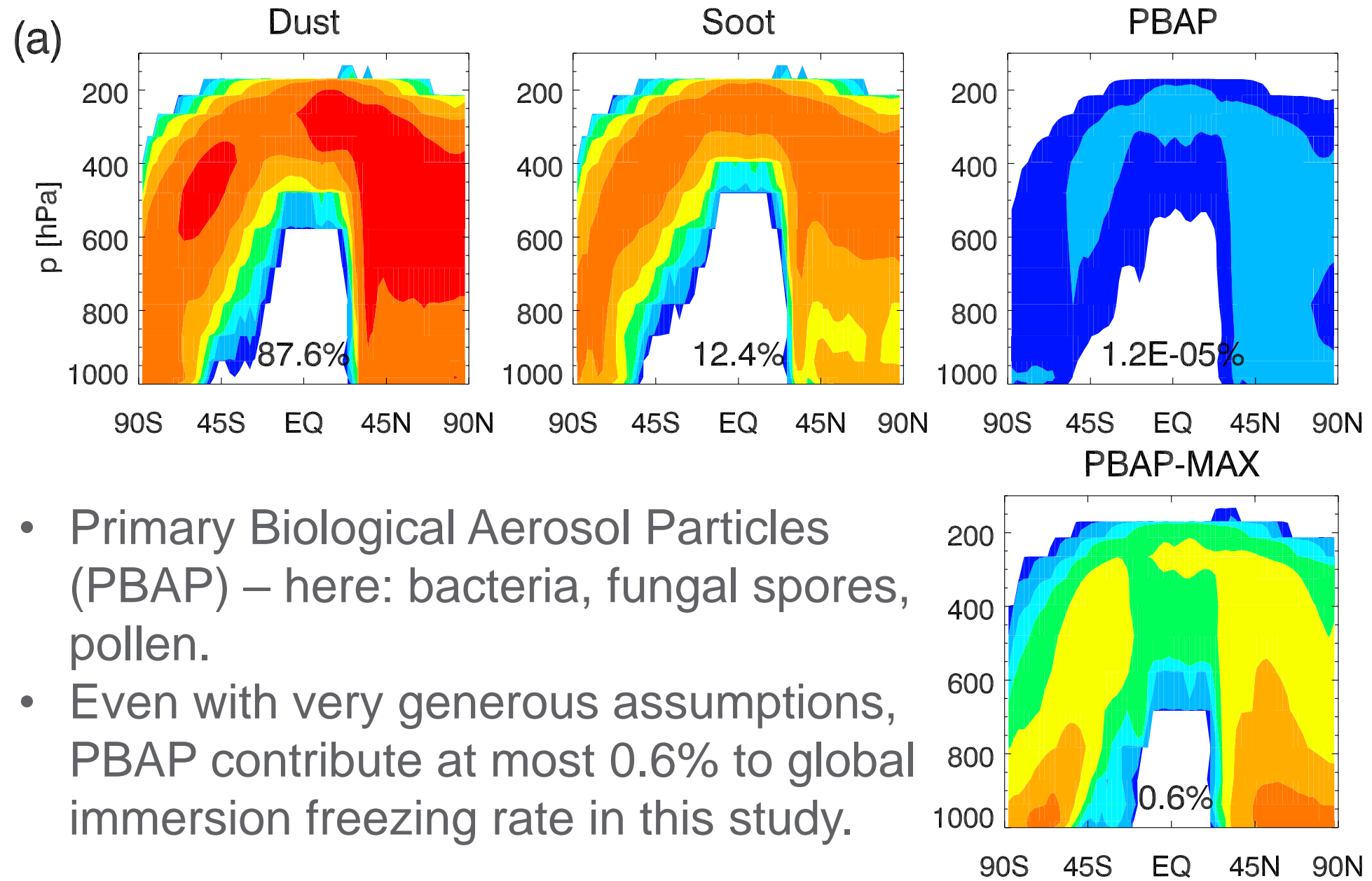
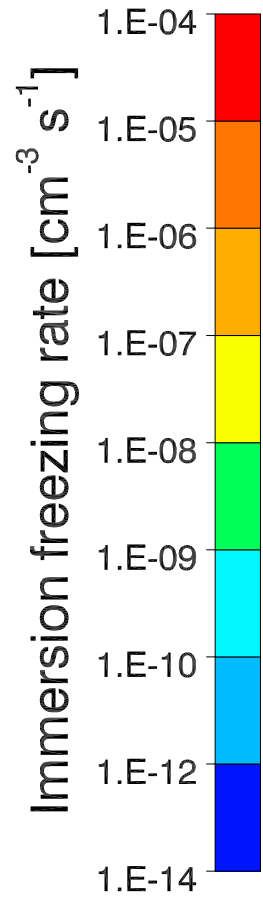


Simulated  
freezing rate

**Uppermost estimate:**  
Mean contribution of  
0.6% to  
global ice nucleation

Generous assumptions  
about IN activity  
(100% Ps. syr.-like,  
higher emissions)





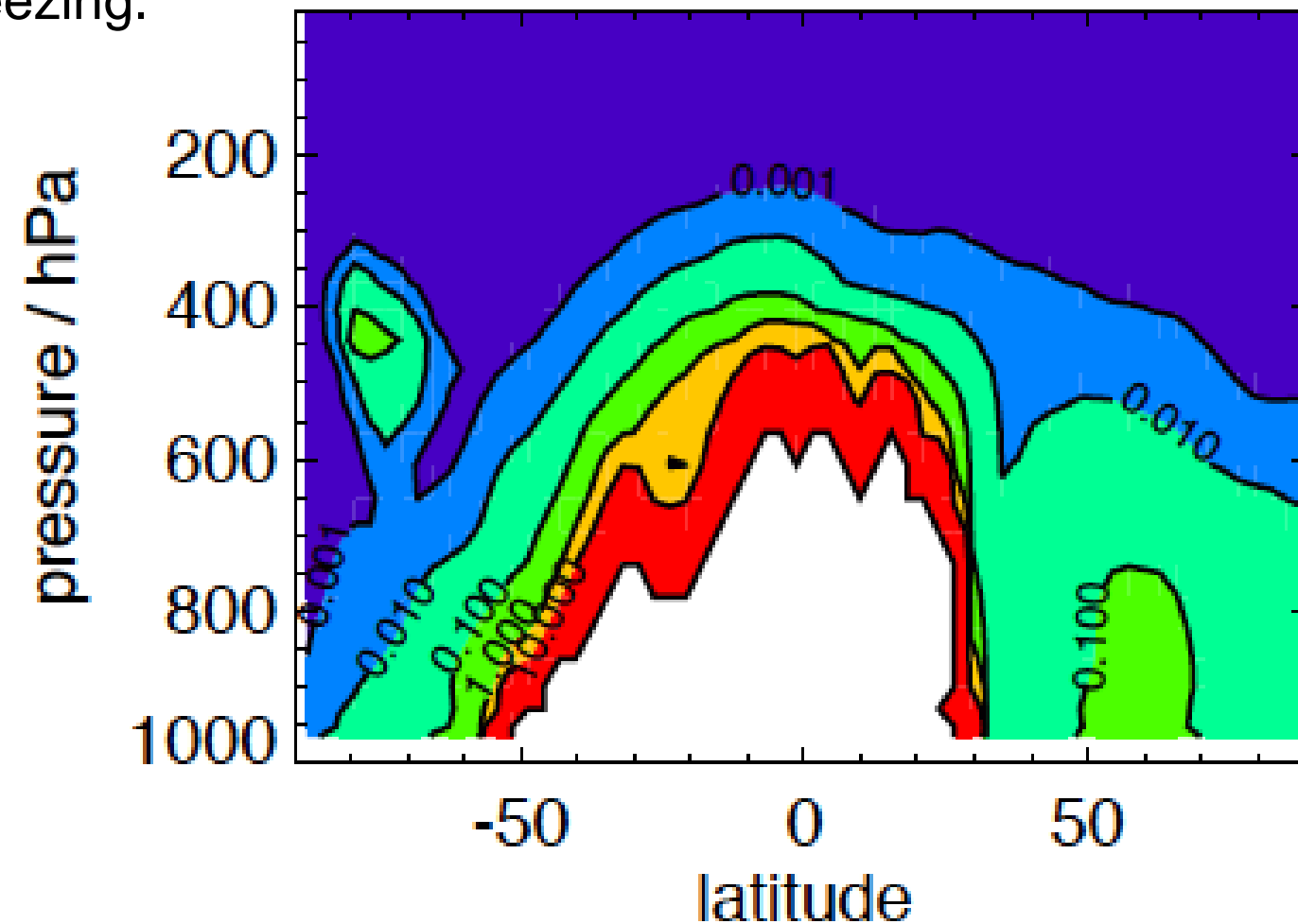
- Primary Biological Aerosol Particles (PBAP) – here: bacteria, fungal spores, pollen.
- Even with very generous assumptions, PBAP contribute at most 0.6% to global immersion freezing rate in this study.

When and where might PBAP matter as IN?

- In regions where dust, soot influence is low (e.g. Amazon, Southern Ocean)?
- In low-lying, warmer parts of mixed-phase clouds

# Primary Biological Aerosol Particles may be important in initiating cloud drop freezing at lower altitudes

Upper limit contribution of PBAP to immersion freezing as a percentage of total immersion freezing.

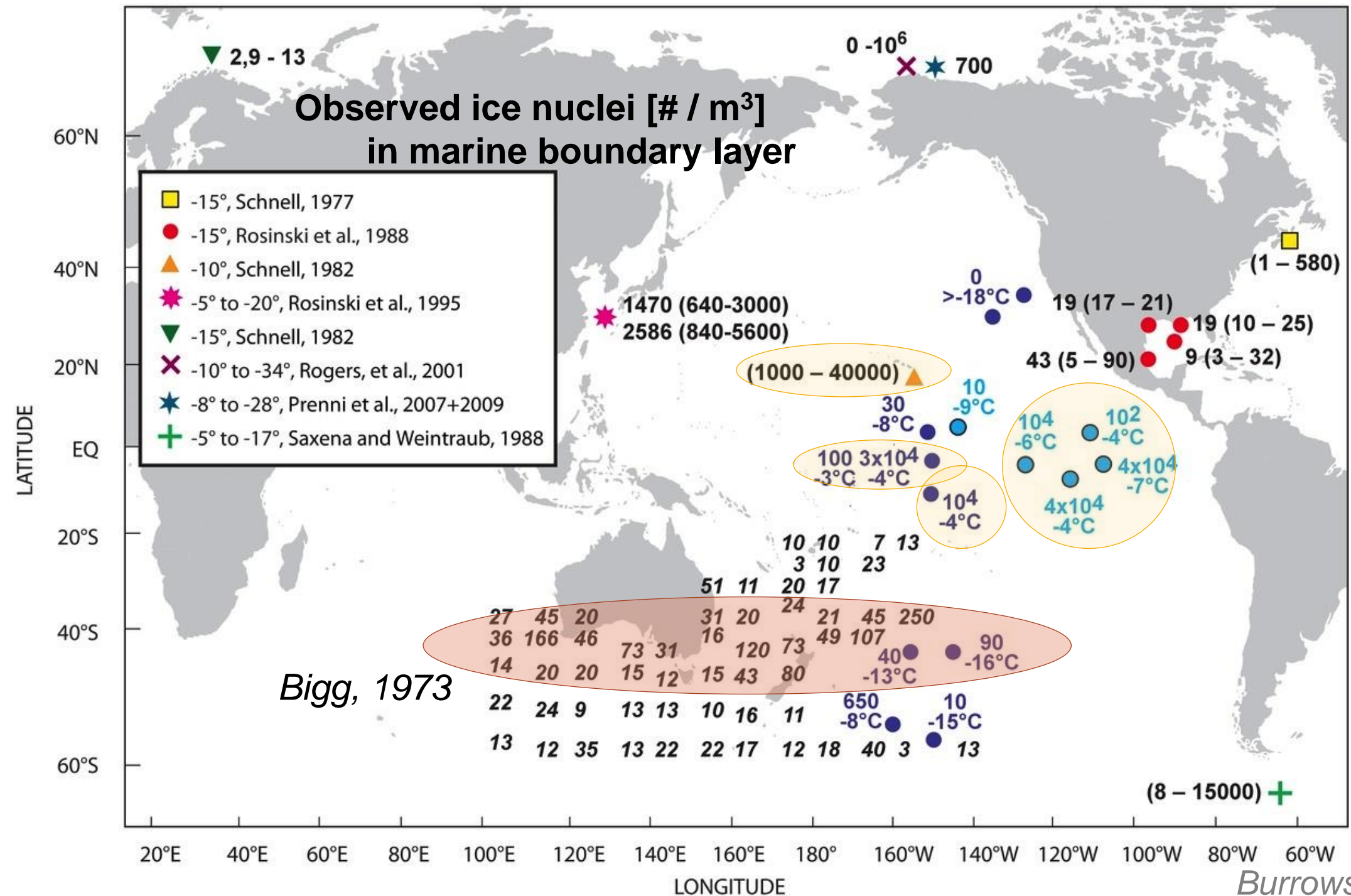


PBAPs may be important in initiation ice formation in the warmer part of low-level clouds.



# Missing piece 2: sea spray organic matter

Observed marine IN concentrations are higher over phytoplankton bloom regions. Occasionally, very high IN concentrations have been reported over active blooms.

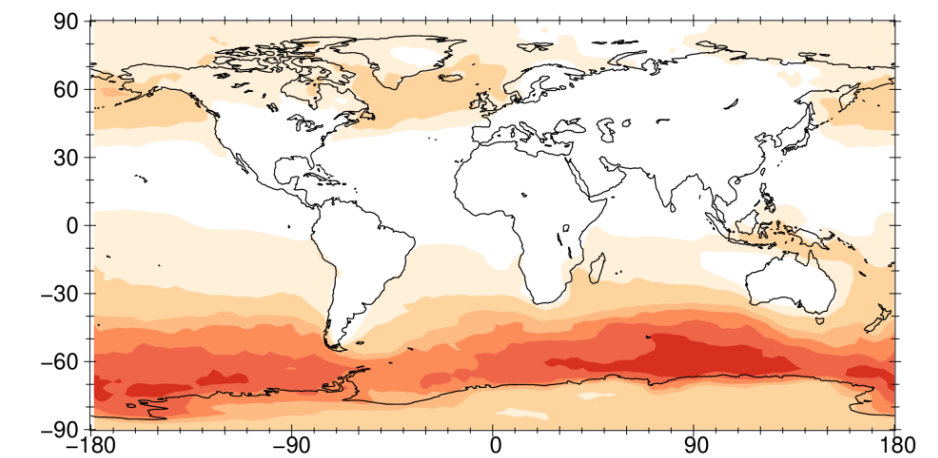
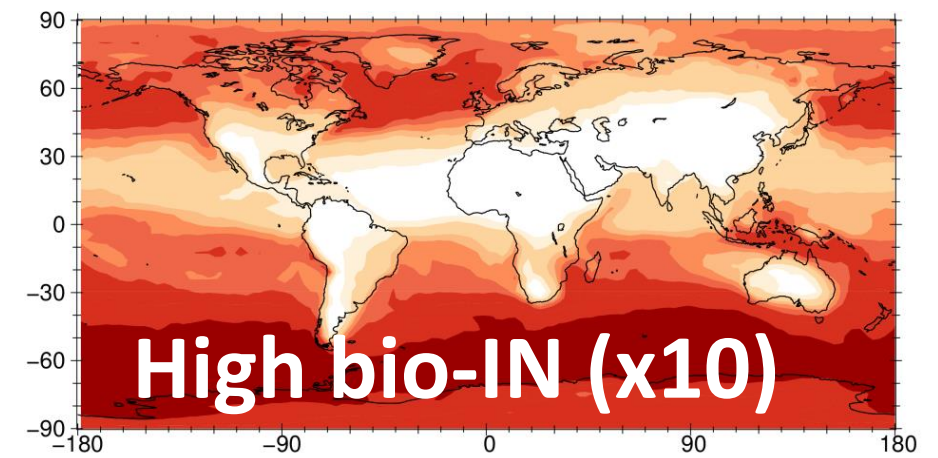




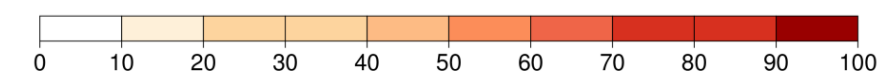
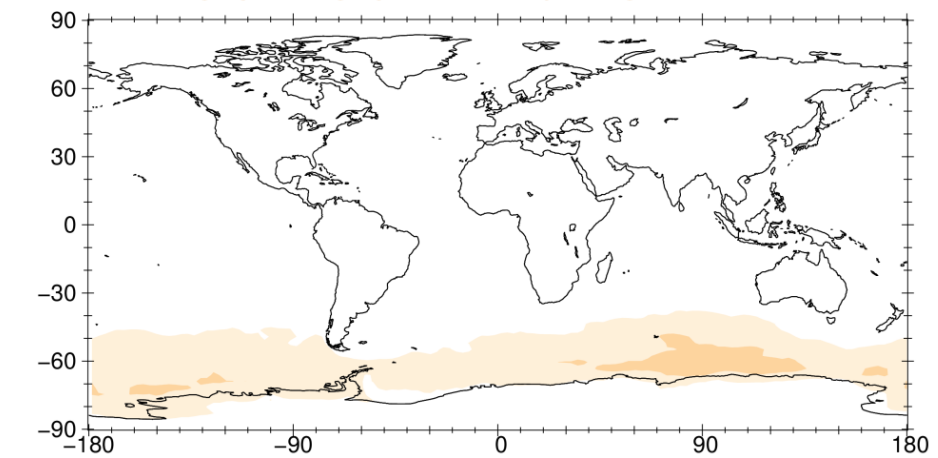


# Percent contribution of sea spray organic INP to total INP in the marine boundary layer

- Best “bounding” estimate using observational data available at the time.
- Relied on a single, decades-old measurement of ocean surface material INP efficiency.
- No modern observations of INP in marine air were available for evaluation of this estimate.



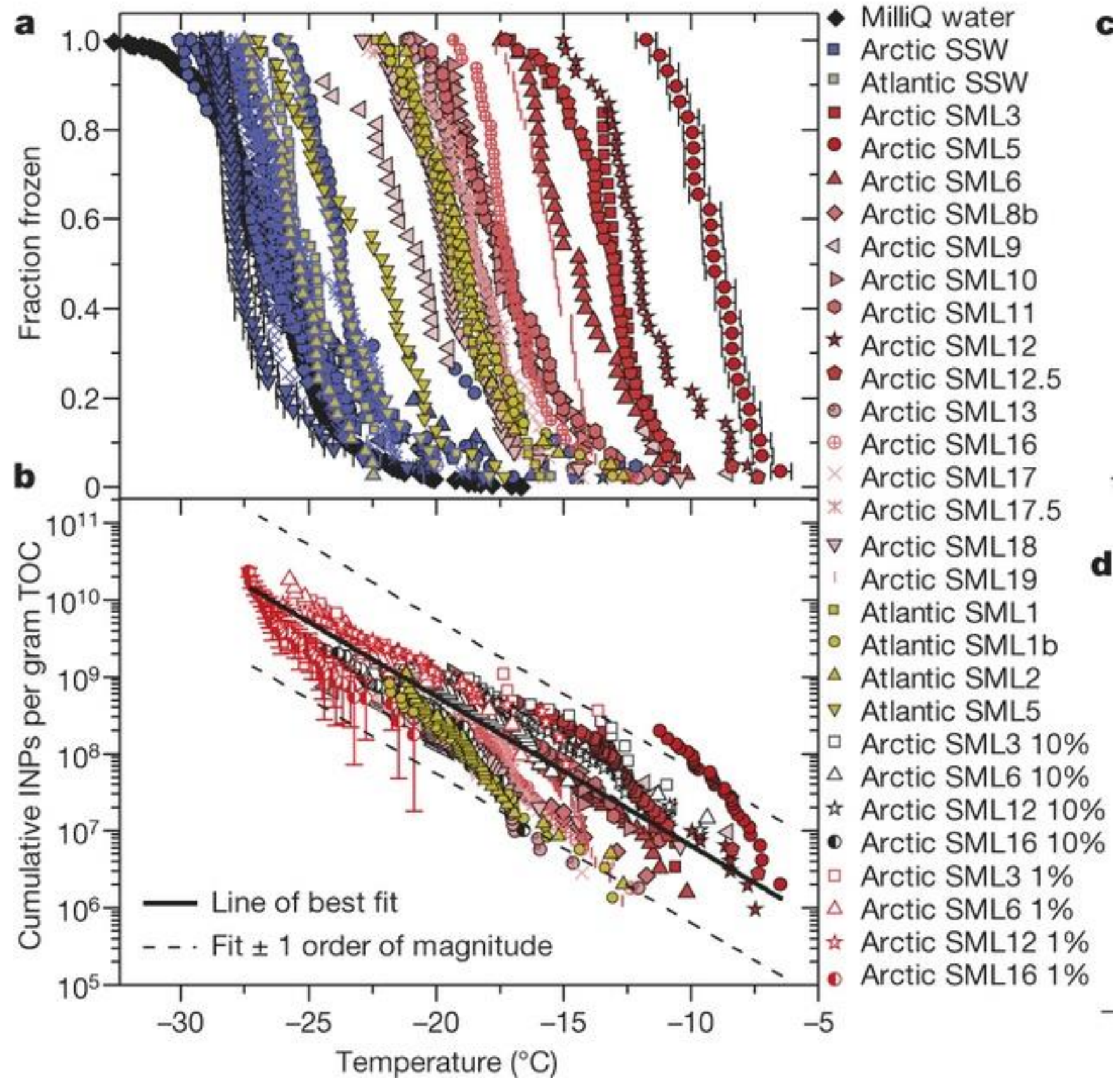
**Best estimate**



**Low bio-INP (x0.1)**

# Sea surface microlayer material as a source of ice nucleating particles

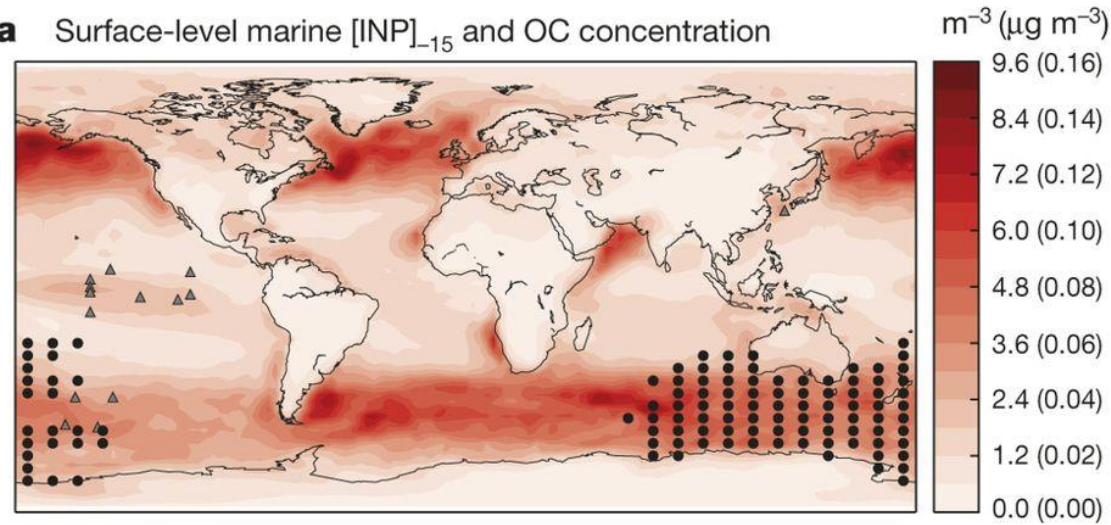
Wilson et al.,  
2015, *Nature*.



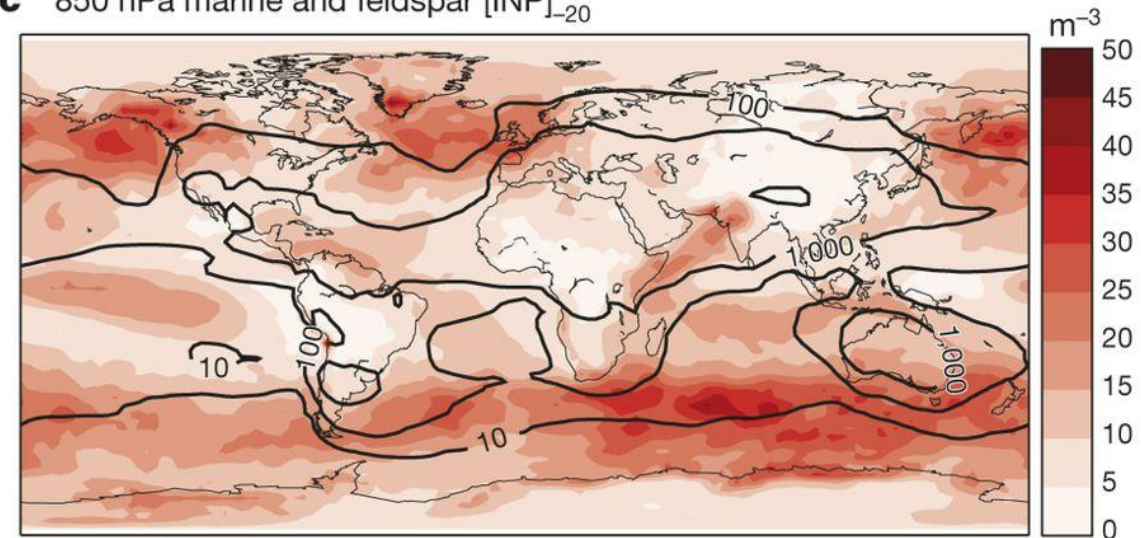


# New measurements of INP in ocean surface microlayer material enable first estimate of sea spray INP using modern measurements of source material

**a** Surface-level marine  $[INP]_{-15}$  and OC concentration

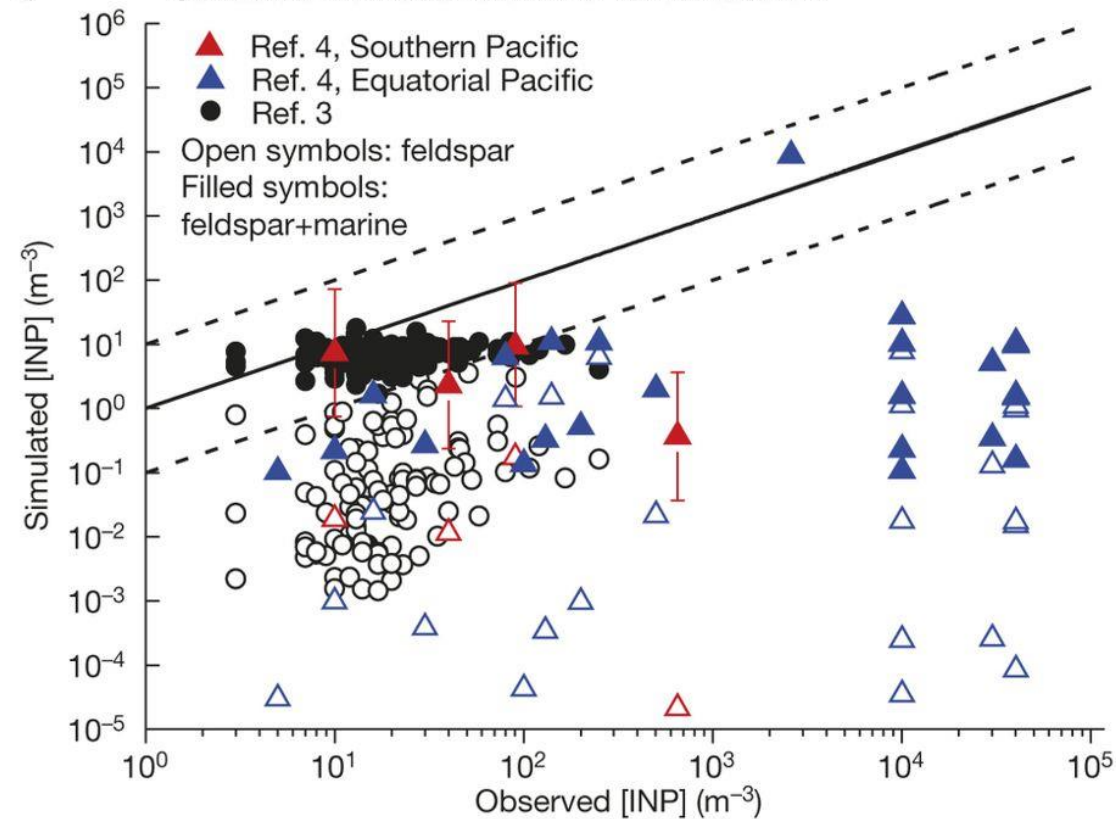


**c** 850 hPa marine and feldspar  $[INP]_{-20}$

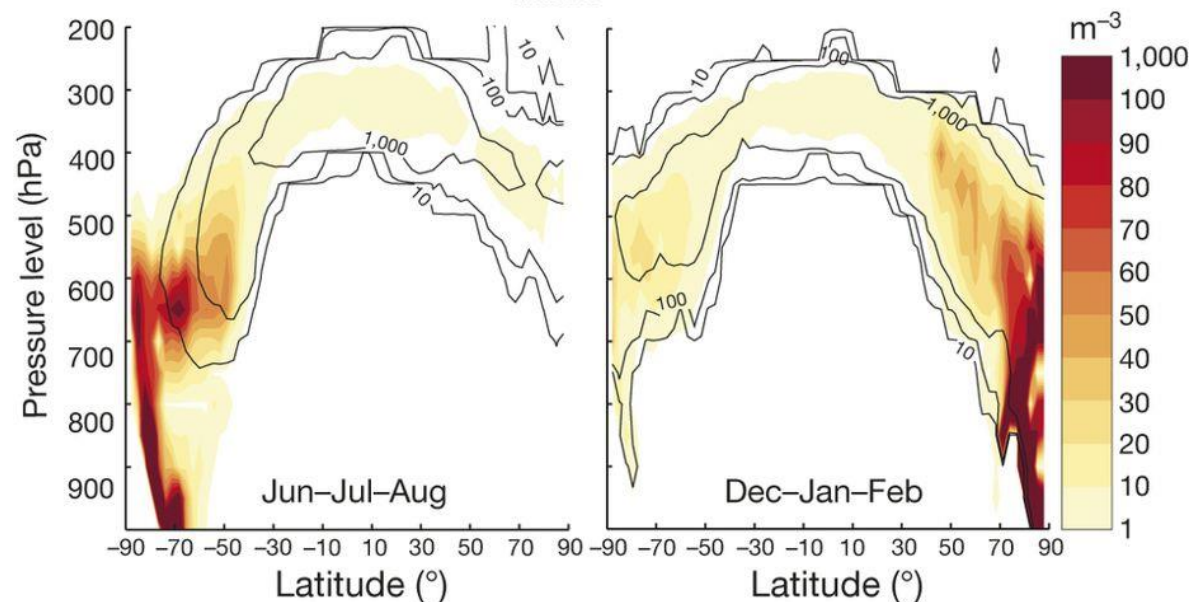


*Wilson et al.  
(2015)*

**b** Simulated versus observed INP concentrations



**d** Marine and feldspar  $[INP]_{ambient}$  seasonal Atlantic transect



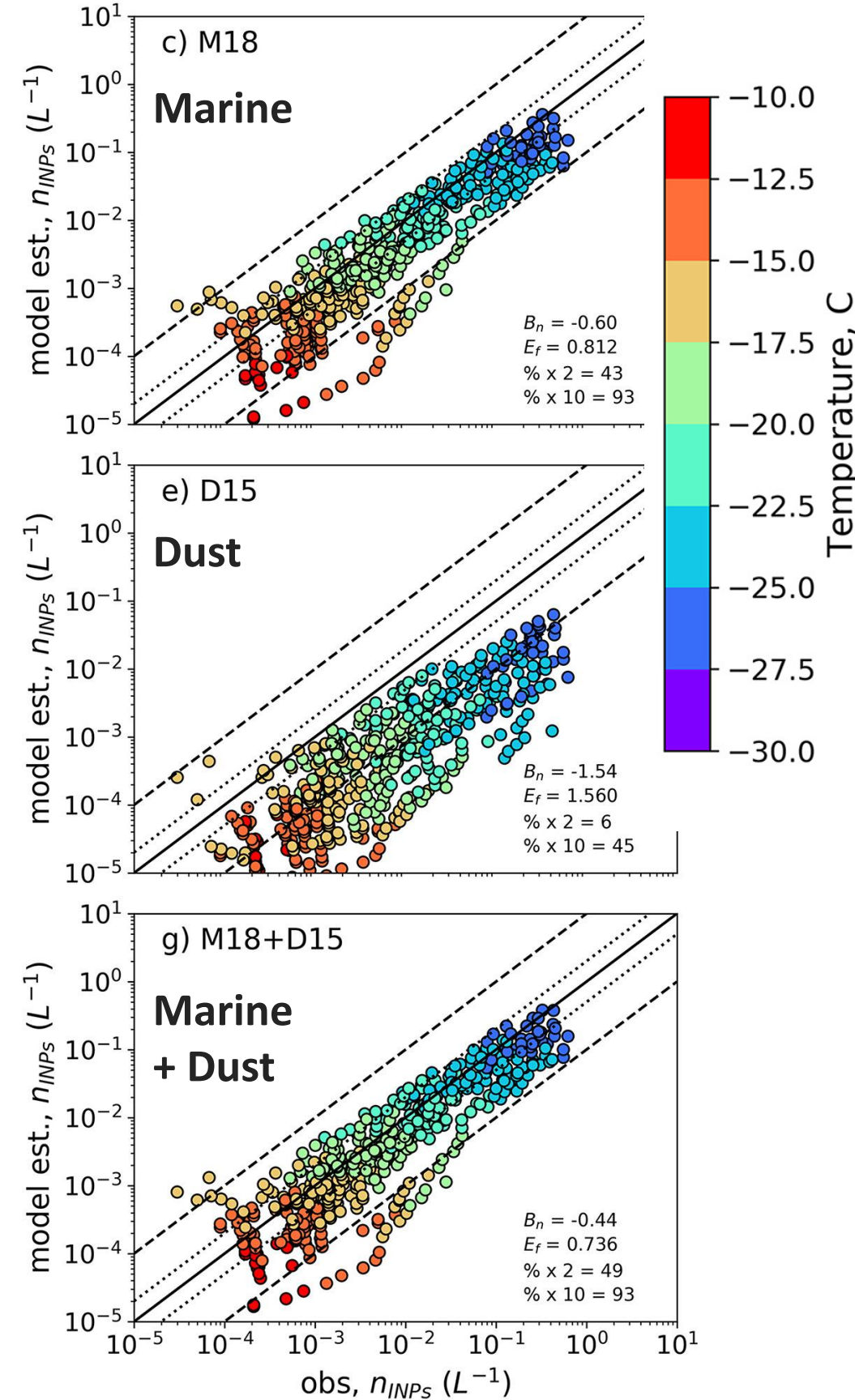
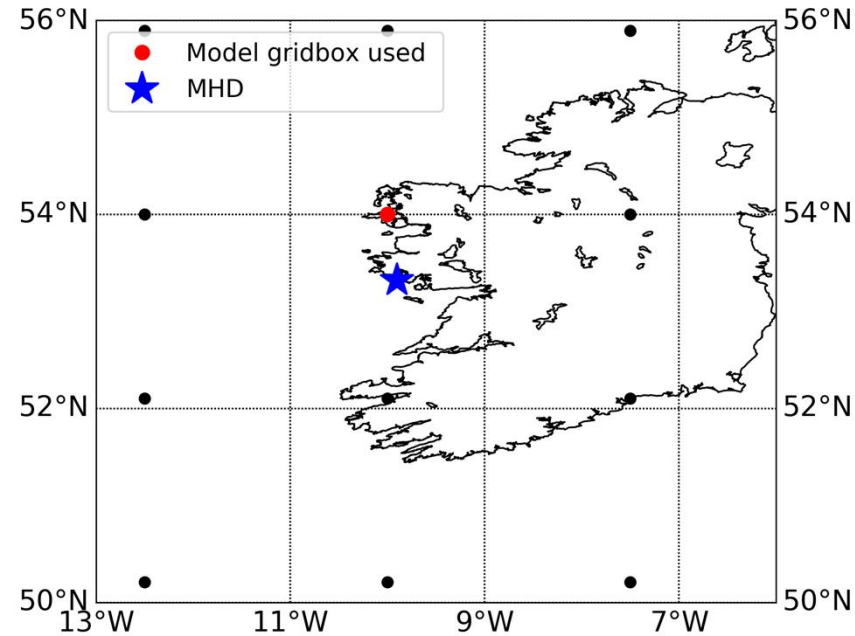


# Accounting for sea spray improves model agreement with observed INP number



DOE SCGSR graduate student fellowship award to Christina McCluskey, CSU

## Mace Head, Ireland: August 2015



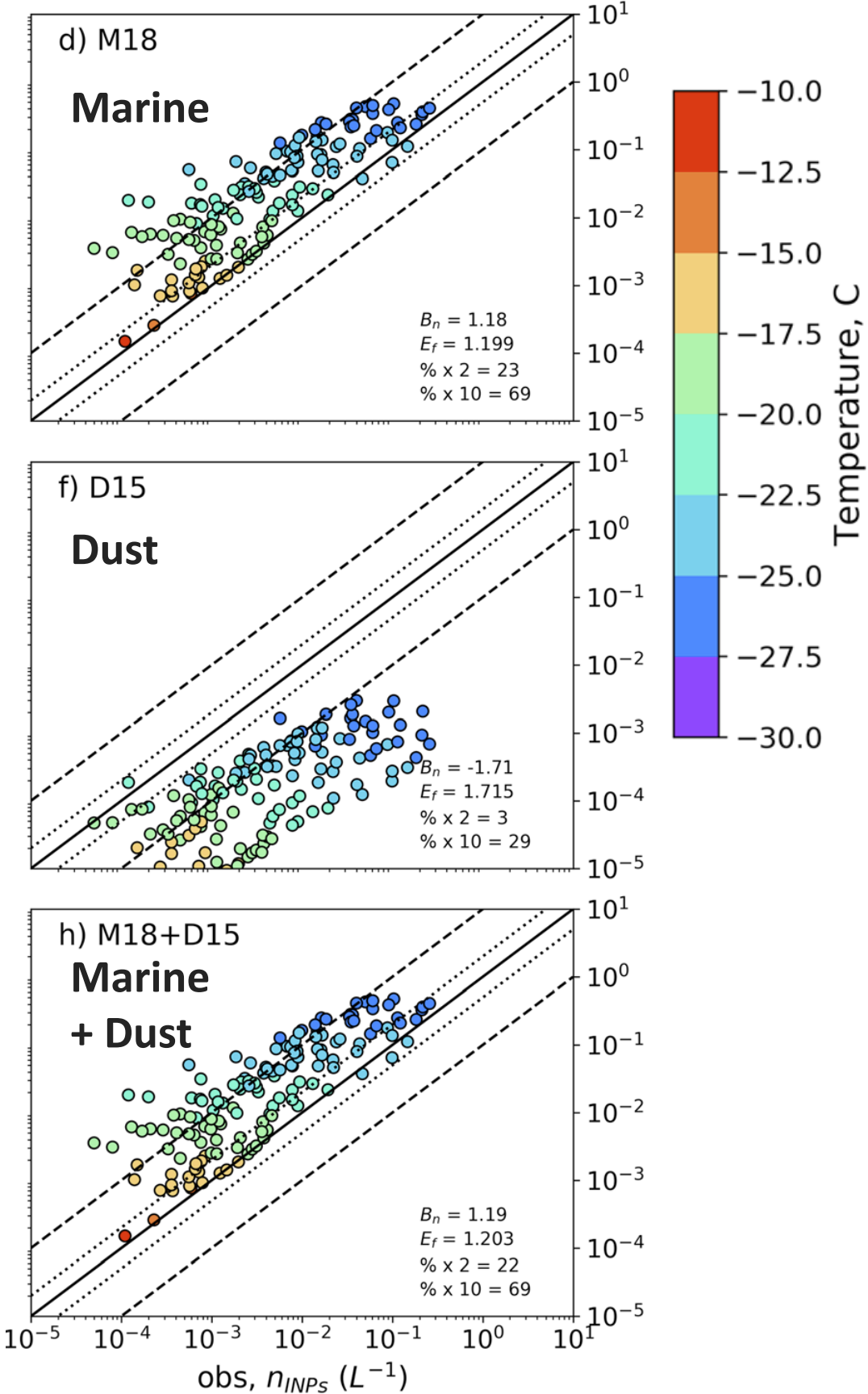
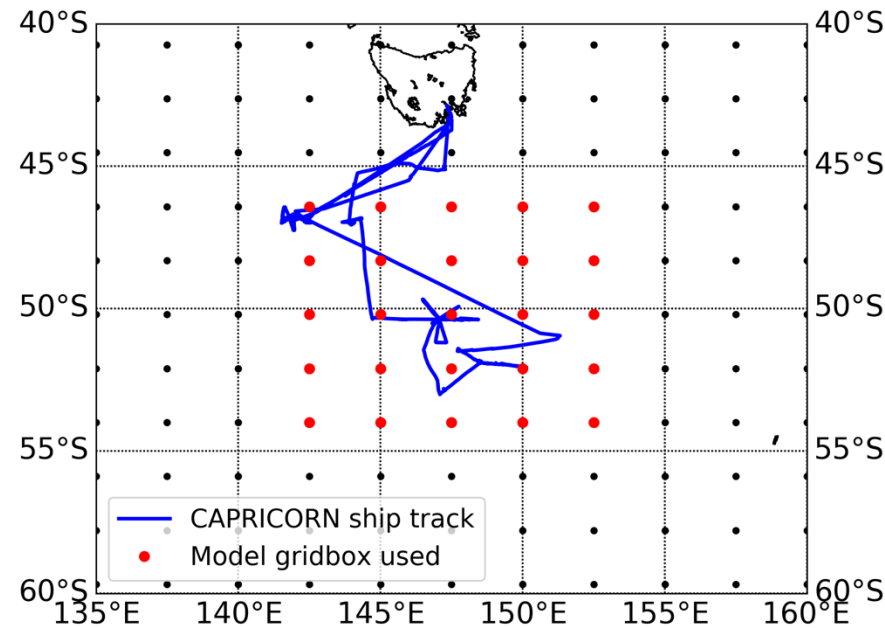


# Accounting for sea spray improves model agreement with observed INP number



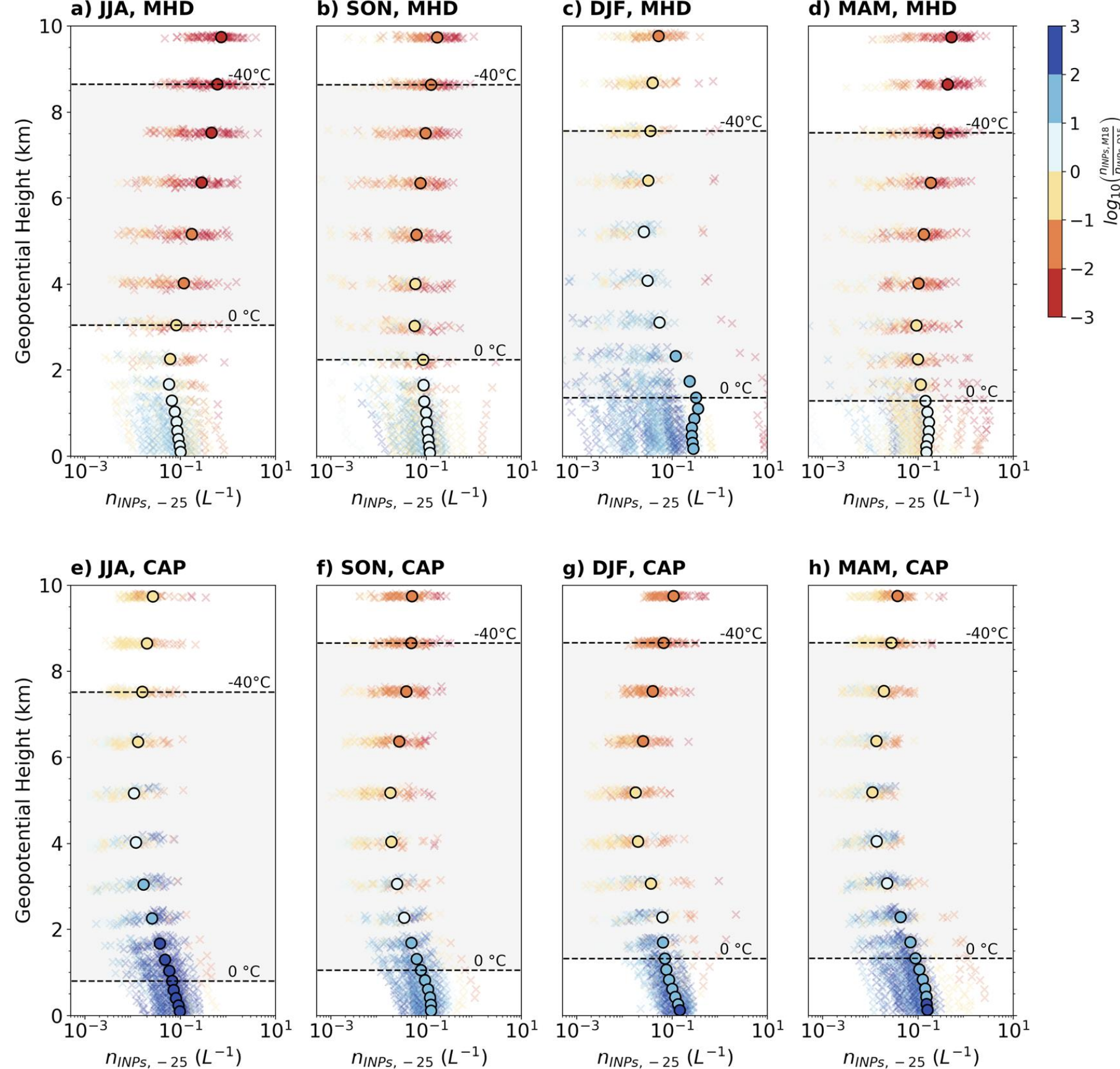
DOE SCGSR graduate student fellowship award to Christina McCluskey, CSU

## CAPRICORN ship campaign, Southern Ocean: Mar-Apr 2016



# Seasonal variability in INP vertical profiles

$n_{INPs} (L^{-1}) @ -25^{\circ} C$



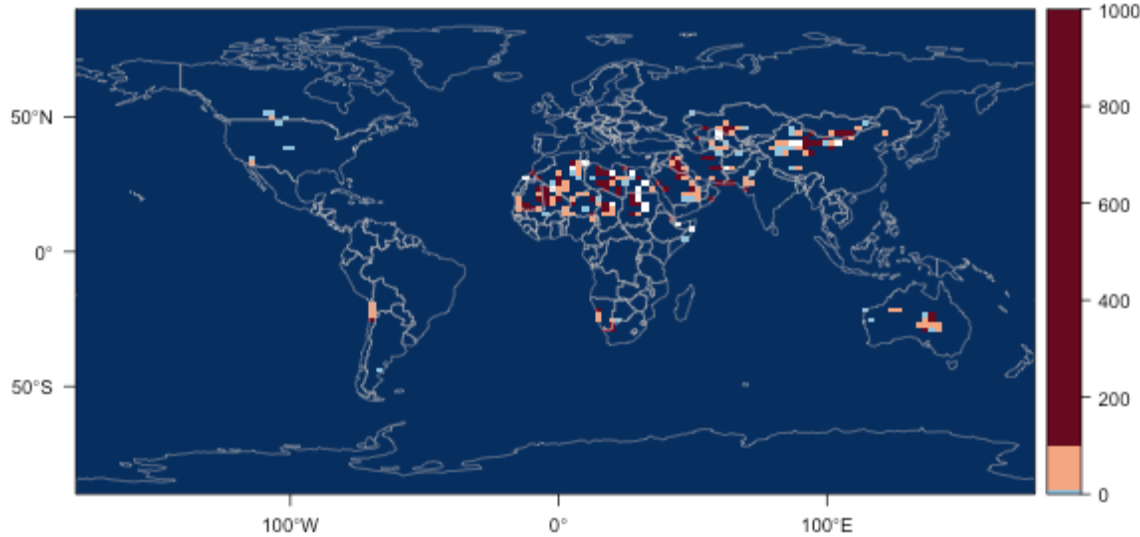




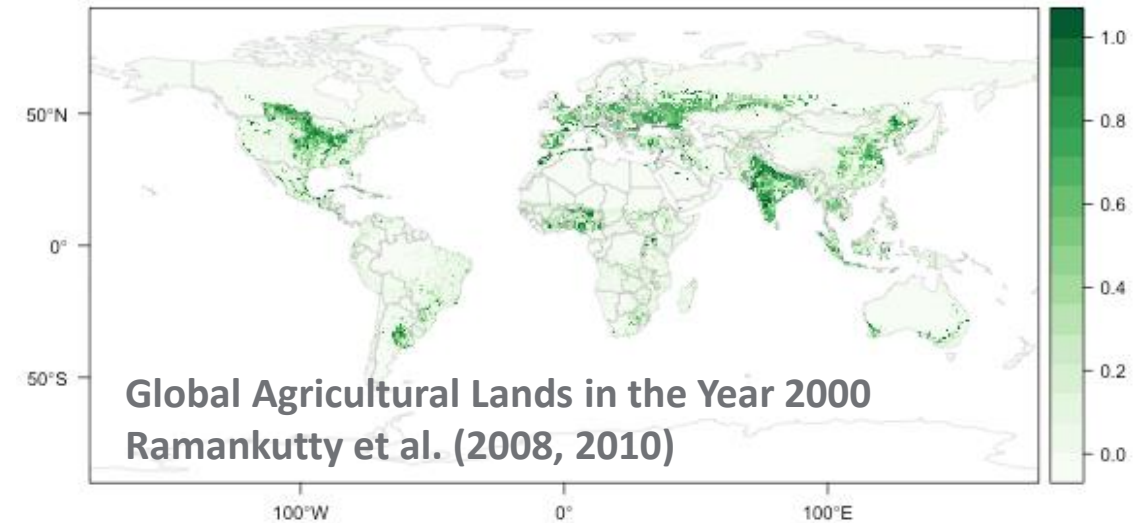
Pacific  
Northwest  
NATIONAL LABORATORY

# Missing piece 3: Organic-rich agricultural soils

Annual mean CESM dust emissions [ $\text{g m}^{-2} \text{yr}^{-1}$ ]



Crops fraction (0.5 x 0.5 deg)

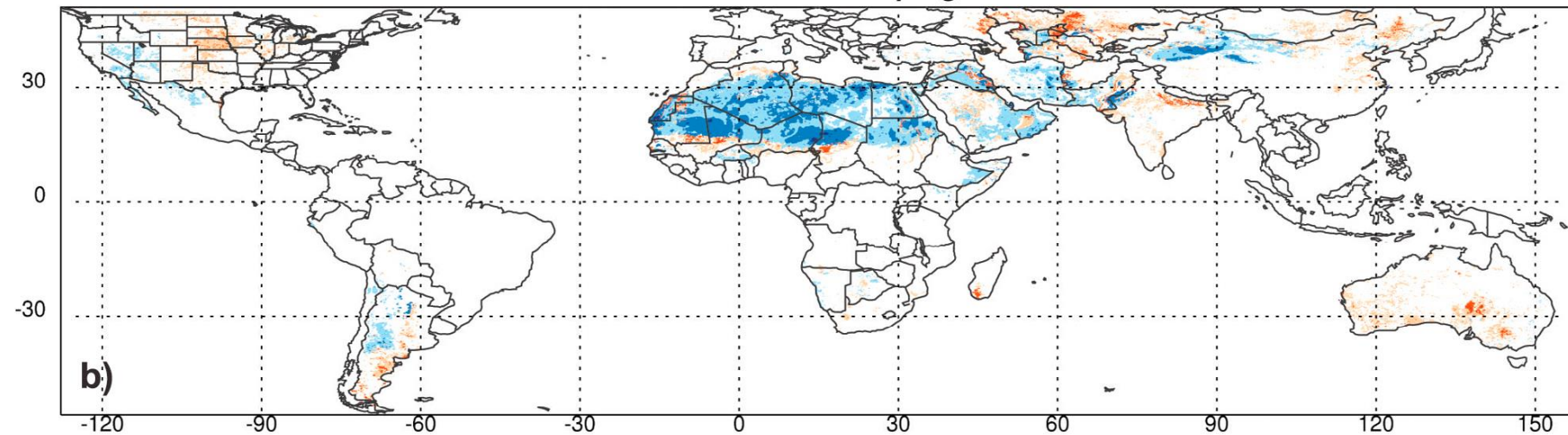


Natural=1173  $\text{Tg.yr}^{-1}$

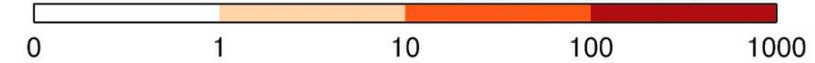
Natural & Anthropogenic

Anthro=363  $\text{Tg.yr}^{-1}$

"natural" (blue) vs  
"anthropogenic" (red;  
mostly agriculture)

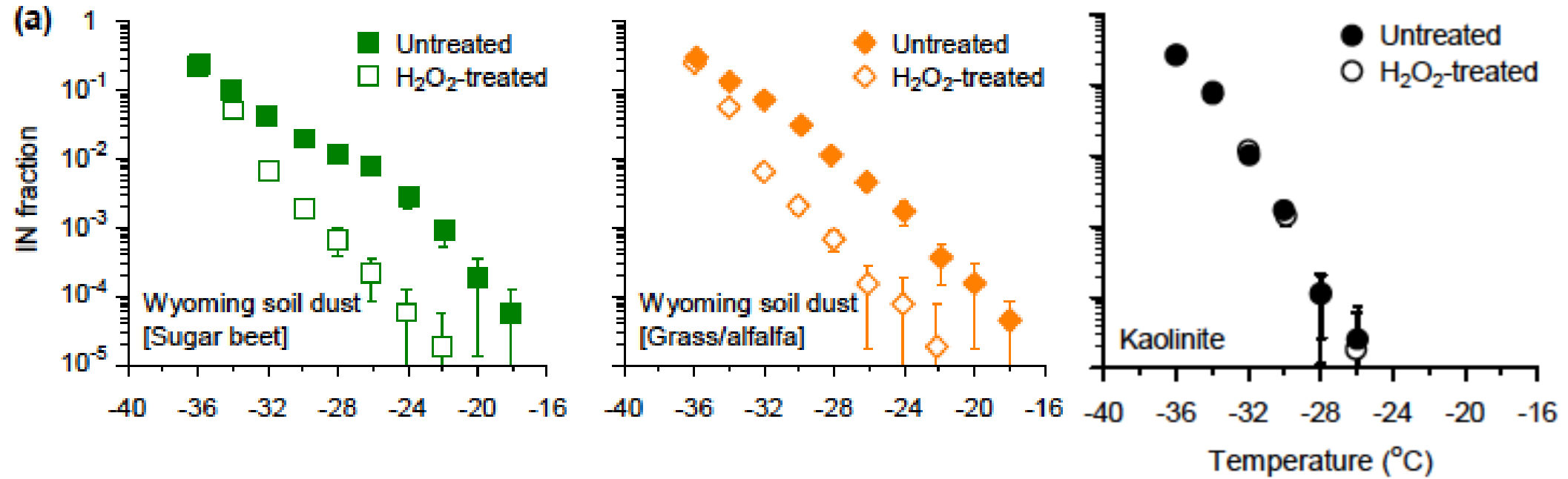


Non-hydro or Anthro emission ( $\text{g.m}^{-2}.\text{yr}^{-1}$ )

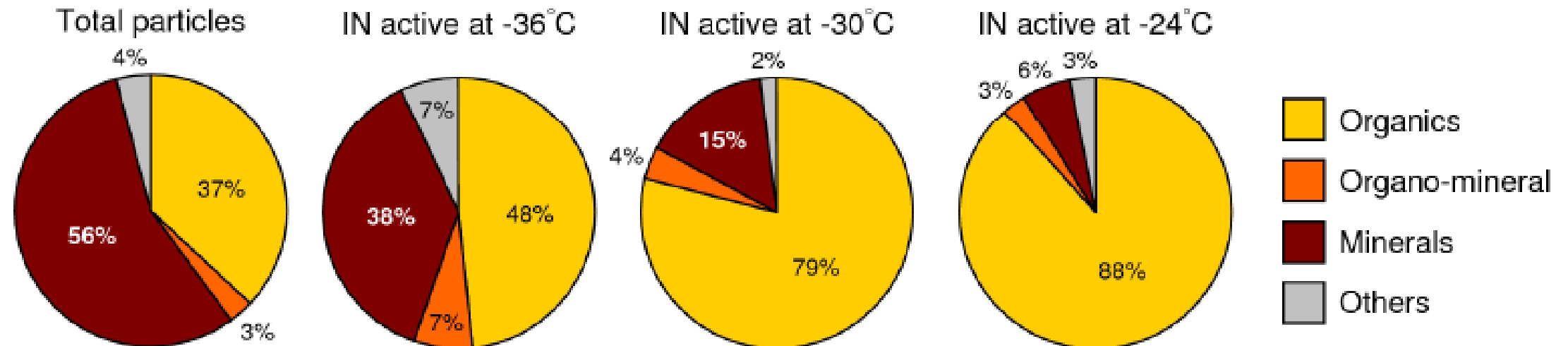


*Ginoux et al. (2012)*

## INP fraction from Continuous Flow Diffusion Chamber (CFDC)

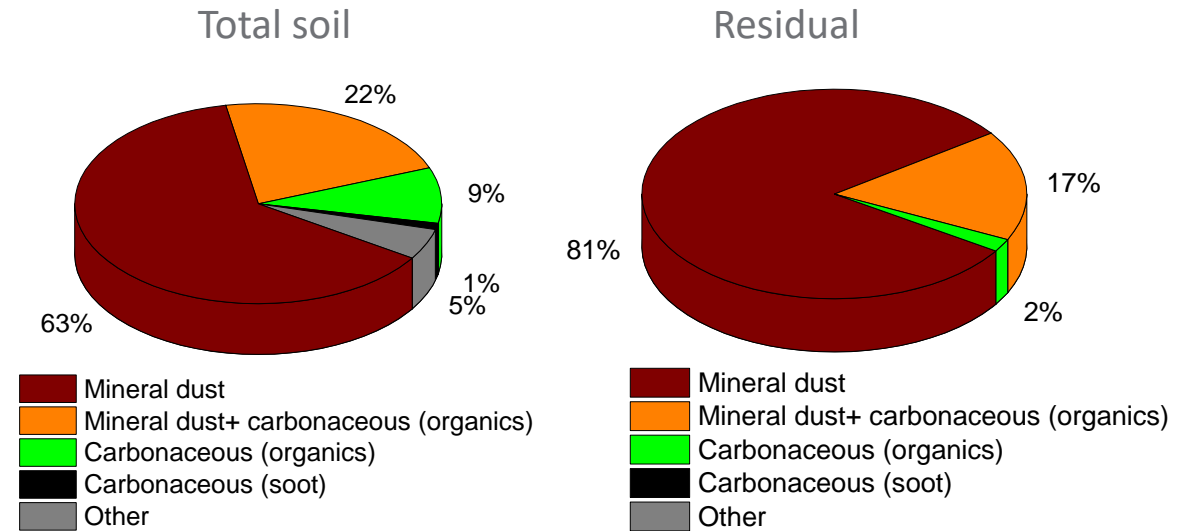
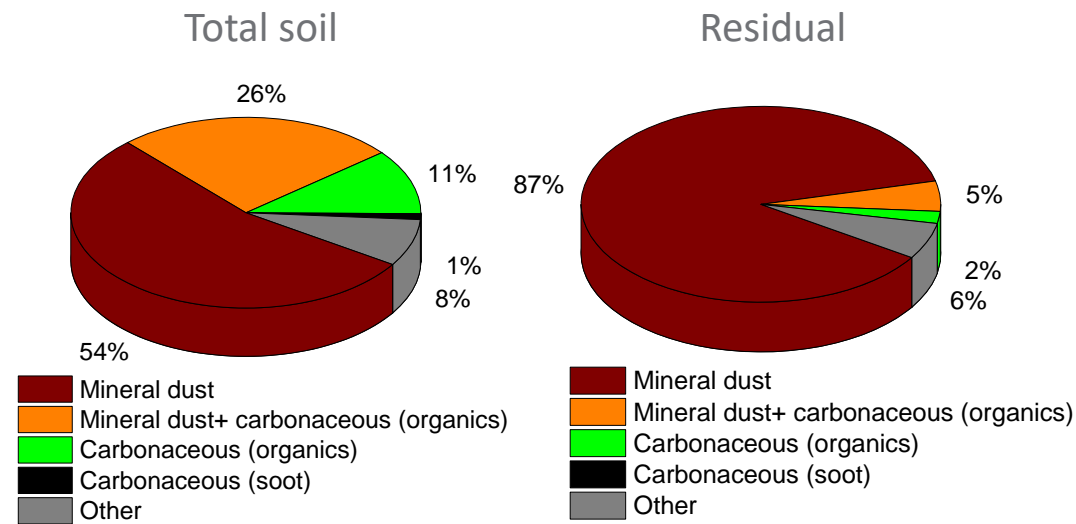
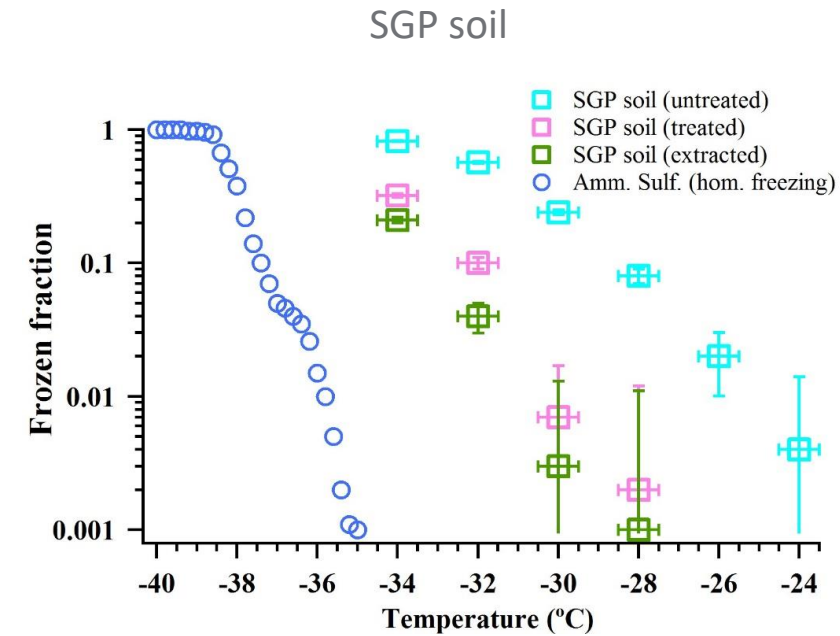
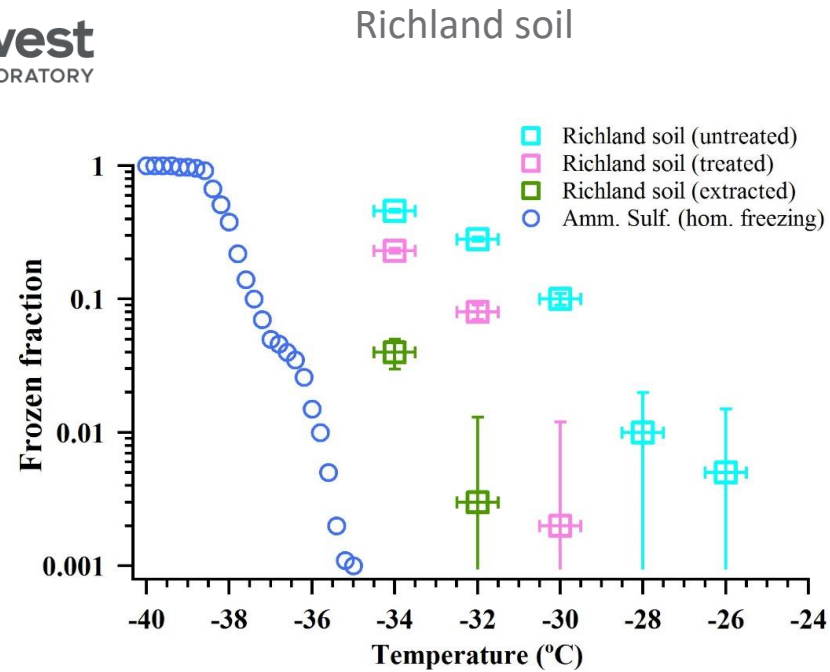


## Composition of total particles and INPs from SEM-EDX





# IN-activity of agricultural soil samples from Richland and SGP



Measurements: Swarup China and Gourihar Kulkarni

Funding: FICUS (Joint EMSL-JGI) User Proposal



# Building a Comprehensive Understanding of Ice-Nucleating Particles from the Ground Up: Establishing the Impact of Sea Spray and Agricultural Soils

U.S. Department of Energy (DOE) Early Career project



PD: Isabelle Steinke



PD: Gavin Cornwell



PD: Aish Raman

Collaborators:



Alla Zelenyuk, PNNL (miniSPLAT: single-particle mass spectrometer)



Gourihar Kulkarni, PNNL (INP Chamber)



Alex Laskin, Purdue (Offline analyses: IN-ESEM, SEM-EDS, Raman, etc.)



Swarup China, PNNL



Alex Huffman, U. Denver (Fluorescent particles)



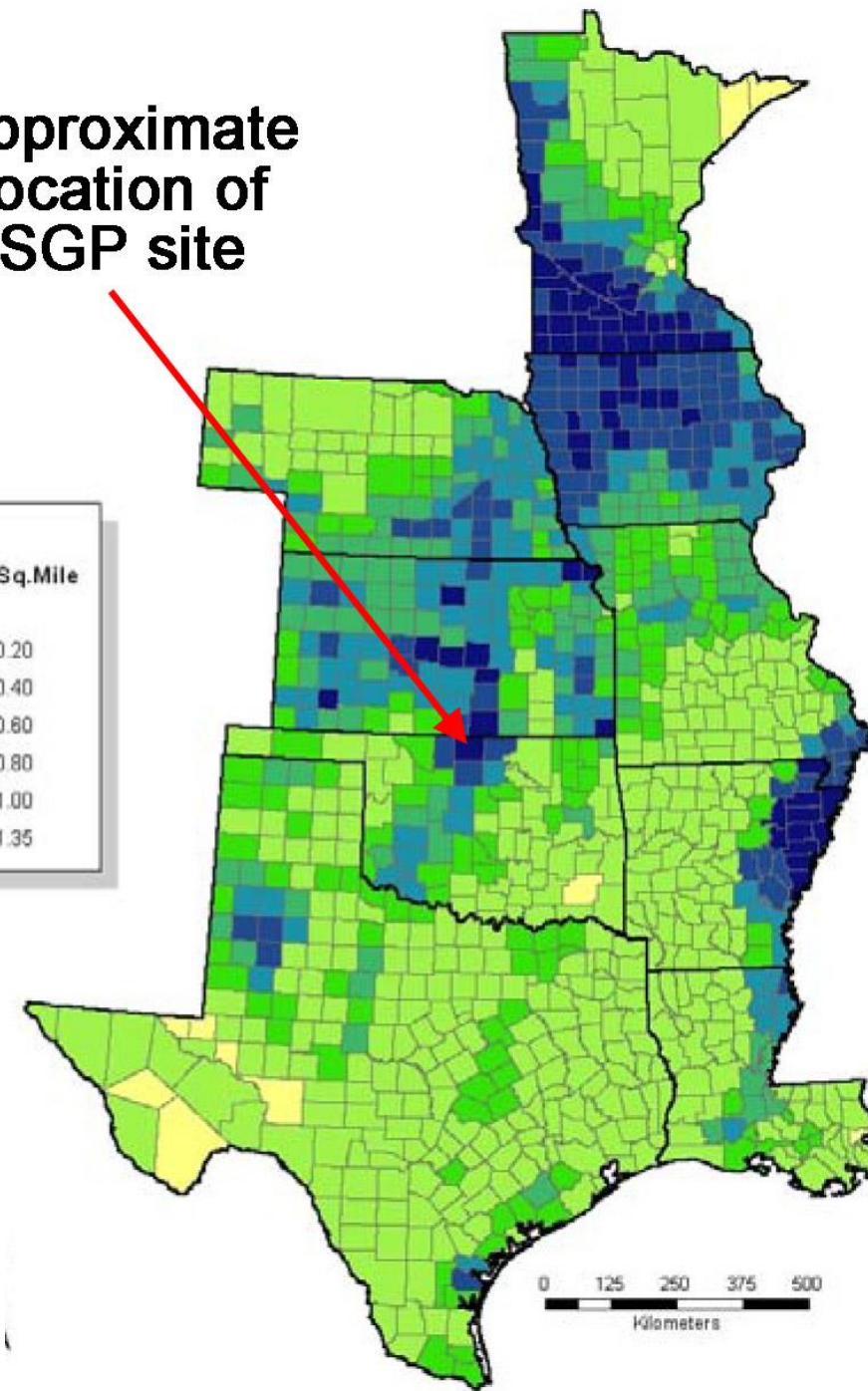
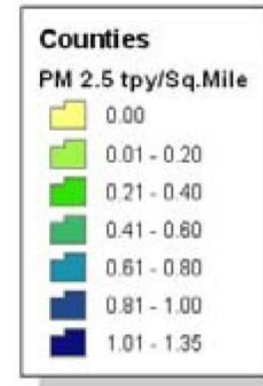
Paul DeMott, CSU (worldwide INP observations)



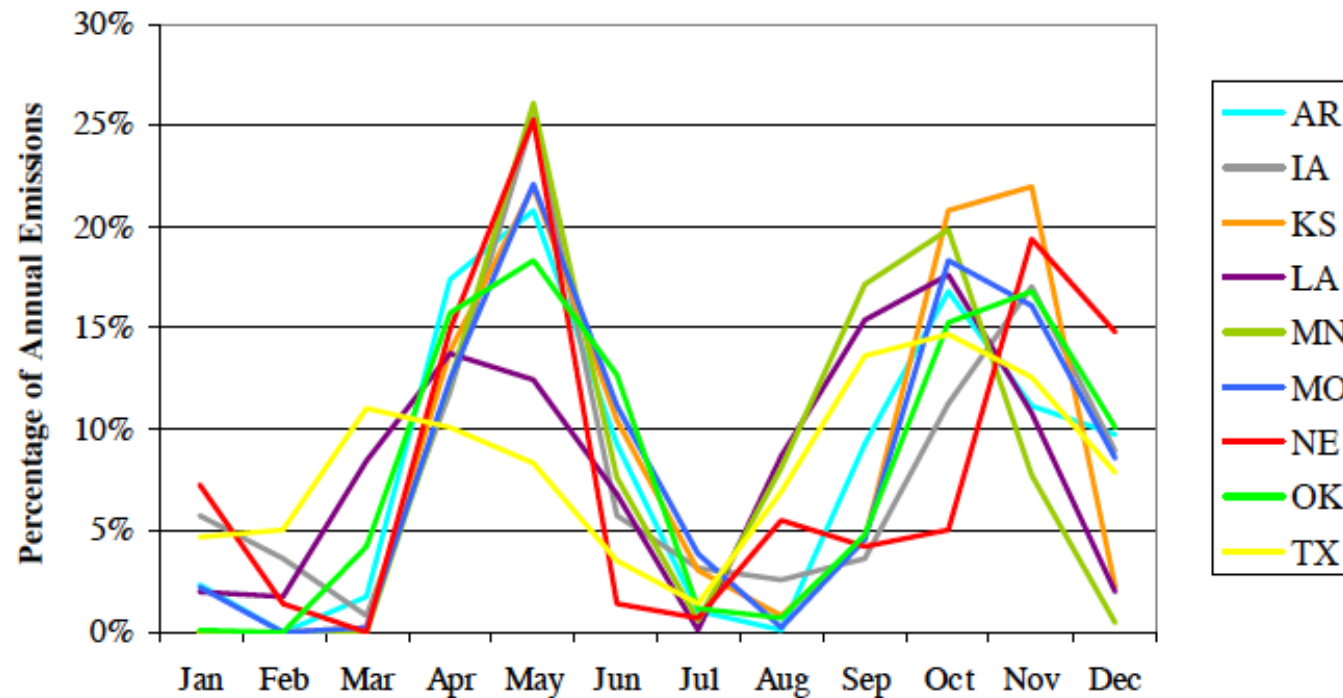
# Upcoming field campaign will target agricultural dust INPs

Emissions of soil dusts in the Great Plains are mainly associated with agricultural activities (tilling, harvesting)

Approximate location of SGP site



## Seasonal cycle of tilling emissions



# Reflections on a decade+ of work on modelling INP sources

- We have learned a lot in the past 10 years!
  - Biological particles can now be better-measured and characterized through fluorescence measurements
  - Sea spray particles are now understood to be weak INPs, and to contribute to INP populations in remote marine air
  - Increasingly, improved INP parameterizations are being tested in models and shown to have skill in predicting INP number
- Bold claim! We are on the cusp of greatly improving the integration of INP measurements into atmospheric modelling
  - More measurements globally in more environments (although still few long-term)
  - Greater understanding of measurement uncertainties
- Progress comes from working together across expertise and disciplinary boundaries.
  - How can modelers more effectively provide guidance to the experimental community?



# Thank you

NSF graduate research fellowship  
Max Planck Institute for Chemistry /  
Max Planck Graduate School

DOE Office of Science:

- Earth and Environmental System Modelling (EESM)
- Atmospheric System Research (ASR)
- Early Career Research Program
- Office of Science Graduate Student Research (SCGSR)