Overview of our atmospheric INP measurements from polar to tropical regions and what we might learn from them.

Heike Wex

and many colleagues from TROPOS and cooperating research groups

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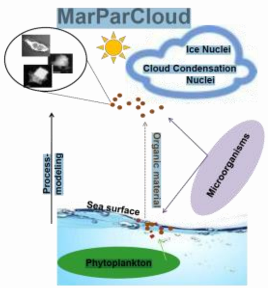

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overview

- (very) short introduction on INP in general

- a bit about INP measurements & data evaluation

- where / what we measured, wrt. INP in
    - Beijing
    - the Arctic
    - Cabo Verde

- what I think might be true about INP in general

ice nucleating particles (INP)

a first review on materials that may occur in the atmosphere and have shown ice nucleating activity:

Szyrmer, W., and I. Zawadzki (1997), Biogenic and anthropogenic sources of ice-forming nuclei: A review, *BAMS, 78(2), 209-228.*

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more recent reviews:

- Murray et al. (2012), Ice nucleation by particles immersed in supercooled cloud droplets, *Chem. Soc. Rev.*

- Hoose, C., and O. Möhler (2012), Heterogeneous ice nucleation on atmospheric aerosols: A review of results from laboratory experiments, *Atmos. Chem. Phys.*

- Kanji et al. (2017), Chapter 1: Overview of Ice Nucleating Particles, in *Ice Formation and Evolution in Clouds and Precipitation: Measurement and Modeling Challenges, edited, Meteor. Monogr.*

- Coluzza et al. (2017), Perspectives on the future of ice nucleation research: Research needs and unanswered questions identified from two international workshops, *Atmosphere*

ice nucleating particles (INP)

some basics known today (some of them corroboating what was formerly known already):

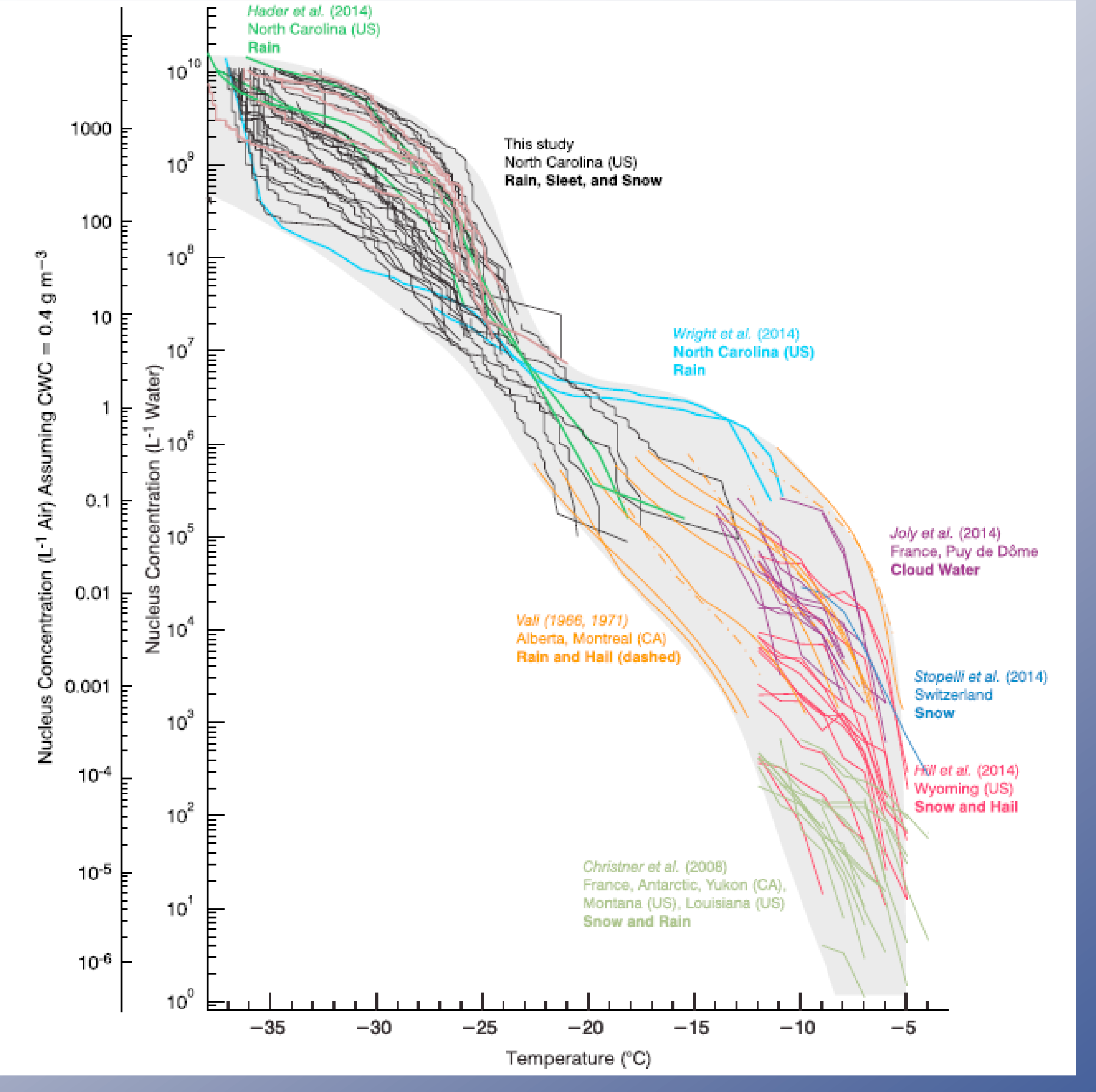
- mineral dust particles are important atmopsheric INP

- among mineral dusts, K-feldspar particles may play the most important role

- biogenic INP are macromolecules (proteins or polysaccarides) found on microorganisms

- sea spray may contribute to atmospheric INP, but to what extent is still open

concentrations of ice nucleating particles (N_{INP})



Petters & Wright (2015), Revisiting ice nucleation from precipitation samples, GRL.

measuring ice nucleating particles (N_{INP})

in-situ

- continuous flow diffusion chambers (CFDC, with names like PINC, SPIN, HINC, INKA, …)

- laminar diffusion tube (LACIS)

- expansion chambers (e.g., AIDA, CLOUD-Chamber, PINE)

- characteristic: optical detection of the ice crystals; due to this and the limited flow, comparably high INP number concentrations have to be present (~ > 1/L); often inlet cut-off of a few micrometers

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off-line

- collecting particles on a filter (or using a batch sample)

- analyzing them off-line in “cold-stages” or “freezing arrays”; different types are now operated by a growing number of different groups

- characteristic:
  + comparably low concentrations can be detected
  - contaminations limit the lowest temperature to which measurements can be made (depending on droplet size and other factors, > -25°C; exception: pico-liter droplets, but droplet production and optical detection much more expensive)
  - long sampling times -> bad time resolution

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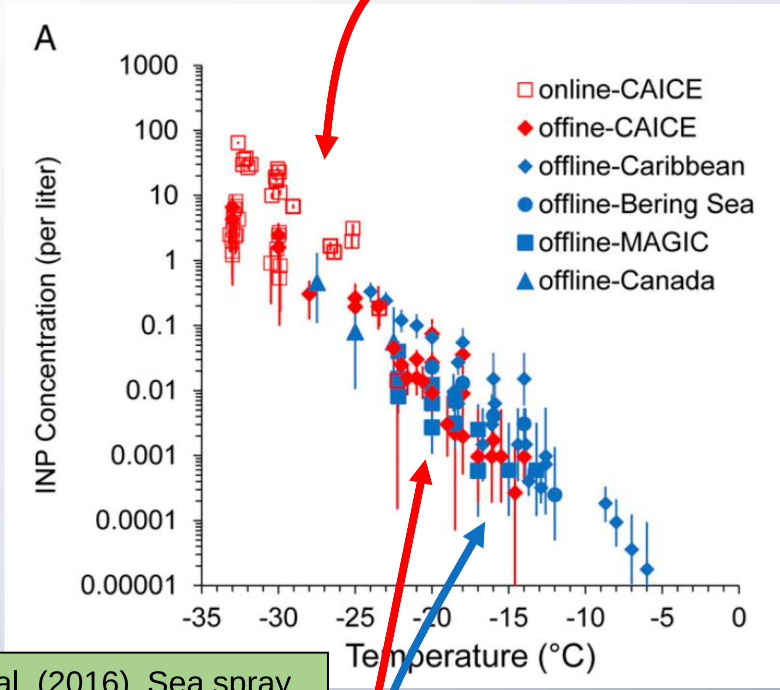
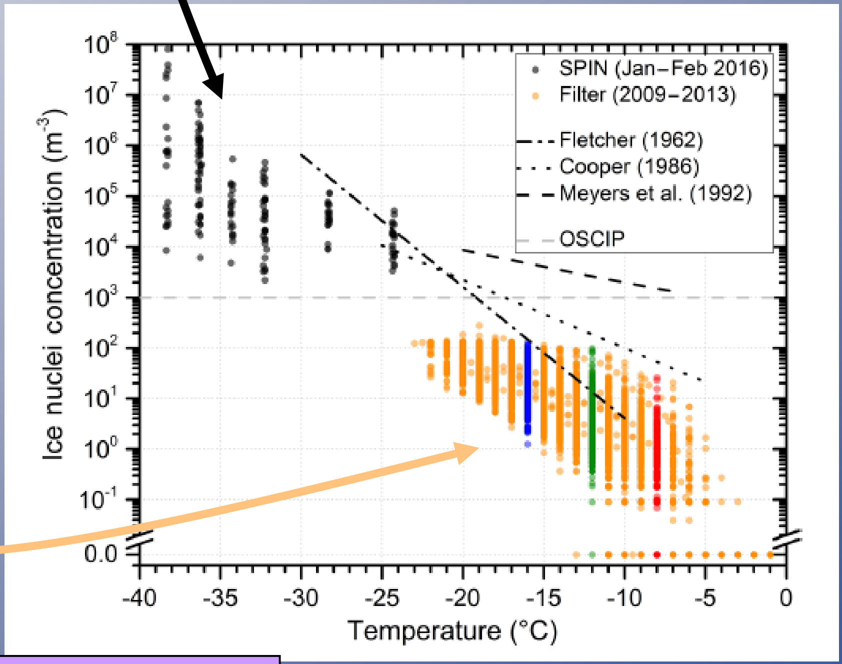
measuring ice nucleating particles (N_{INP})

in-situ



DeMott et al. (2016), Sea spray aerosol as a unique source of ice nucleating particles, PNAS.

off-line



Welti et al. (2018), Concentration and variability of ice nuclei in the subtropical maritime boundary layer, ACP.

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measuring INP

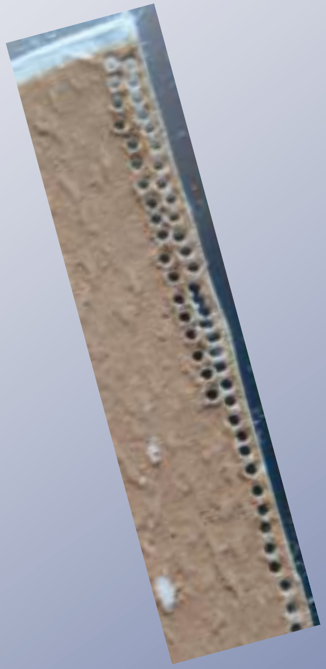
* Leipzig Ice Nucleation Array
** Ice Nucleation Droplet Array

measurement principles
of our cold-stage / freezing array:

INDA and LINA are used for suspensions (e.g., washed polycarbonate filters),

INDA also for filter punches (quartz-fiber filters),

LINA reaches lower temperatures



LINA *

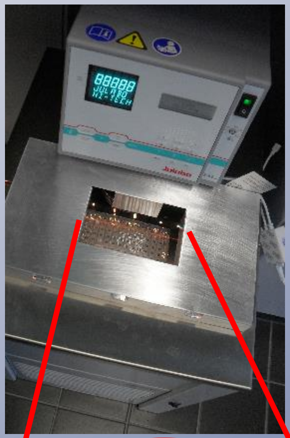
V= 1 μL in one droplet, 90 droplets

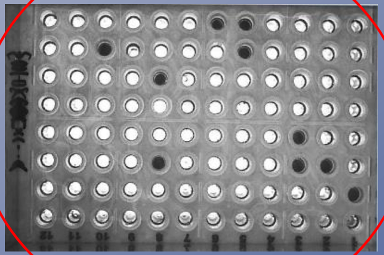
droplets on a glass slide, cooled by a Peltier element

e.g., Budke & Koop et al. (2015), AMT

INDA **

V=50 μL in one droplet, 96 droplets



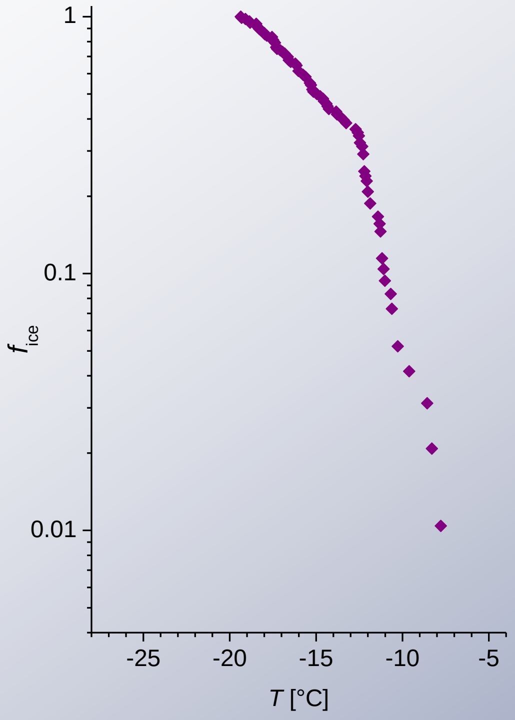


PCR-trays in a thermostat

Conen et al. (2012), ACP
Hill et al. (2014), AEM

measuring INP

cumulative distribution

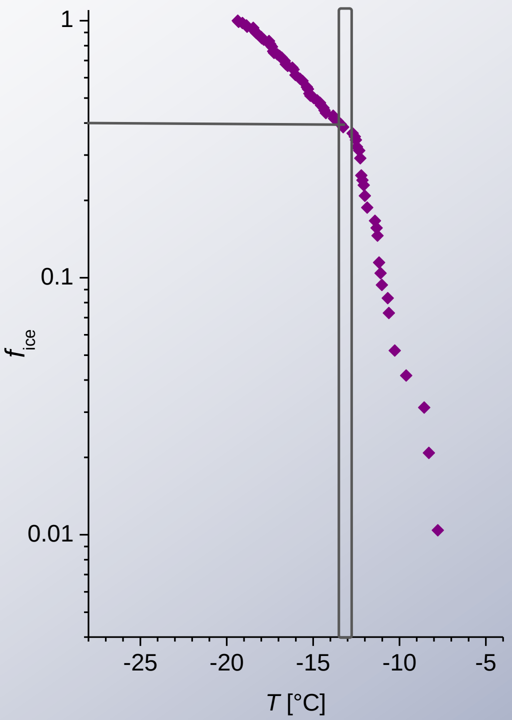


Vali, J. Atmos. Sci. (1971)

f_{ice} -> fraction of frozen droplets from all droplets

measuring INP

cumulative distribution



Vali, J. Atmos. Sci. (1971)

Poisson distribution:

$$P_\lambda(k) = \frac{\lambda^k}{k!}\,\mathrm{e}^{-\lambda}$$

for $k = 0$ ->

$\exp(-\lambda) = f_{unfrozen} = 1 - f_{ice}$

or

$\lambda = -\ln(1 - f_{ice})$ (e.g., $f_{ice} = 0.4 \rightarrow \lambda = 0.51$)

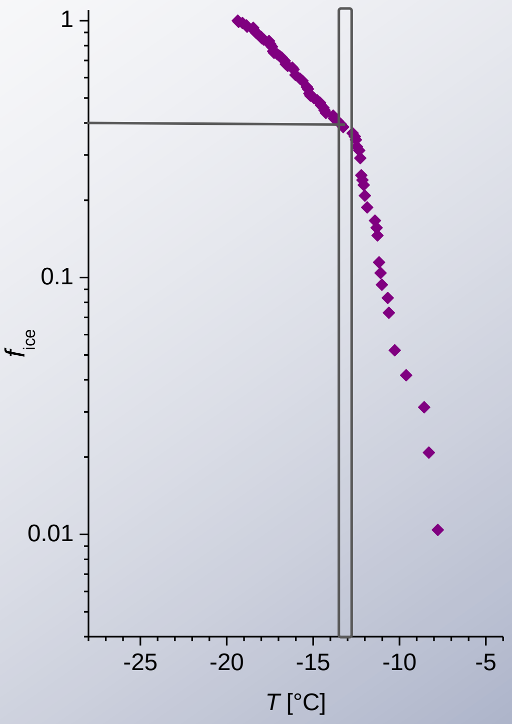
λ -> for us: average number of ice active entities per droplet

$f_{unfrozen}$ -> fraction of frozen droplets from all droplets

f_{ice} -> fraction of frozen droplets from all droplets

measuring INP

cumulative distribution



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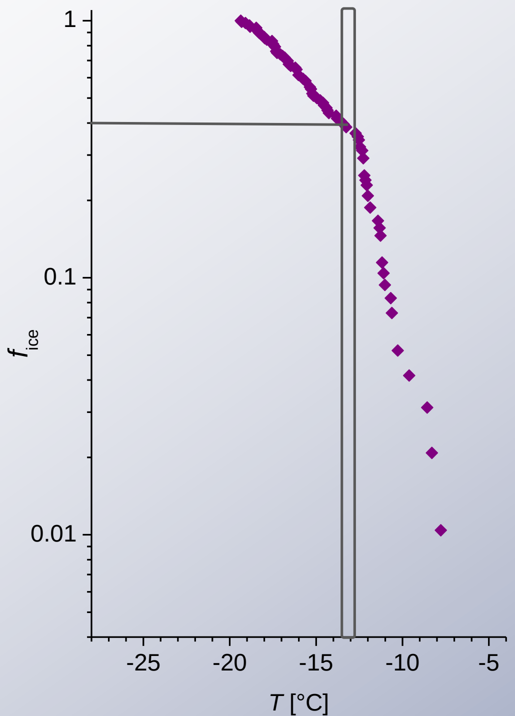

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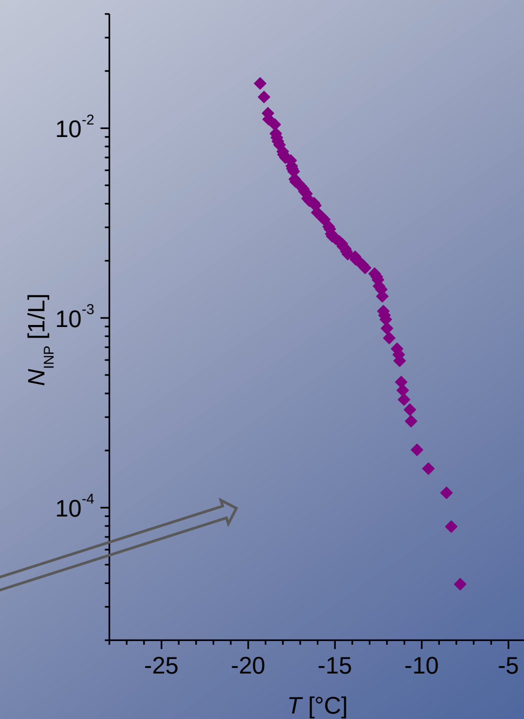


Normalization:

$N_{INP} = \lambda / V_{droplet} = -\ln(1 - f_{ice}) / V_{droplet}$

Vali, J. Atmos. Sci. (1971)

λ -> for us: average number of ice active entities (INP) per droplet

f_{ice} -> fraction of frozen droplets from all droplets

measuring INP

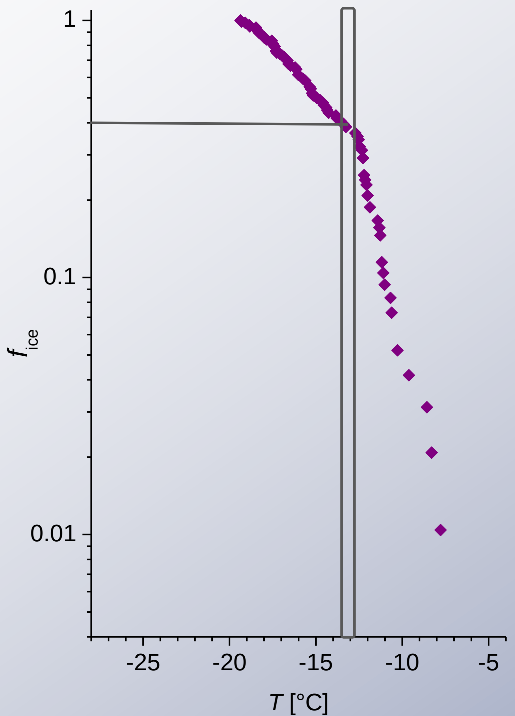
Polen et al. (2018), Cleaning up our water: …, AMT.

see supplement of Wex et al. (2019), ACP.

$$N_{INP,corr} = (-ln(1 - f_{ice,s}) + ln(1 - f_{ice,b}))/V$$

subtraction of background can not be done for f_{ice}

cumulative distribution



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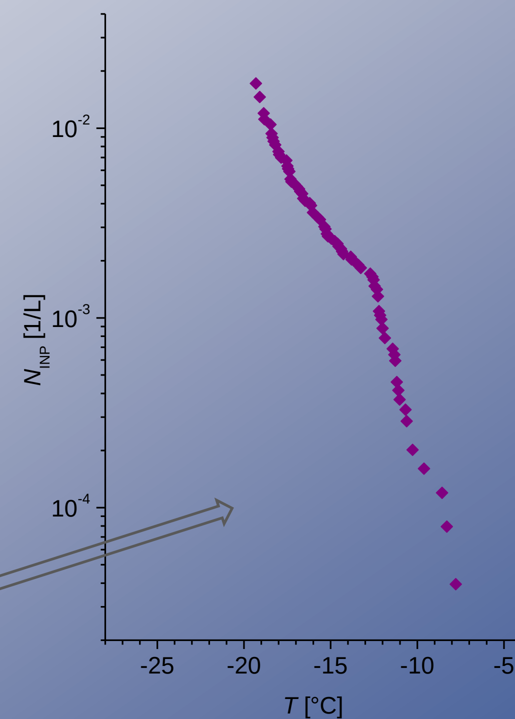

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measuring INP

Normalization:

$N_{INP} = \lambda / V_{droplet} = - \ln(1 - f_{ice}) / V_{droplet}$

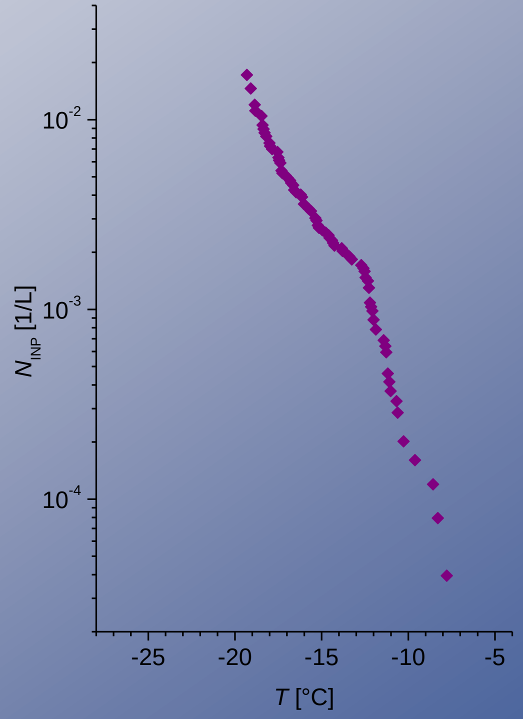
Normalization can be done for:

- per volume of suspension (e.g., „K(T)“ has been used)
  -> $V_{droplet}$ = volume of liquid in one droplet
  (e.g., for sea water samples)

- per volume of sampled air (airborne INP conc. N_{INP})
  -> $V_{droplet}$ = volume of air collected into one droplet
  (all that I will show today)

- per surface area (surface site density n_s)
  -> $V_{droplet}$ = surface area of material in one droplet

- …



measuring INP

Normalization:

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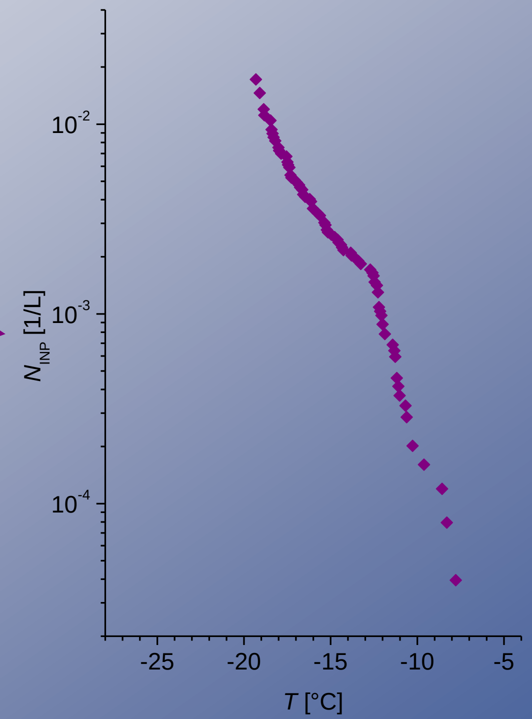
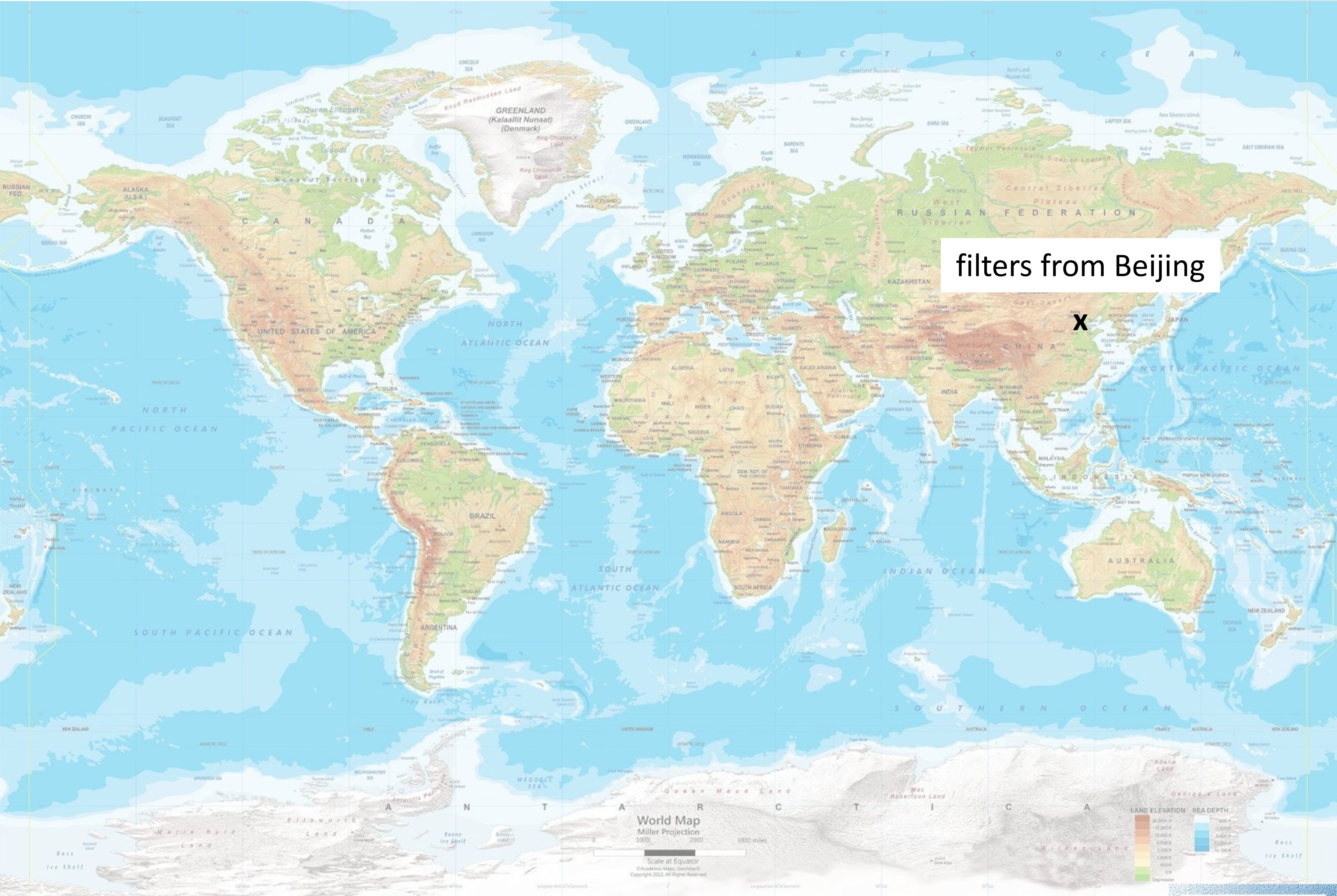
- per surface area (surface site density n_s)
  -> $V_{droplet}$ = surface area of material in one droplet, $S_{droplet}$)



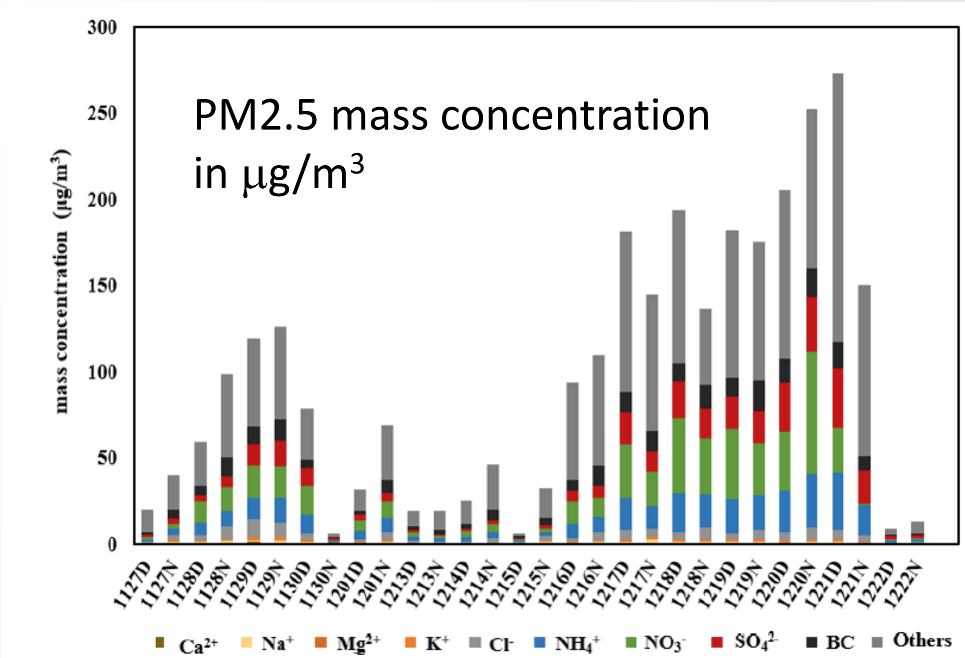
but: $$n_s(T) = \frac{N_i}{S_{tot}}^{*} = \frac{f_{ice}}{S_{droplet}}$$ results from above equation after a Taylor series expansion and is only valid for small f_{ice}

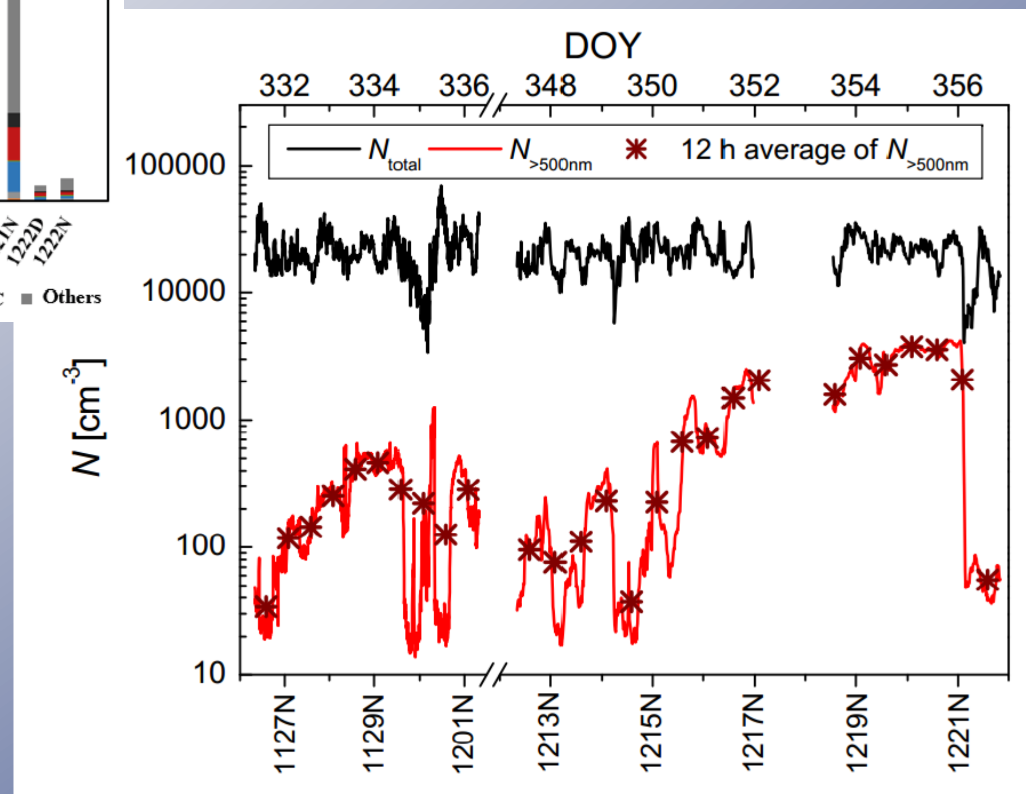
* Note that N_i is small compared with N_d (e.g., $N_i \approx 10^6\ \mathrm{m}^{-3}$ and $N_d \approx 10^8\ \mathrm{m}^{-3}$ at −28°C, corresponding to a frozen fraction f_i of about 1%),

* as introduced in Niemand et al. (2012), A particle-surface-area-based parameterization of Immersion freezing on desert dust particles, JAS.


filters from Beijing
x
ARCTIC OCEAN
GREENLAND (Kalaallit Nunaat) (Denmark)
ALASKA (U.S.A.)
CANADA
UNITED STATES OF AMERICA
NORTH ATLANTIC OCEAN
NORTH PACIFIC OCEAN
RUSSIAN FEDERATION
KAZAKHSTAN
CHINA
INDIA
JAPAN
BRAZIL
ARGENTINA
AUSTRALIA
SOUTH ATLANTIC OCEAN
INDIAN OCEAN
SOUTH PACIFIC OCEAN
SOUTHERN OCEAN
ANTARCTICA
World Map
Miller Projection
Scale at Equator
LAND ELEVATION
SEA DEPTH

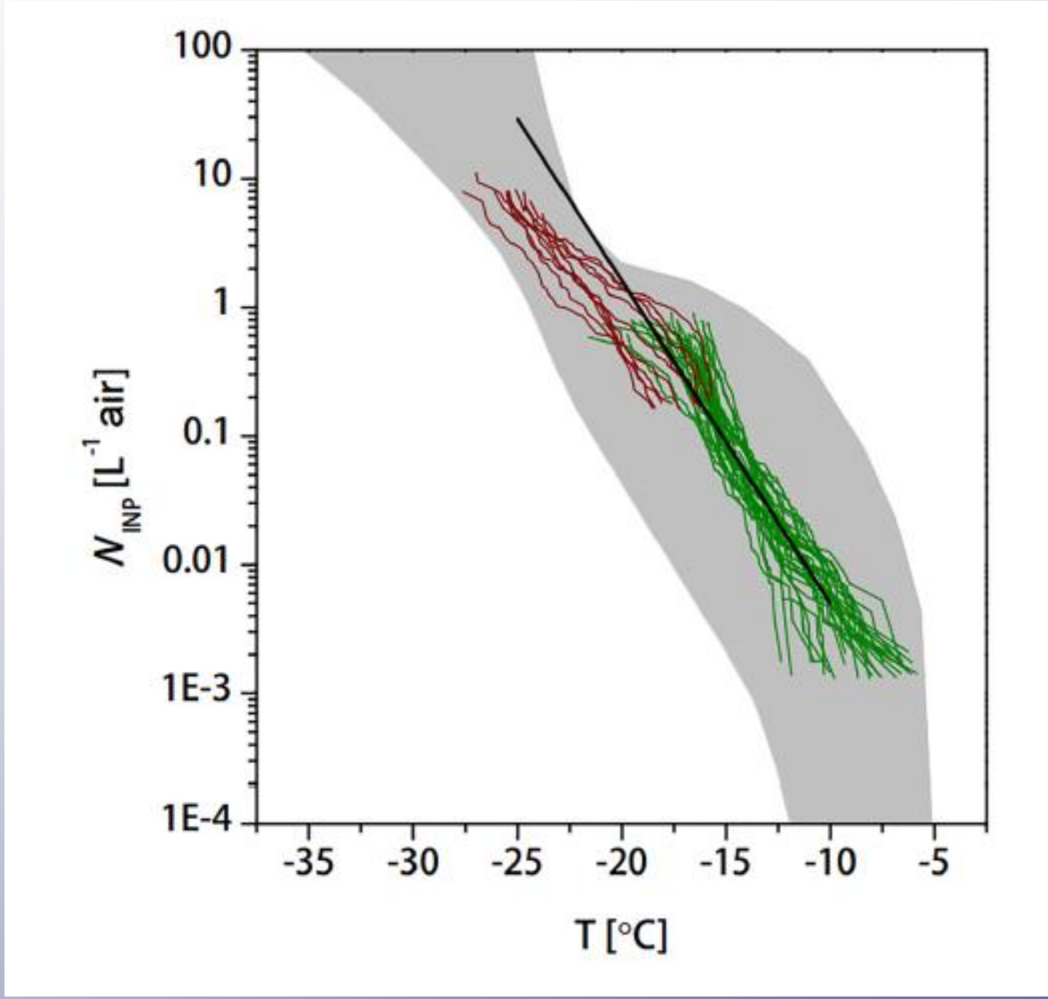
N_{INP} in Beijing during clean and polluted conditions





Chen et al. (2018), Ice nucleating particle concentrations unaffected by urban air pollution in Beijing, China, ACP.

N_{INP} in Beijing during clean and polluted conditions



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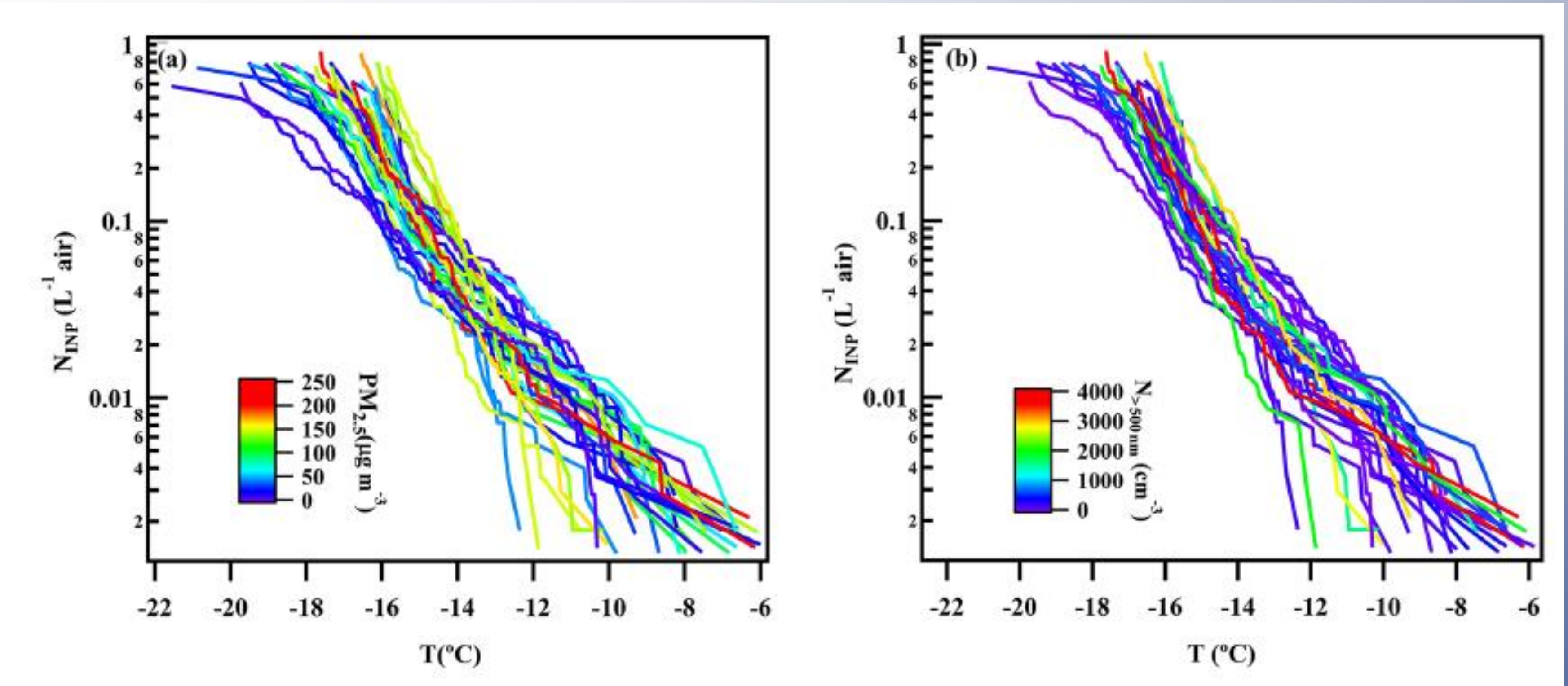
N_{INP} in Beijing during clean and polluted conditions



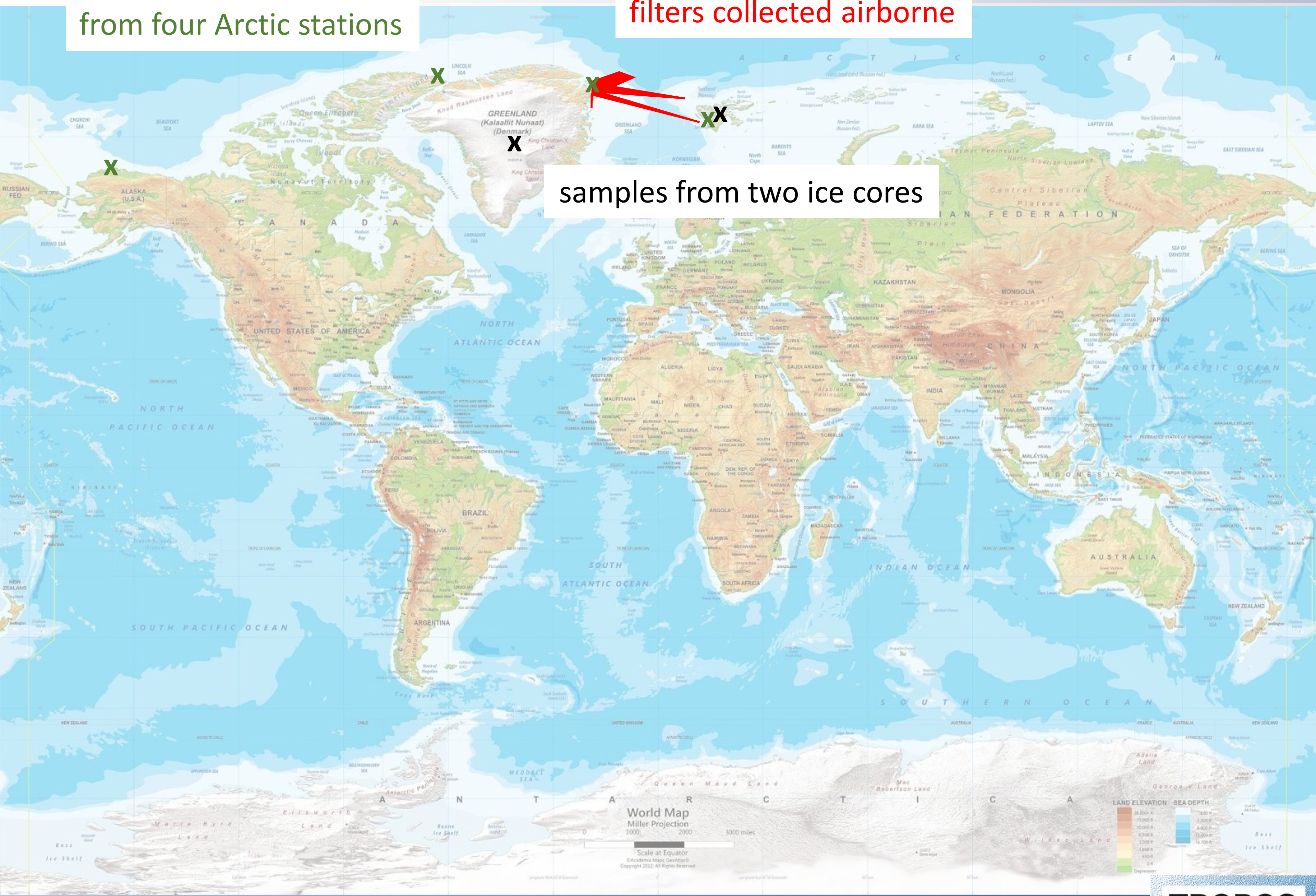
- pollution in Beijing did not add INP that are ice active in the temperature range above -25°C

Chen et al. (2018), Ice nucleating particle concentrations unaffected by urban air pollution in Beijing, China, ACP.

annual series of filters
from four Arctic stations

filters collected airborne

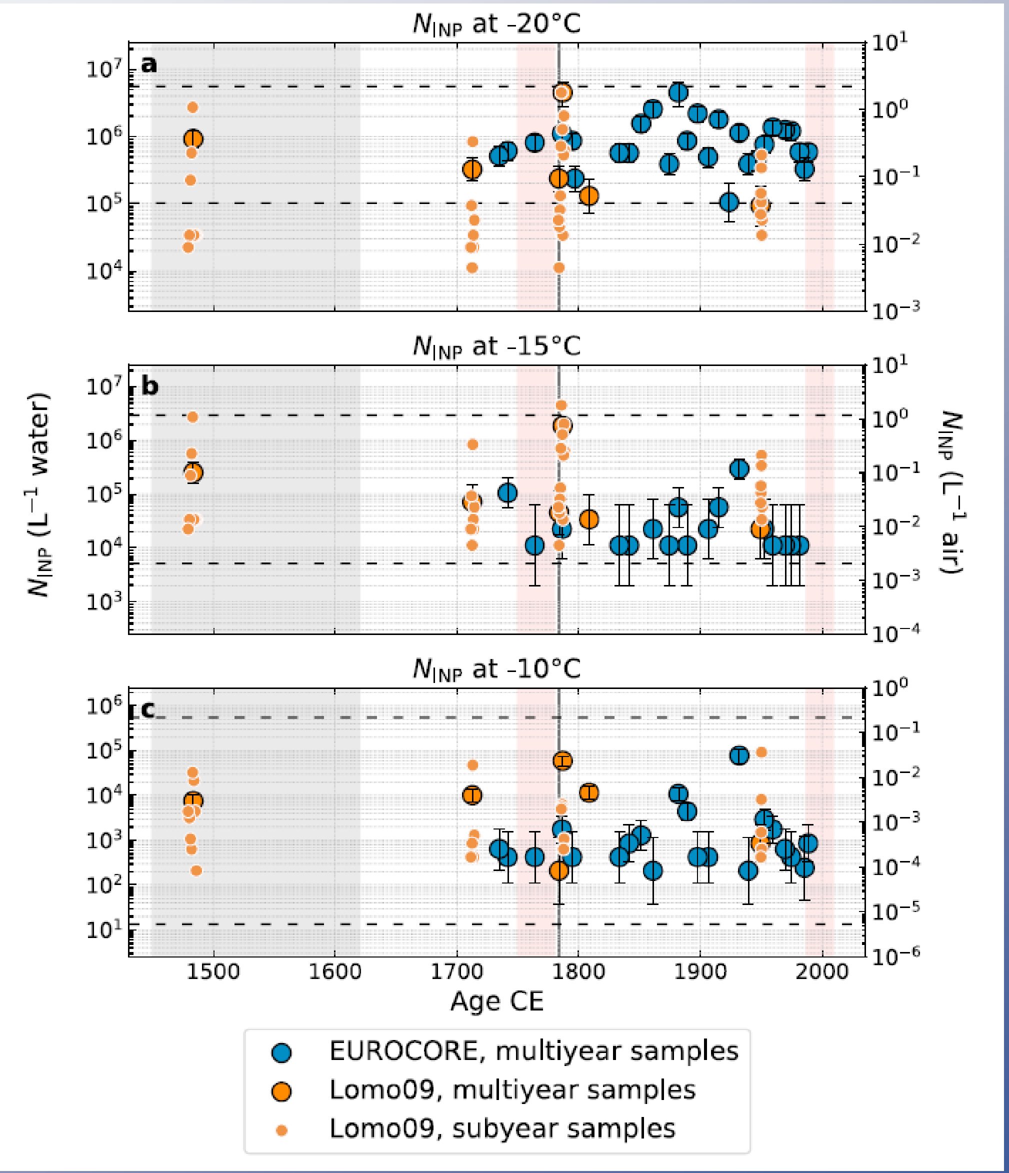
samples from two ice cores



N_{INP} from two Arctic glaciers

two Arctic ice cores
(Greenland and Svalbard)
spanning the years 1478 to 1989

-> no increase in N_{INP} active down to -20°C



Hartmann et al. (2019), Variation of ice nucleating particles in the European Arctic over the last centuries, GRL.

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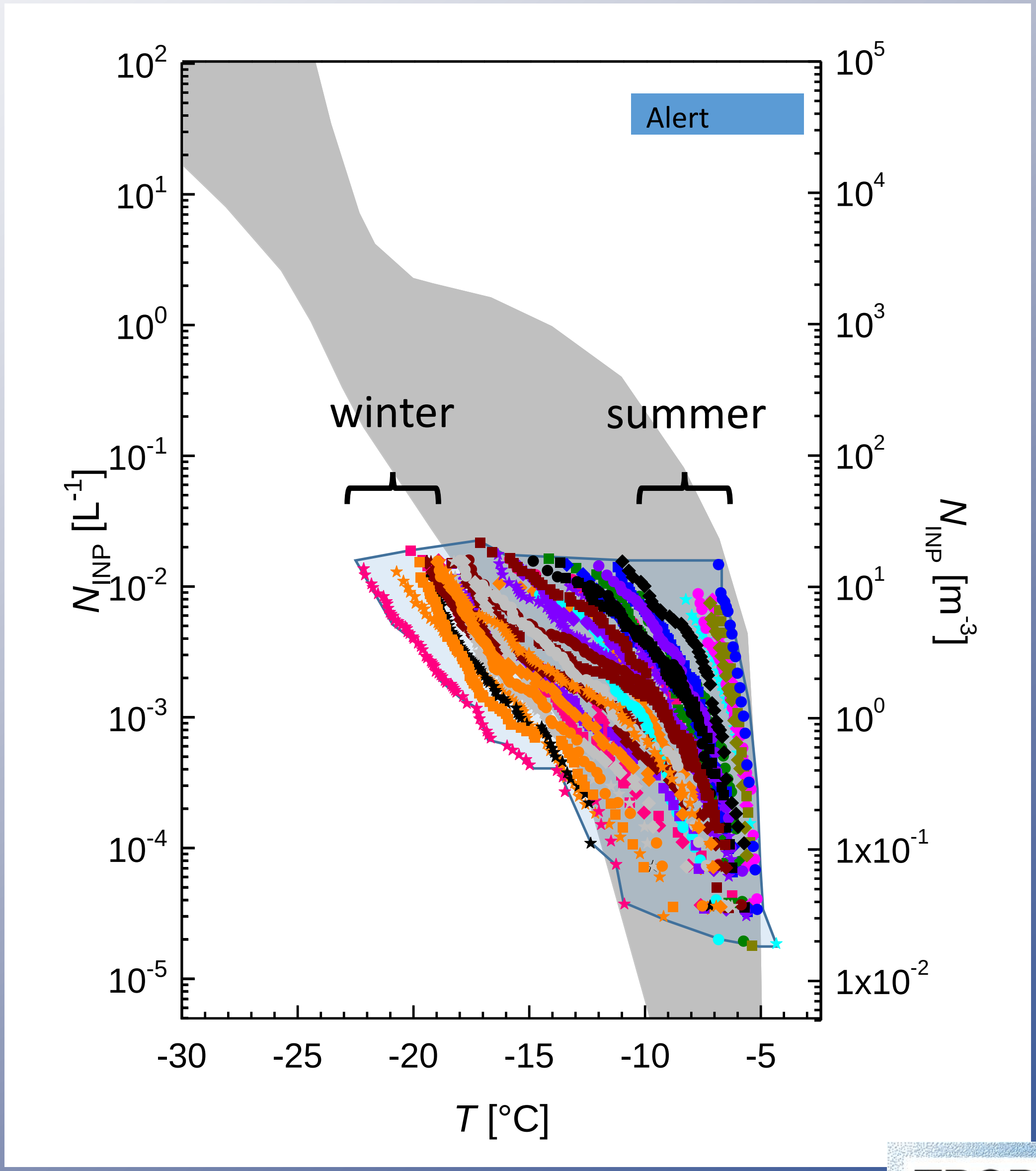
annual N_{INP} from four Arctic stations

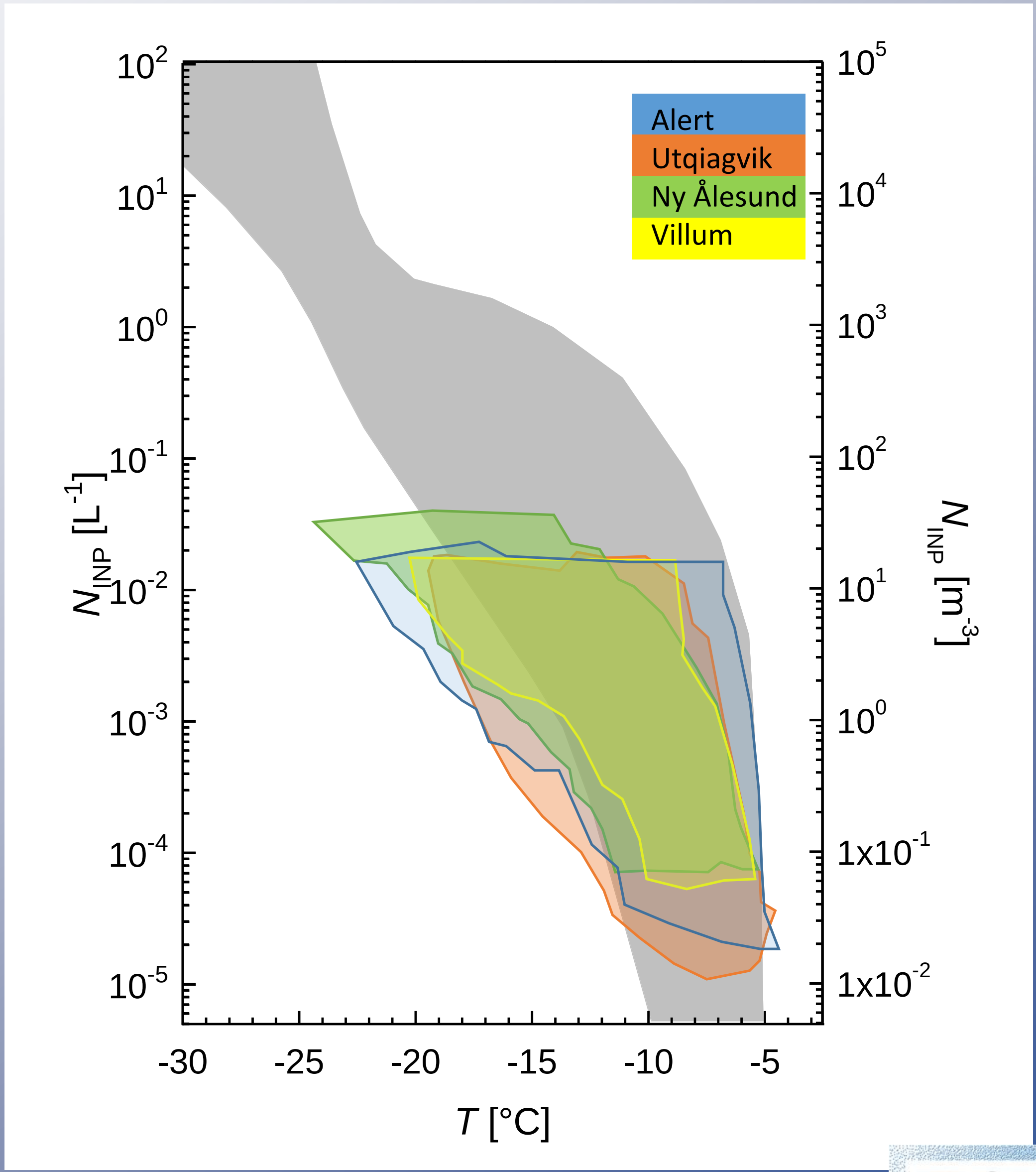


Wex et al. (2019), Annual variability of ice nucleating particle concentrations at different Arctic locations, ACP.

annual N_{INP} from four Arctic stations

high concentrations in summer, origin terrestrial or marine

more indications, that human pollution does not add INP (active > -20°C)



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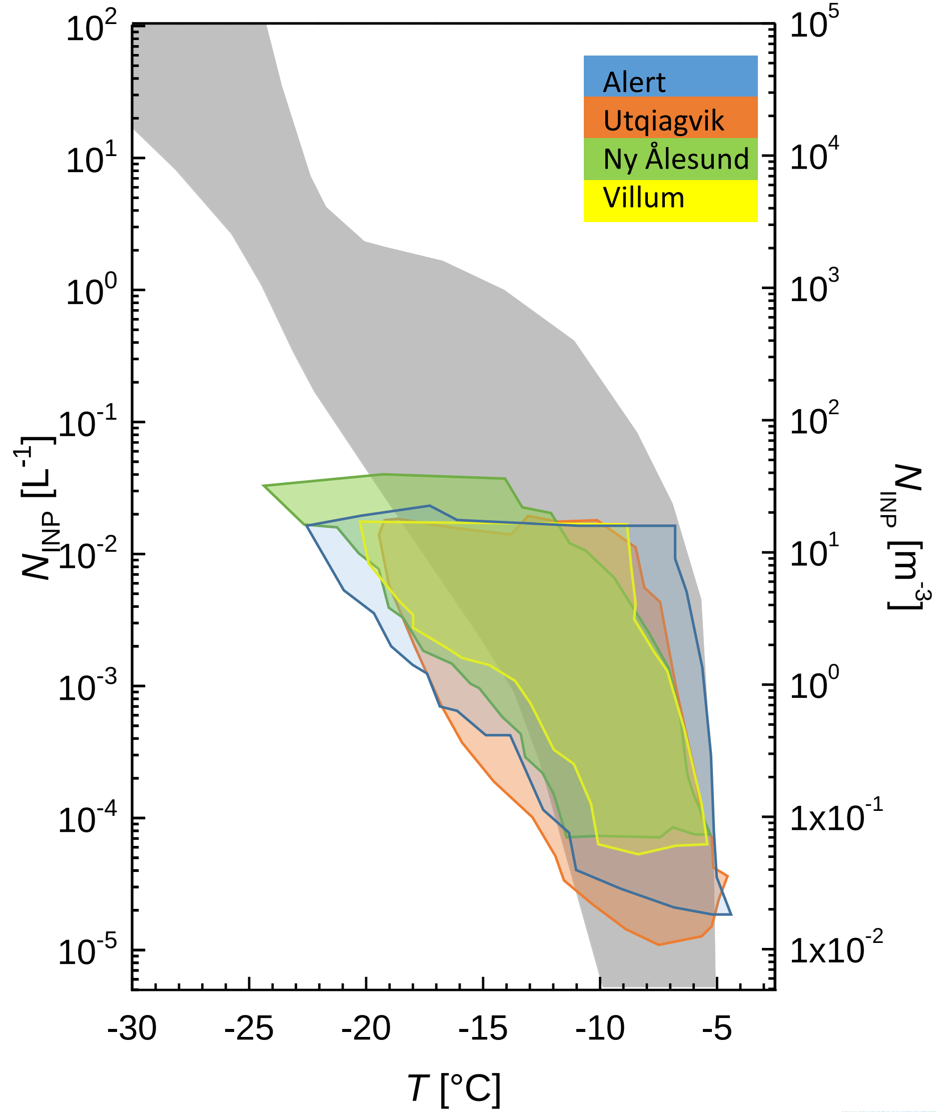


Borys (1983), The effects of long-range transport of air pollutants on Arctic cloud-active aerosol, PhD thesis.

Bigg & Leck (2001), Cloud-active particles over the central Arctic Ocean, JGR.

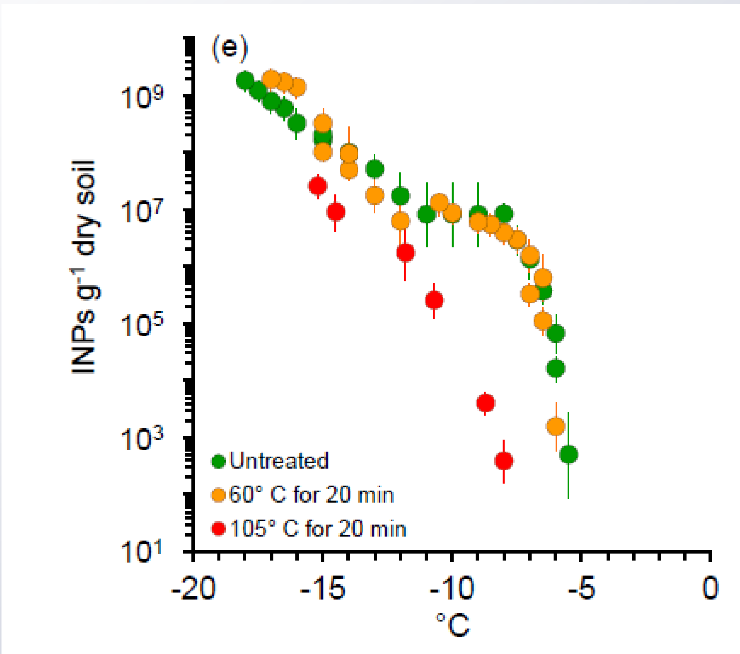
Creamean et al. (2018), Marine and terrestrial influences on ice nucleating particles during continuous springtime measurements in an Arctic oilfield location, ACP.

Tobo et al. (2019), Glacially sourced dust as a potentially significant source of ice nucleating particles, Nat. Geosci.

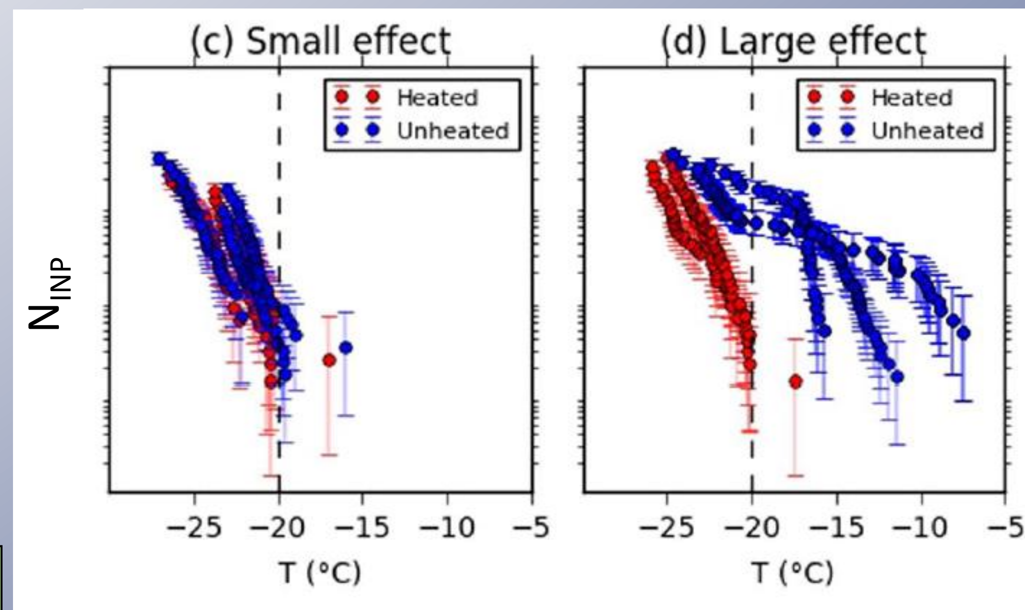
Wex et al. (2019), Annual variability of ice nucleating particle concentrations at different Arctic locations, ACP.

intermezzo: biogenic INP (proteins)

heating samples to test for proteinaceous INP



Hill et al. (2016), Sources of organic ice nucleating particles in soils, ACP.



O'Sullivan et al. (2018), Contributions of biogenic material to the atmospheric ice-nucleating particle population in North Western Europe, Sci. Rep.

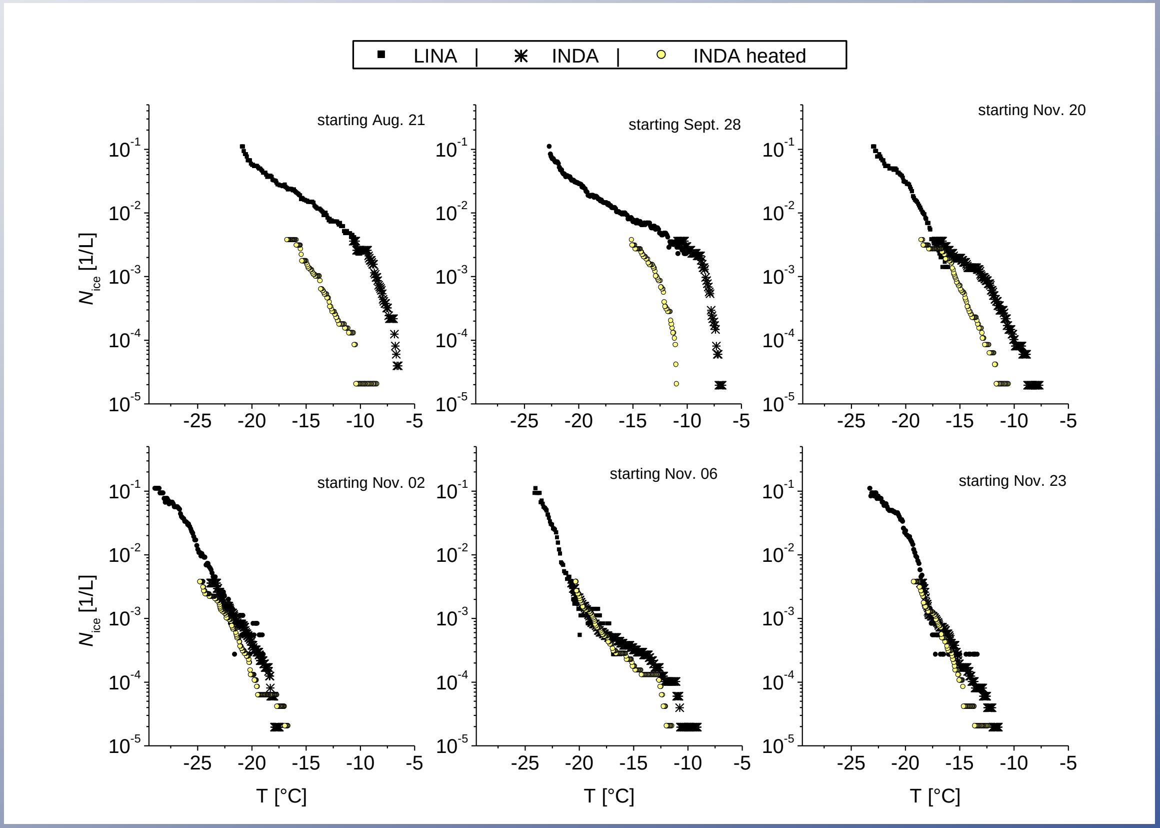
annual N_{INP} from four Arctic stations

high concentrations in summer, origin terrestrial or marine

more indications, that human pollution does not add INP (active > -20°C)

biogenic

for VRS:
unheated and heated (95°C for 1 h) samples from INDA and the unheated samples from LINA



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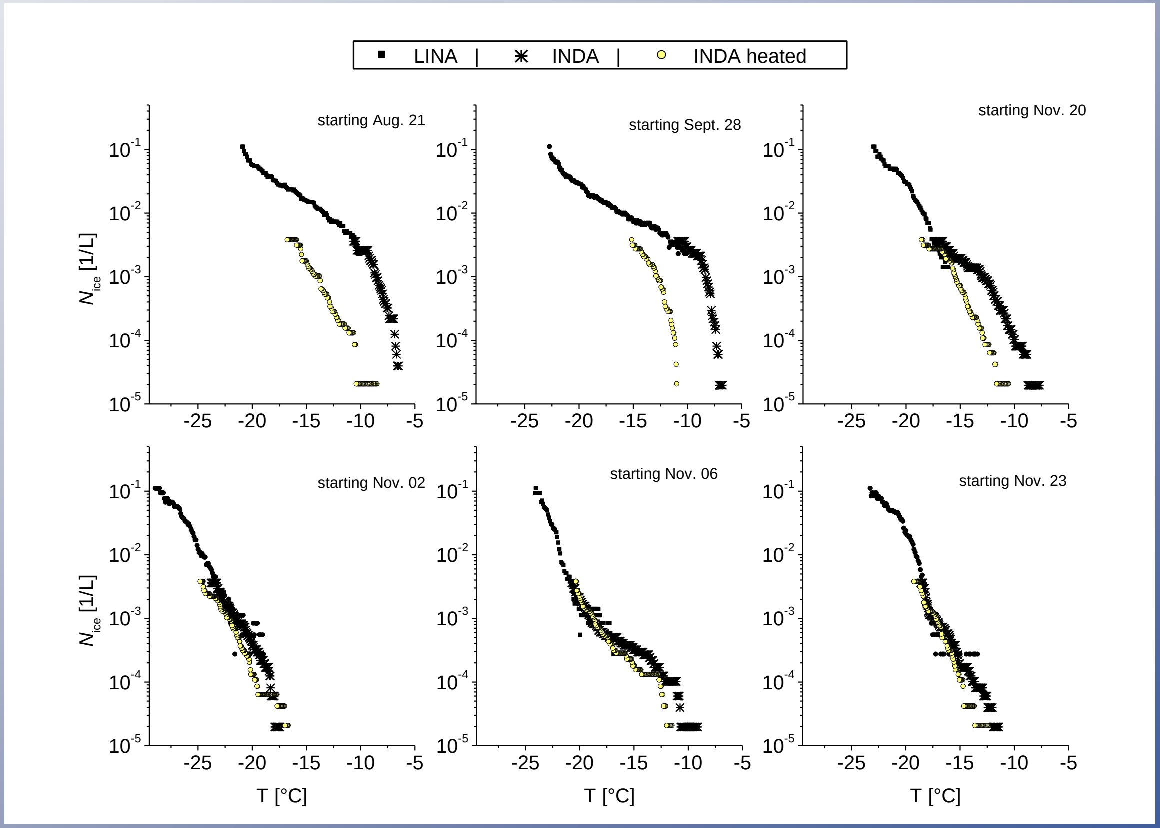

Zeppenfeld et al. (2019), Glucose as a potential chemical marker for ice nucleating activity in Arctic seawater and melt pond samples, ES&T.

Šantl-Temkiv et al. (2019), Biogenic sources of Ice Nucleation Particles at the high Arctic site Villum Research Station, ES&T.

Wex et al. (2019), Annual variability of ice nucleating particle concentrations at different Arctic locations, ACP.

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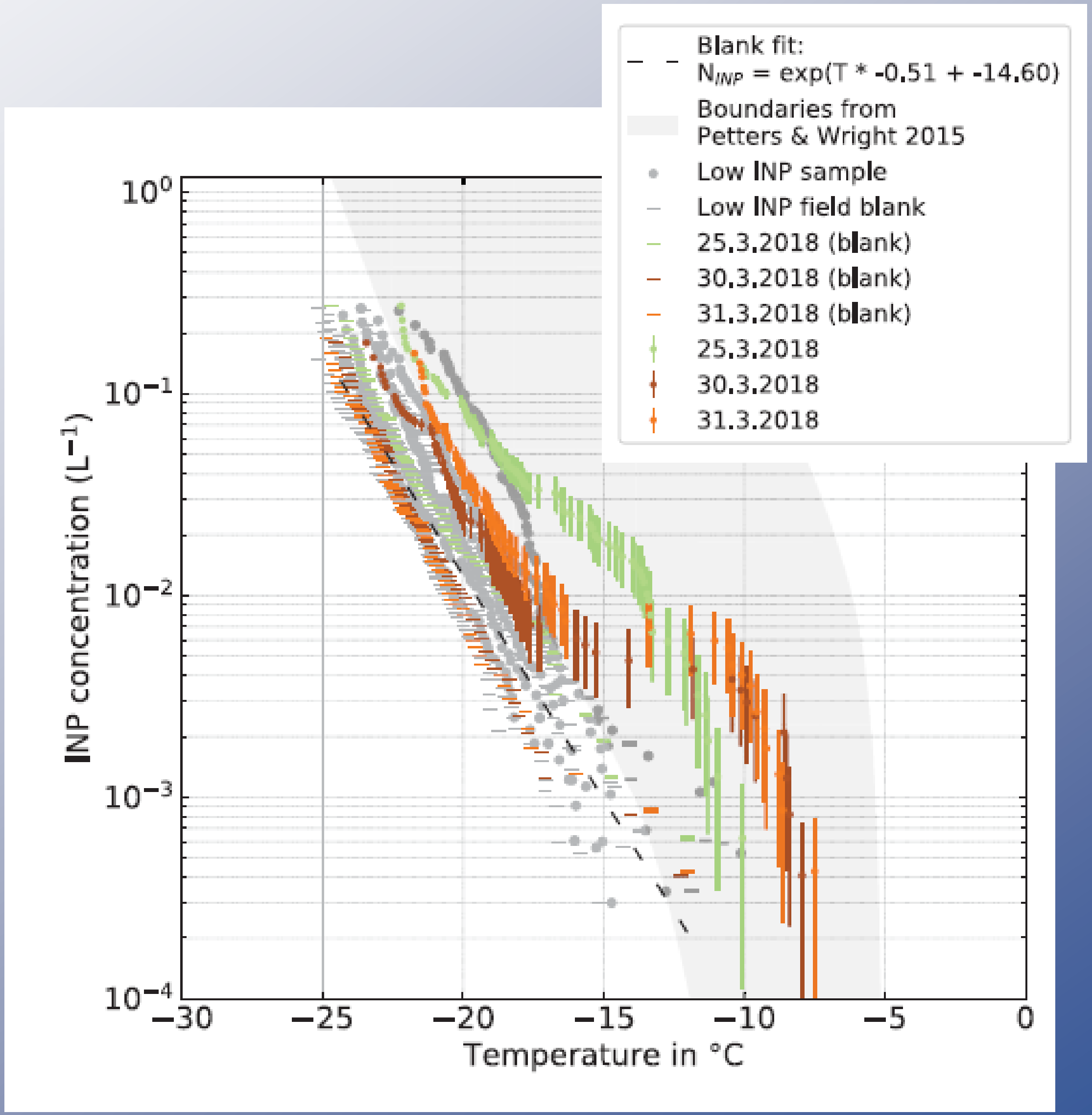
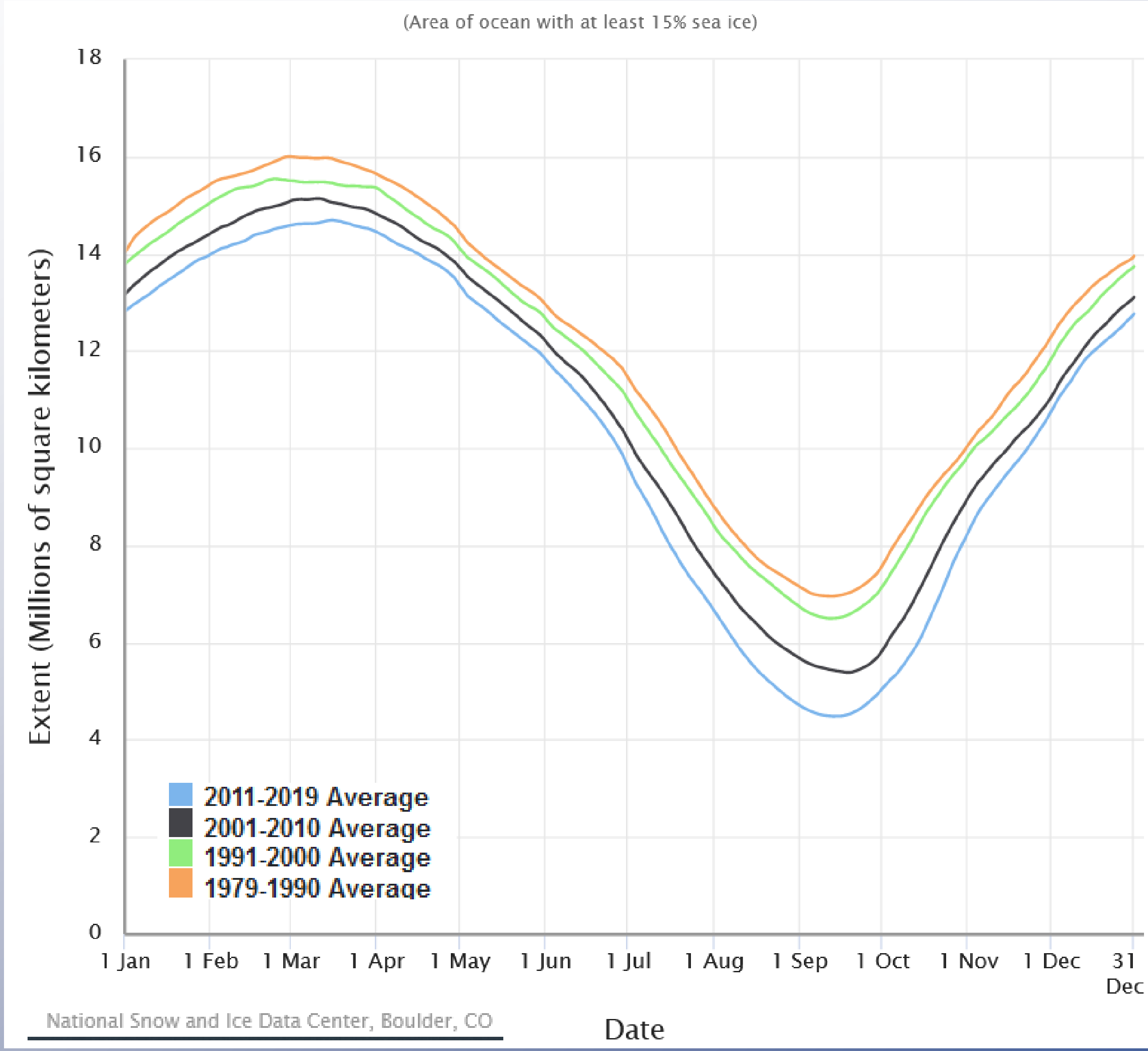
airborne Arctic N_{INP}



at least in March over polynyas, INP (likely biogenic) originated locally from the ocean area

Hartmann et al. (2020), Wintertime airborne measurements of ice nucleating particles in the high Arctic: a hint to a marine, biogenic source for Ice Nucleating Particles, GRL.

intermezzo: Arcic Sea Ice Extent



https://nsidc.org/arcticseaicenews/charctic-interactive-sea-ice-graph/

N_{INP} in the Arctic

- annual cycle of N_{INP} with high values in summer, likely biogenic

- more indications, that human pollution does not add INP (active > -20°C)

    - two ice cores
    - high concentrations in summer

- origin terrestrial or marine

- at least in March over polynyas, INP originated locally from the ocean area

Hartmann et al. (2019), Variation of ice nucleating particles in the European Arctic over the last centuries, GRL.

Hartmann et al. (2020), Wintertime airborne measurements of ice nucleating particles in the high Arctic: a hint to a marine, biogenic source for Ice Nucleating Particles, GRL.

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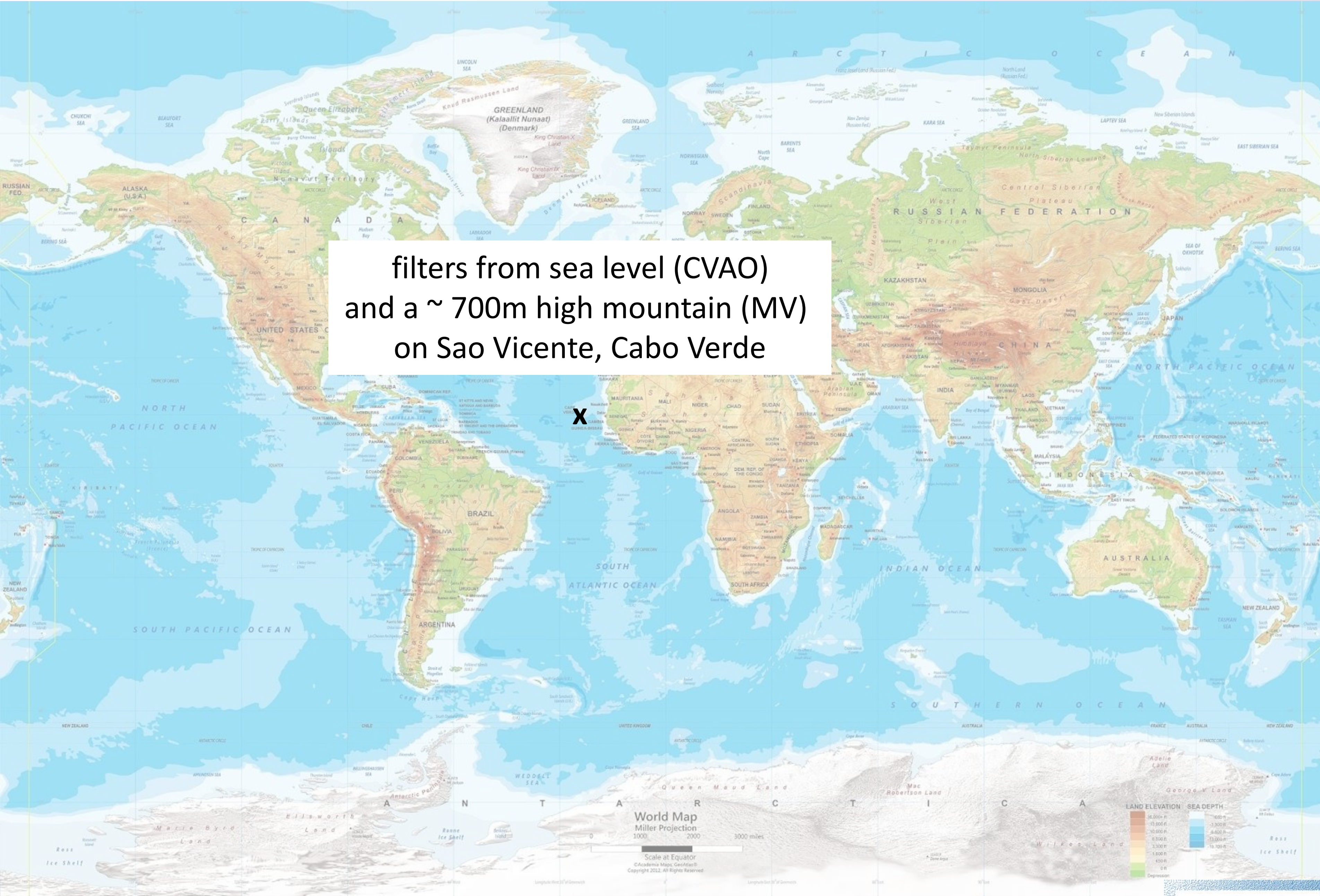
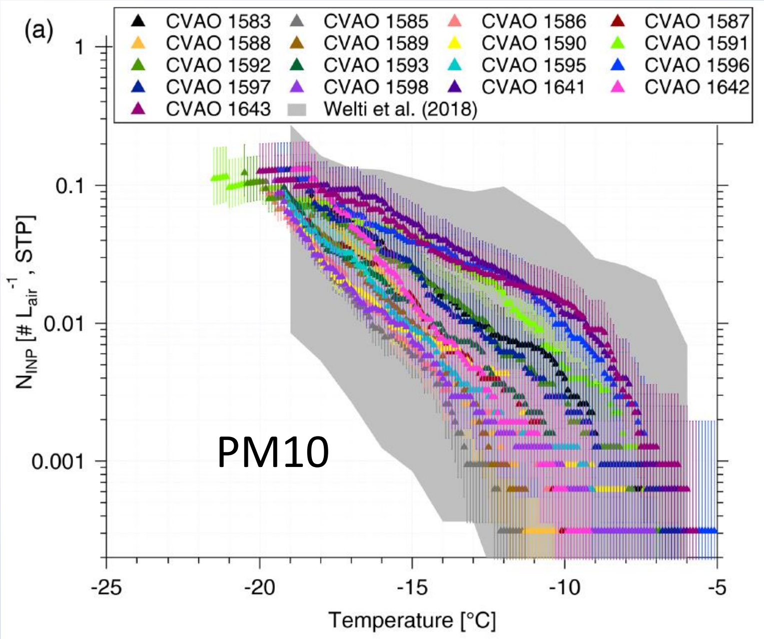
filters from sea level (CVAO)
and a ~ 700m high mountain (MV)
on Sao Vicente, Cabo Verde



N_{INP} in the tropical Atlantic region, Cabo Verde



Welti et al. (2018), Concentration and variability of ice nuclei in the subtropical maritime boundary layer, ACP.

Gong et al. (2020), Characterization of aerosol particles at Cape Verde close to sea and cloud level heights - Part 2: ice nucleating particles in air, cloud and seawater, ACP.

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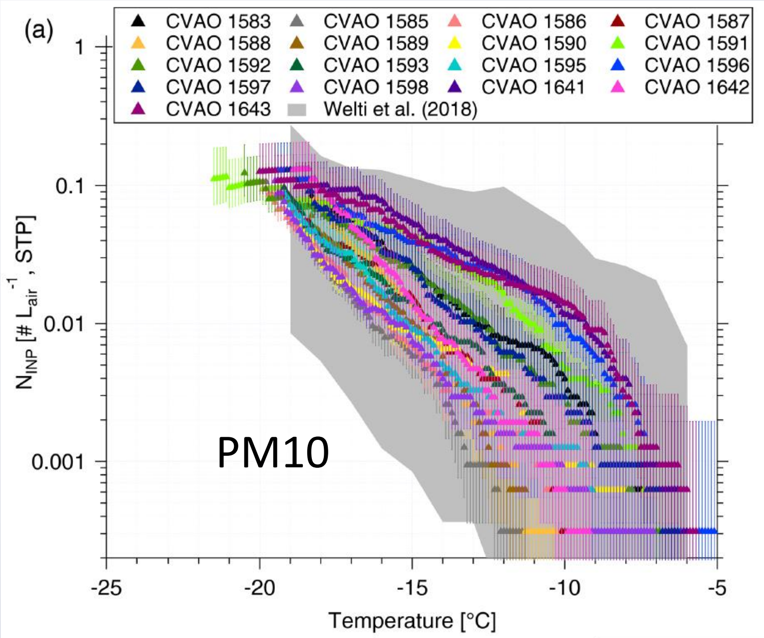
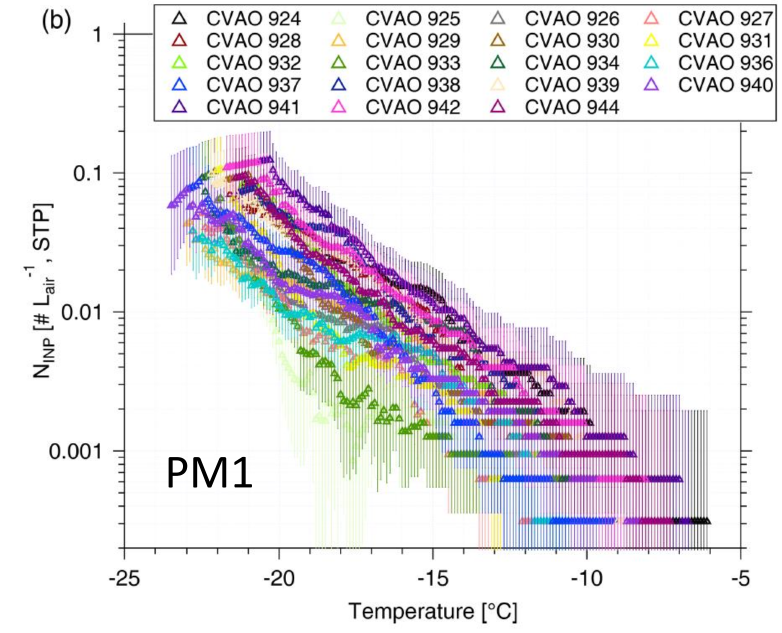
N_{INP} in the tropical Atlantic region, Cabo Verde





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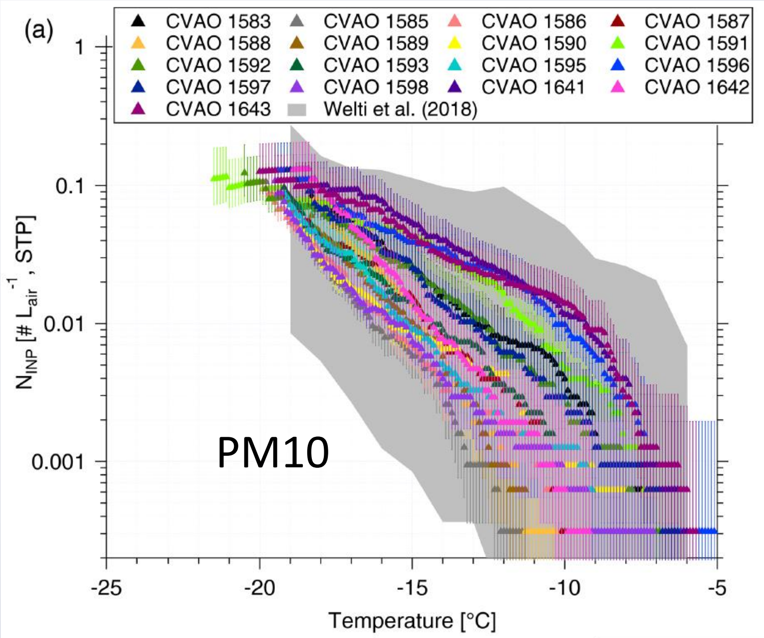
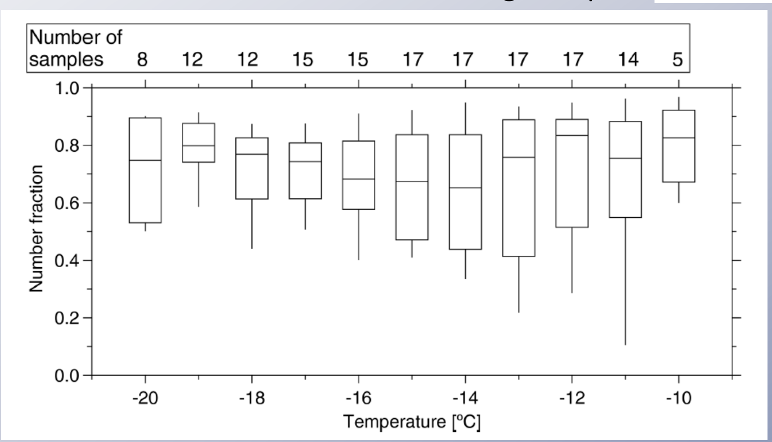
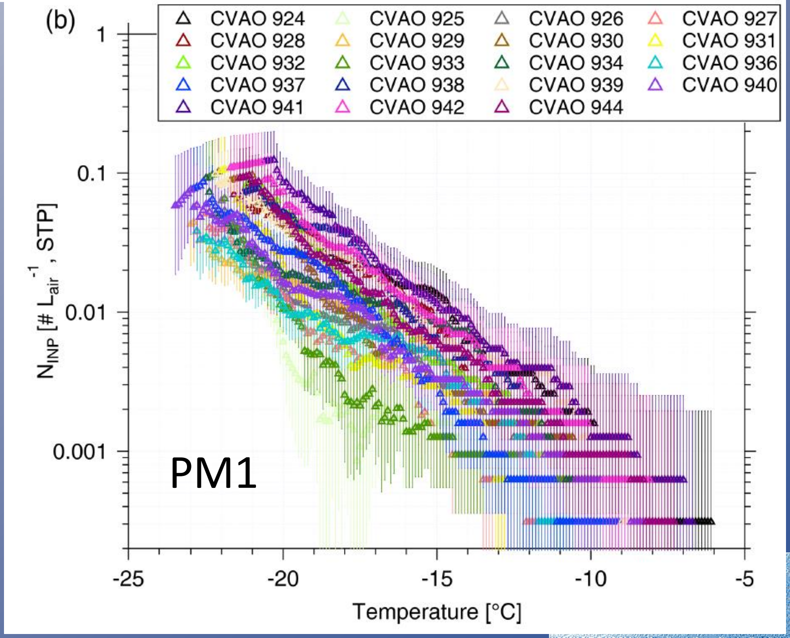
N_{INP} in the tropical Atlantic region, Cabo Verde

most (> 70%) of all INP were supermicron in size



number fraction of INPs in the size range > 1 µm

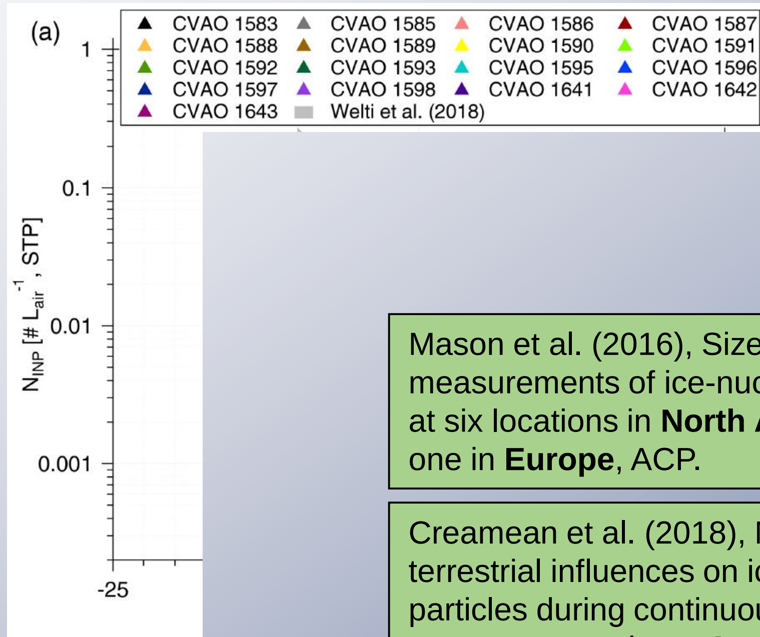




Gong et al. (2020), Characterization of aerosol particles at Cape Verde close to sea and cloud level heights - Part 2: ice nucleating particles in air, cloud and seawater, ACP.

N_{INP} in the tropical Atlantic region, Cabo Verde

most (> 70%) of all INP were supermicron in size



Mason et al. (2016), Size-resolved measurements of ice-nucleating particles at six locations in **North America** and one in **Europe**, ACP.

Creamean et al. (2018), Marine and terrestrial influences on ice nucleating particles during continuous springtime measurements in an **Arctic** oilfield location, ACP.

Si et al. (2018), Ice-nucleating ability of aerosol particles and possible sources at three coastal marine sites, ACP.

(**in Canada, one site Arctic**)

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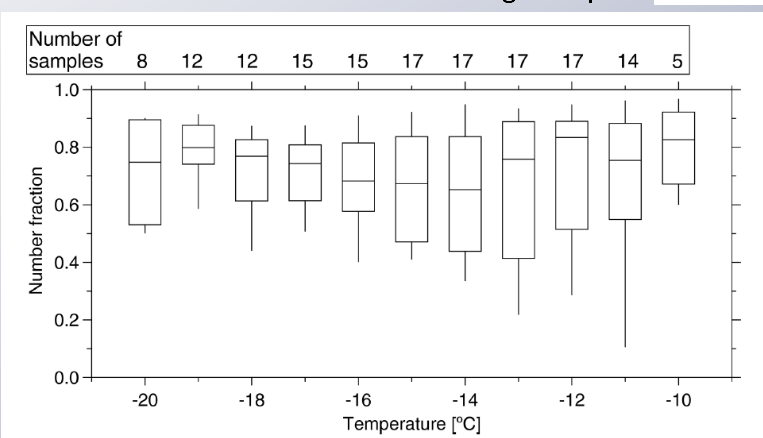

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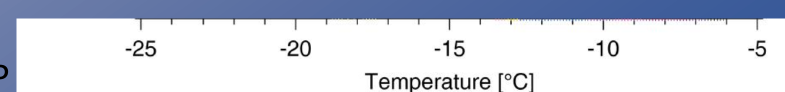
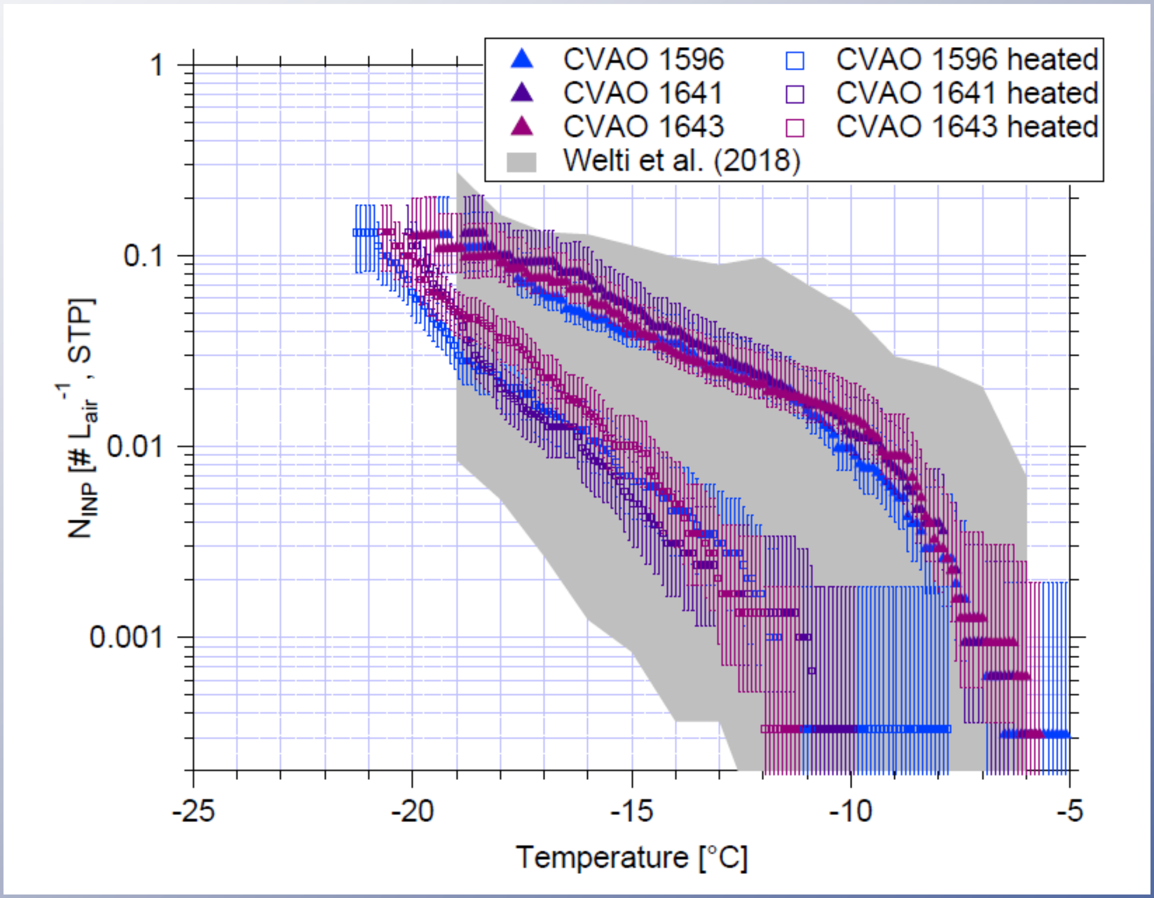


N_{INP} in the tropical Atlantic region, Cabo Verde

biogenic INP were present

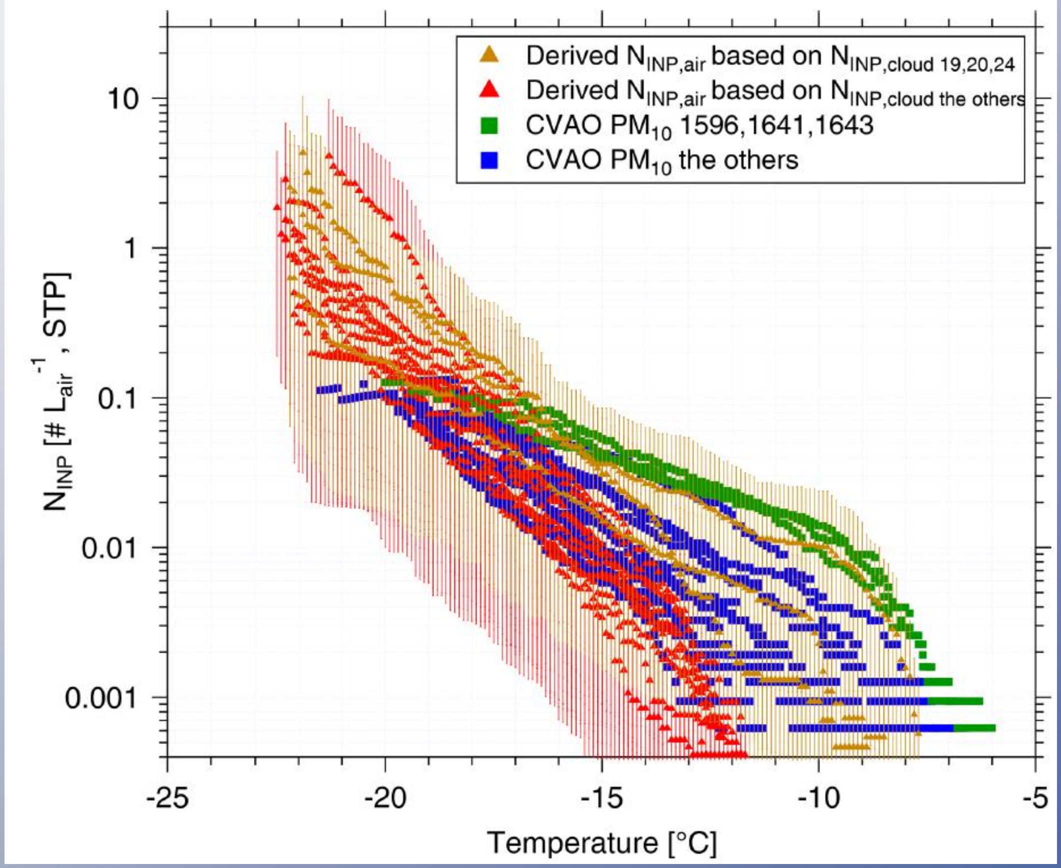


Gong et al. (2020), Characterization of aerosol particles at Cape Verde close to sea and cloud level heights - Part 2: ice nucleating particles in air, cloud and seawater, ACP.

N_{INP} in the tropical Atlantic region, Cabo Verde

at Cape Verde, the marine boundary layer was well mixed

N_{INP} in air and cloud water fit well



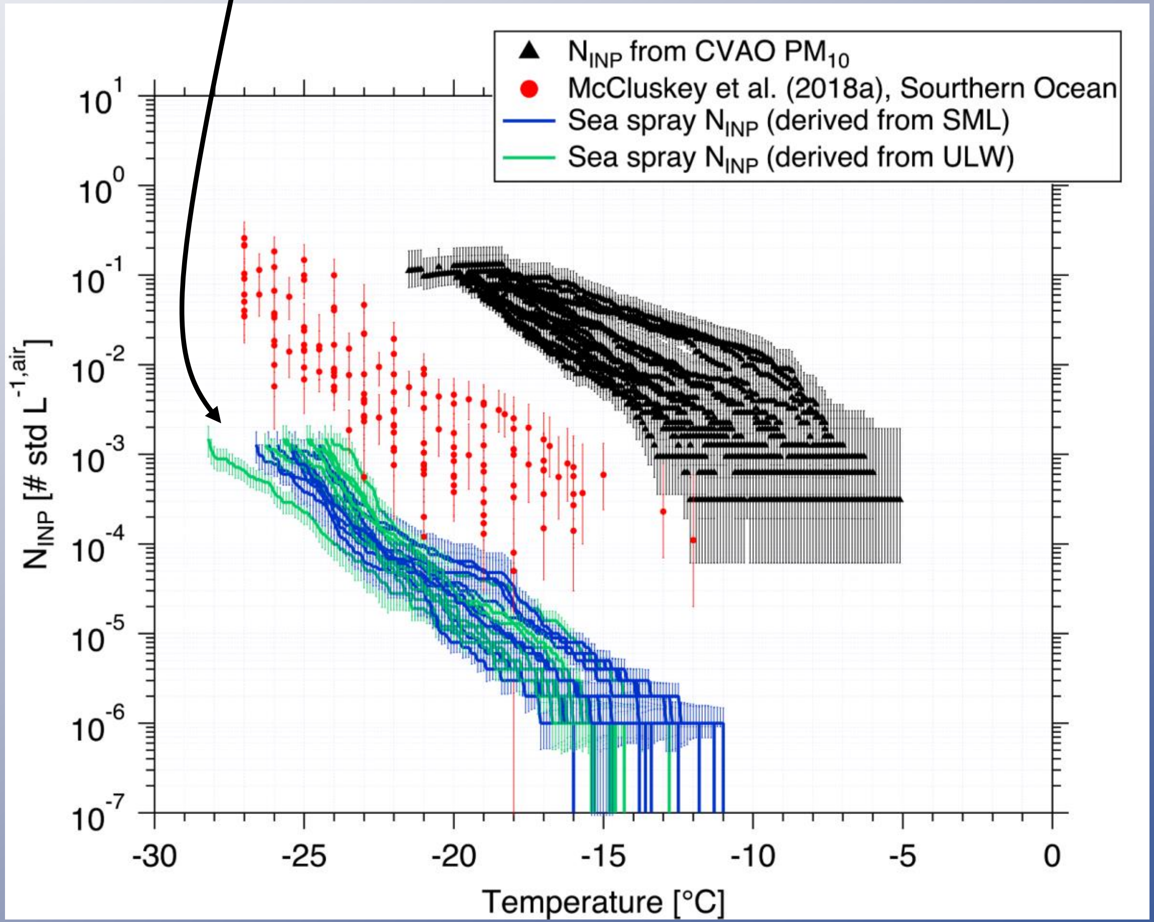
compared by scaling with CCN-concentrations and an assumed droplet size

Gong et al. (2020), Characterization of aerosol particles at Cape Verde close to sea and cloud level heights - Part 2: ice nucleating particles in air, cloud and seawater, ACP.

N_{INP} in the tropical Atlantic region, Cabo Verde

$$N_{INP}^{\text{sea spray,air}} = \frac{\text{NaCl}_{\text{mass,air}}}{\text{NaCl}_{\text{mass,seawater}}} \cdot N_{INP}^{\text{seawater}}$$

INP from the ocean likely only contributed a small fraction of all INP



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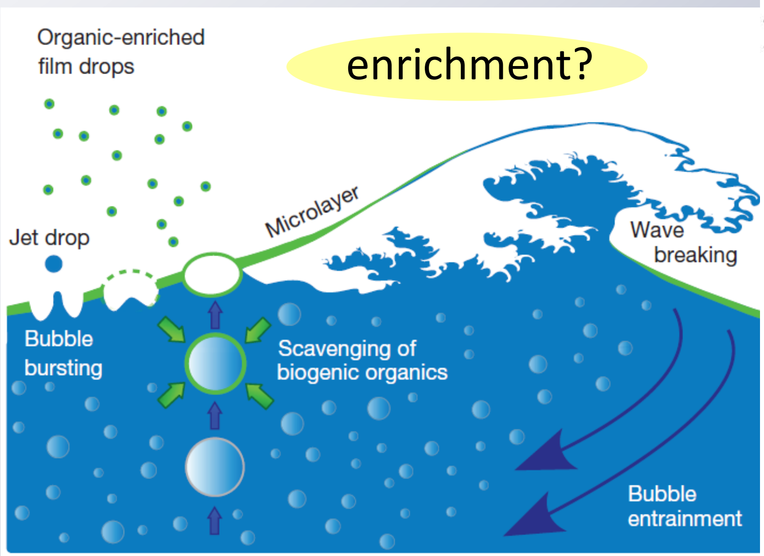
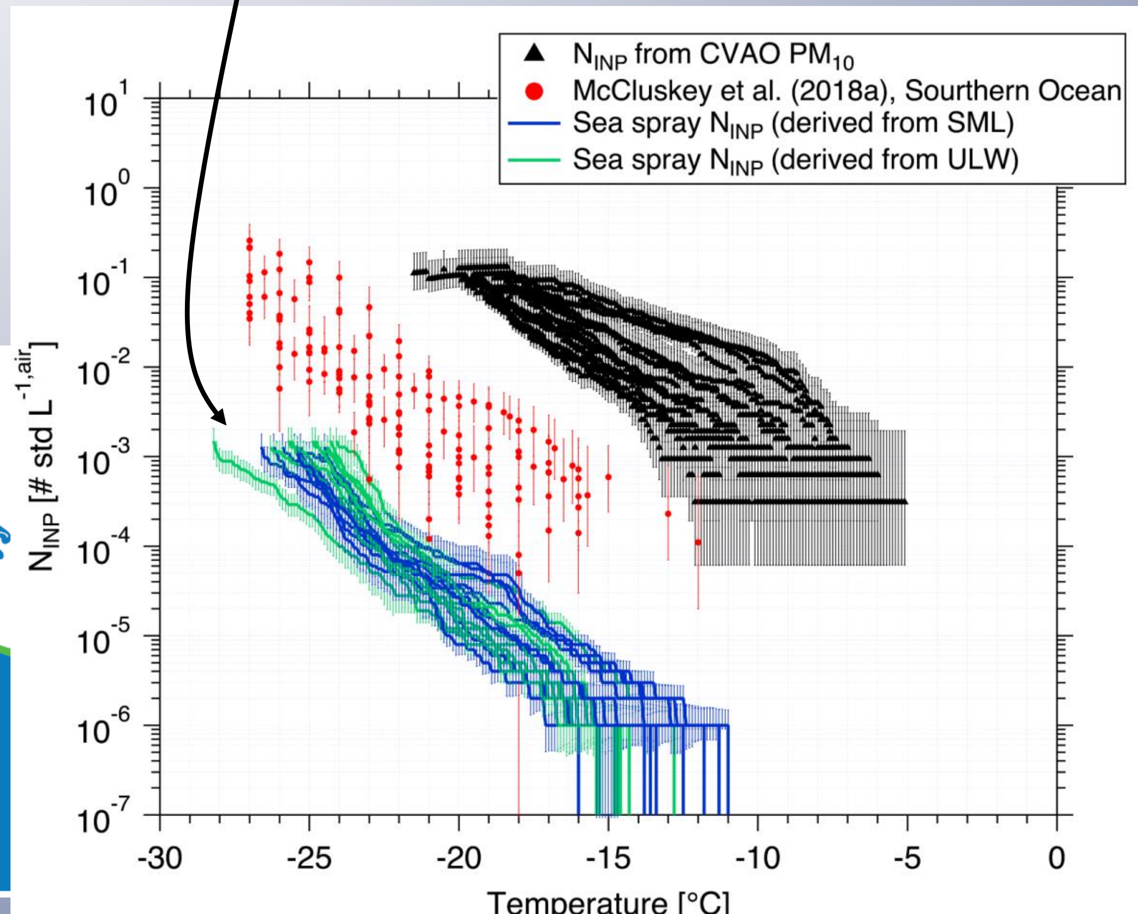


Wilson et al. (2015), A marine biogenic source of atmospheric ice-nucleating particles, Nature.



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N_{INP} in the tropical Atlantic region, Cabo Verde

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- biogenic INP were present
- marine boundary layer was well mixed
- N_{INP} in air and cloud water fit well
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the picture I have in my head

- human pollution does not add INP (active > -25°C)

- supermicron INP are important in the atmosphere

- high ice activity at high ice nucleation temperatures is (often) related to biogenic INP

- INP origin terrestrial or marine

  –> it is often assumed that terrestrial sources are over-whelming, but the source strength of the ocean is still unknown, as well as biogenic contributions from terrestrial areas

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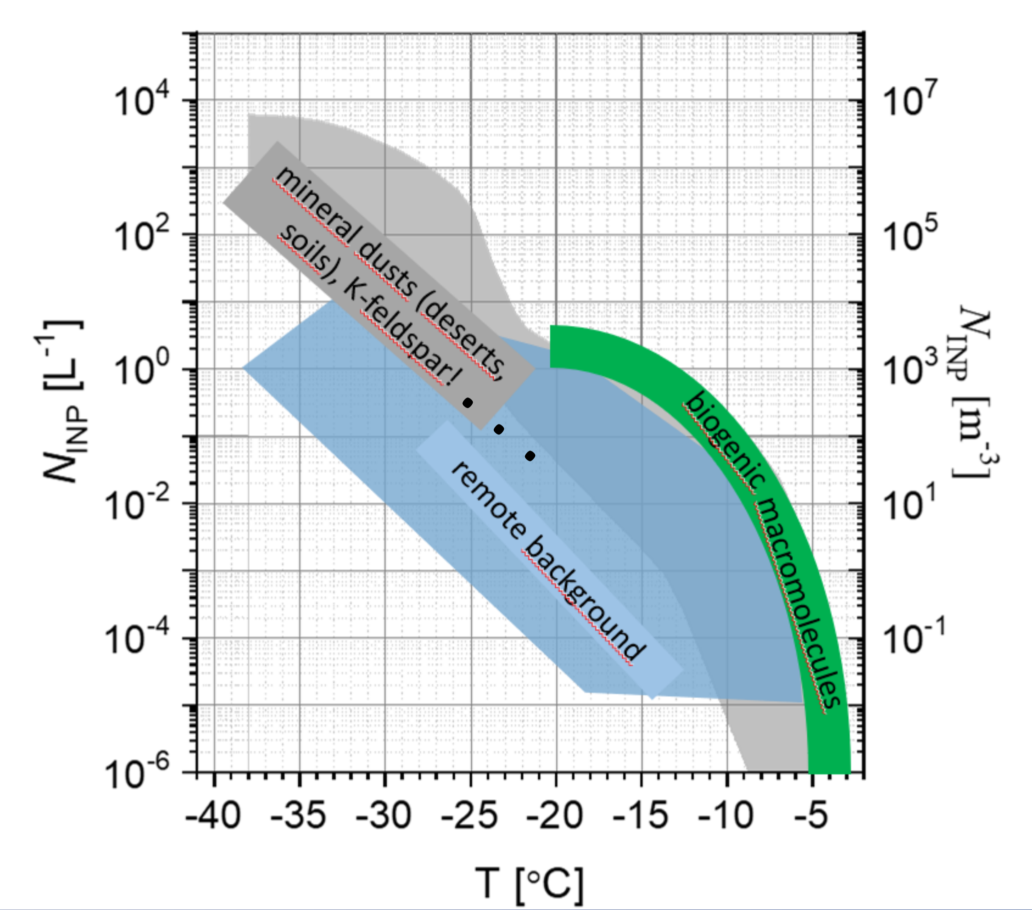

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Thank you for listening