Ice nucleating particles (INP)

in the atmosphere

by

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Leibniz Association

TROPOS
Leibniz Institute for
Tropospheric Research

overview

- general introduction

- basic knowledge

- result from recent field studies

each roughly 1/3 of the talk
ALL will need to stay on the surface

atmospheric aerosol particle sources








primary: particles are emitted **directly**

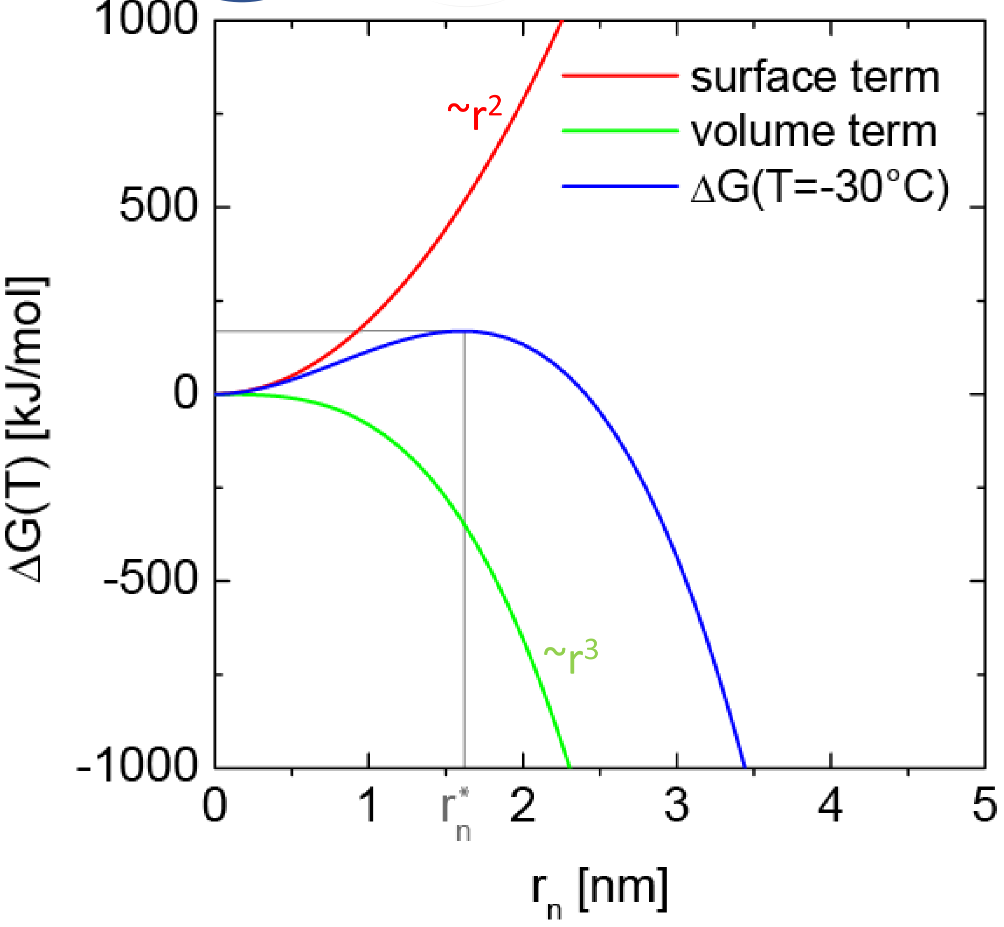
secondary: particulate mass formed from **gaseous precursors**

particle ageing

particle concentrations: **roughly 100 to 10 000 cm^{-3}**

INP (ice nucleating particles): **VERY rare** subgroup, with some ten to hundred L^{-1} or m^{-3}

homogeneous freezing



- below 0°C, ice is the thermodynamic stable phase, BUT:
- for ice formation, energy barrier needs to be overcome
- ice germ: needs a critical size (r_n*), once this is reached, droplet freezes
- r_n* is temperature dependent

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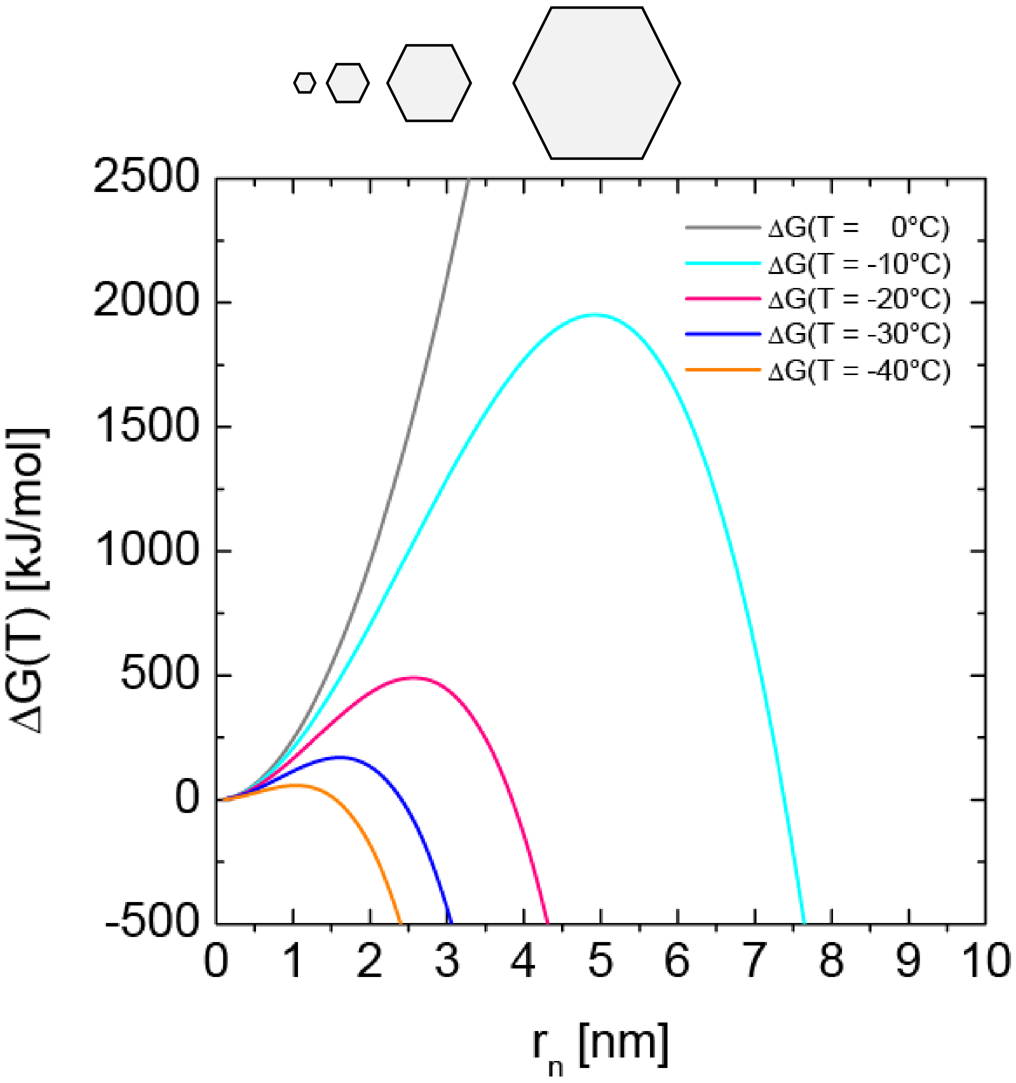

look out for a good additional explanation in lecture from Thomas Koop on:
https://iac.ethz.ch/group/atmospheric-physics/research/ice-nucleation-colloquium.html

- below 0°C, ice is the thermodynamic stable phase, BUT:
- for ice formation, energy barrier needs to be overcome
- ice germ: needs a critical size (r_n*), once this is reached, droplet freezes
- r_n* is temperature dependent
- homogeneous freezing: below -38°C, critical germ size can randomly be reached
- above -38°C: INP „stabilize“ or „arrange“ water molecules and fill part of the void

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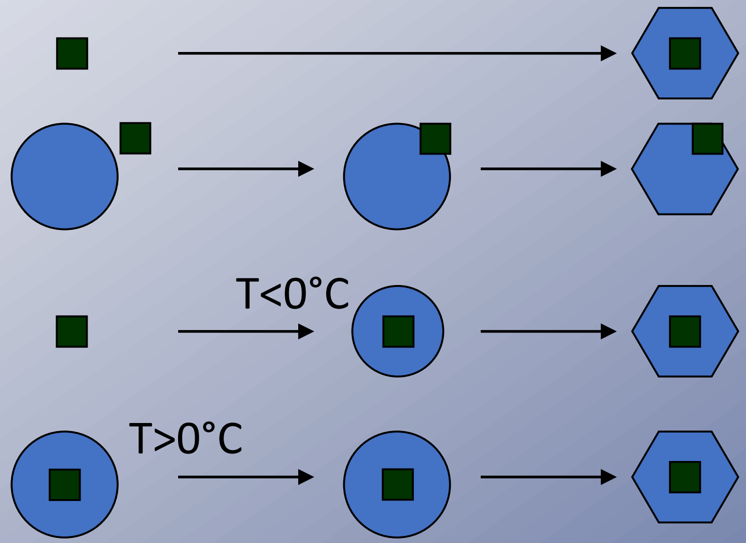
heterogeneous freezing

⇨ heterogenous freezing, i.e., nucleation is aided by a „catalyst“

(a surface on which water molecules can arrange in an ice like manner)

different heterogenous freezing processes:

- deposition ice nucleation
- contact freezing
- condensation freezing
- immersion freezing



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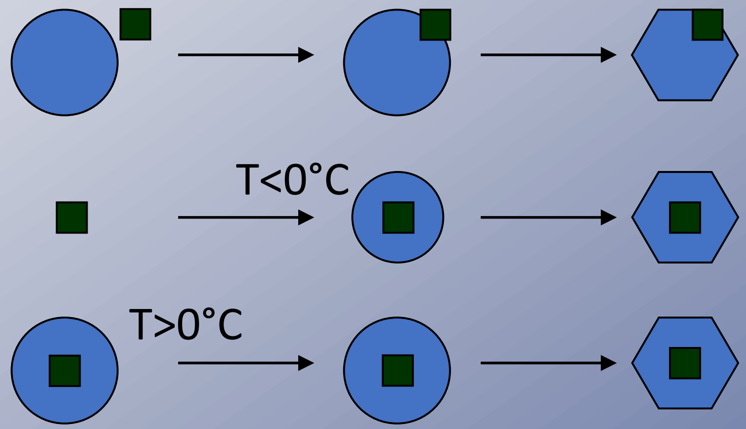

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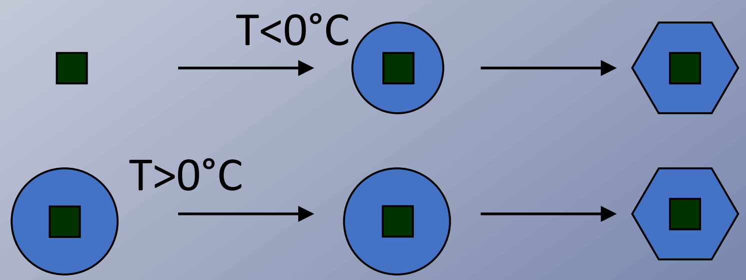

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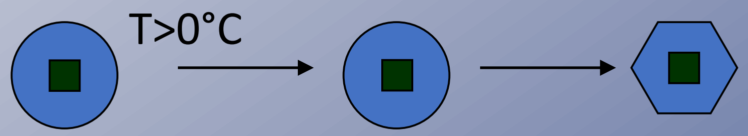

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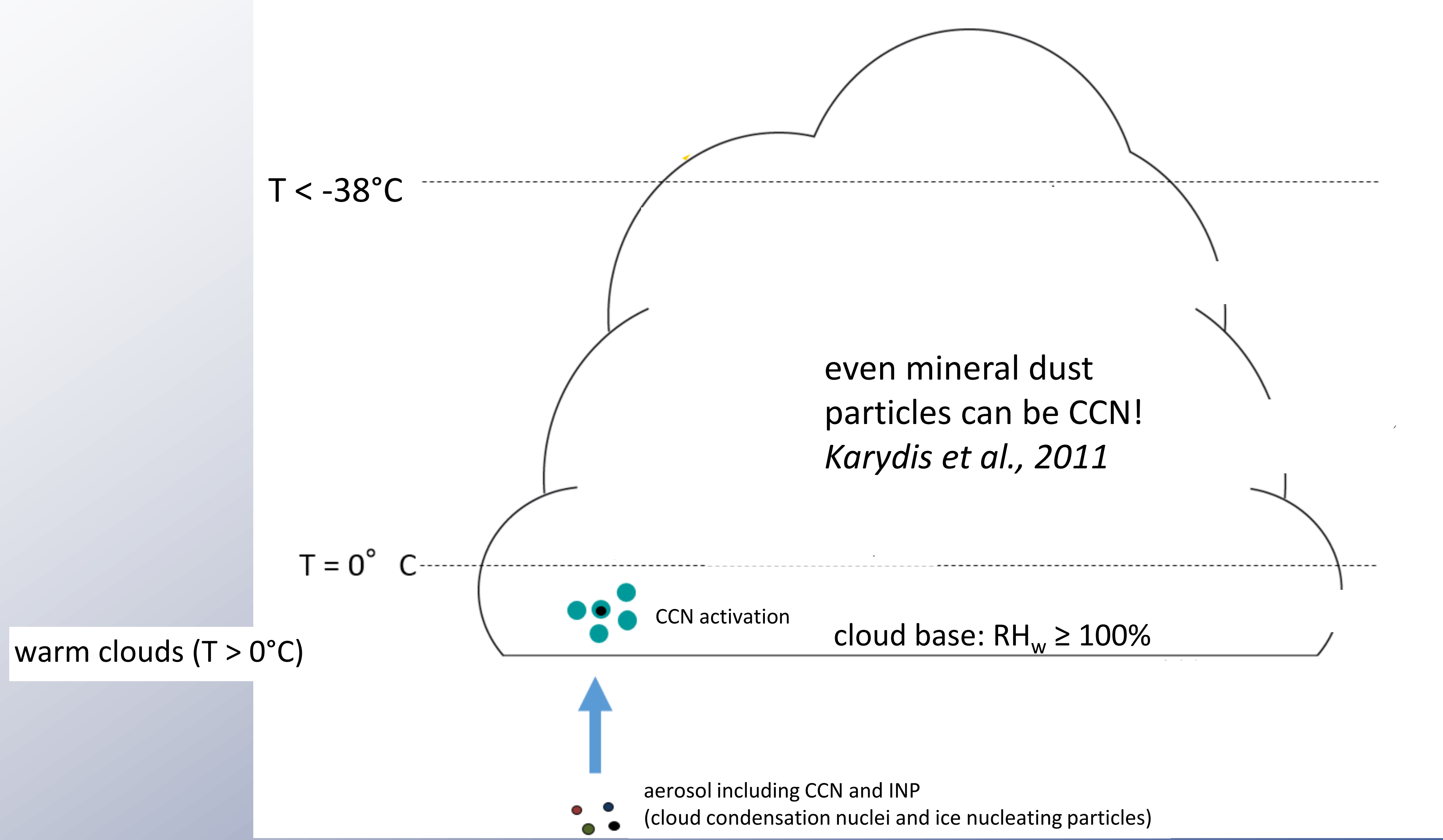
- contact freezing (*Hoffmann et al., 2013*, BUT: in atmosphere, **collisions too seldom**)
- condensation freezing (**immersion freezing in concentrated solutions**, *Wex et al., 2014*)
- immersion freezing (most important freezing process in **mixed phase clouds (at T between 0°C and -38°C)**, e.g., *Ansmann et al., 2008; de Boer et al. 2011; Westbrook and Illingworth, 2013*)

clouds



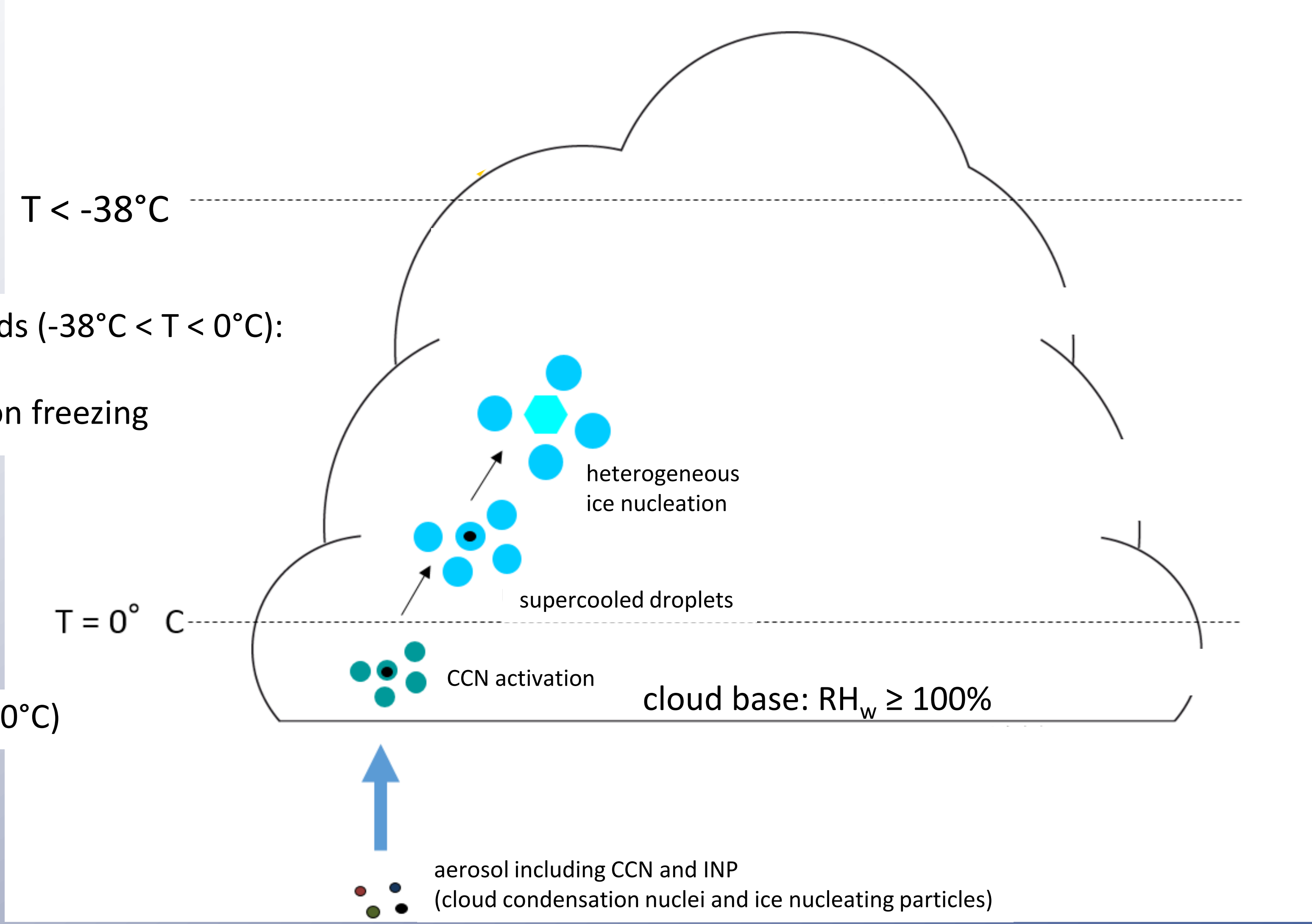
adjusted from sketch
by Dennis Niedermeier

clouds

mixed phase clouds (-38°C < T < 0°C):

- mainly immersion freezing

warm clouds (T > 0°C)



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ice clouds (cirrus) (T < -38°C)
- radiative effects
- homogeneous freezing
- different types of hete-rogeneous freezing

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- mainly immersion freezing
- precipitation formation
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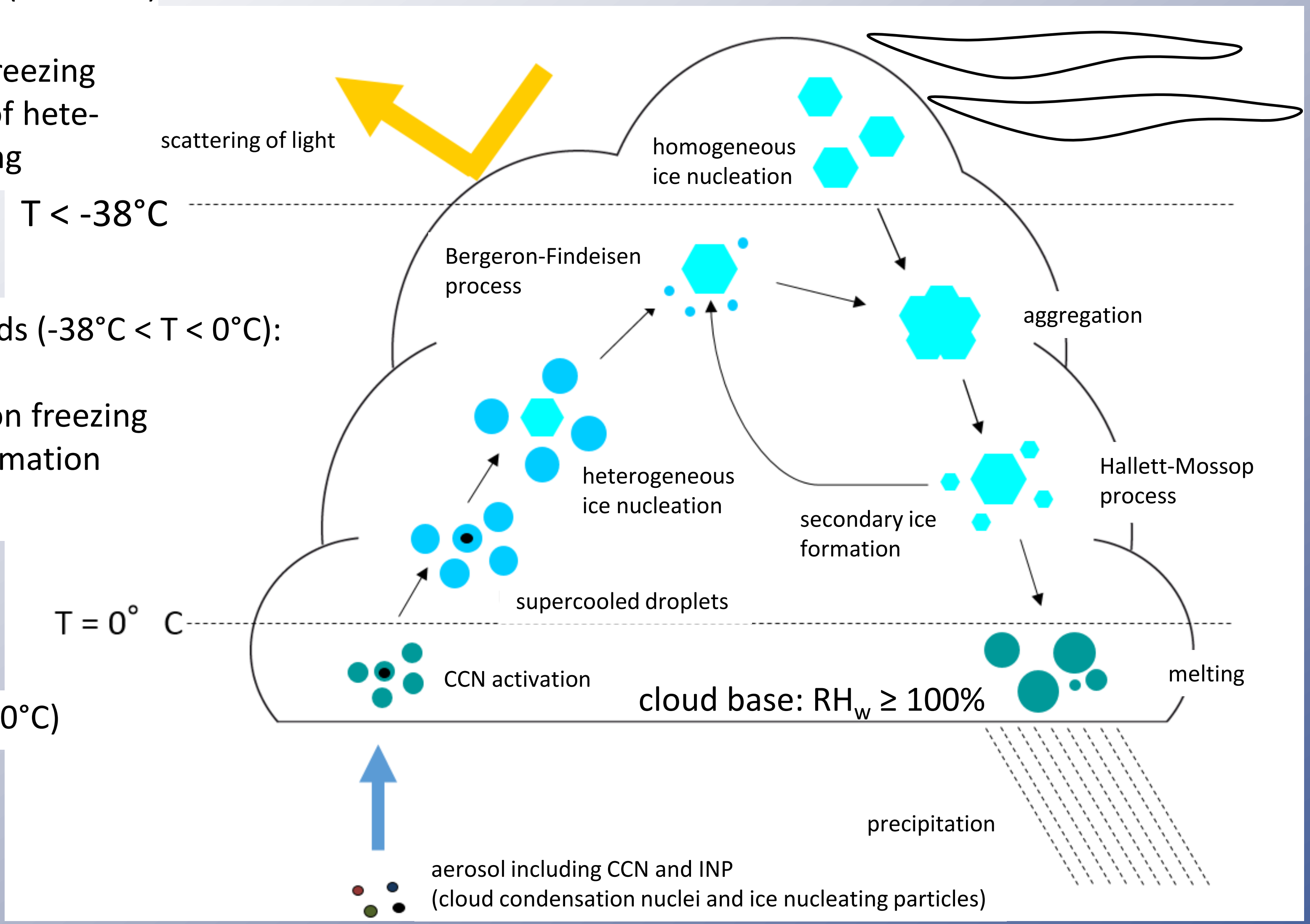

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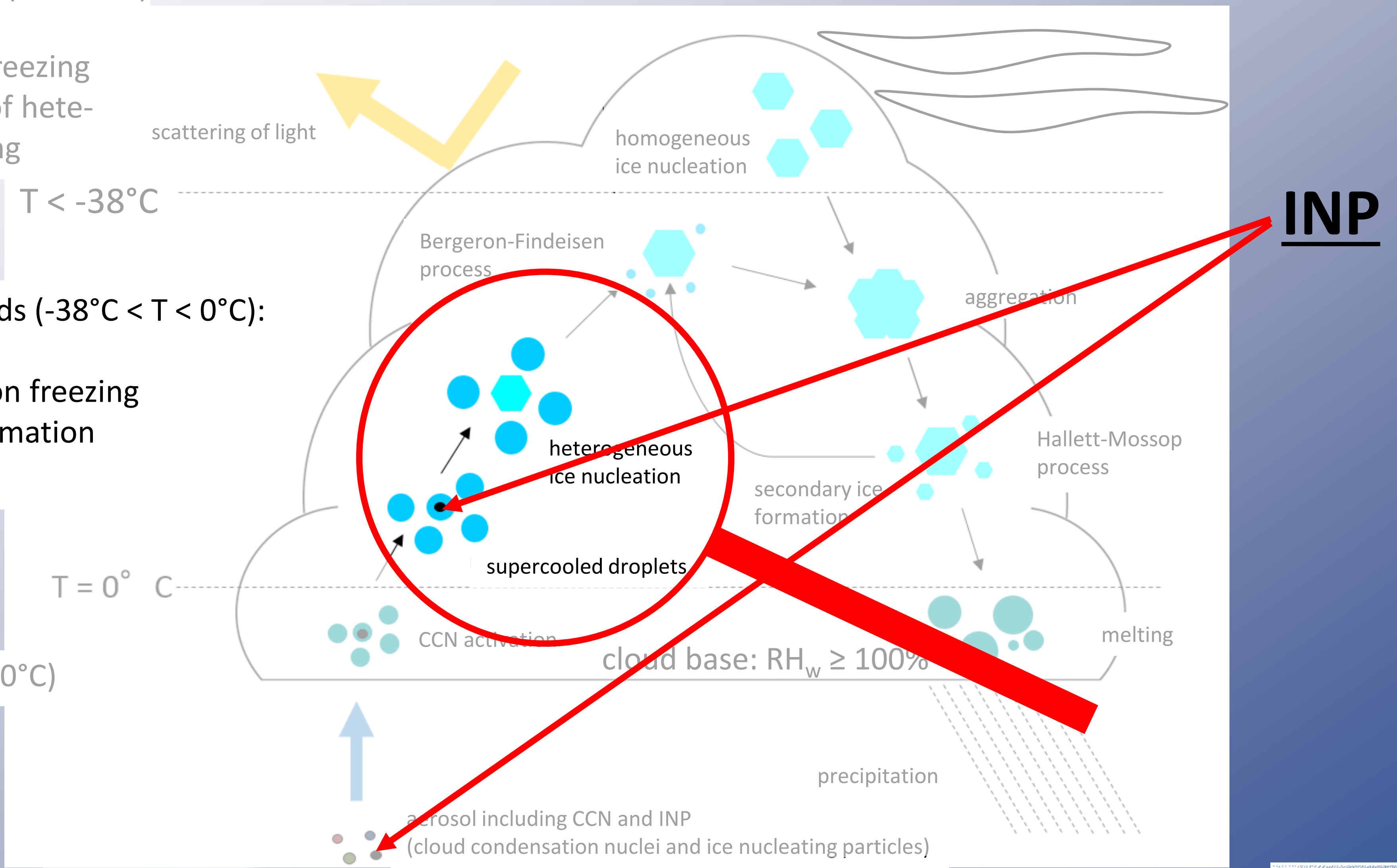

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re-emergence of INP research:

- review by *Szyrmer & Zawadzki (1997)* on „biogenic and anthropogenic sources“ of INP
- increased activity in the field of INP starting ~ 2010
- newer reviews: *Hoose & Möhler (2012); Murray et al. (2012); Coluzza et al. (2017); Kanji et al. (2017)*

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Annual sum of publications relevant to atmospheric ice nucleation

Year	Paper # - Annual Sum
2021	187
2020	222
2019	204
2018	238
2017	178
2016	230
2015	283
2014	216
2013	186
Annual Average	**216**

tables from Naruki Hiranuma, who publishes a monthly summary on new publications related to INP

(nhiranuma@wtamu.edu)

summarizing the general introduction

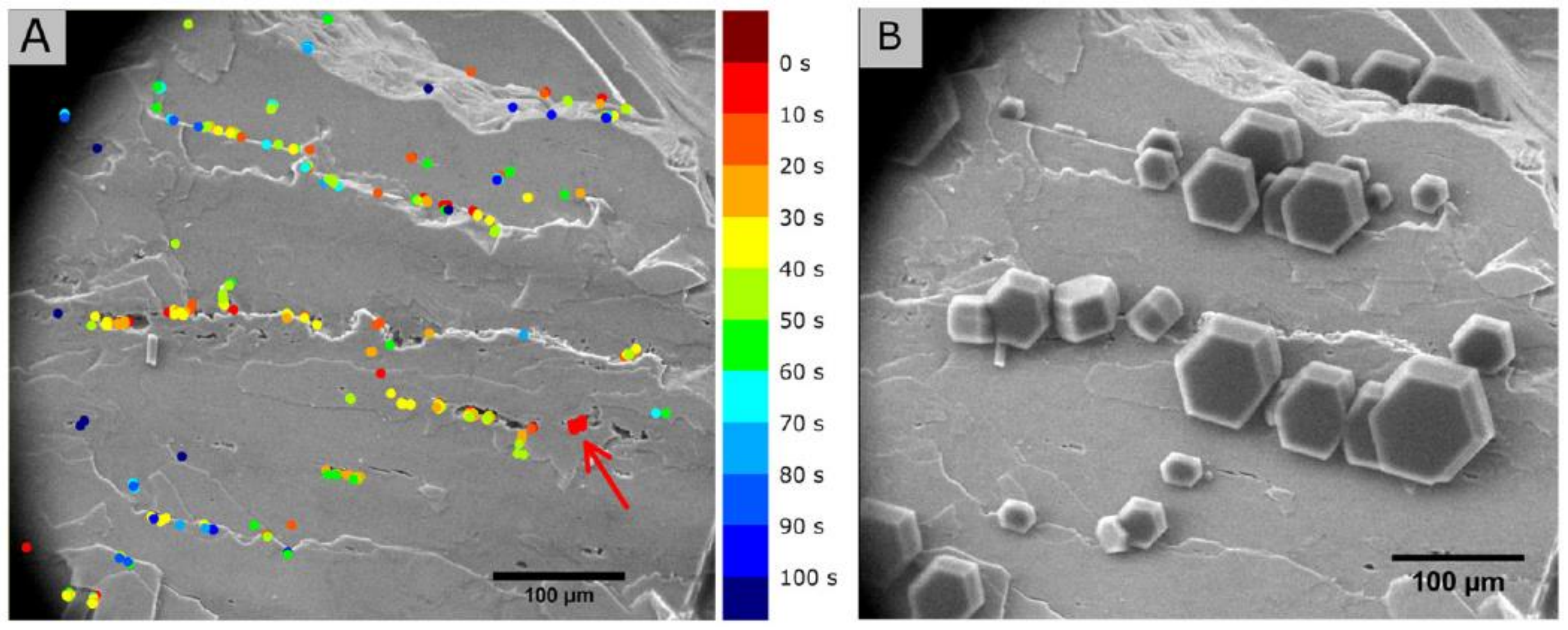
INP are

- needed for ice nucleation in mixed phase clouds
- important for cloud radiative effects and precipitation formation
- very rare among atmospheric aerosol particles
- immersion freezing is the most important process for ice nucleation in mixed phase clouds

ice nucleation by mineral dust particles

- surface sites, nicely demonstrated by

  Kiselev et al. (2016)



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- size dependence

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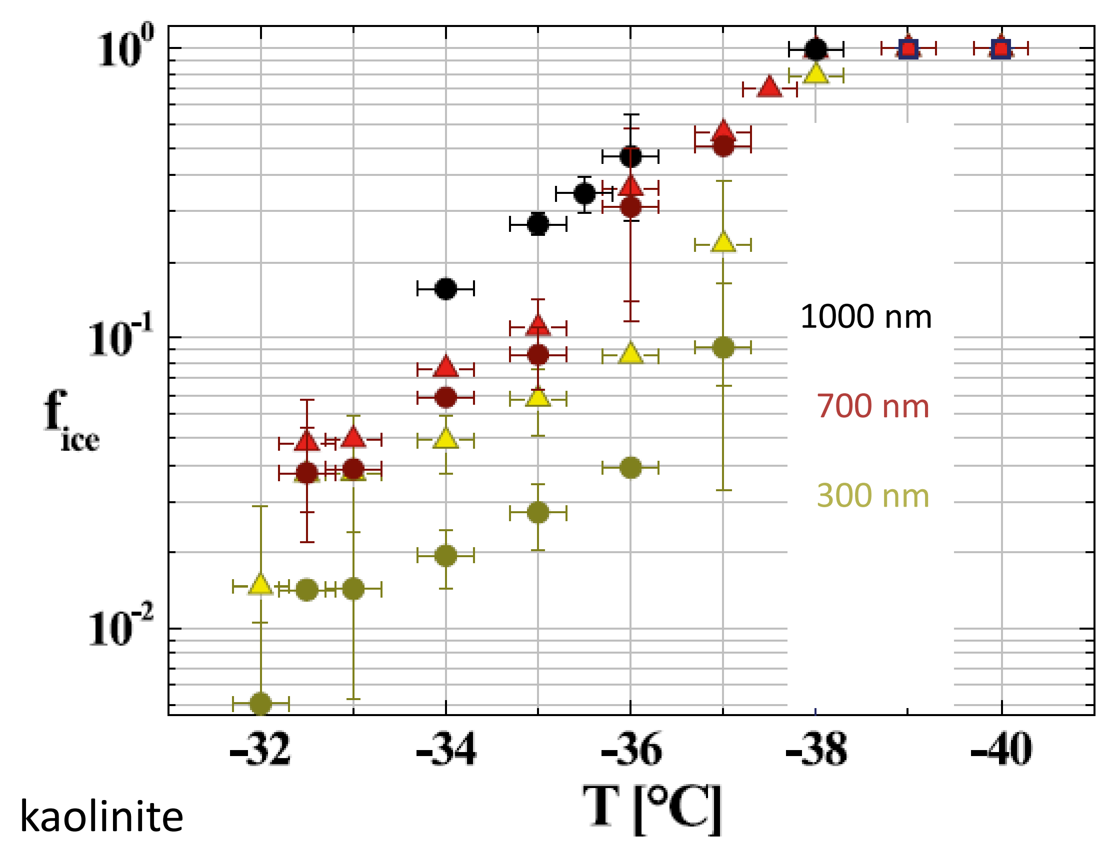

frozen fraction: f_{ice}

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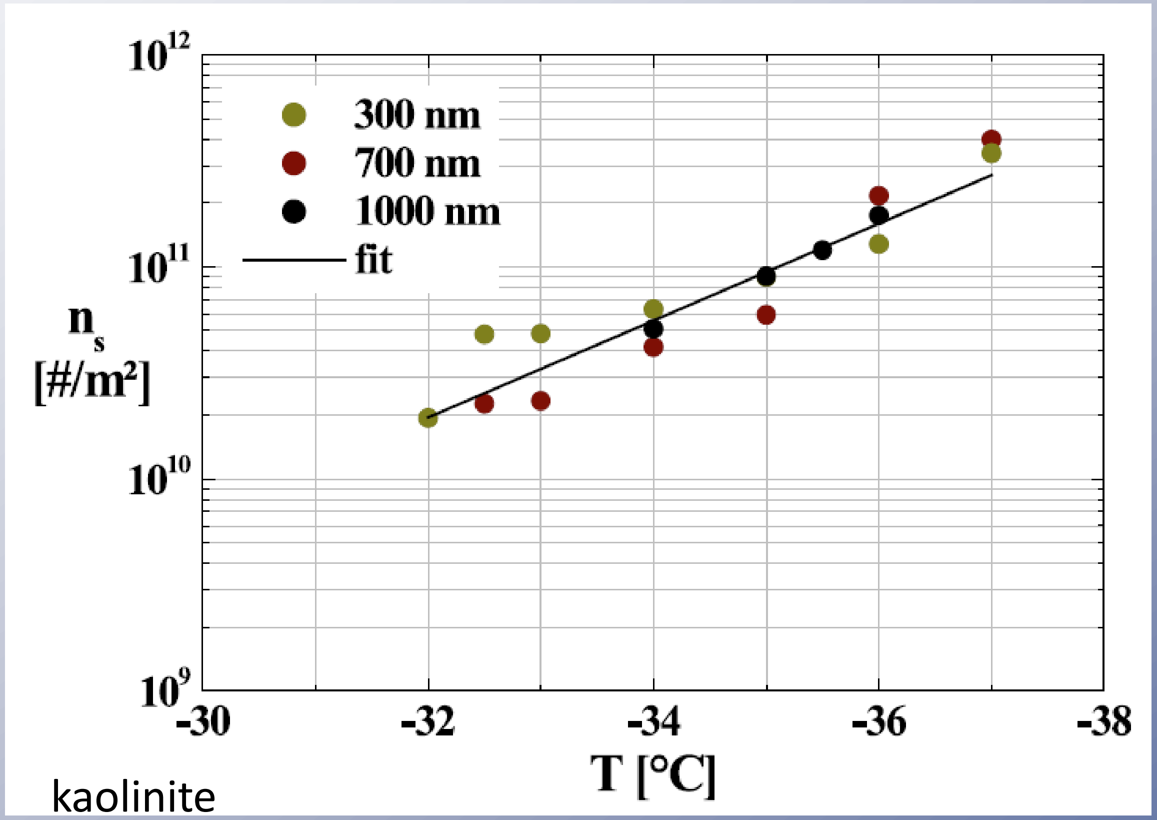

surface site density n_s accounts for that (*Niemand et al., 2012*)

$n_s = -\ln(1 - f_{ice}) / S$

frozen fraction: f_{ice}
particle surface area per droplet S

ice nucleation by mineral dust particles

- surface sites, nicely demonstrated by

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- size dependence

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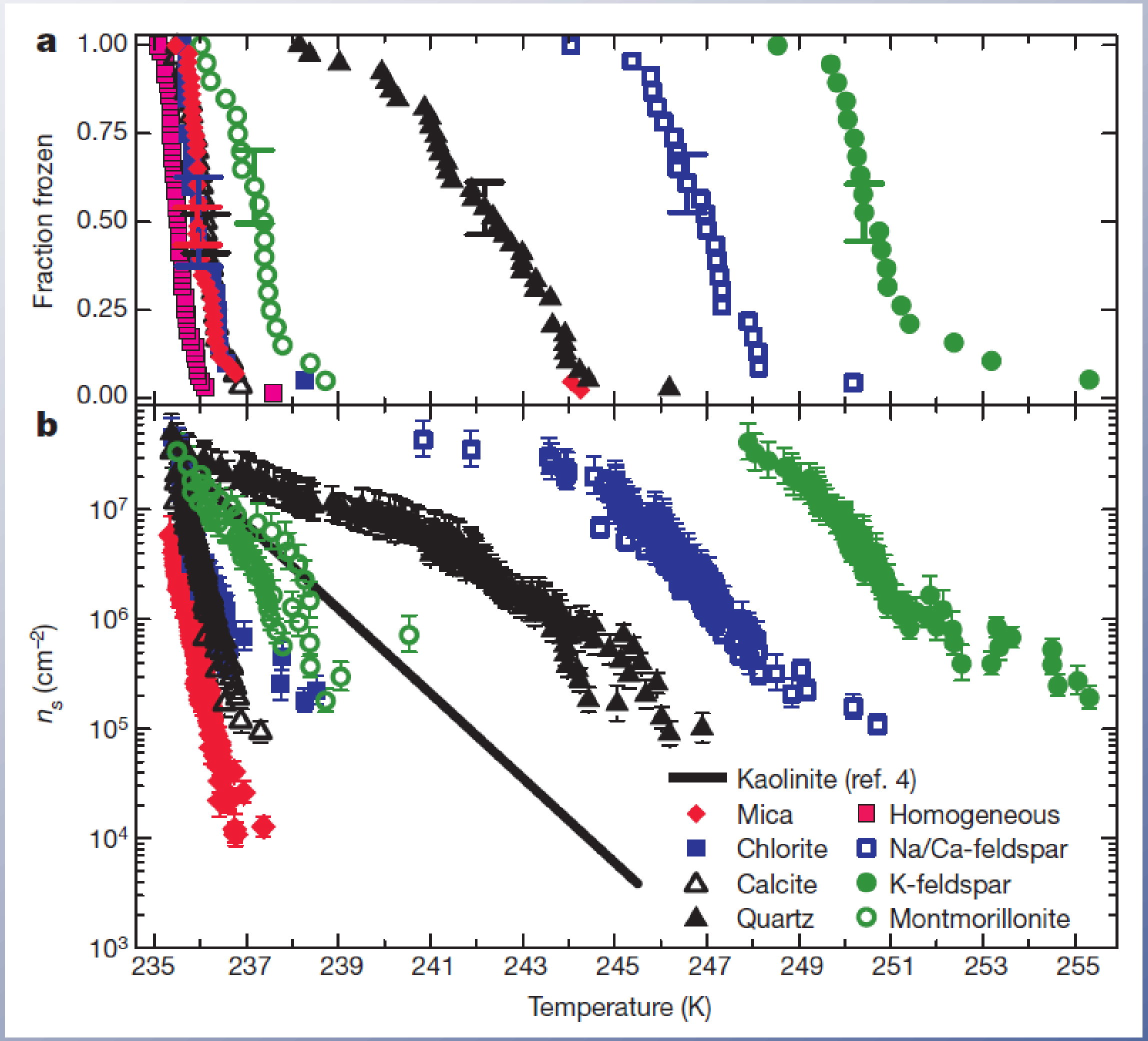
- K-feldspar is the most ice active mineral

  Atkinson et al. (2013)
  Augustin-Bauditz et al. (2014)
  Peckhaus et al. (2016)

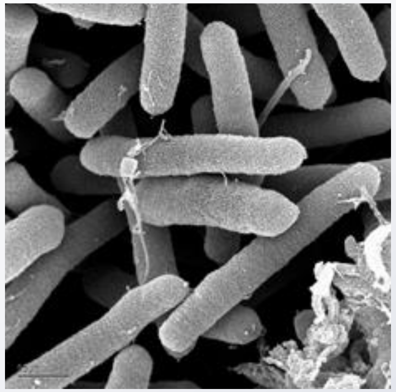


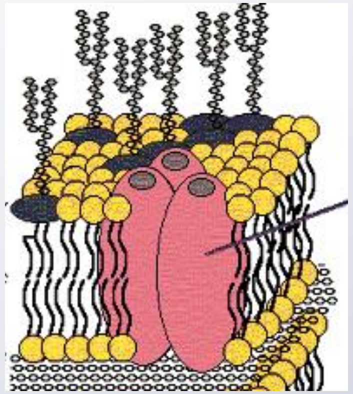
ice nucleation by biological particles

bacteria

pollen

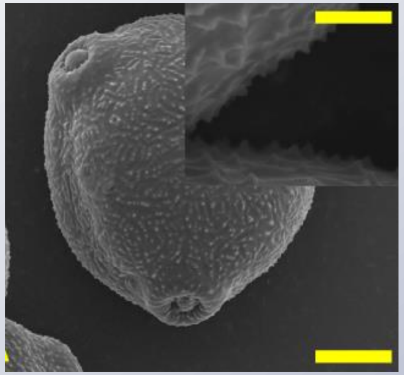
fungal spores



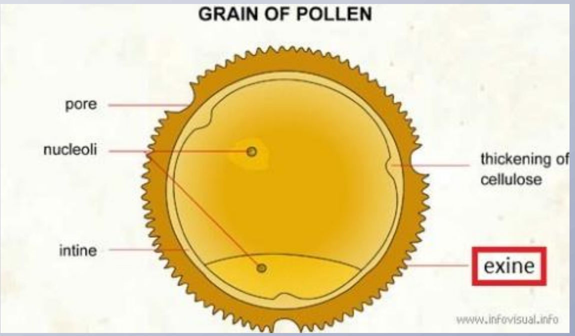


Pseudomonas syringae
foto: G. Vrdoljak, U.C. Berkeley

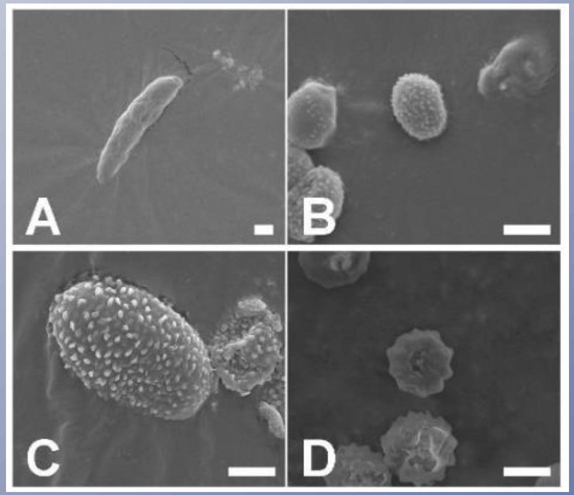
sketch: hawashpharma. blogspot.de



Birch pollen grain, Pummer et al. (2012)



www.infovisual.info



Fungal spores from different species, Pummer et al. (2013)

cell: 10 μm length, 2 μm width

grain: 20 to 30 μm in diameter

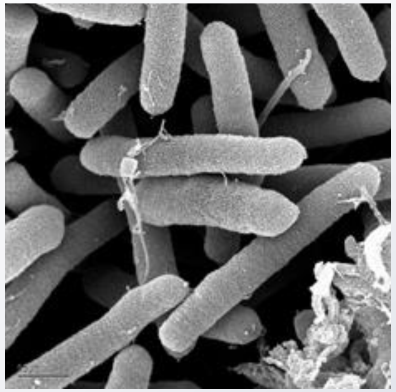
spore: dimensions from 2 to10 μm

ice nuc.: protein complexes
in the cell membrane

ice nuc.: polysaccharides but also proteins,
can easily be washed off

ice nucleation by biological particles

bacteria



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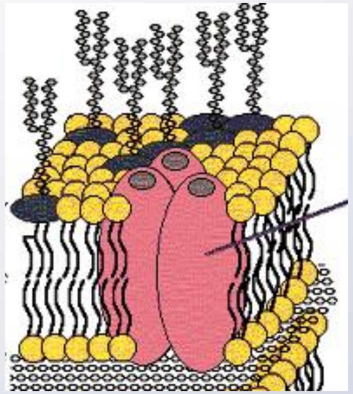

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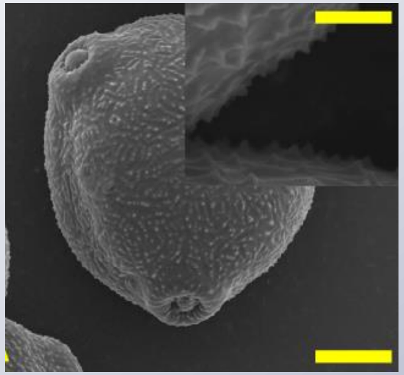

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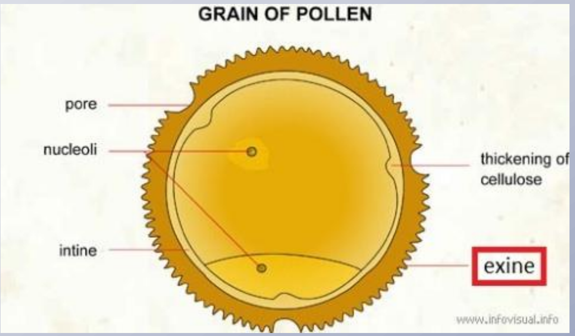

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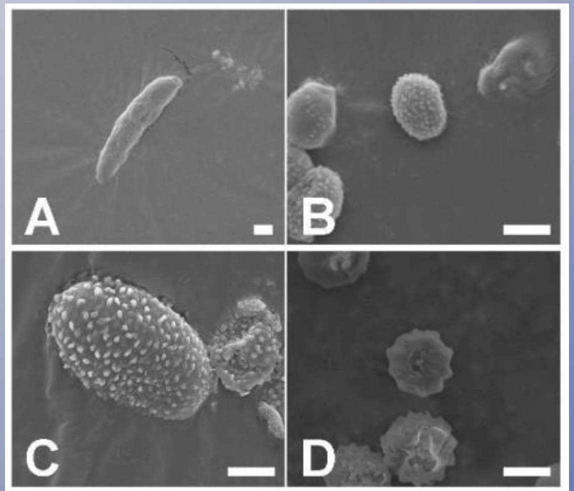


Fungal spores from different species, Pummer et al. (2013)

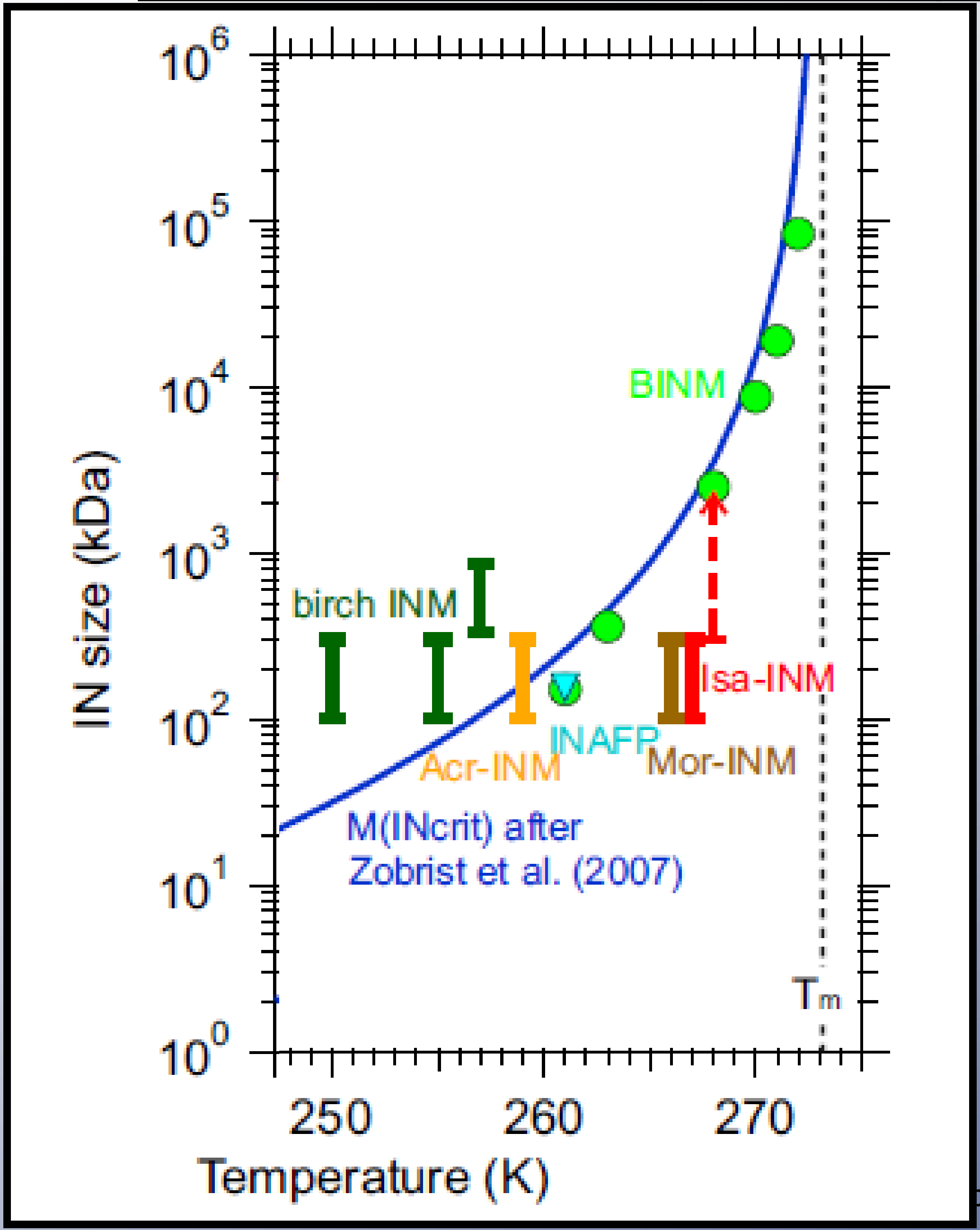
spore: dimensions from 2 to10 μm

ice nuc.: polysaccharides but also proteins, can easily be washed off

-> ice nucleation rates for single macromolecules from bacteria, pollen and fungal spores

Hartmann et al. (2013) *Augustin et al. (2013)* *Pummer et al. (2015)*

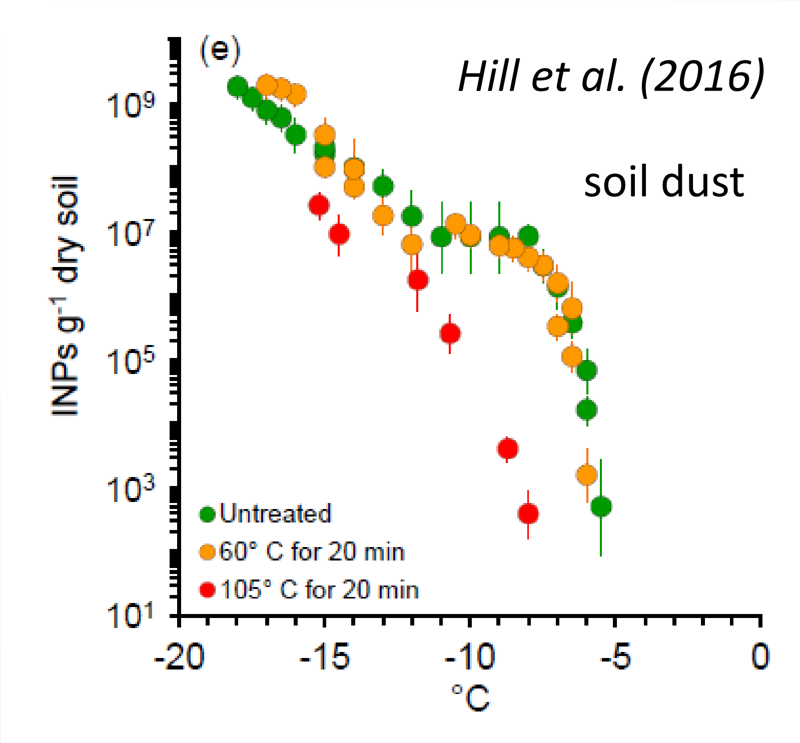
ice nucleation by biological particles



single macromolecules

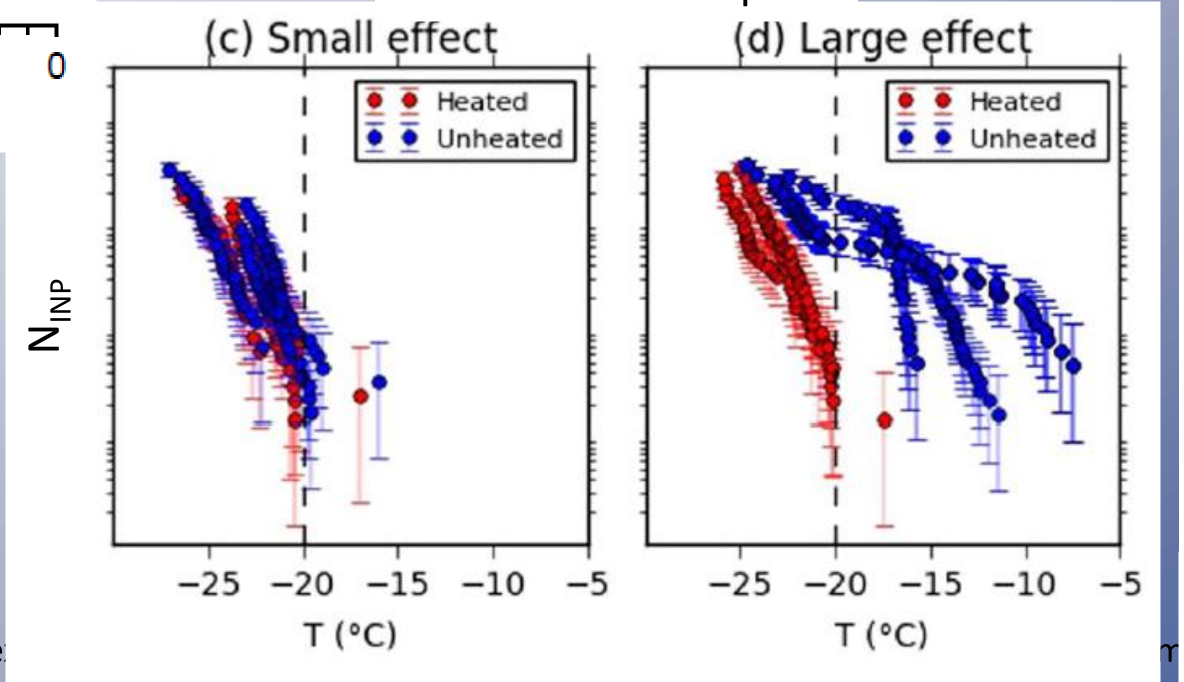
Pummer et al. (2015)

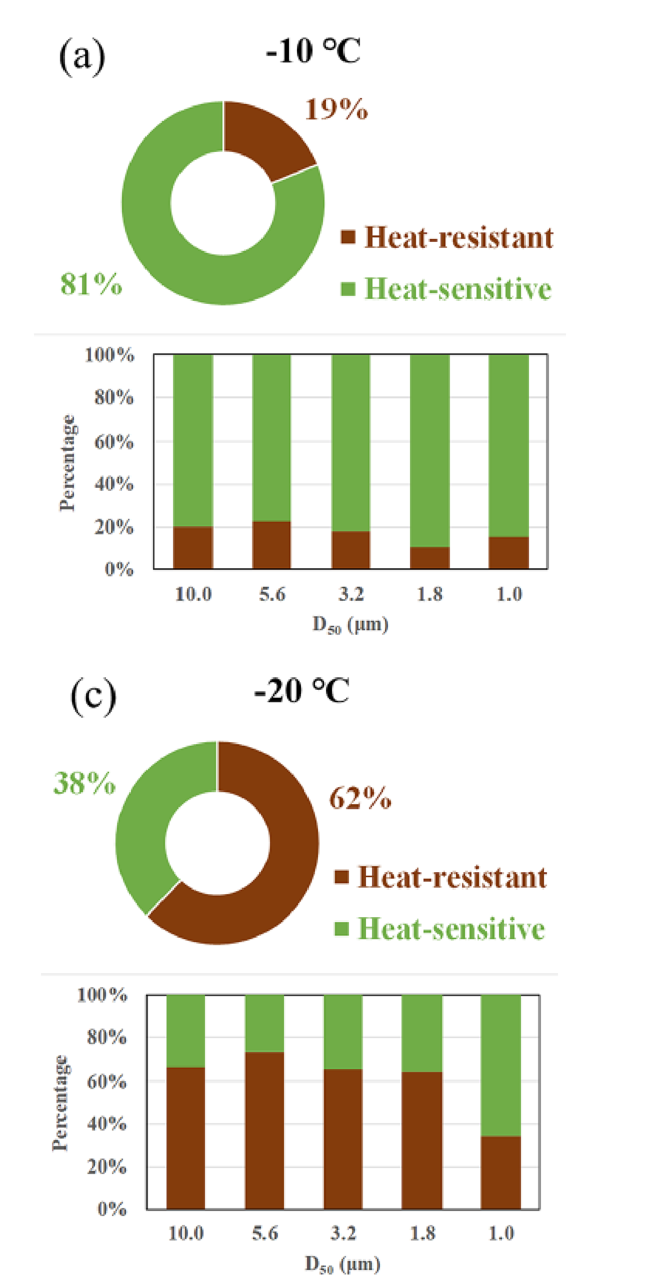
testing for biogenic INP (proteins)



heating samples
to test
for proteinaceous INP

O'Sullivan et al. (2018)
UK airborne samples



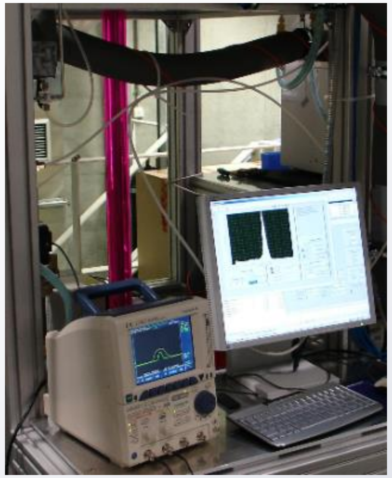


Chen et al. (2021)
Beijing dust storms

measuring INP in-situ

laminar flow tube

LACIS



Hartmann et al. (2011)

expansion chambers

PINE,
AIDA, …

Möhler et al. (2021)

continuous flow diffusion chambers

CFDC, PINC, SPIN, HINC, INCA, …

Rogers et al. (1988)
Stetzer et al. (2008)
Garimella et al. (2016)

typically **singe particle optical detection**

different operating principles, some can be bought

measuring INP in-situ

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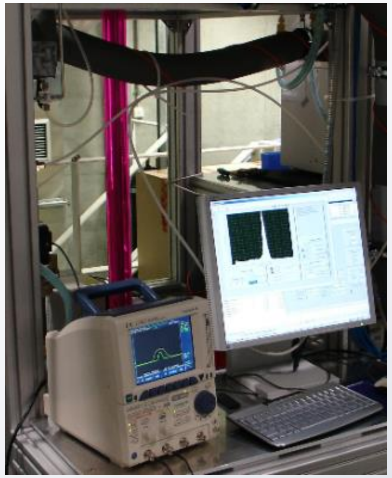

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different operating principles, some can be bought

advantage:
- high time resolution
- closer to what happens in atmosphere

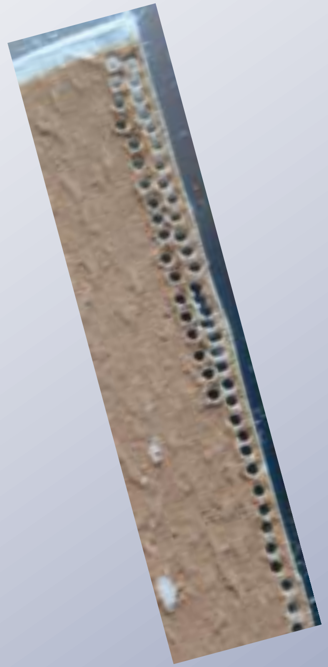
disadvantage:
- comparably **high detection limit** (low INP concentrations cannot be detected)
- typically **more difficult to operate**
- large particles may need to be **rejected** at inlet

measuring INP off-line

* Leipzig Ice Nucleation Array
** Ice Nucleation Droplet Array

cold-stage and freezing array for suspensions (e.g., washed filters, suspensions of samples, rain- or ocean water, … ; INDA also for filter punches)

a large number of these instruments has started to be operated in different groups



working cleanly:

Polen et al. (2018)

Barry et al. (2021)

LINA *

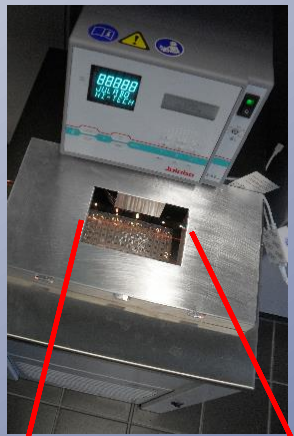
V= 1 μL in one droplet, 90 droplets

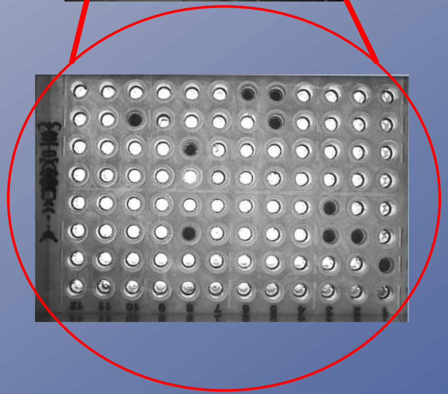
droplets on a glass slide, cooled by a Peltier element

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

INDA **

V=50 μL in one droplet, 96 droplets





PCR-trays in a thermostat

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

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a large number of these instruments has started to be operated in different groups

advantage:
- detection of **lower INP concentrations**
- **additional analysis** possible

disadvantage:
- **contamination** -> difficult to reach down to low temperatures (exception: pico-liter droplets, but droplet production and optical detection much more expensive)
- long sampling times -> **bad time resolution**

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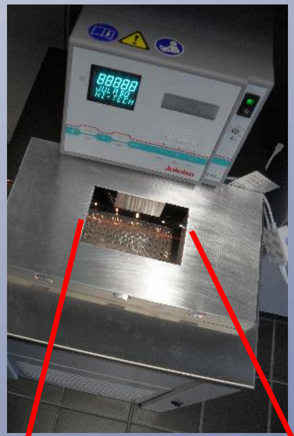

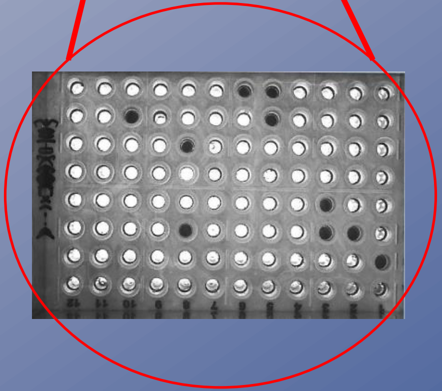

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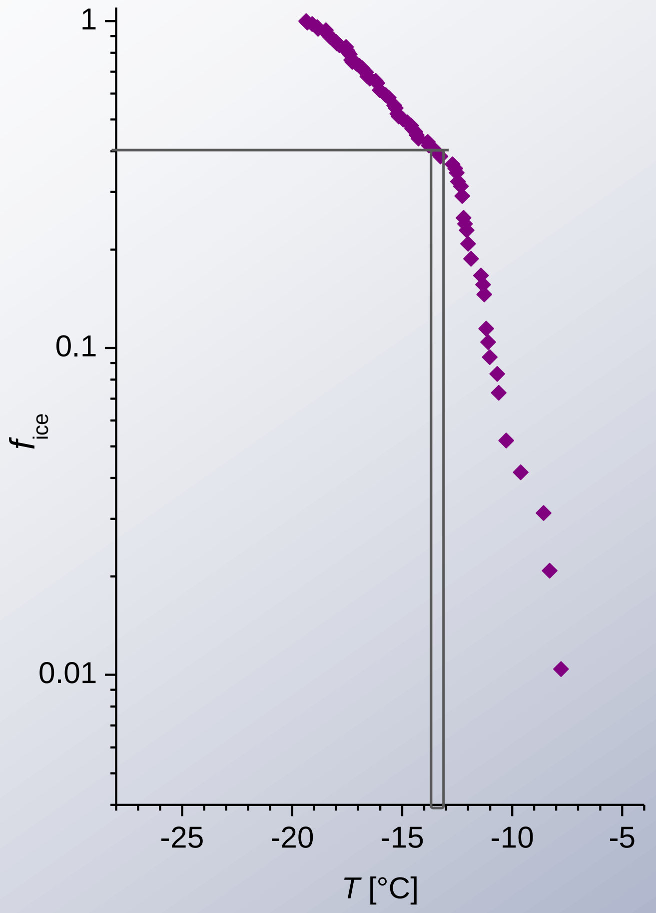
evaluating off-line INP data

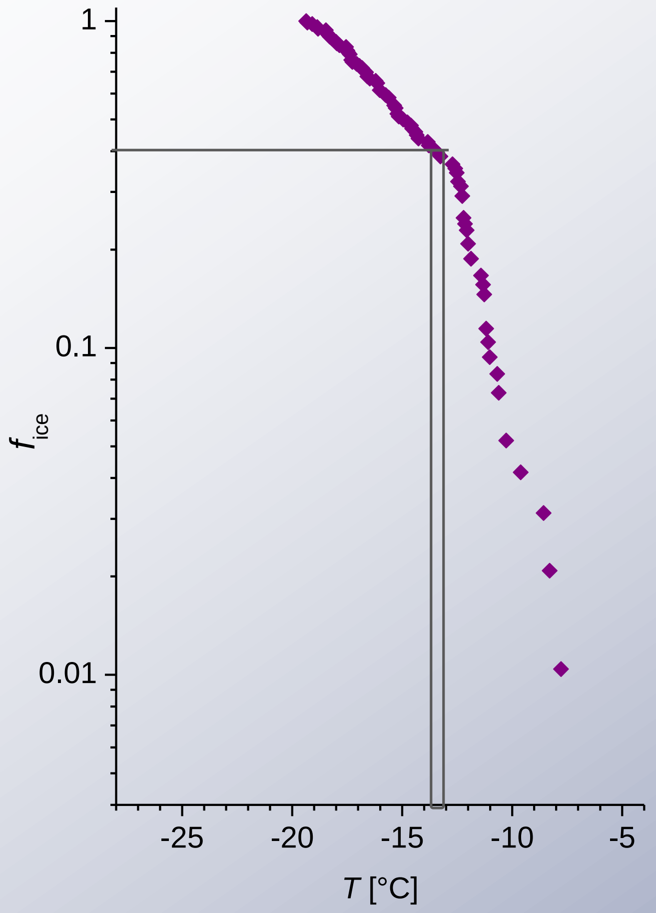
cumulative distribution



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

evaluating off-line INP data

cumulative distribution



Poisson distribution:

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

(e.g., f_{ice} = 0.4 -> λ = 0.51)

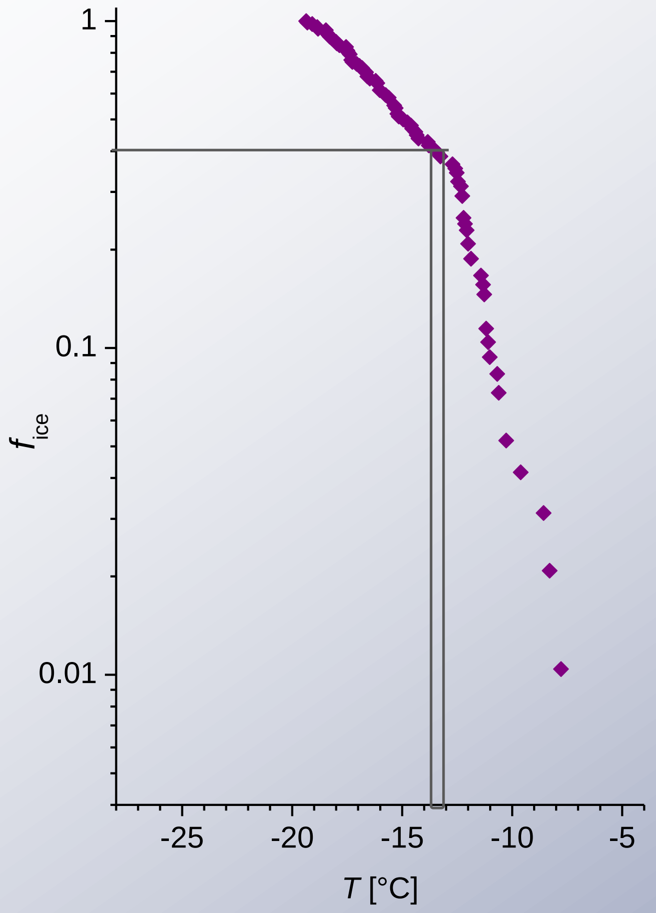
λ -> for us: average number of ice active entities per droplet

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

Vali (1971)

evaluating off-line INP data

cumulative distribution



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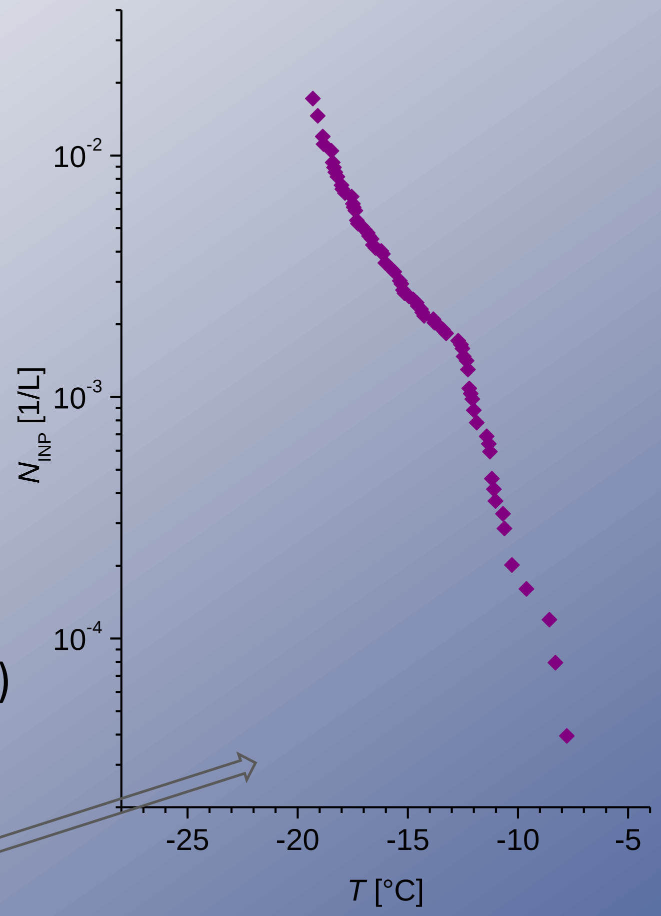
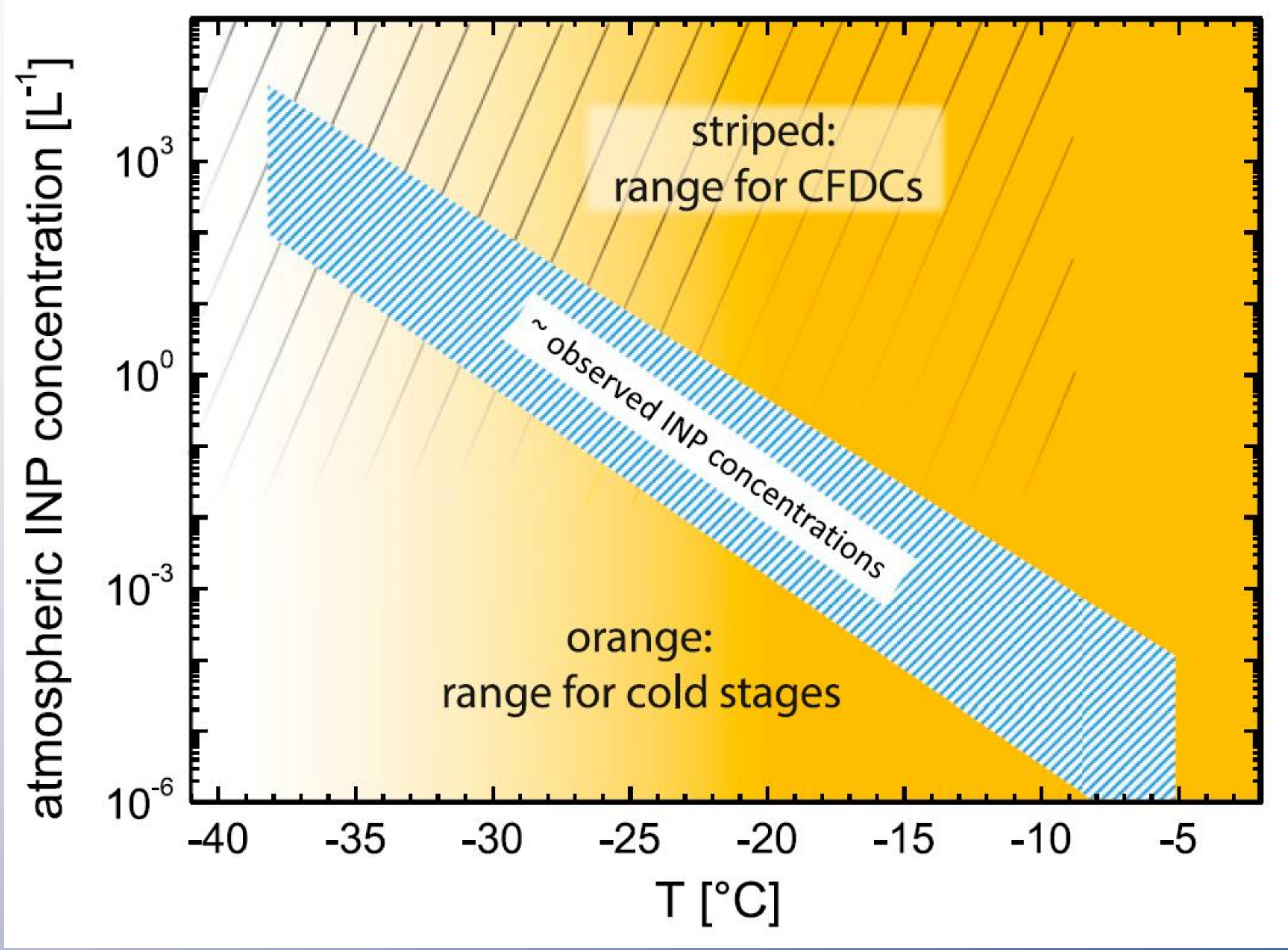


Normalization:

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

Vali (1971)

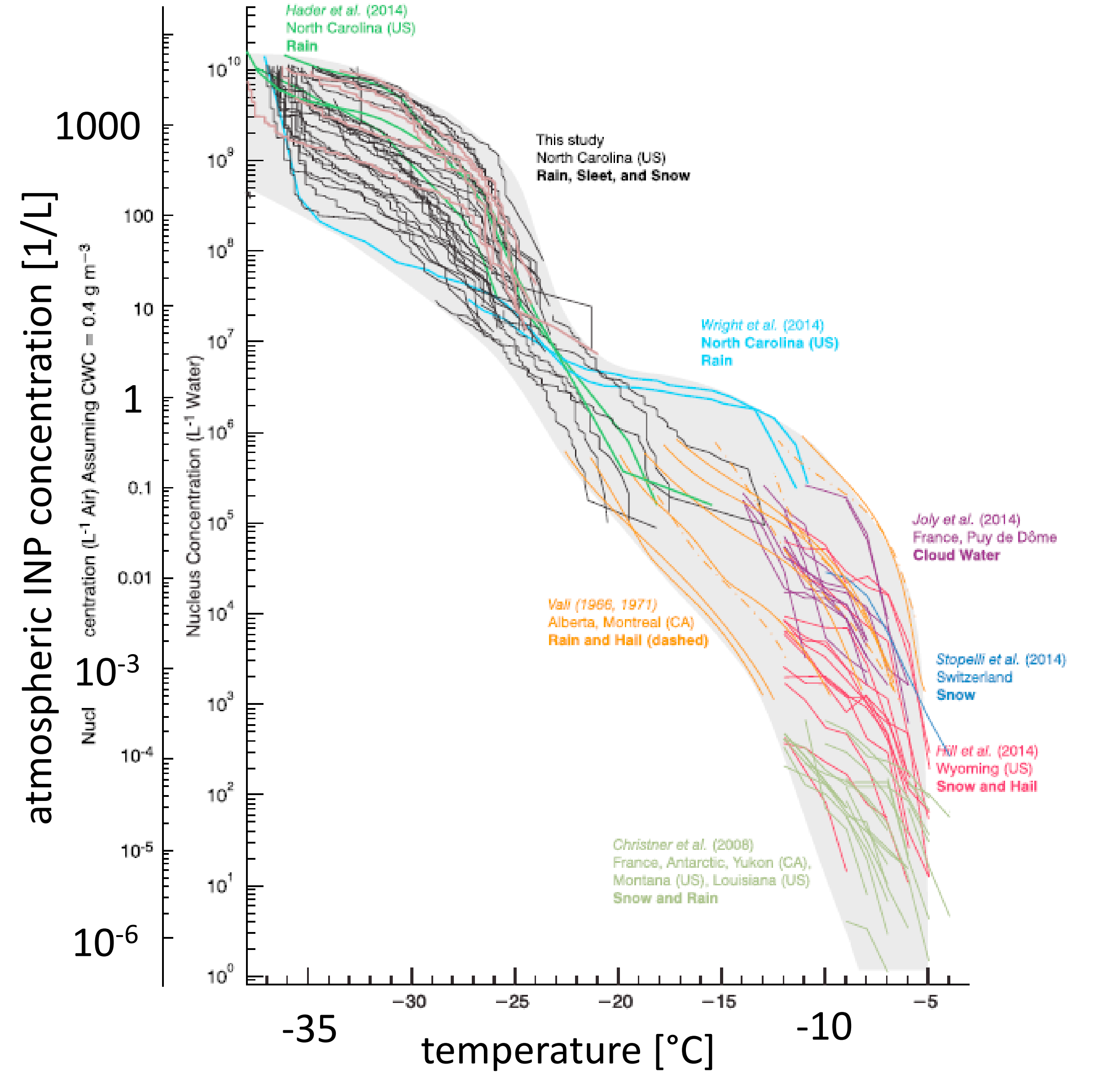
where to measure with which instrument



Cziczo et al. (2017)

concentrations of ice nucleating particles (N_{INP})

Petters & Wright, (2015)



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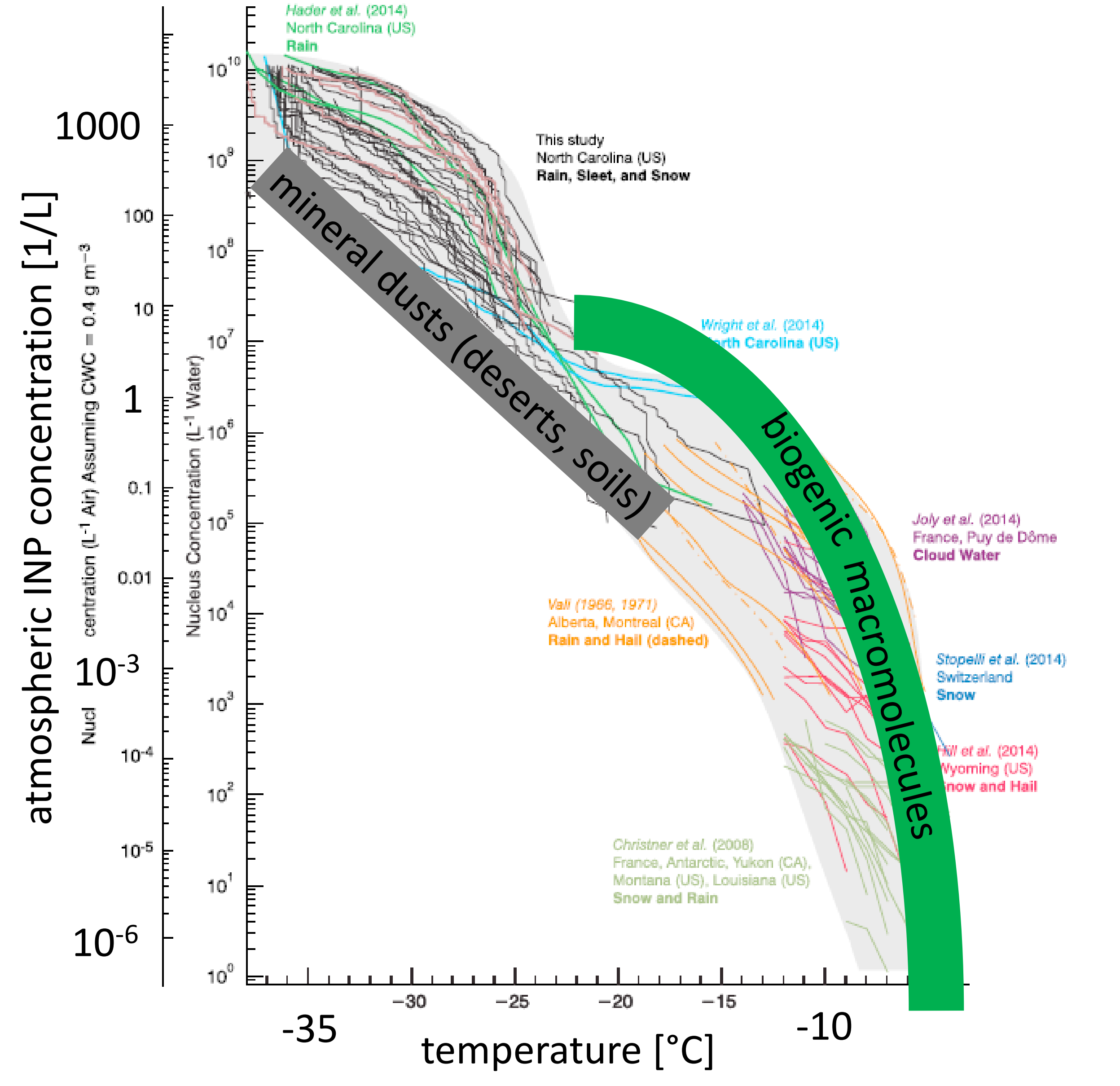

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summarizing basic knowledge

- in the atmosphere, mainly two main INP types contribute:
  mineral dust particles and microorganisms

- microorganisms have macromolecules causing the ice activity (proteins or polysaccarides)
- they are very ice active, but VERY rare
- heat can destroy bio-INP (proteins)

- mineral dust particles have ice active sites
- they are ice active at lower temperatures but are more abundant, however, still rare
- K-feldspar is the most ice active mineral dust

- a multitude of instruments exist to measure INP in-situ or off-line; all with strength and weaknesses

(topic of parameterization will be left out due to time limits -> additional backup slides)

upcoming results from recent field studies

Where do we have which INP concentrations?

Where do these INP come from?

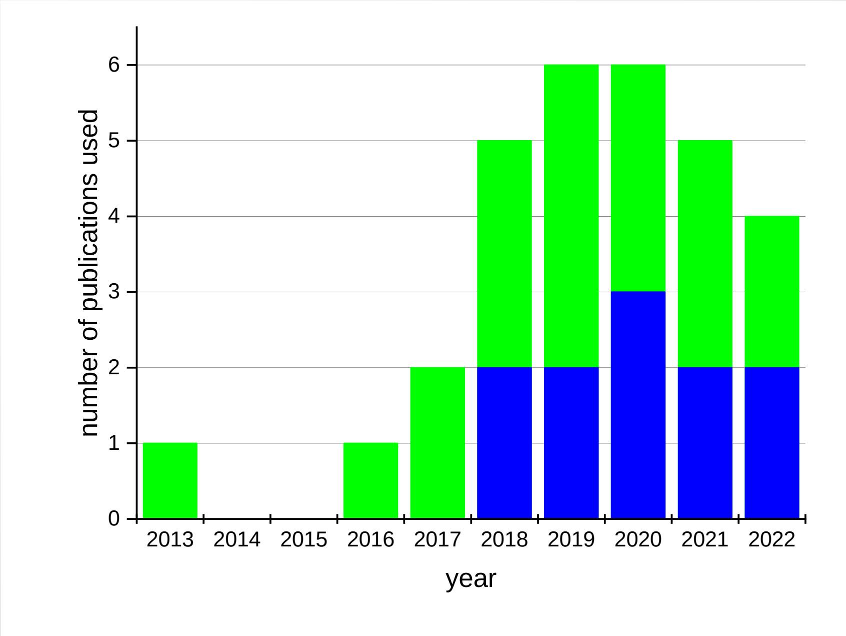
-> sources and occurrences

31 publications from 2013 until 2022

only few main messages

mostly based on off-line methods &

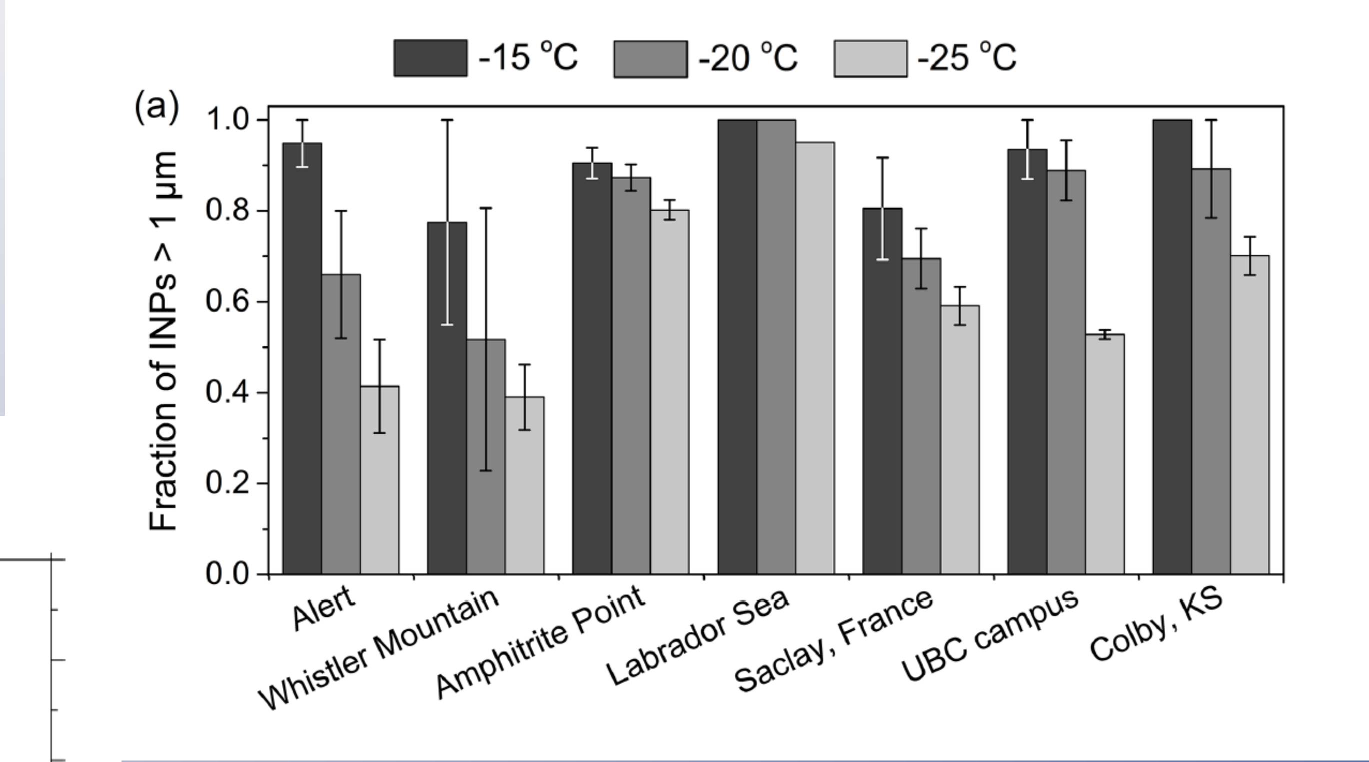
mostly ground based measurements

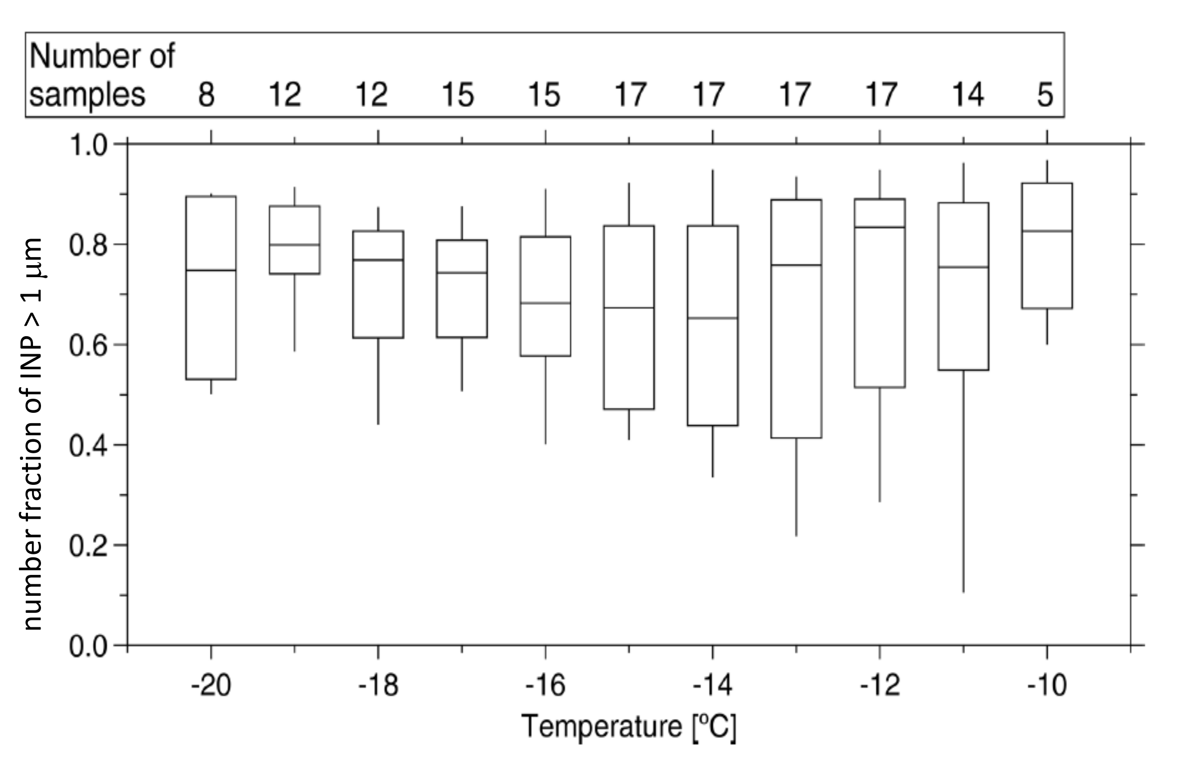


TROPOS cloud group

highly ice active atmospheric INP often supermicron

- *Mason et al. (2016)*
- *Creamean et al. (2018),* Arctic
- *Gong et al. (2020),* Cabo Verde

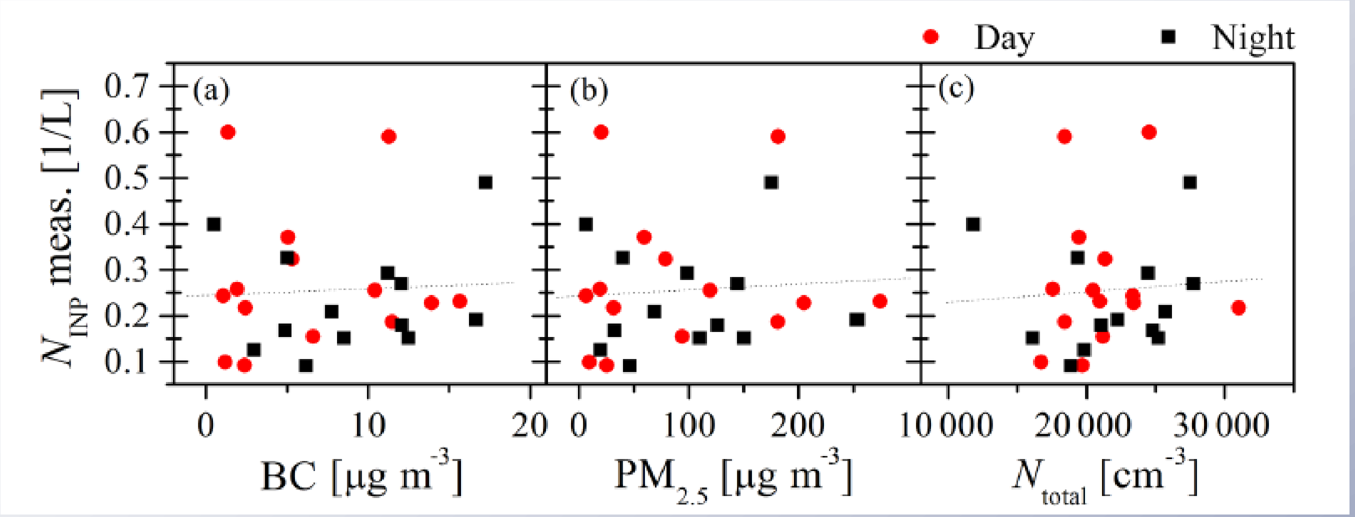


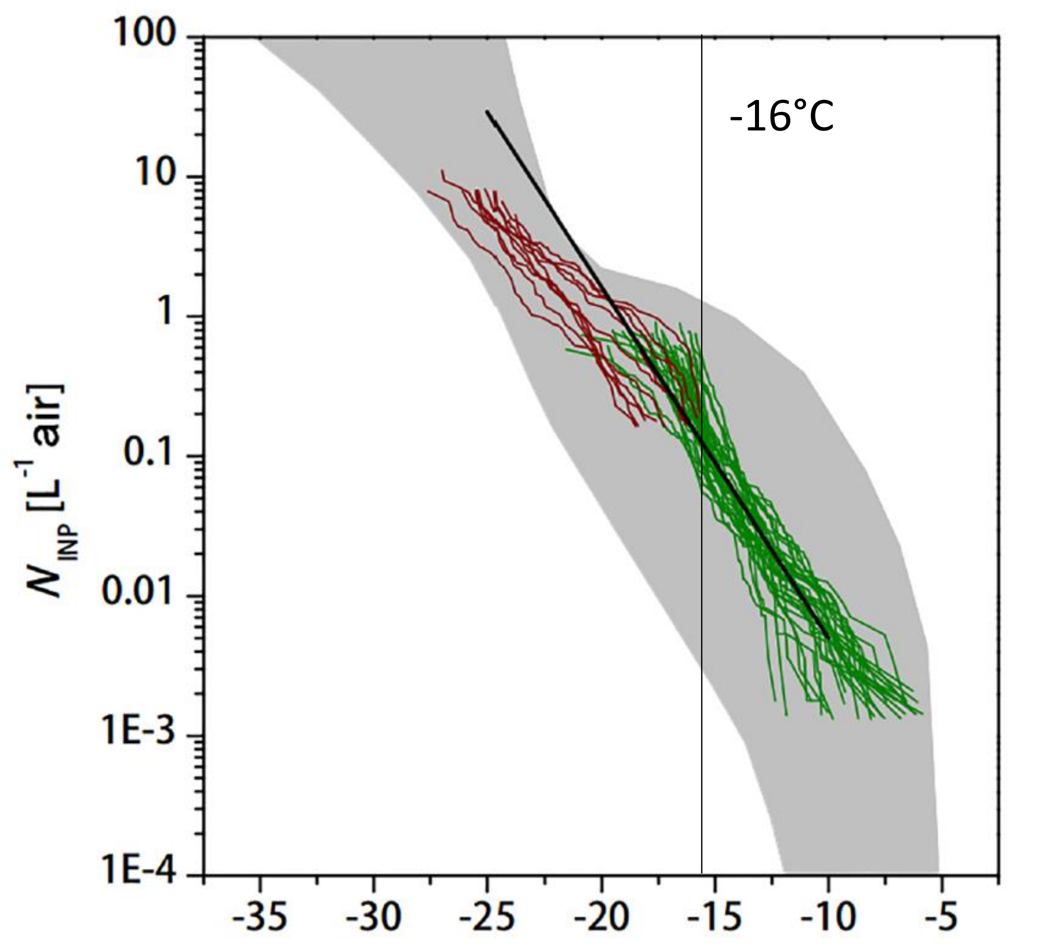


TROPOS

INP and anthropogenic pollution

- *Chen et al. (2018):*
  Beijing air pollution did not contribute INP (down to -25°C)





gray area:
Petters & Wright (2015)

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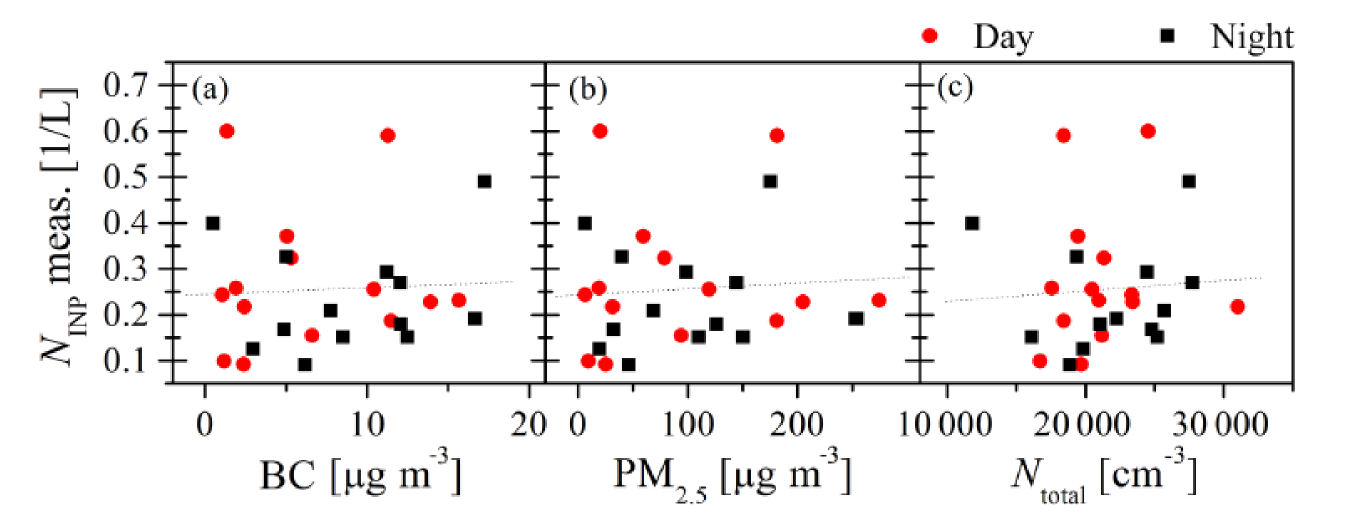
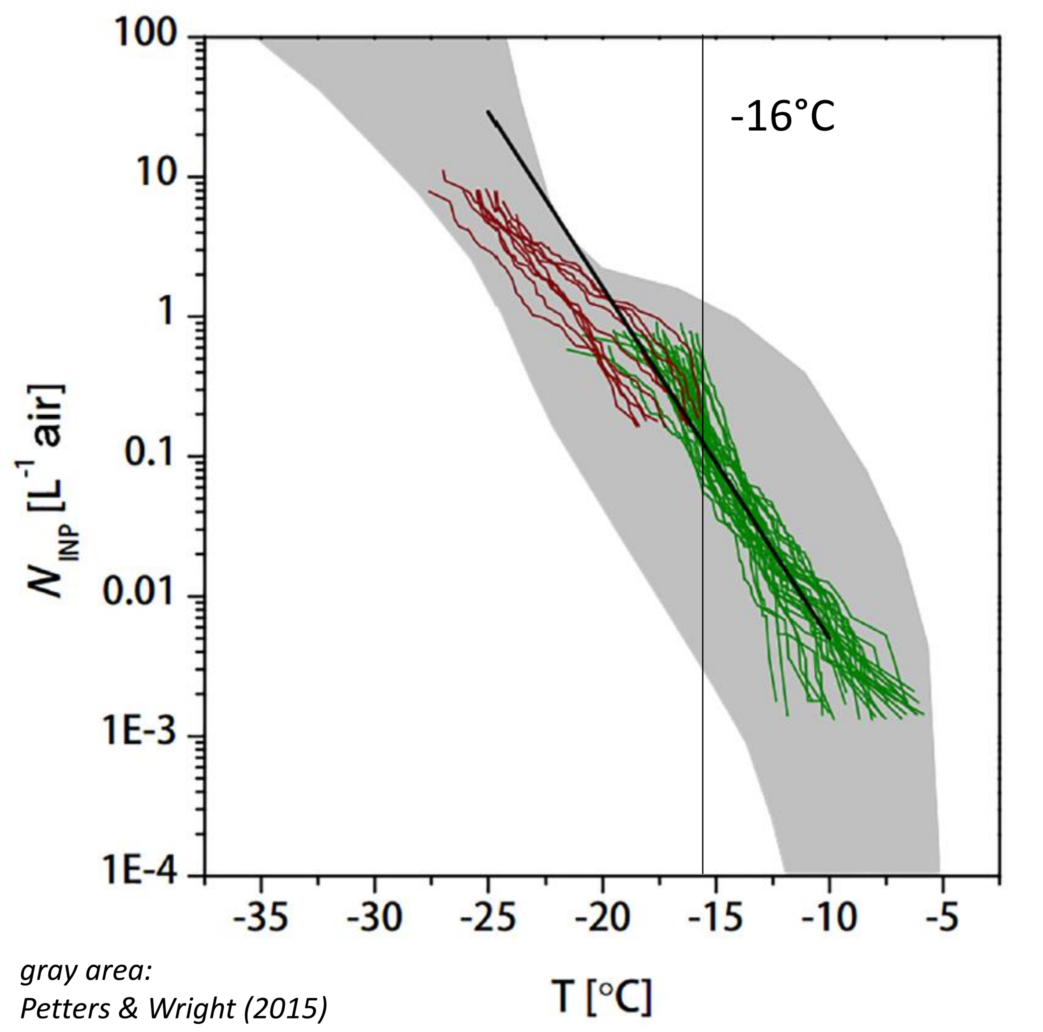


however, there is a clear increase in INP in Beijing during dust events
Chen et al. (2021)



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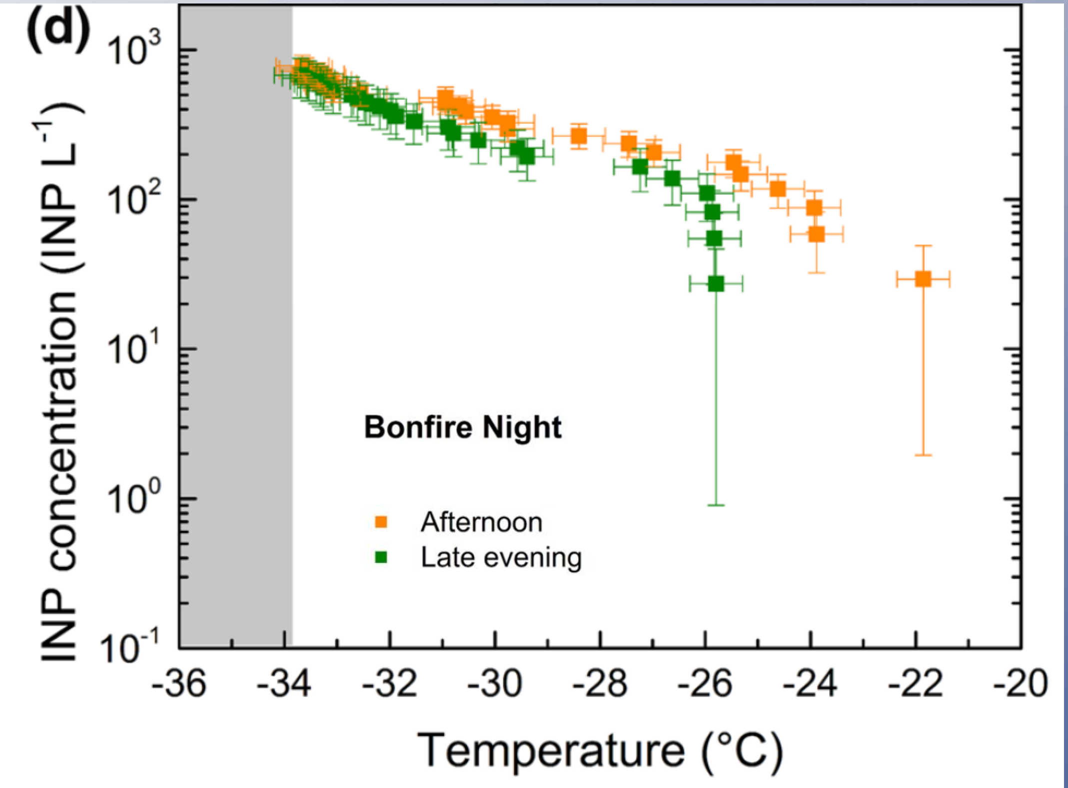

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  potential contribution of **ship exhaust** to INP only below -36 °C

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- *Tobo et al. (2020):*
  year round data-set **in Tokyo**, INP variations from long range transported dust and biological INP

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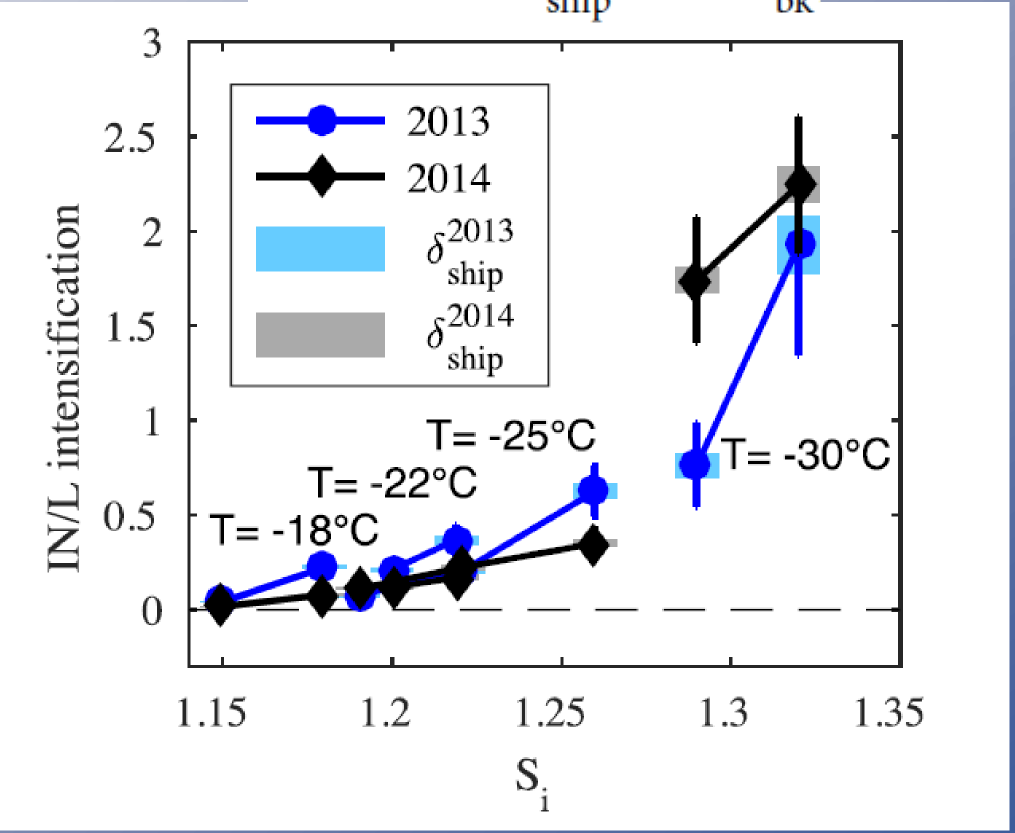
- *Thomson et al. (2017):*
  ship emmission plumes in habor of Gothenburg, Sweden showed increased INP concentrations

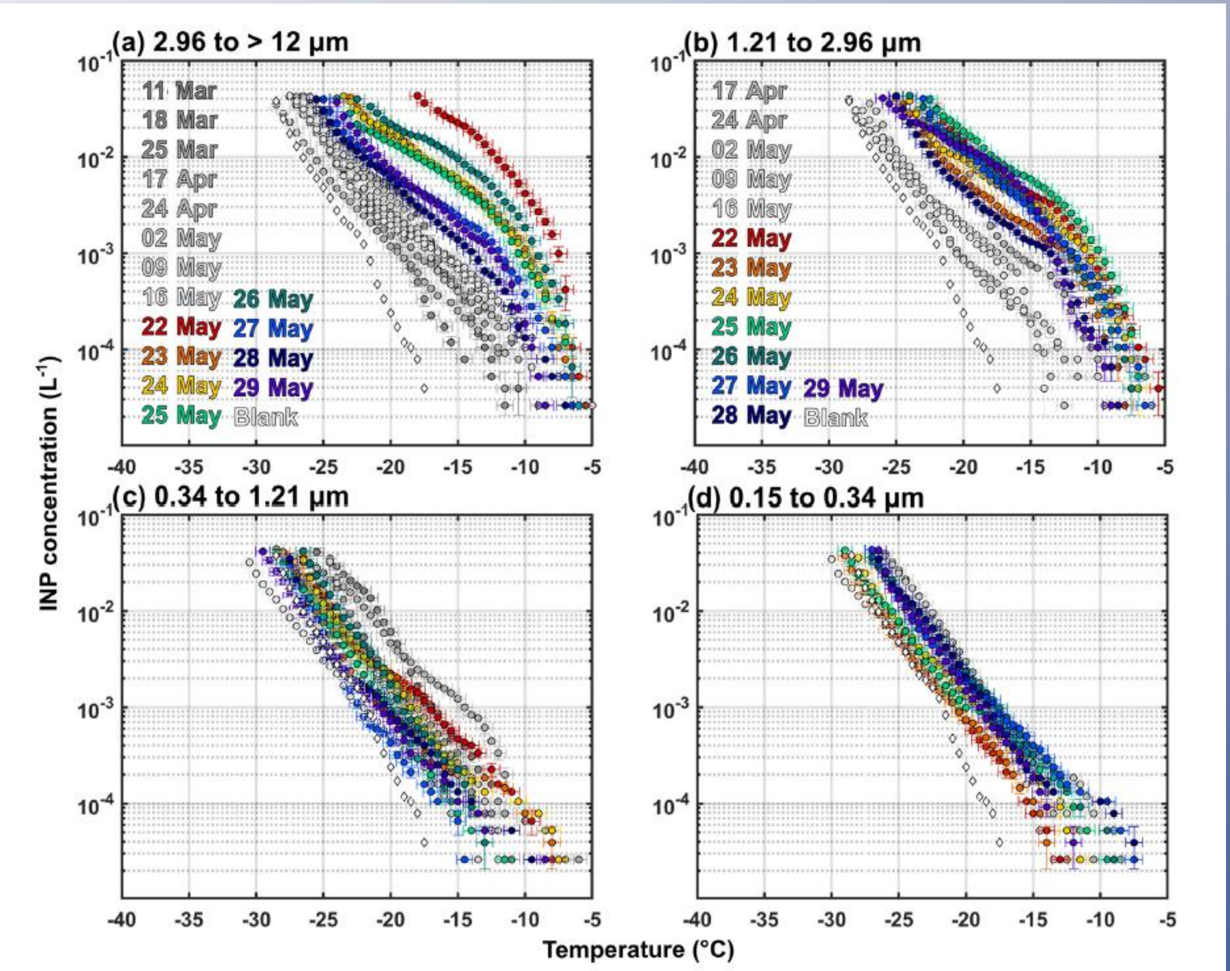
$$\Delta INP = \overline{INP}_{ship} - \overline{INP}_{bk}$$



TROPOS

INP in the Arctic

- *Creamean et al. (2018):*
  supermicron particles contribute strongly **increased INP concentrations in May**



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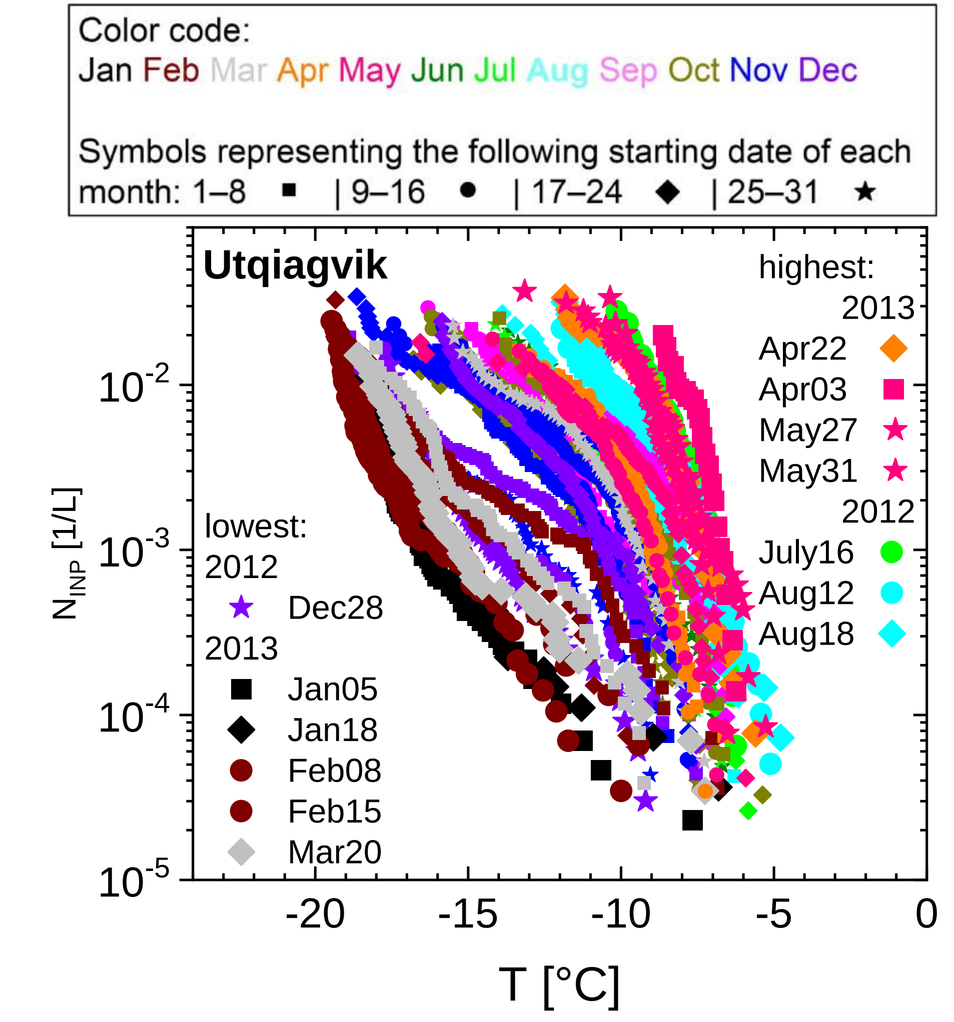

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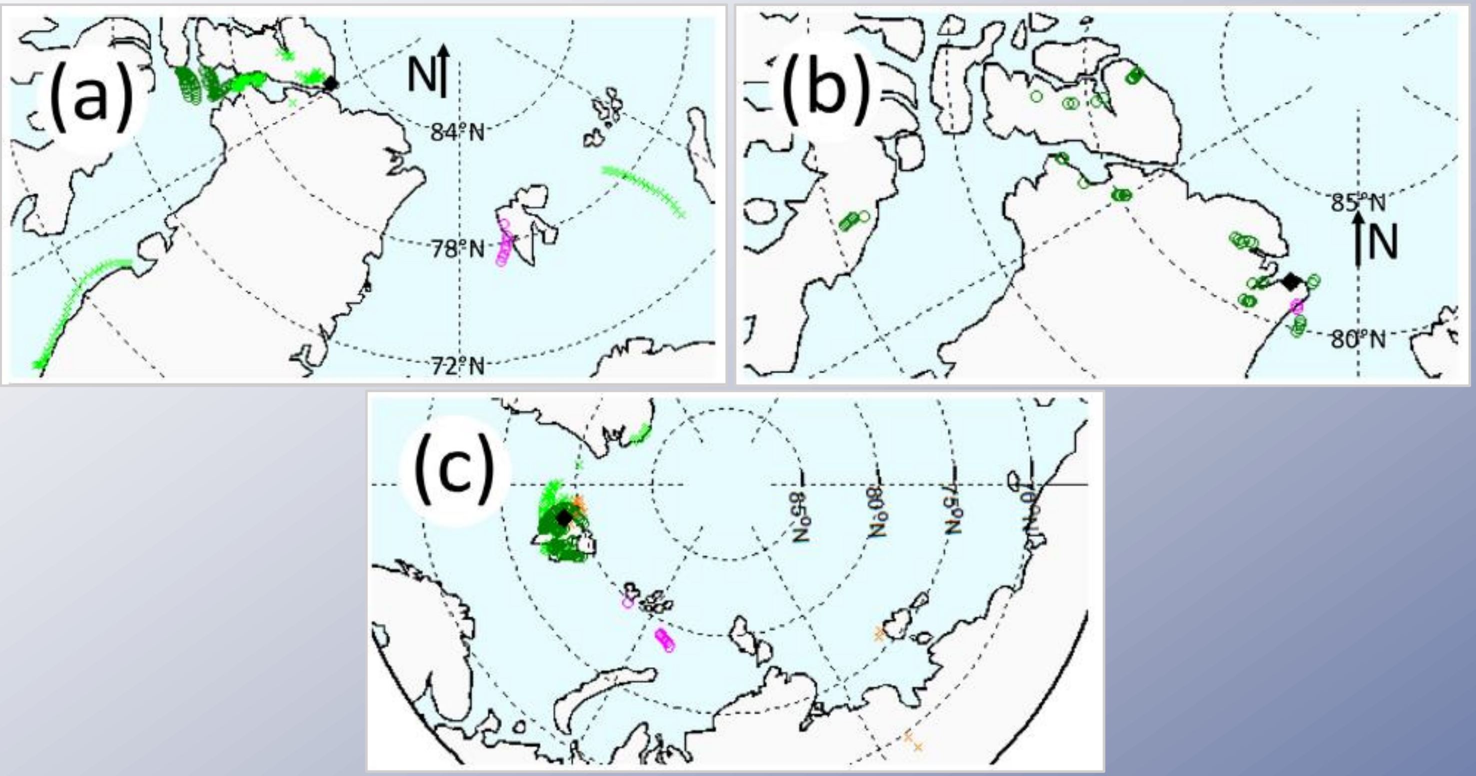

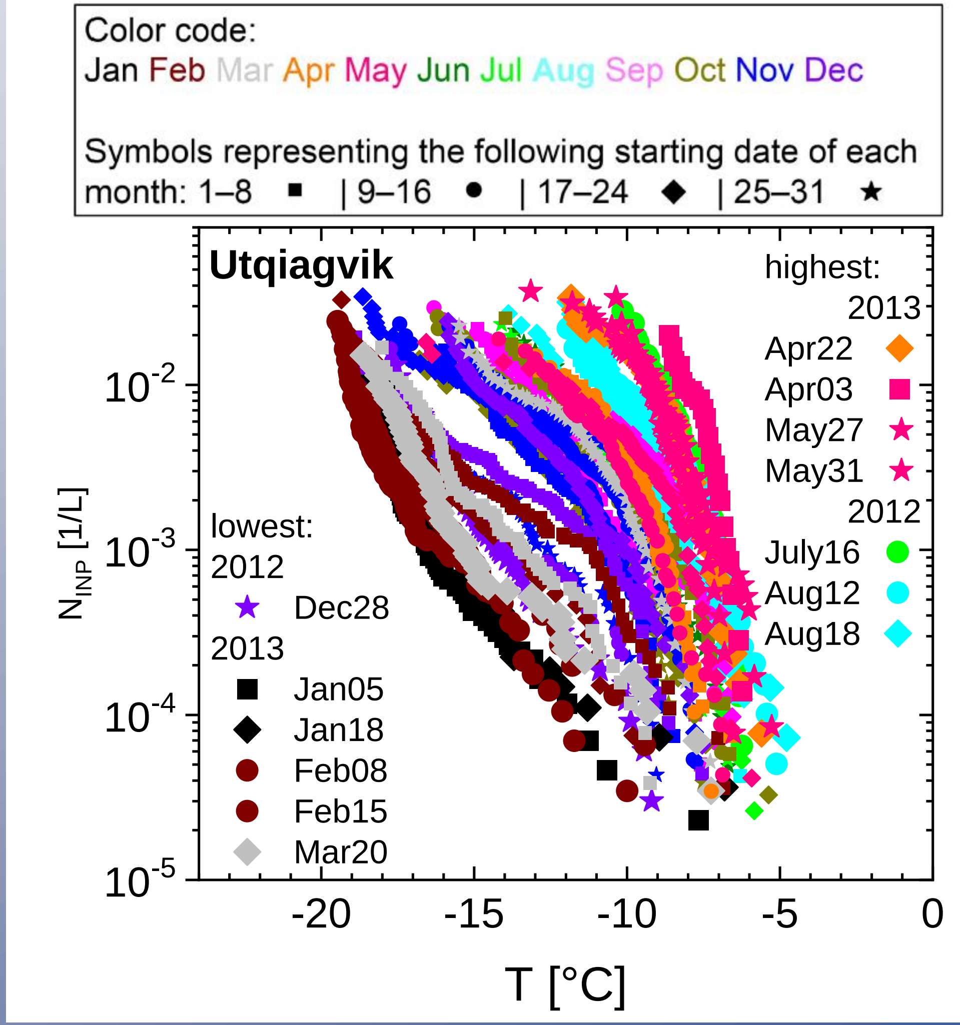

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- *Hartmann et al. (2019)* & *Schrod et al. (2020)*
  no increase in INP from ice cores (some hundred years back, going up to 1990)
  (the latter: maybe a small increase after 1960,
  due to land-use change)

https://archaeologynewsnetwork.blogspot.com/2015/09/ice-sample-from-greenland-and-russia.html

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  due to land-use change)

-> **antropogenic pollution does not contribute INP**

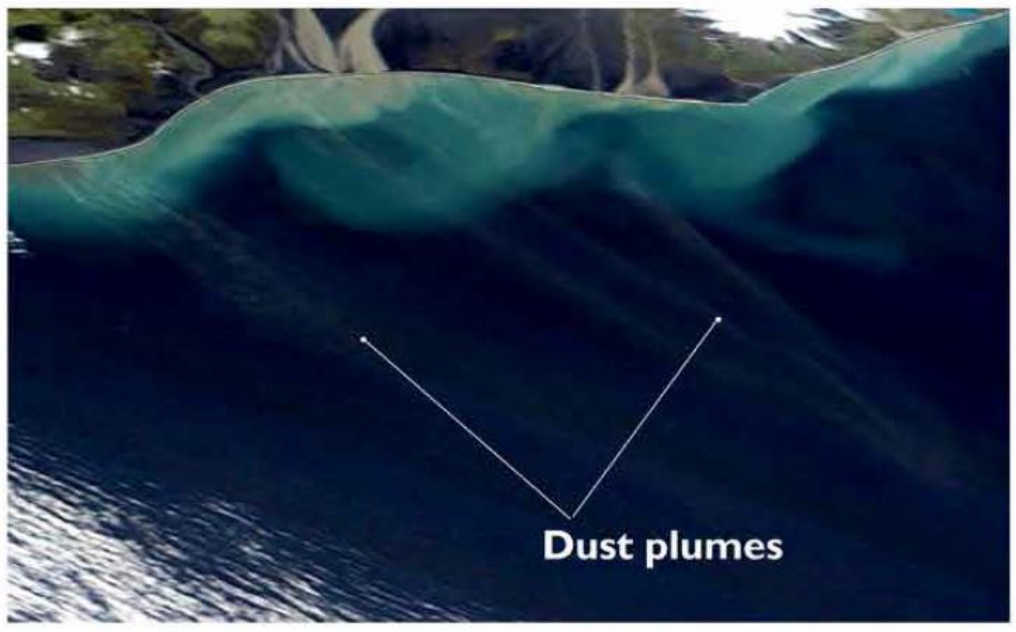
-> BUT: **future changes** due to **Arctic Amplification** may change INP concentrations and therewith clouds

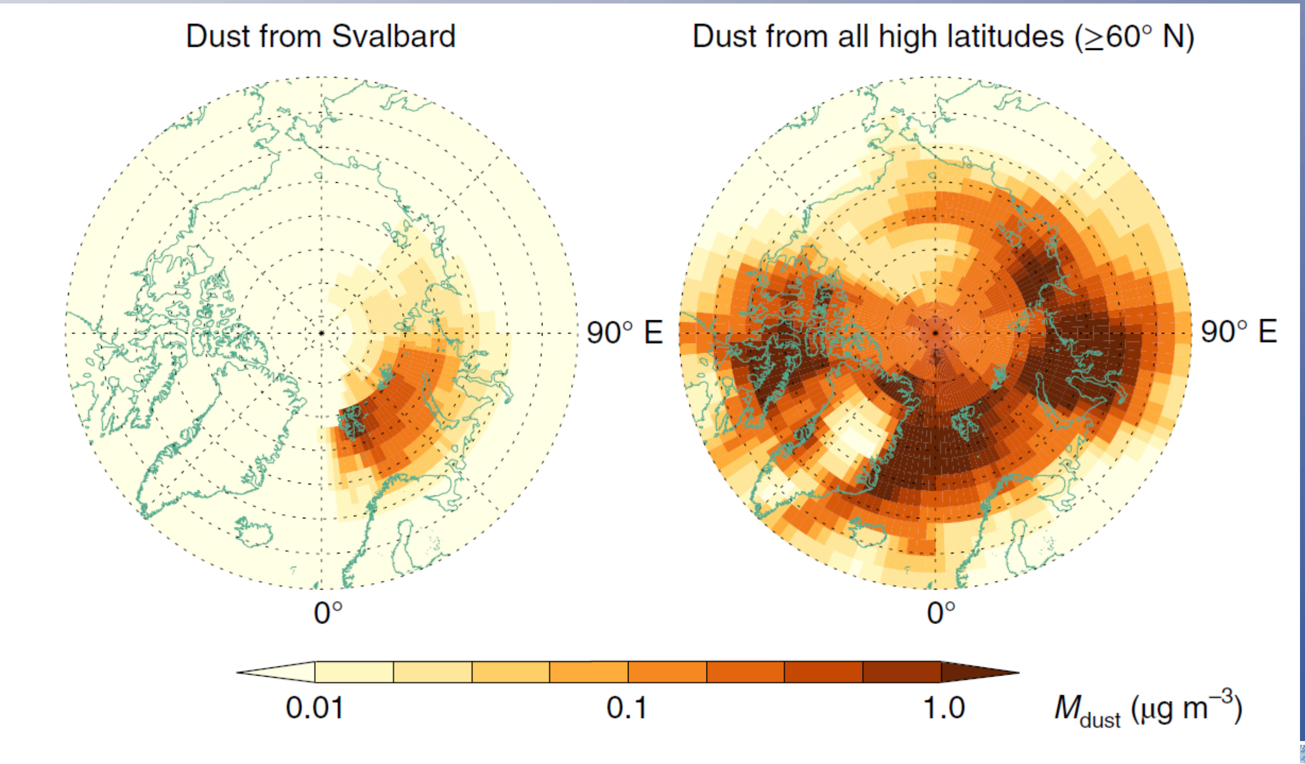
https://archaeologynewsnetwork.blogspot.com/2015/09/ice-sample-from-greenland-and-russia.html

INP in the Arctic - sources

- possible terrestrial sources:

Santl-Temkiv et al. (2019) – Villum, northern Greenland: **local dust** source with **biogenic contribution**
Si et al. (2019) – Alert, Canada: INP @ -25°C -> **long range dust transport** from Gobi desert
Tobo et al (2019) – Svalbard: local dust source, **glacial outwash**, connected to **organic material**
Sanchez-Marroquin et al. (2020) – Iceland: **local dust source**





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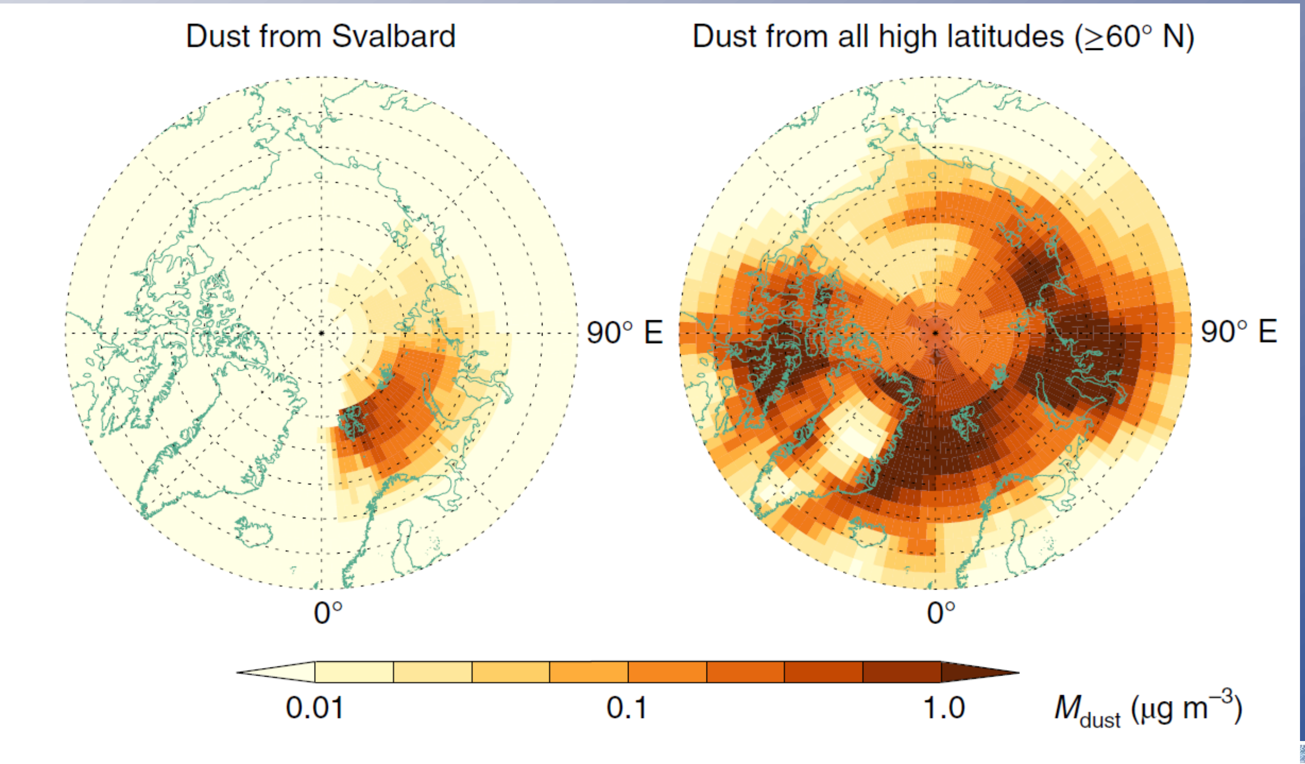
-> there are **Arctic dust sources** which may be **connected to biogenic INP**
-> **biogenic INP** may be emitted from the terrestrial **biosphere**
-> **long range transport** may also contribute INP

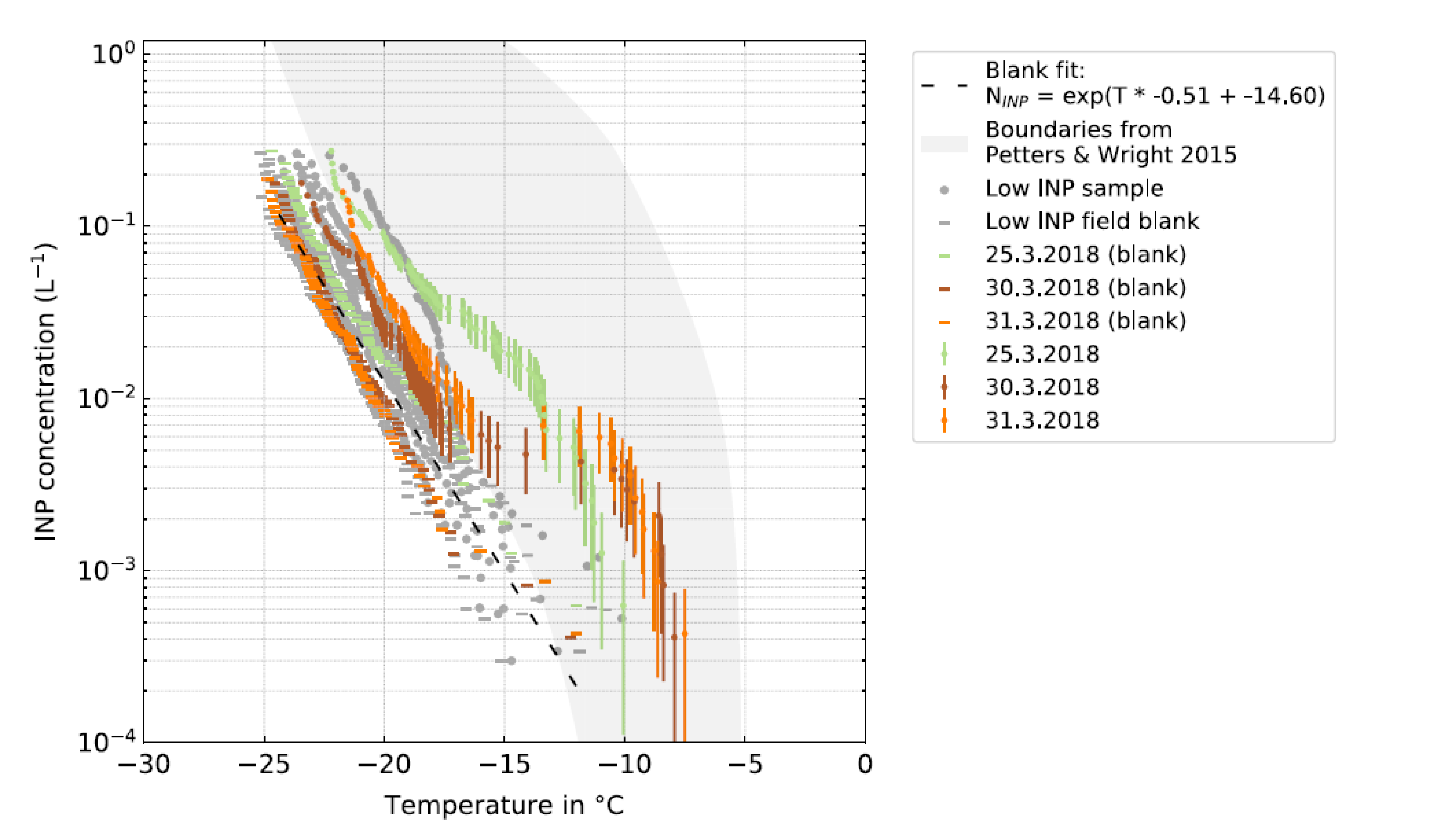


TROPOS

INP in the Arctic - sources

- possible marine sources:

Irish et al. (2017) – **INP in Arctic ocean**, particularly **close to melting sea ice**
Hartmann et al. (2020) – airborne sampling over ice in March, **INP from open polynyas**



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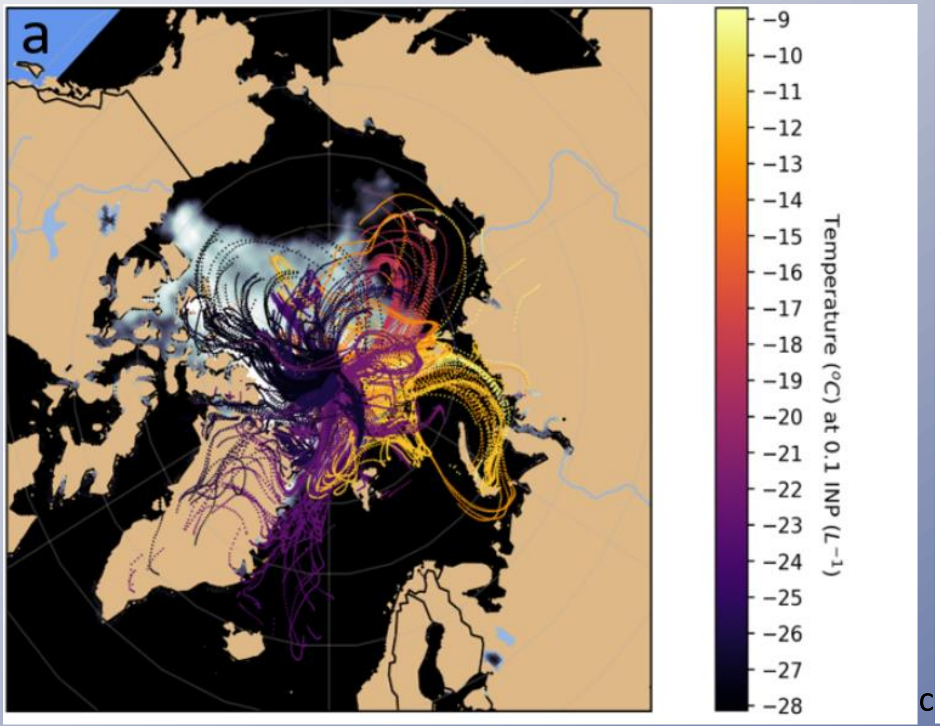
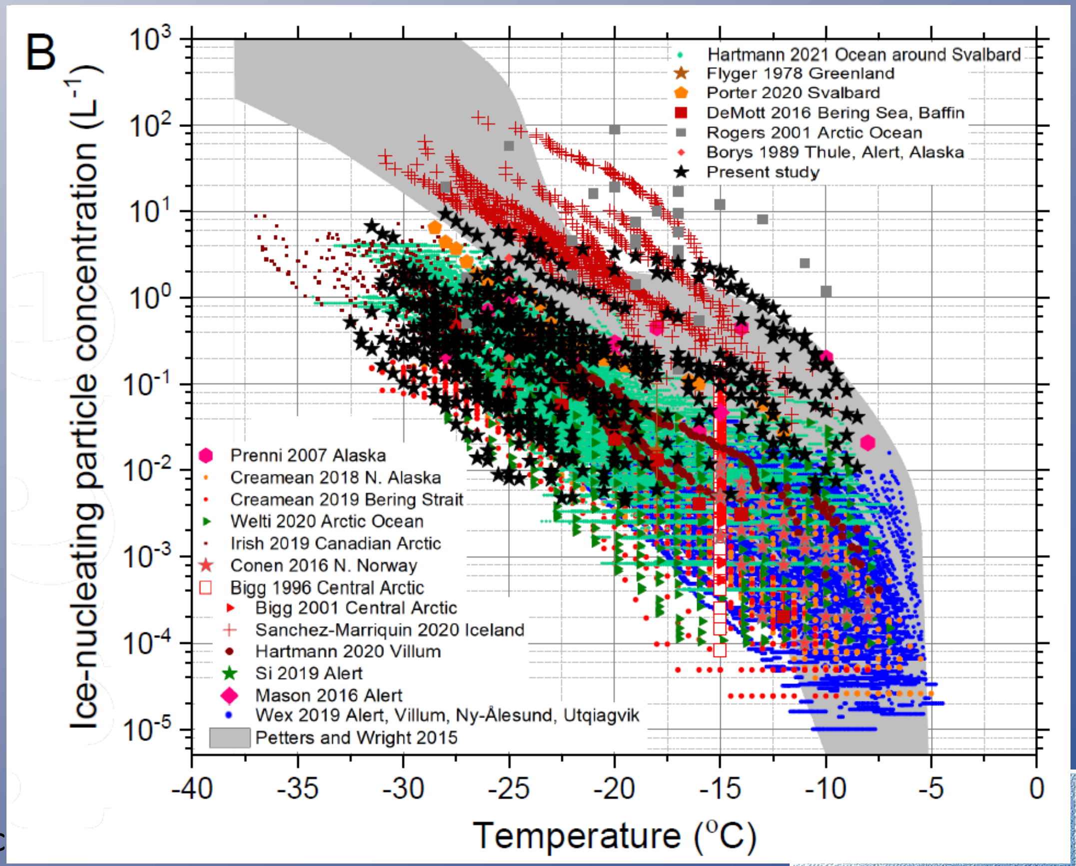
Porter et al. (2022) – **highly varying INP concentrations** at the **North Pole**, up to very high values

-> **marine biogenic sources** may also contribute INP





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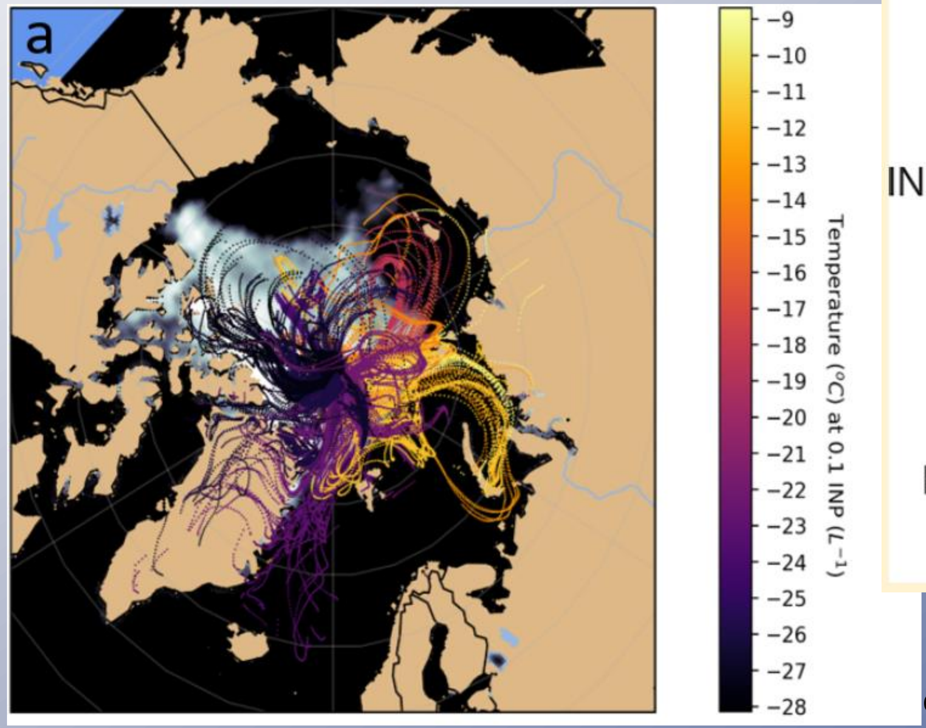
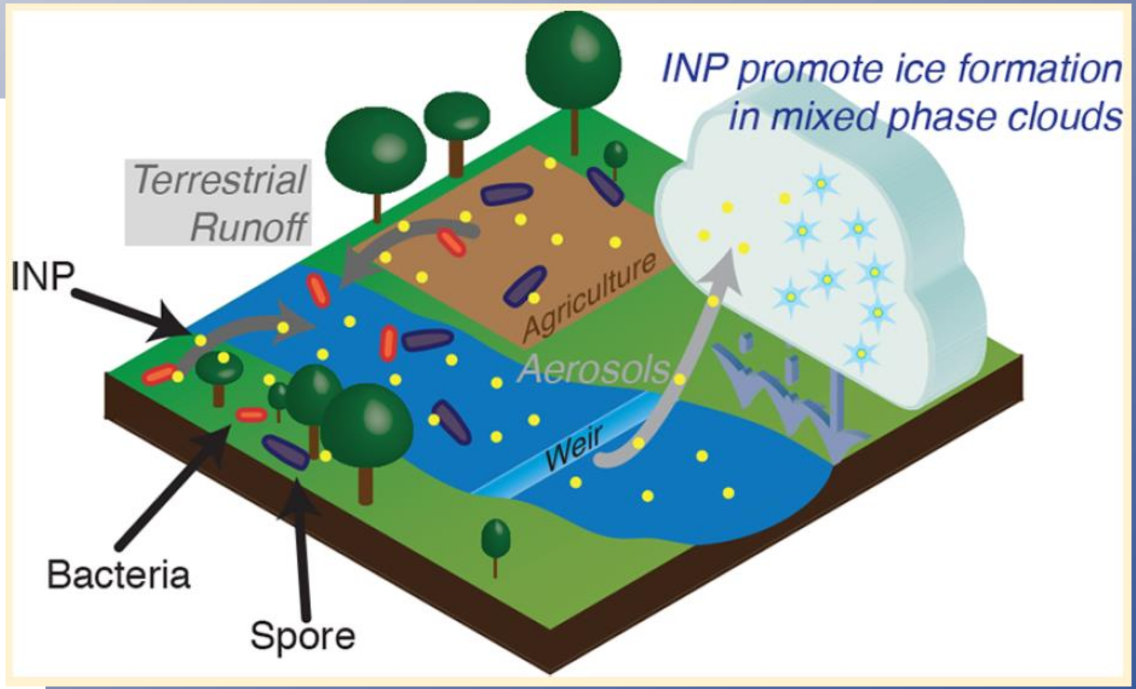
Knackstedt et al. (2018):

rivers contain INP from terrestrial sources

-> **marine (?) biogenic sources** may also contribute INP

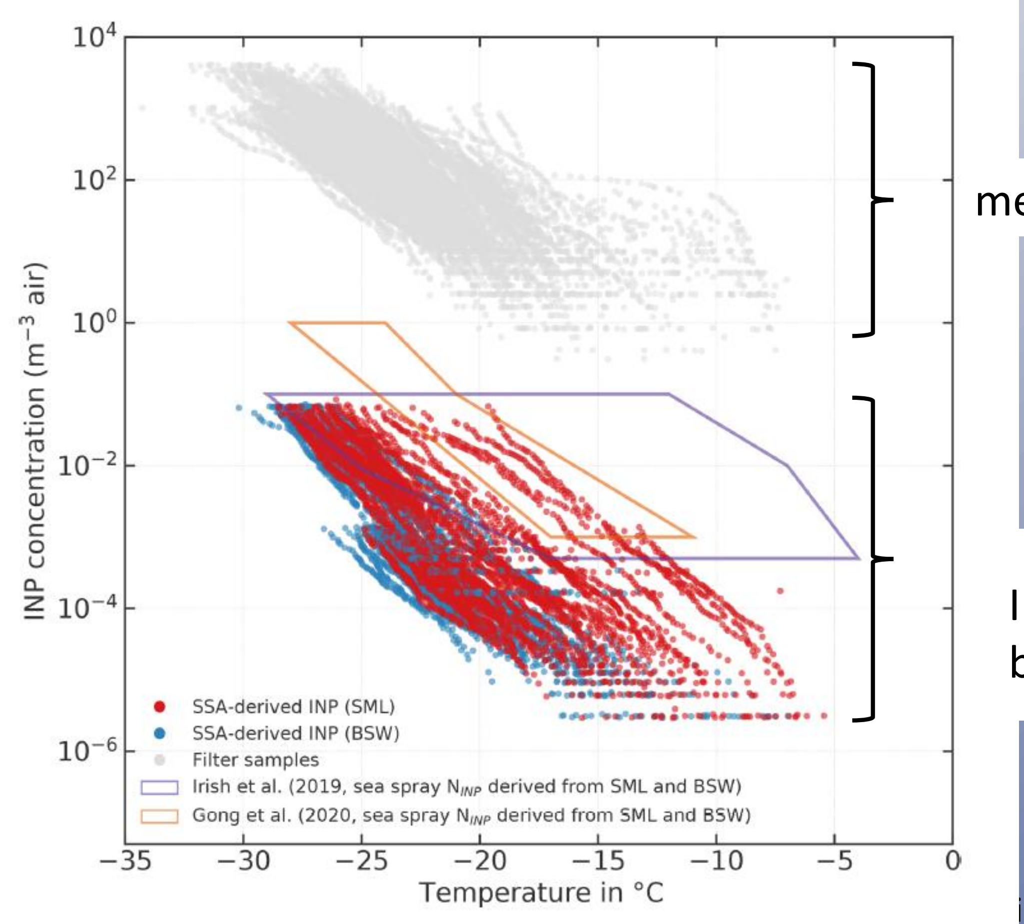




but

- *Gong et al. (2020)* **at Cabo Verde** and *Hartmann et al. (2021)* **during ship cruise in the Arctic**

INP concentrations in the **ocean water** (bulk and surface microlayer (SML) are orders of magnitude **too low to explain related atmospheric INP concentrations**, **UNLESS** there is **strong enrichment** during bubble bursting

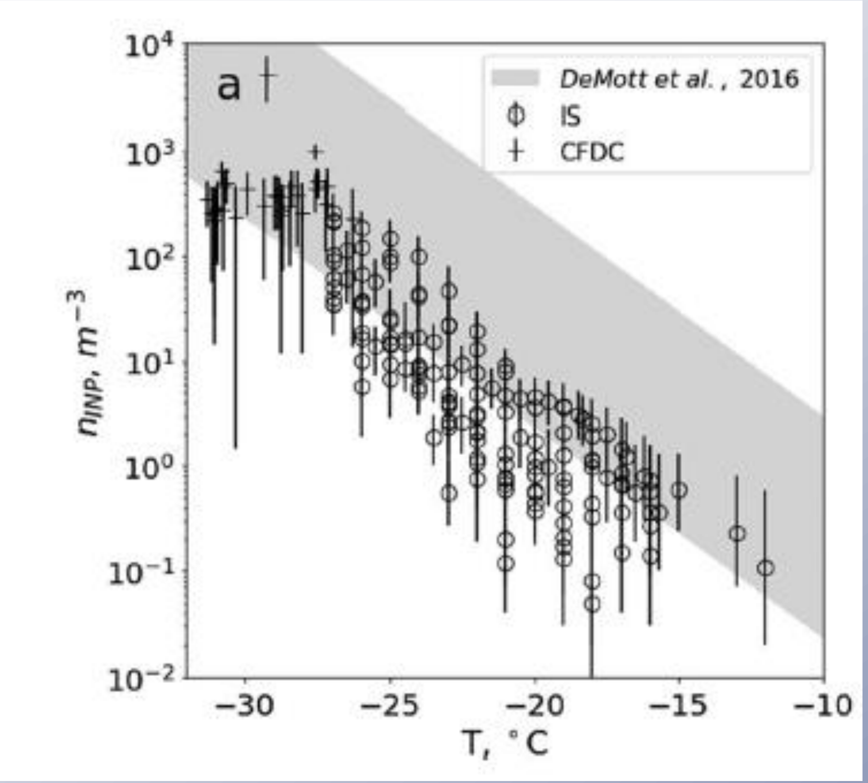


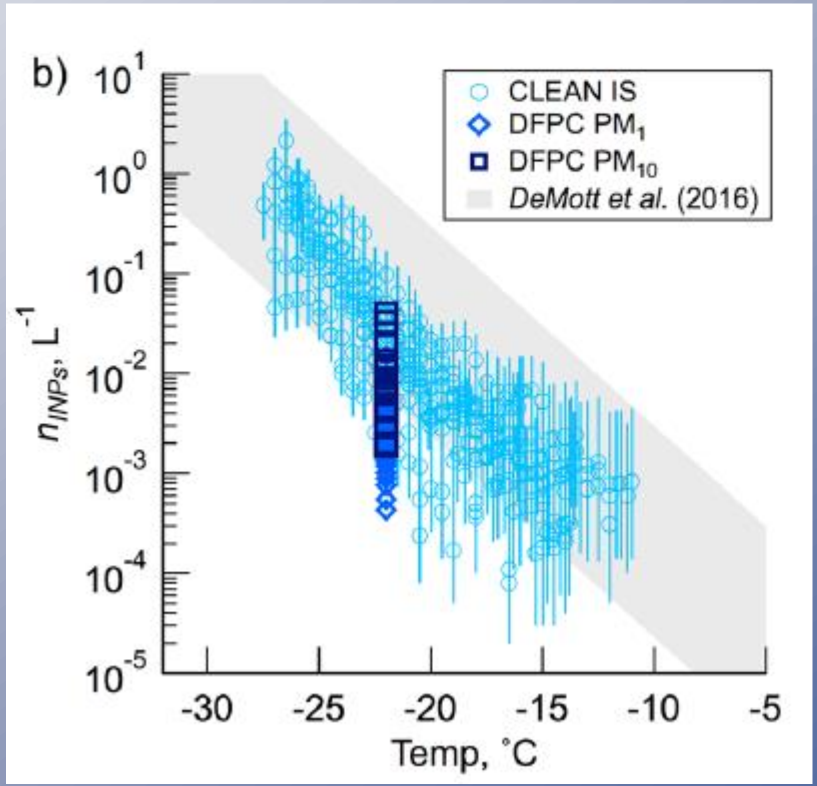
measured atmospheric INP concentration

INP concentration expected from bubble bursting, based on INP concentrations in sea water

INP in remote oceanic regions / Southern Ocean

- *McCluskey et al. (2018a,b):*
  very low INP concentrations in the **Southern Ocean** and the **clean North-East Atlantic**





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  INP concentrations from ship based measurements: lower by 1 order of magnitude for marine, compared to continental, **Southern Ocean** even one order of magnitude **lower**

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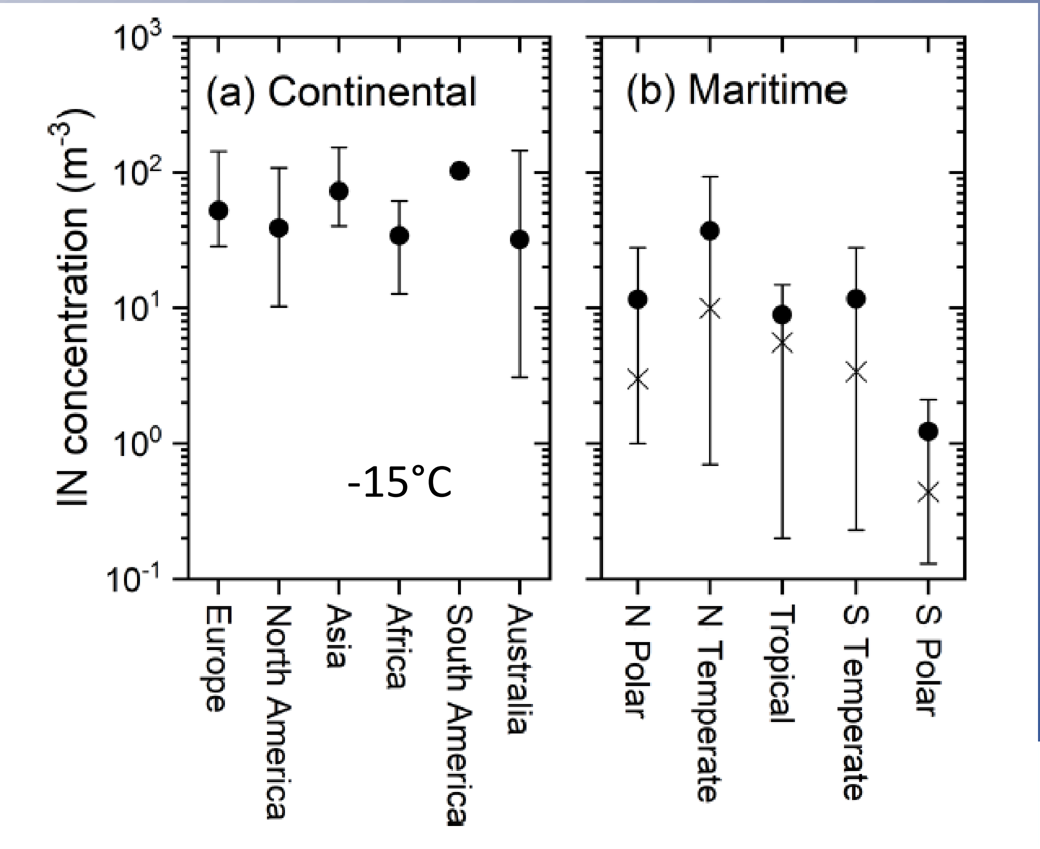
  also **very low INP concentrations** in the **Southern Ocean**



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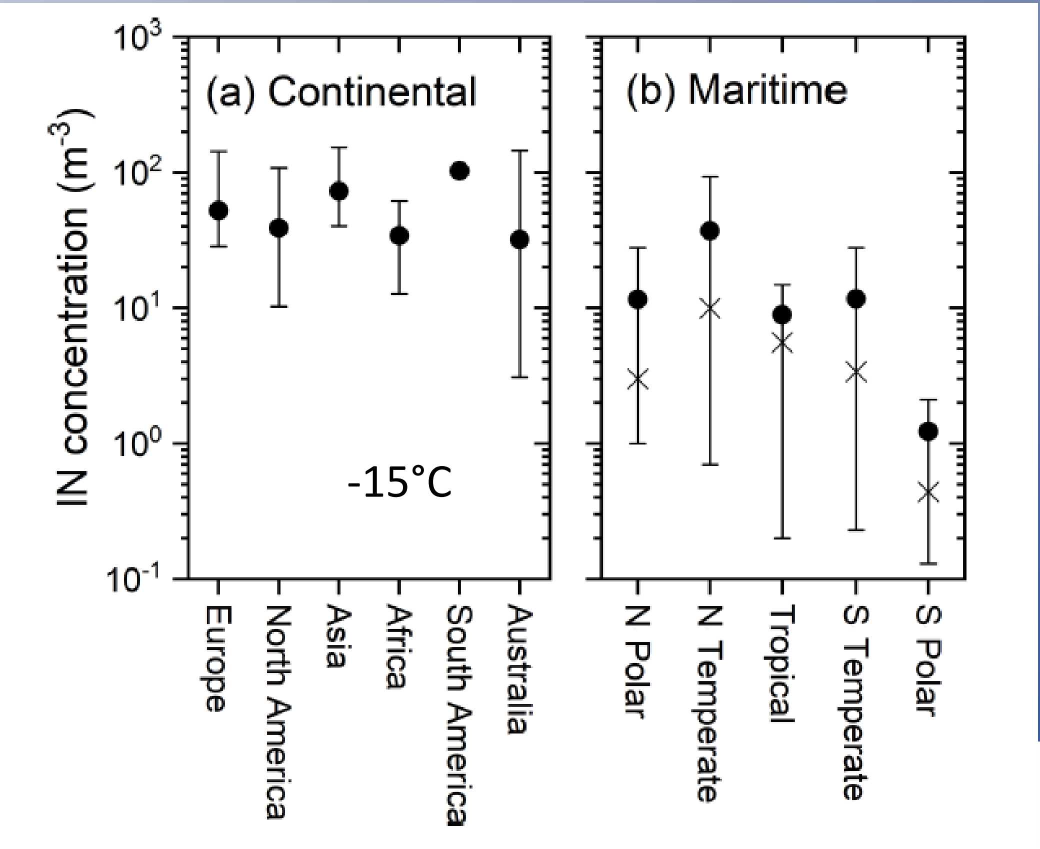
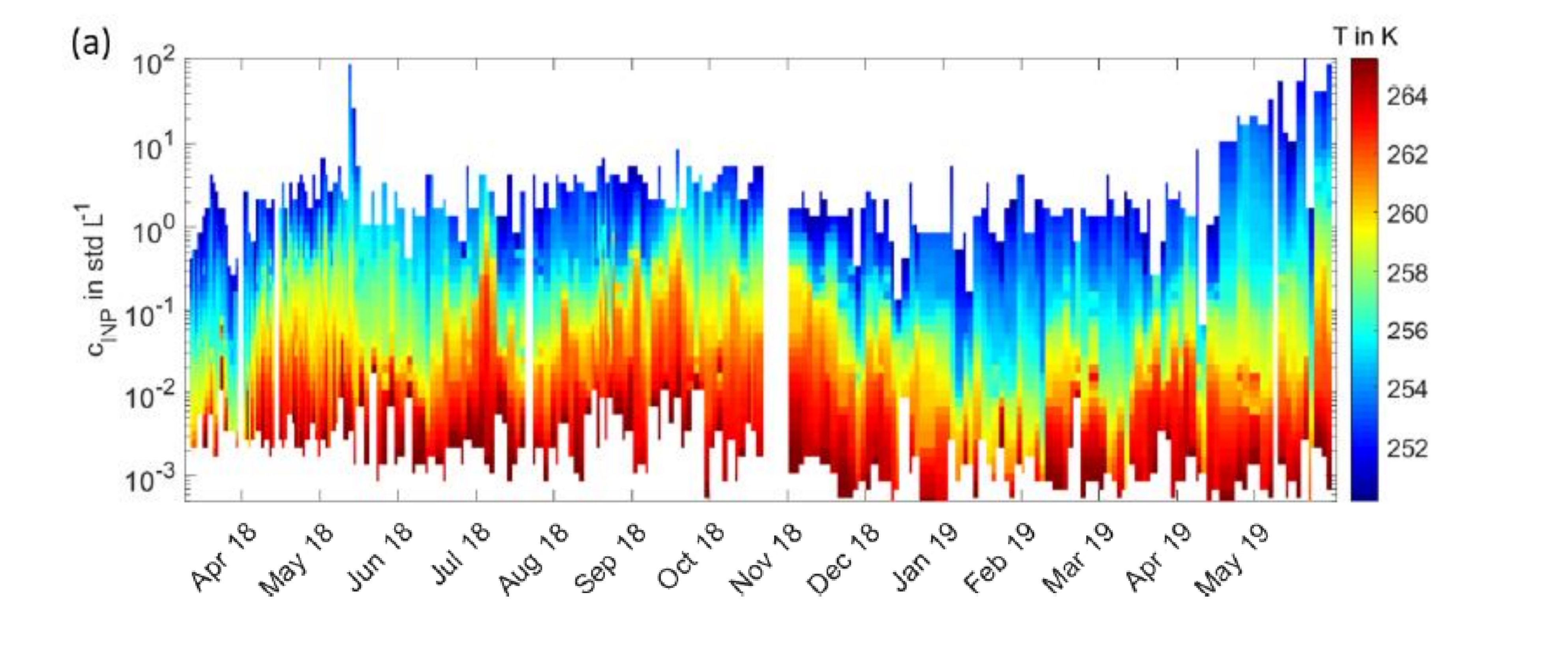
fits to **high fractions of supercooled liquid droplets** in clouds over the **Southern Ocean** observed from **satellite**
Choi et al. (2010), Zhang et al., (2018)



long term studies

- *Schneider et al. (2021):*

boreal forest in Finnland, seasonal **INP cycle** linked to the prevalence of **biogenic** aerosol particles
-> **parameterization wrt. temperature**



more long term studies

- *Schrod et al. (2020):*

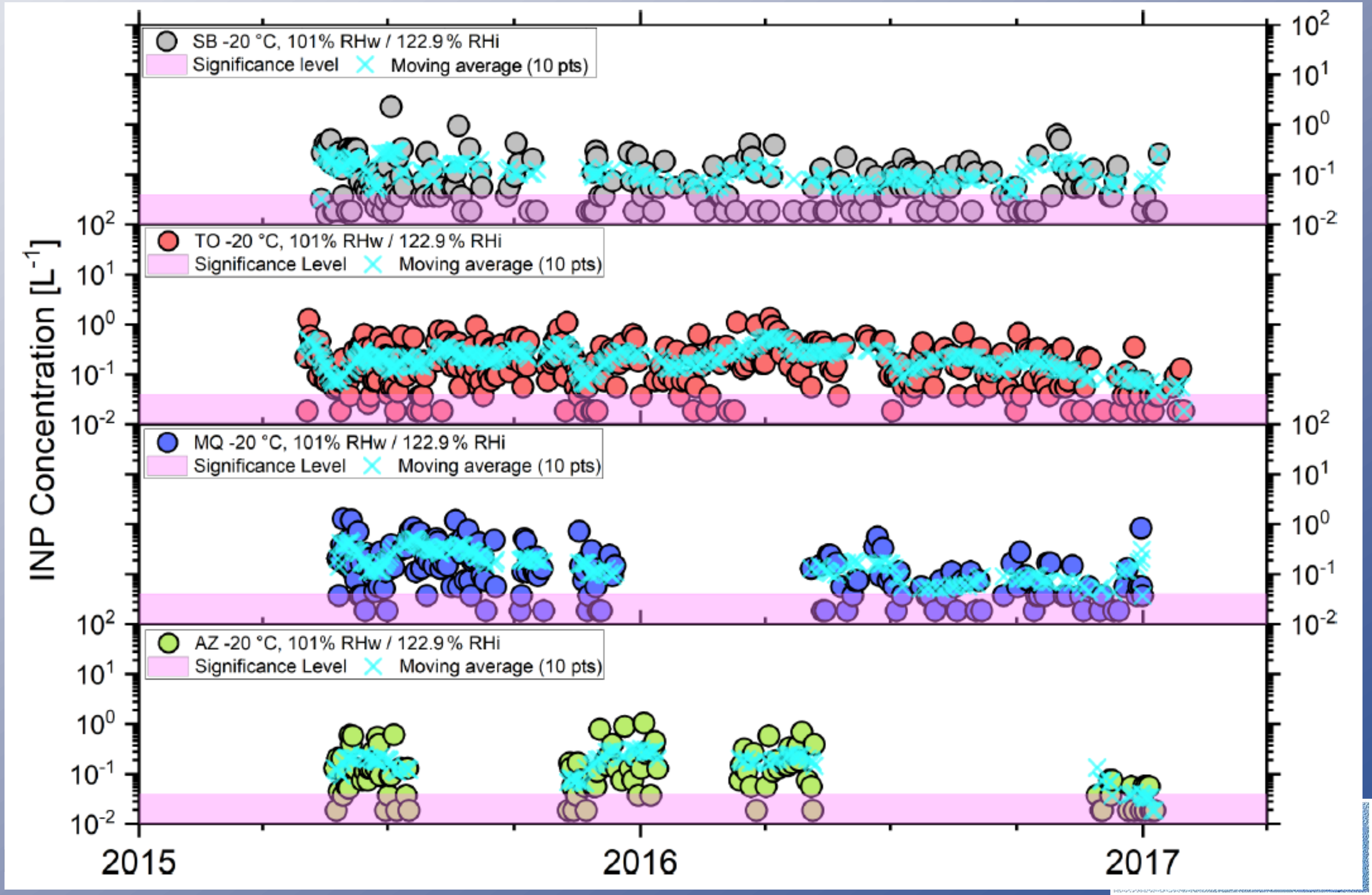
4 stations: Arctic (Svalbard), central Europe (Germany), Caribbean (Martinique) and Amazon (Brazil), **data at -20°C and -25°C**

short-term variability overwhelms all long-term trends and/or seasonality

Amazon mostly **unaffected by biomass burning** season

no anthropogenic influence in Europe or Arctic

Carribean affected by **long range dust transport**



more long term studies

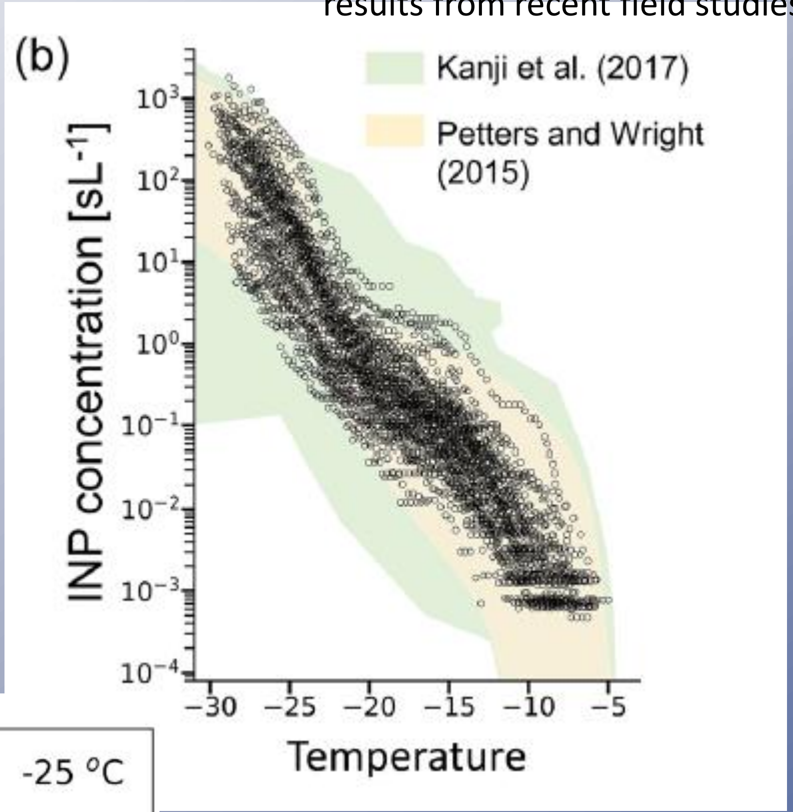
- *Testa et al. (2021):*

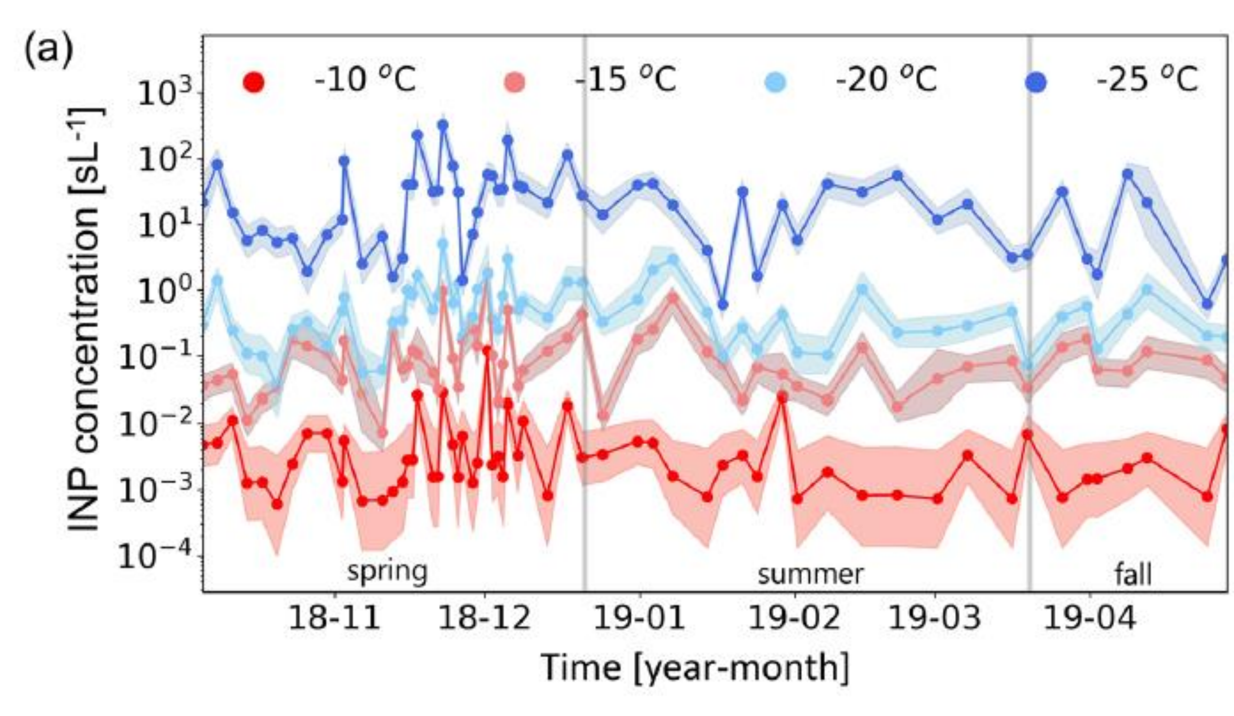
north central Argentina, 7 month (austral spring to mid fall), **no seasonal cycle**

heat labile INPs dominated at -5 to -20°C

non-heat-labile organic INPs (H2O2-treatment) dominated from -20 to -28°C, their ratio to mineral dust was constant
-> **likely regional INP from arable topsoil**

bio INP peaked during rain & high relative himidity





TROPOS

more long term studies

- *Gong et al. (2022):*

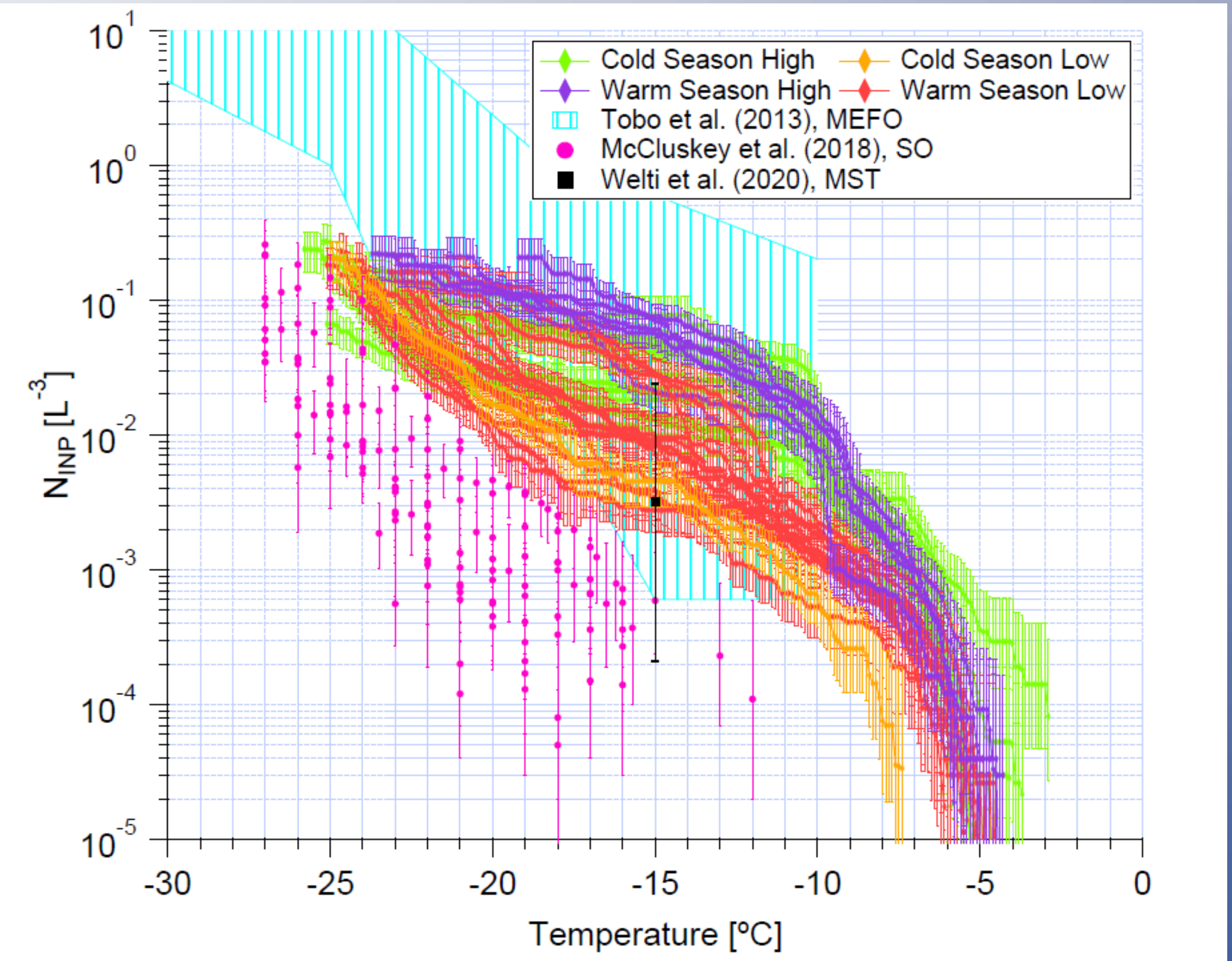
southern Chile (600m high, 8km west of Punta Arenas), 11 month (May to March), no seasonal cycle,

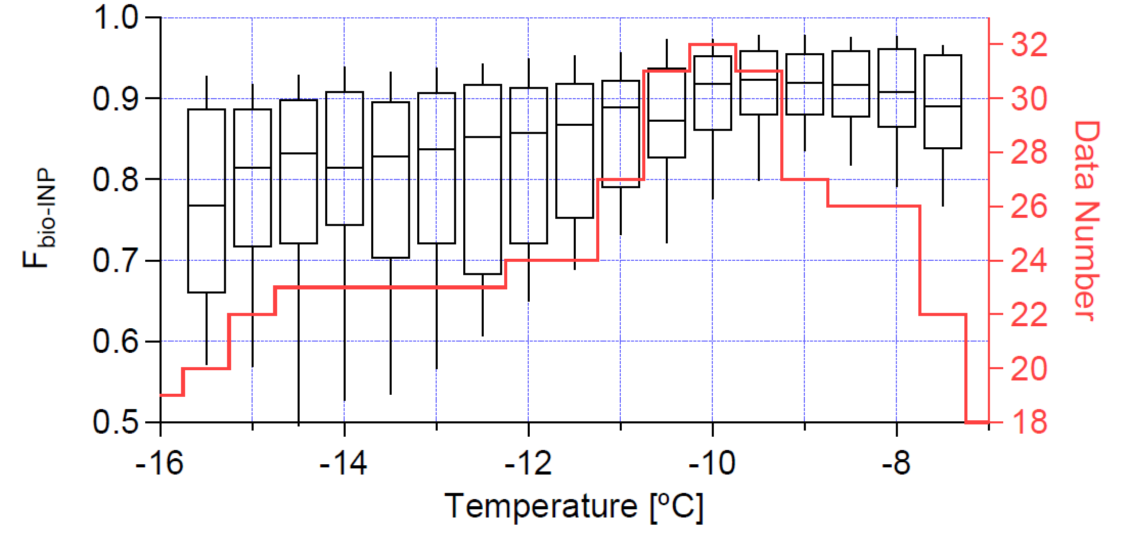
suprisingly high INP concentrations

high fraction of heat liable **(biogenic) INP** down to -16°C

precipitation enhances INP concentrations

-> see also ***Huffman et al. (2013)***





summary for results from recent field studies

- a large number of **atmospheric INP** is **supermicron** in size (down to -25°C)

- **no INP from anthropogenic pollution** for temperatures typical for mixed phase clouds

- **annual cycle** for highly ice active (biogenic) INP **in some locations** (**Arctic, Finnish boreal forest**)
  but **not everywhere** (**North Central Argentina, southern Chile**)
  and also **no annual cycle at -20°C and -25°C**

- **summertime Arctic** can have INP **concentrations as observed over mid-latitude continents**

- **remote marine regions** (**Southern Ocean, clean North West Atlantic**) have **low INP** concentrations

- **INP from sea spray production** can **not explain atmospheric INP** concentrations in marine areas **without INP enrichment during bubble bursting process**

- **enhanced INP concentrations over continents**, maybe **connected to precipitation**

TROPOS

TROPOS

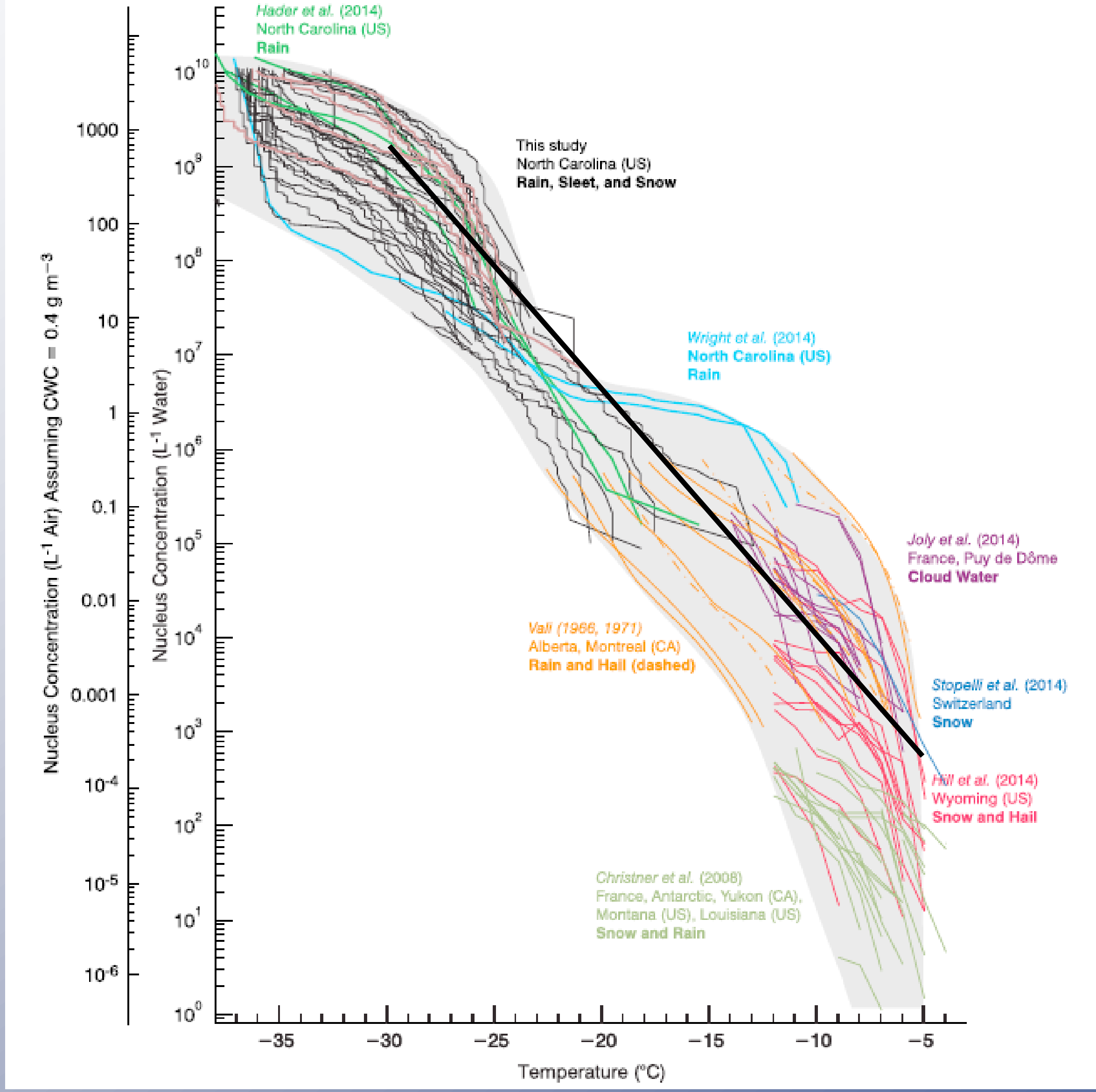
concentrations of ice nucleating particles (N_{INP})

Petters & Wright, GRL (2015)

Fletcher (1962):

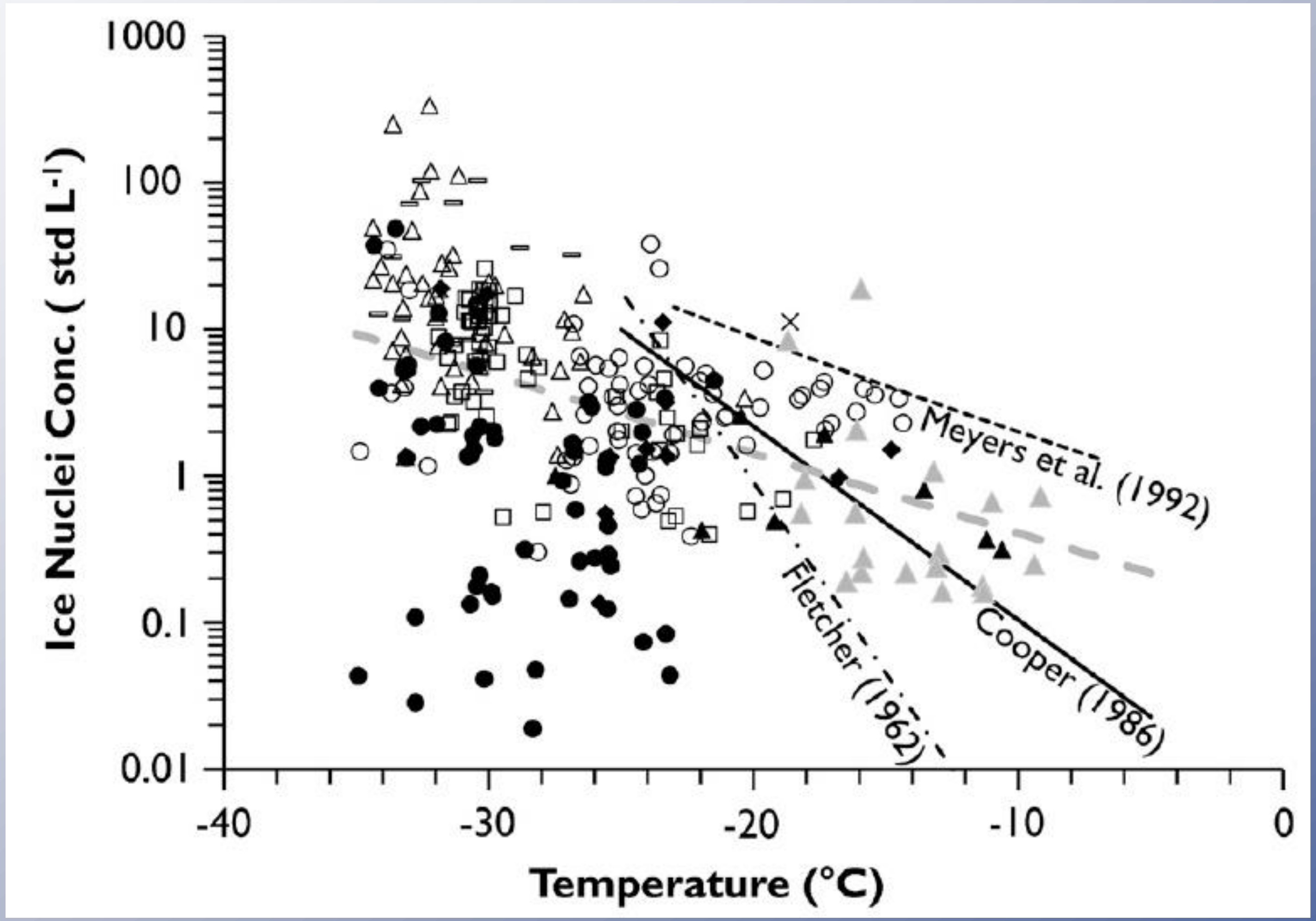
- overestimation at lower temperatures

- does not capture observed variations



parameterizations for N_{INP}

DeMott et al., PNAS (2010)



parameterizations for N_{INP}

DeMott et al.,
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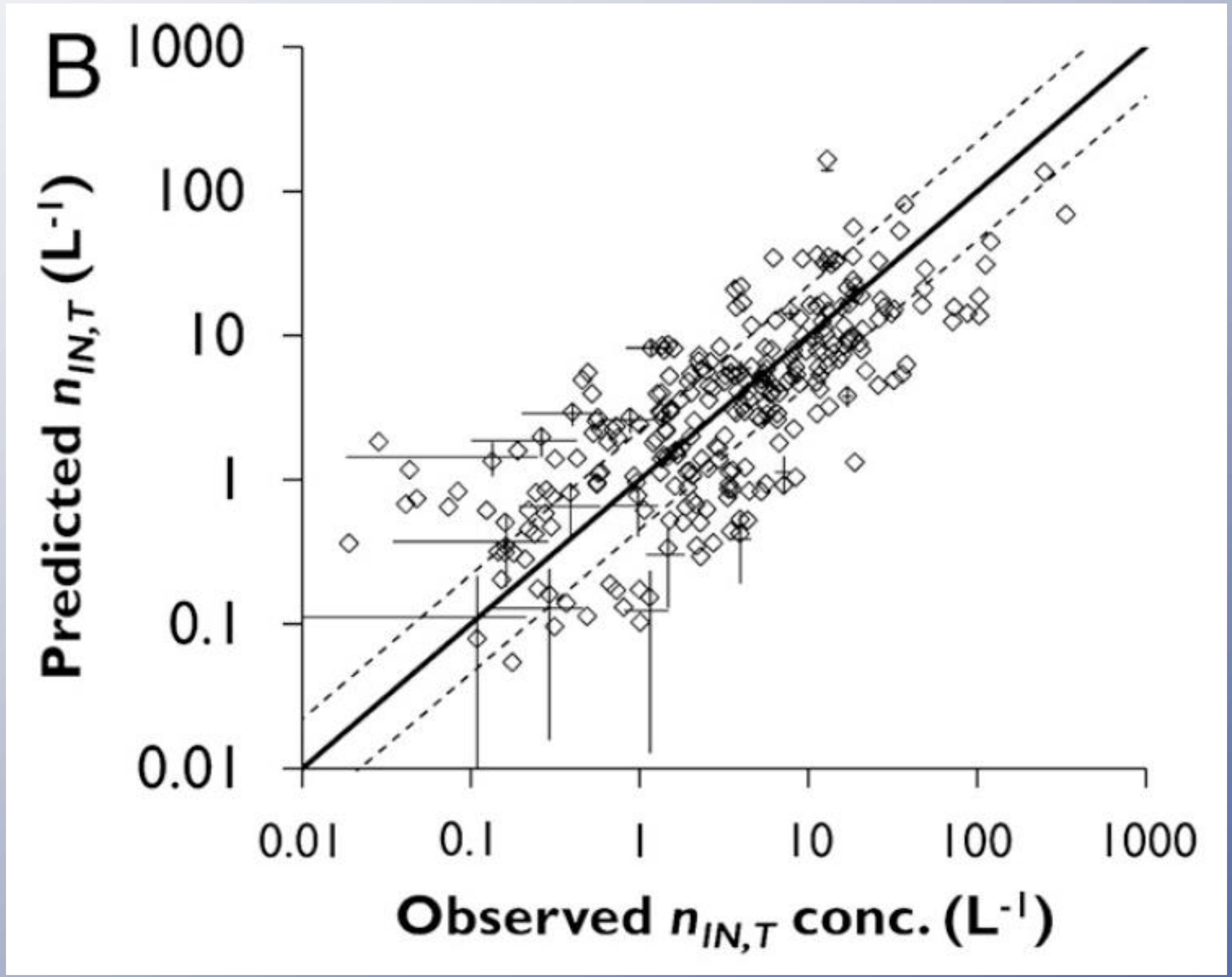
$$n_{\mathrm{IN},T_k} = a(273.16 - T_k)^b (n_{\mathrm{aer},0.5})^{(c(273.16-T_k)+d)}$$



where $a = 0.0000594$, $b = 3.33$, $c = 0.0264$, $d = 0.0033$, T_k is cloud temperature in degrees Kelvin, $n_{\mathrm{aer},0.5}$ is the number concentration (scm^{-3}) of aerosol particles with diameters larger than 0.5 μm and n_{IN,T_k} is ice nuclei number concentration (std L^{-1}) at T_k.

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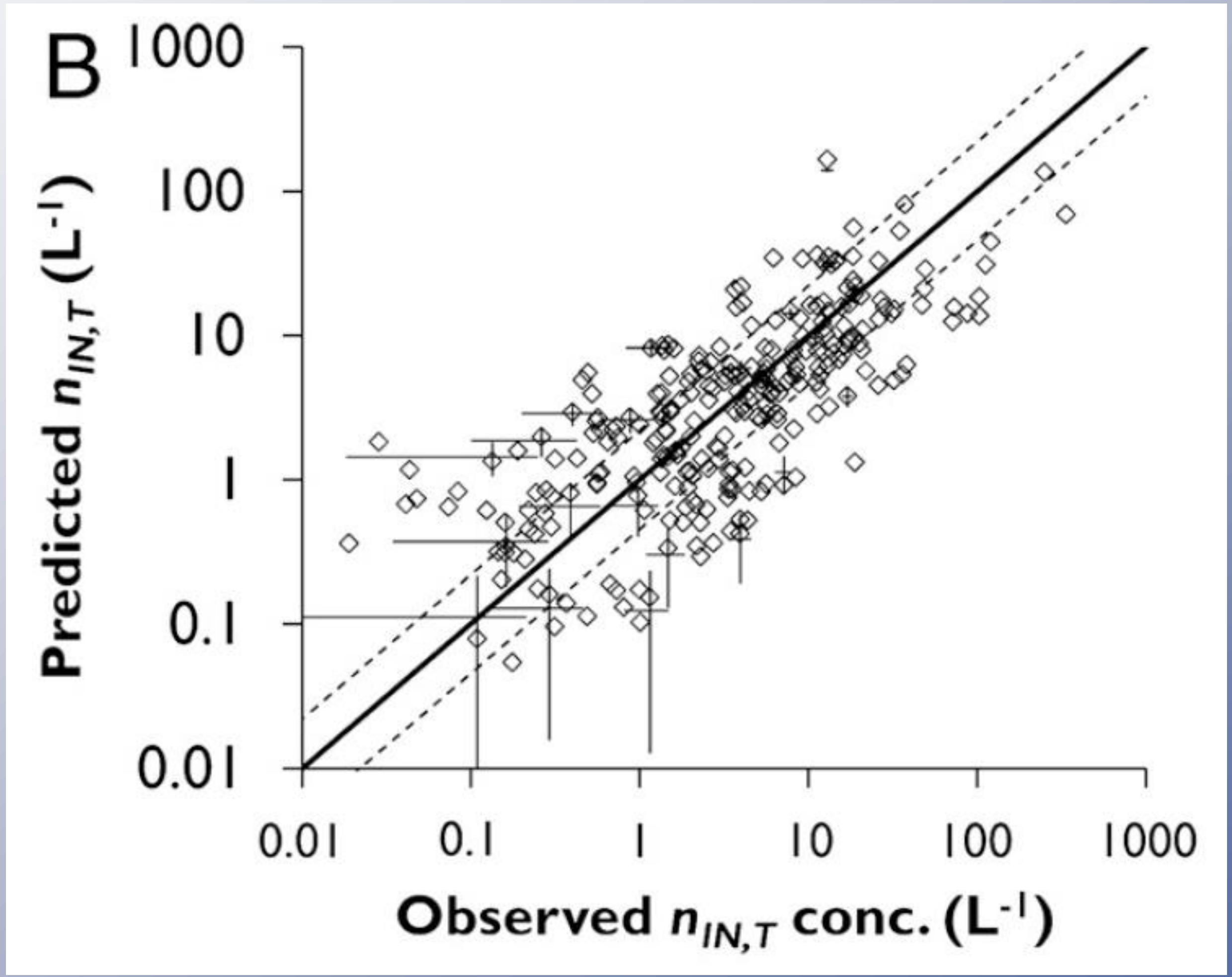
$$n_{\mathrm{IN},T_k} = a(273.16 - T_k)^b (n_{\mathrm{aer},0.5})^{(c(273.16-T_k)+d)}$$

by now, there are a number of updated versions of this fit, e.g., Tobo et al. (2013), DeMott et al. (2015)



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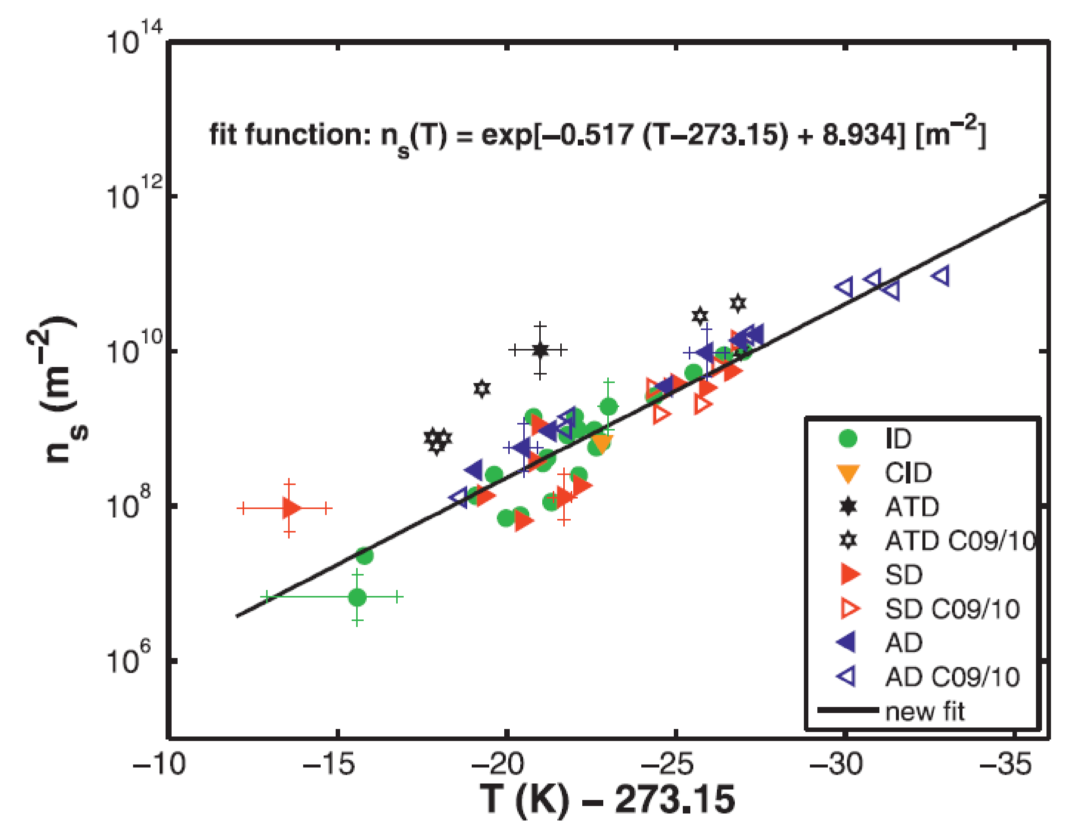
parameterizations for N_{INP}

Niemand et al.,
J. Atmos. Sci. (2012):

- ice nucleation active surface site density n_s

- dependent on available surface area of mineral dust



but: $n_s(T) = \frac{N_i}{S_{tot}}$, * $= \frac{f_{ice}}{S_{droplet}}$

is only valid for small f_{ice}, as stated in the paper!

that comes from using only the first term of a Taylor series expansion

-> full equation: $n_s = - \ln(1 - f_{ice}) / S_{droplet}$

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

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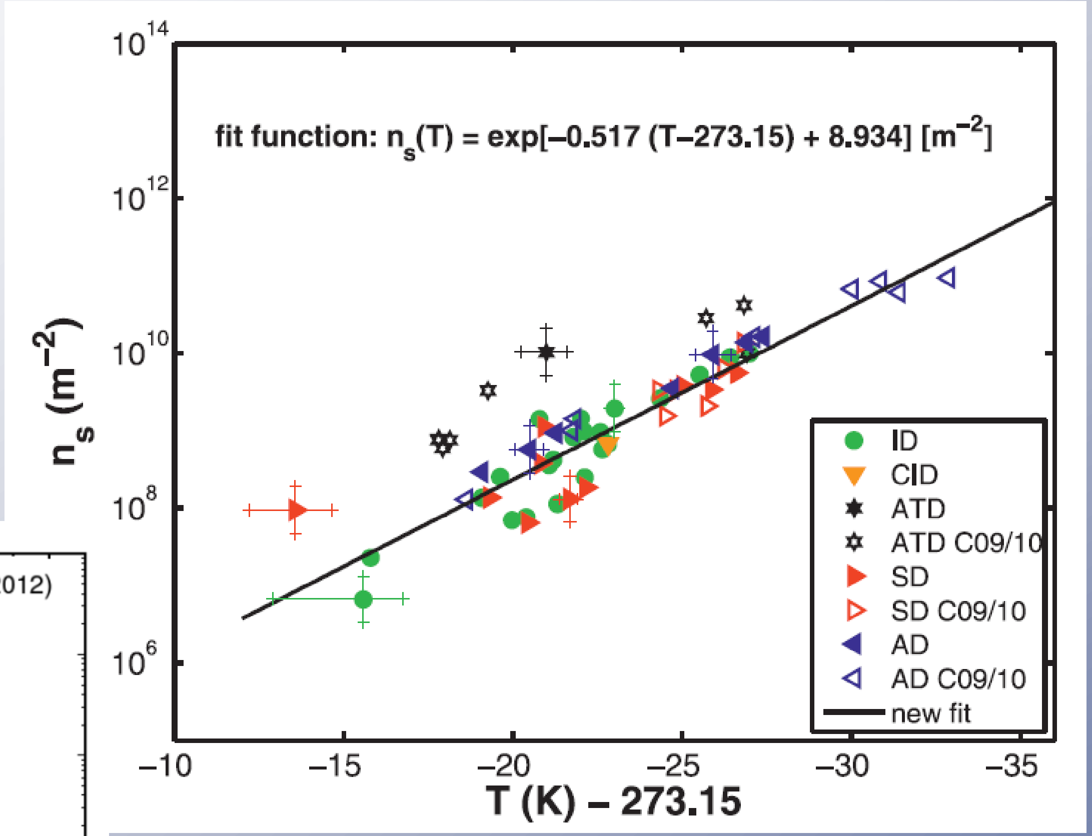
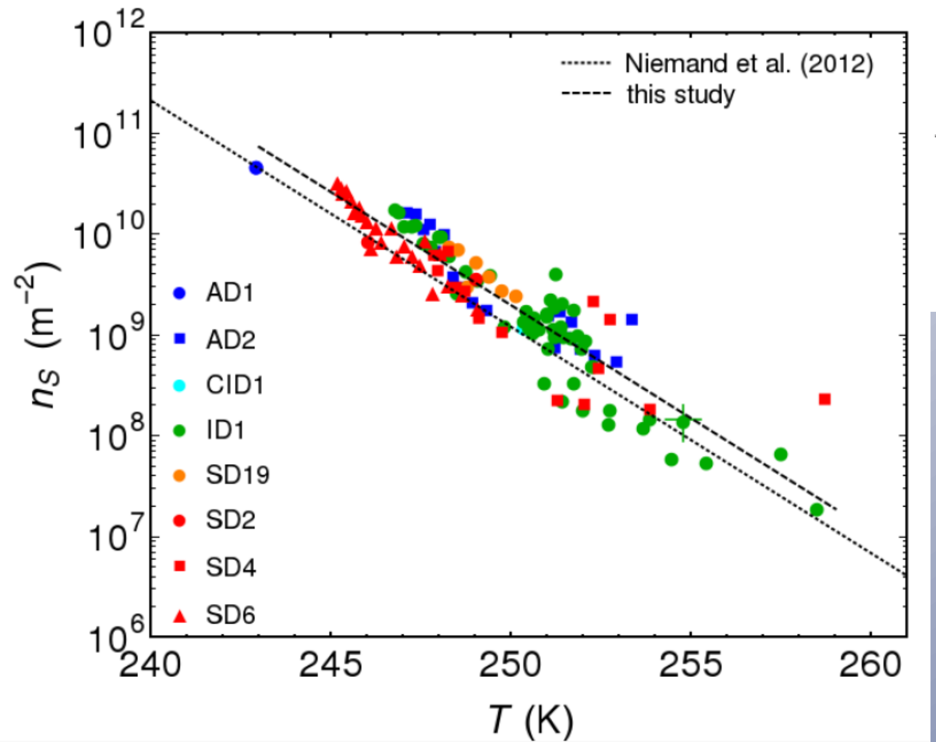
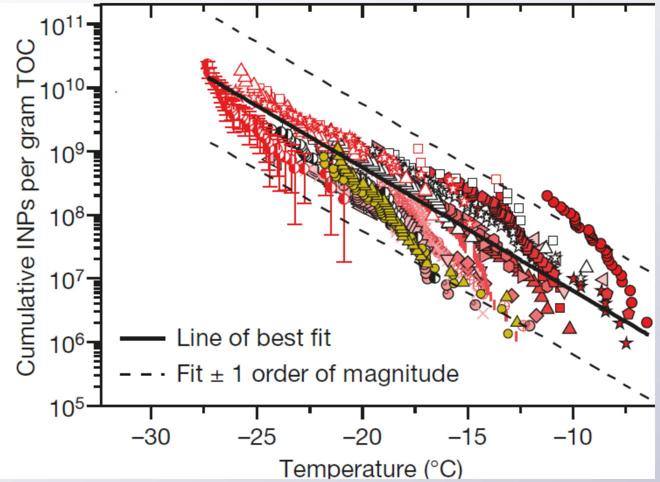
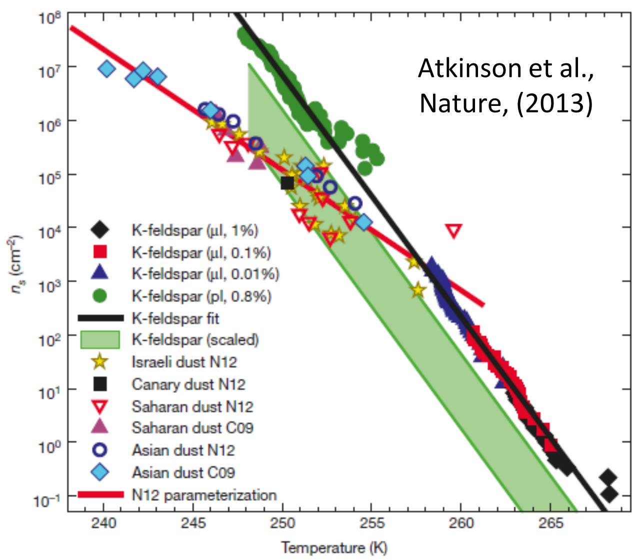




parameterizations for N_{INP}

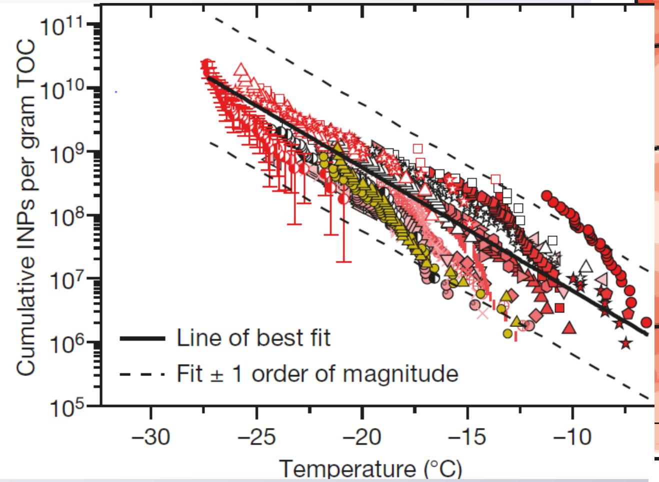
Wilson et al., Nature, (2015)

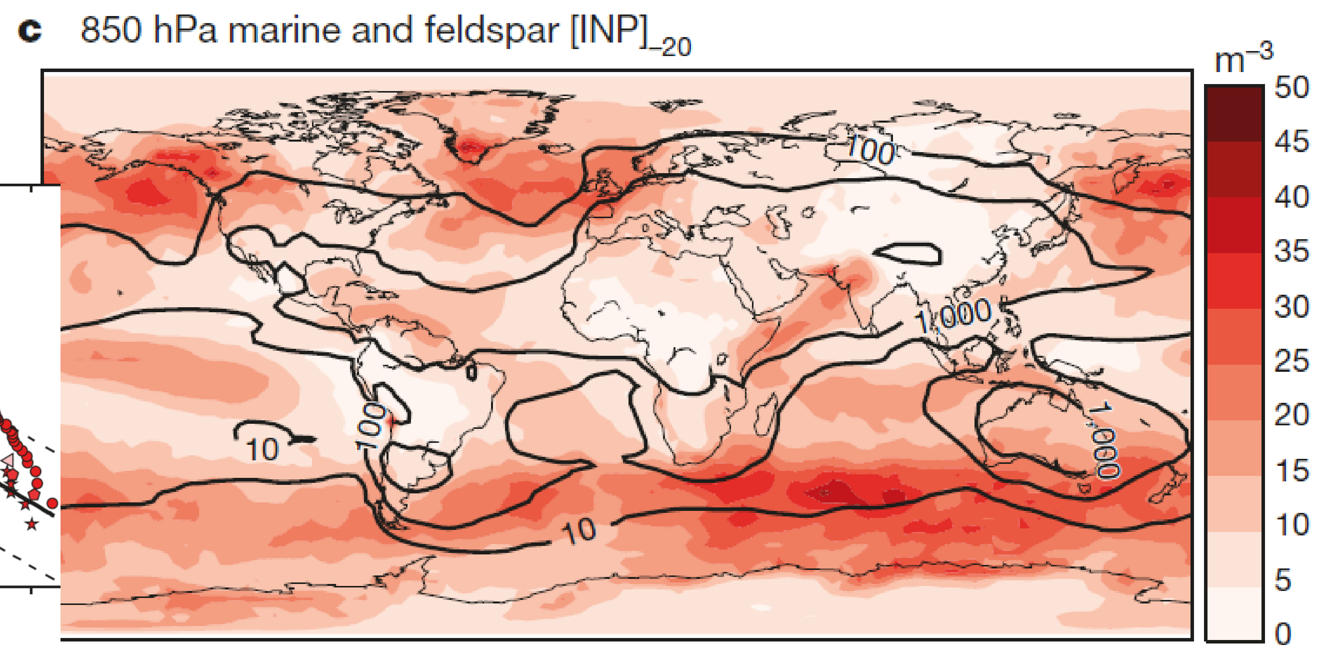




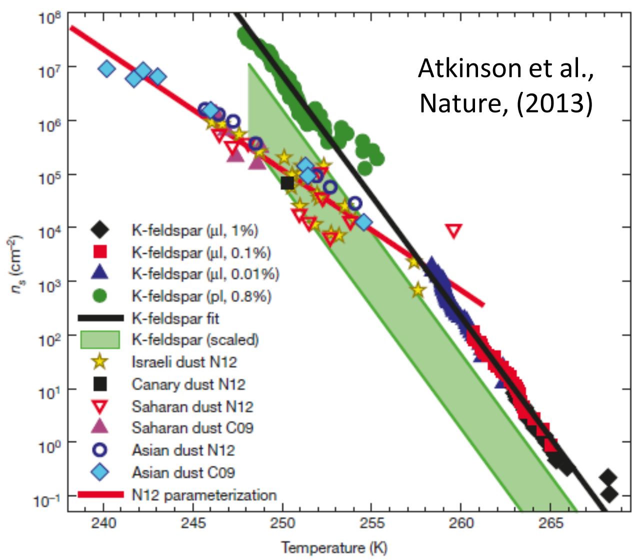
parameterizations for N_{INP}

Wilson et al., Nature, (2015)





c, Modelled distribution of marine biogenic INP concentrations active at −20 °C at 850 hPa (corresponding to the altitude of high-latitude mixed-phase clouds). Black contours indicate the INPs from desert dust based on K-feldspar emissions[30].

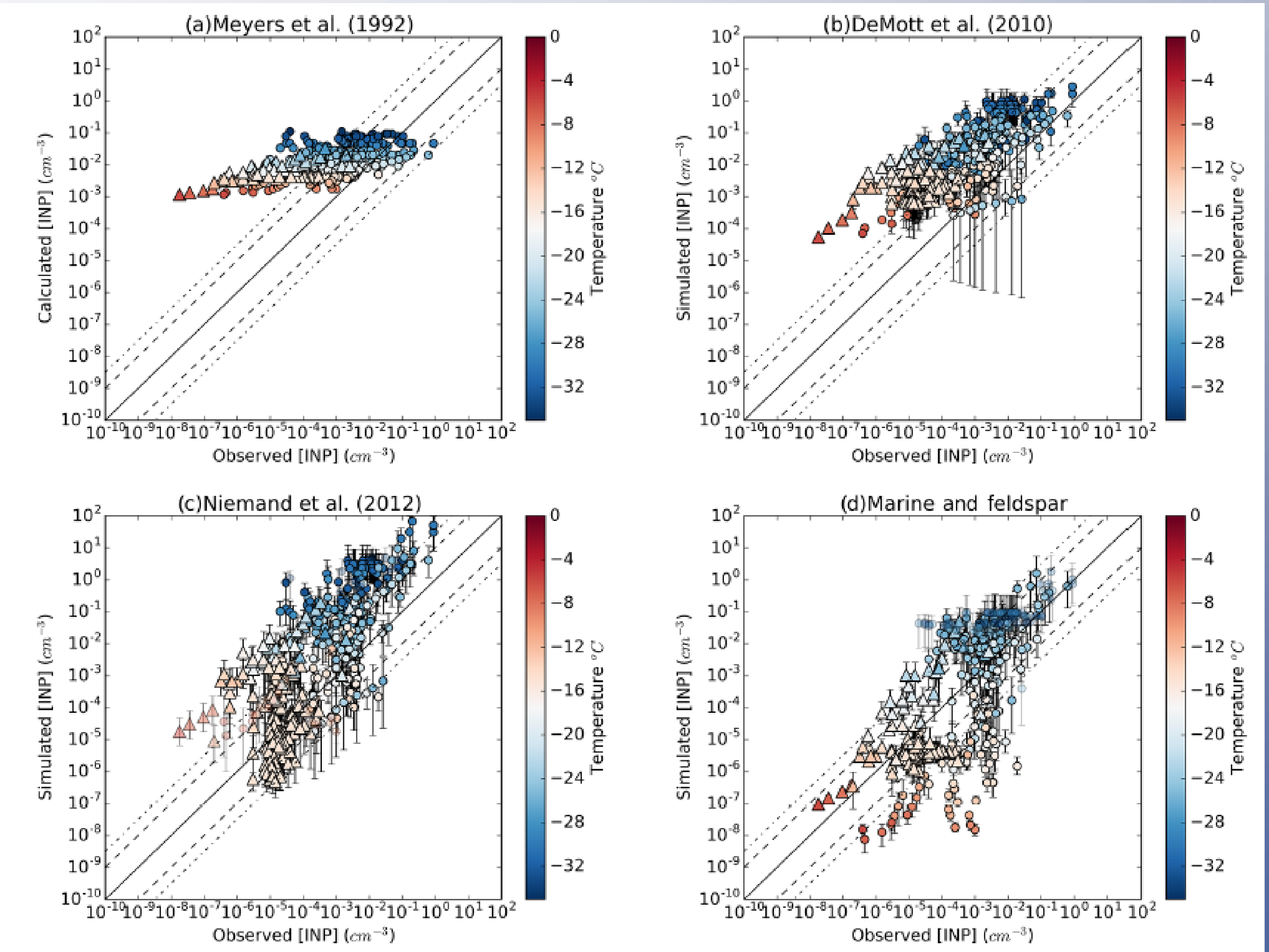


parameterizations for N_{INP}

Wilson et al., Nature, (2015)

developed further: Vergara-Temprado et al., ACP (2017)

-> discrepancies possibly due to missing contributions from terrestrial biosphere



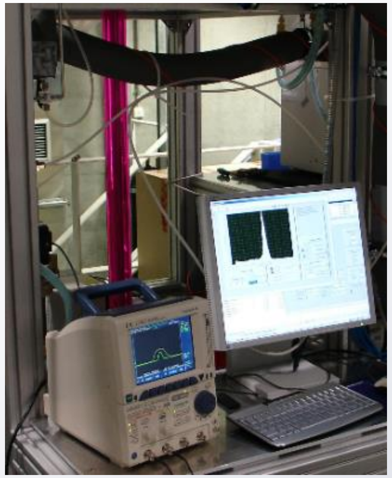
In the temperature range between 0 and -38°C, ice nucleation in atmospheric cloud droplets has to be aided by a catalyst, which is provided by one kind of atmospheric aerosol particles, INP (ice nucleating particles). Therefore, INP are important for mixed phase clouds, but also for some ice formation in cirrus clouds. And the ice formation, in turn, is important for cloud radiative effects, precipitation formation and cloud lifetime.

INP comprise different types of particles, more specifically biological and mineral dust particles. They are very rare in general, but still occur in vastly varying concentrations, depending on factors as location on Earth, season and temperature. Intensive laboratory research on INP was done in the past decade, while now the focus has shifted at understanding atmospheric INP based on atmospheric measurements. This presentation will provide some basic understanding of ice nucleation and measurements principles, will summarize the main findings from laboratory studies and then give an overview of the newest understanding gained from atmospheric measurements in the past years. Due to the vastness of the topic, it is thought as spark that may kindle curiosity and further own research in this important topic.

measuring INP in-situ

laminar flow tube

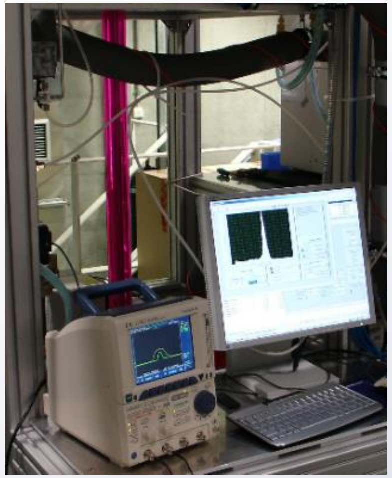
LACIS



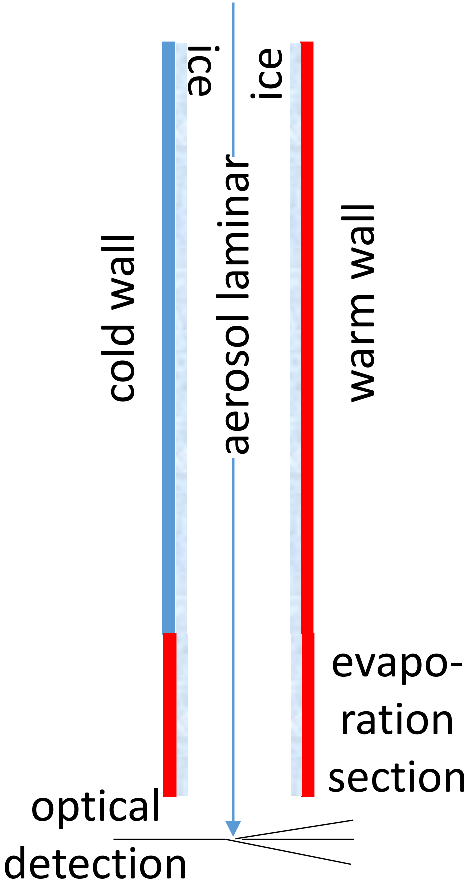
measuring INP in-situ

laminar flow tube

LACIS



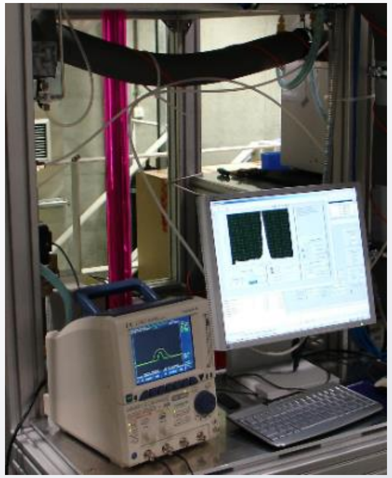
continuous flow diffusion chambers



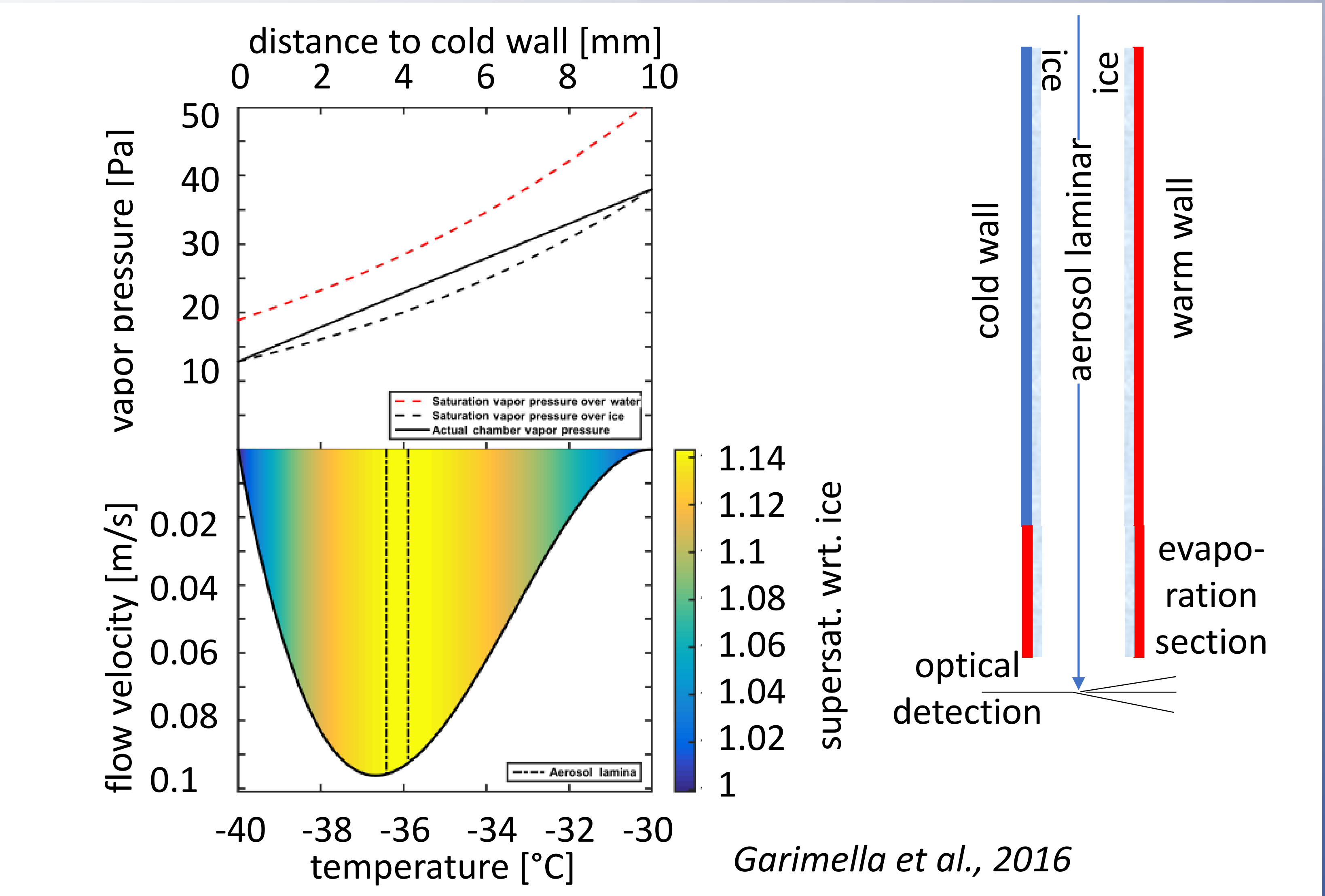
measuring INP in-situ

<u>laminar flow tube</u>

LACIS

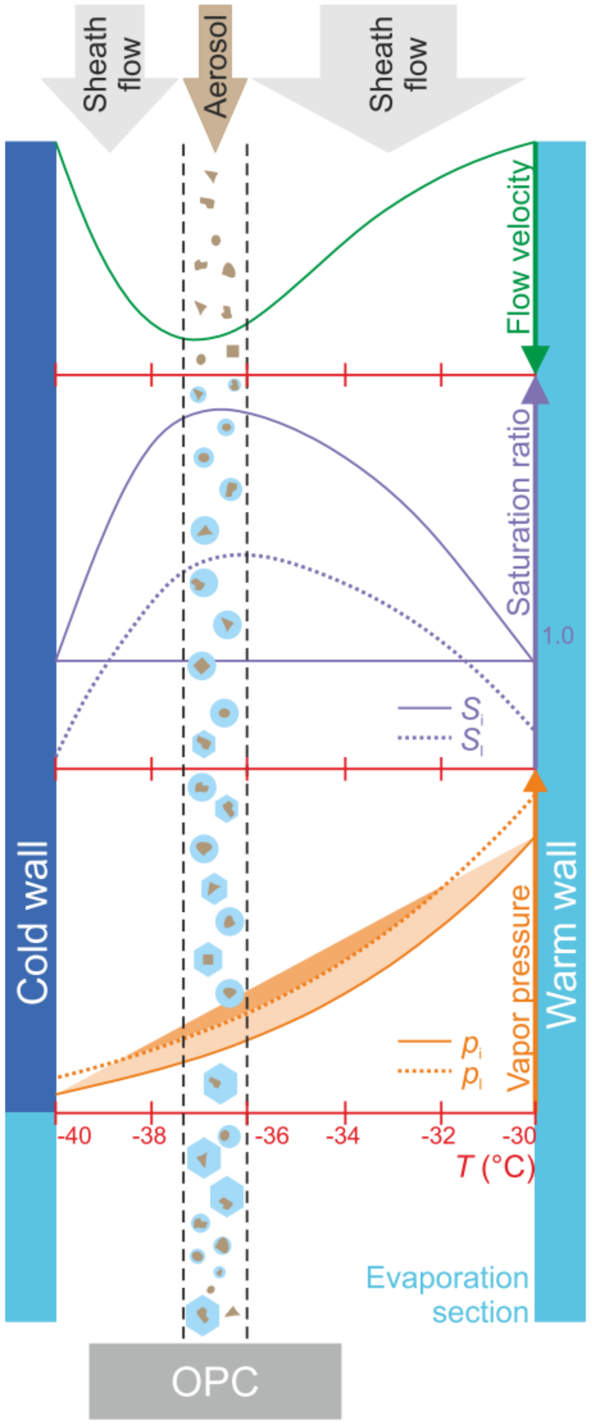


<u>continuous flow diffusion chambers</u>



Garimella et al., 2016

TROPOS

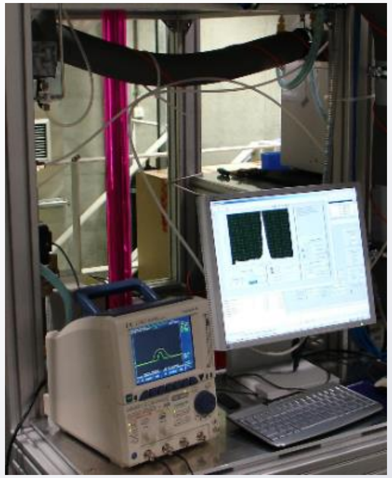


courtesy of Sarah Grawe, adapted from Stetzer et al. (2008)

measuring INP in-situ

laminar flow tube

LACIS



continuous flow diffusion chambers

CFDC, PINC, SPIN, HINC, INCA, …

measuring INP in-situ

laminar flow tube

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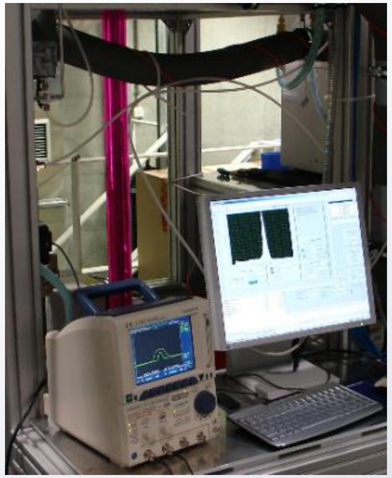

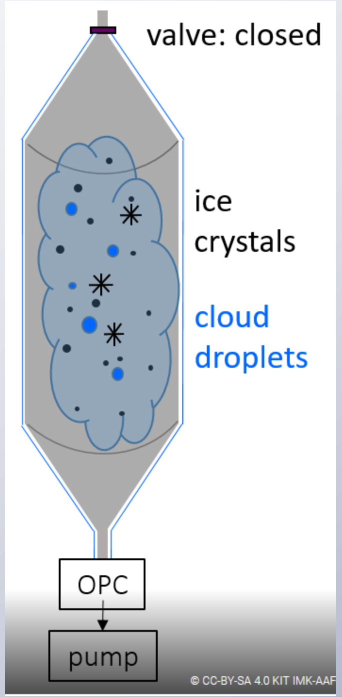

expansion chambers

continuous flow diffusion chambers

CFDC, PINC, SPIN, HINC, INCA, …