Biological ice nucleating particles are dominant and enigmatic over arable regions

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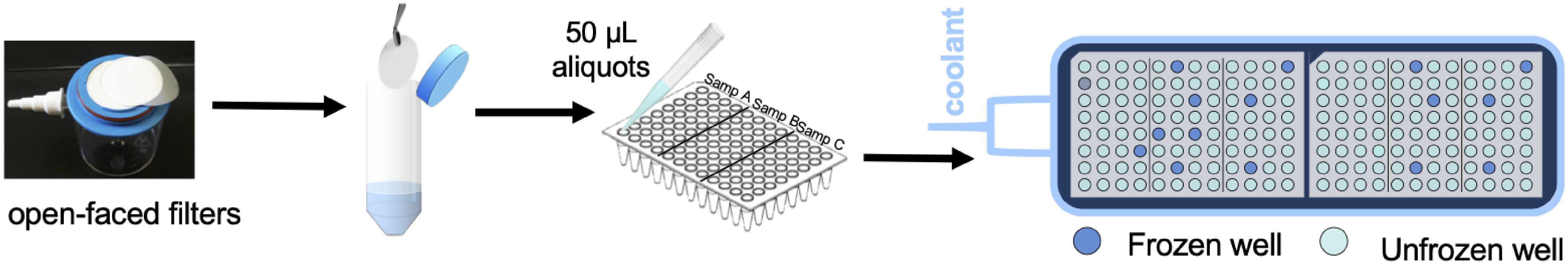
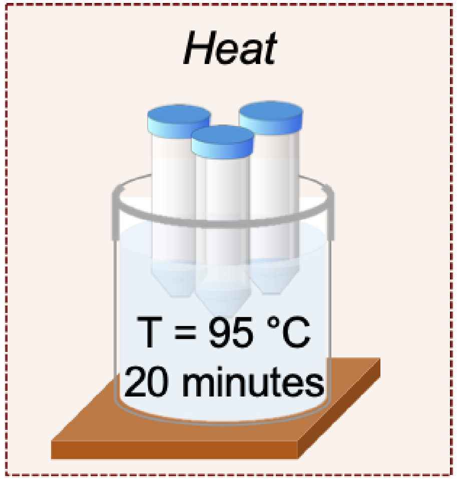
Ways to interrogate the composition of ice nucleating particles

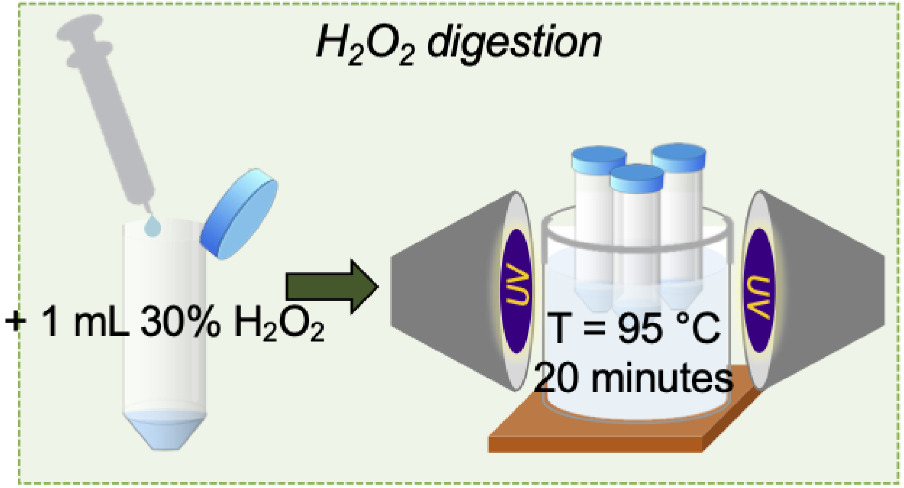
- **Classical immersion freezing studies with bulk ”treatments” to infer compositions – this is the current DOE ARM mentor method**
- Single particle activation
  - Environmental scanning electron microscopy for activation of INPs (e.g., as practiced at PNNL)
  - Continuous flow or expansion chambers using aerodynamic extraction methods on outflow to sample INPs on substrates or in real-time for mass spectrometry
  - Pre-filtering continuous flow chambers with heat, BC removal, etc.

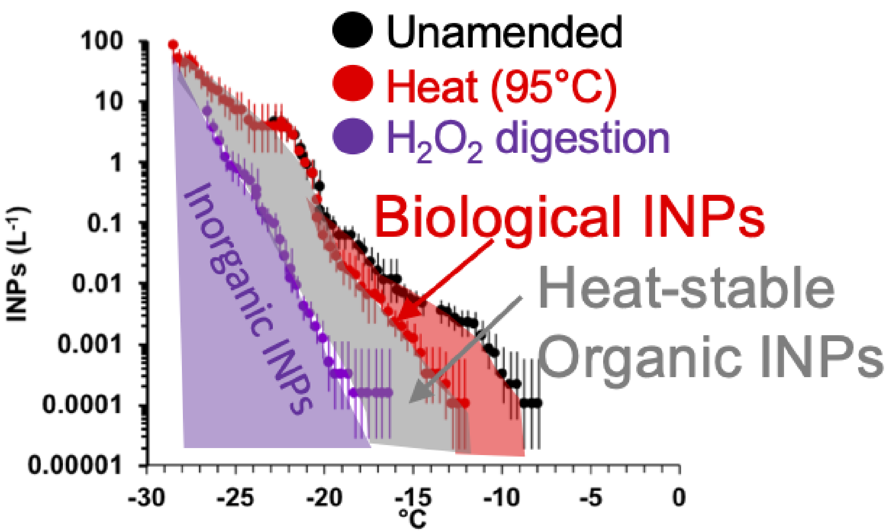
Compositional inferences from offline immersion freezing INP measurement techniques: Here the DOE mentor ice spectrometer (IS)



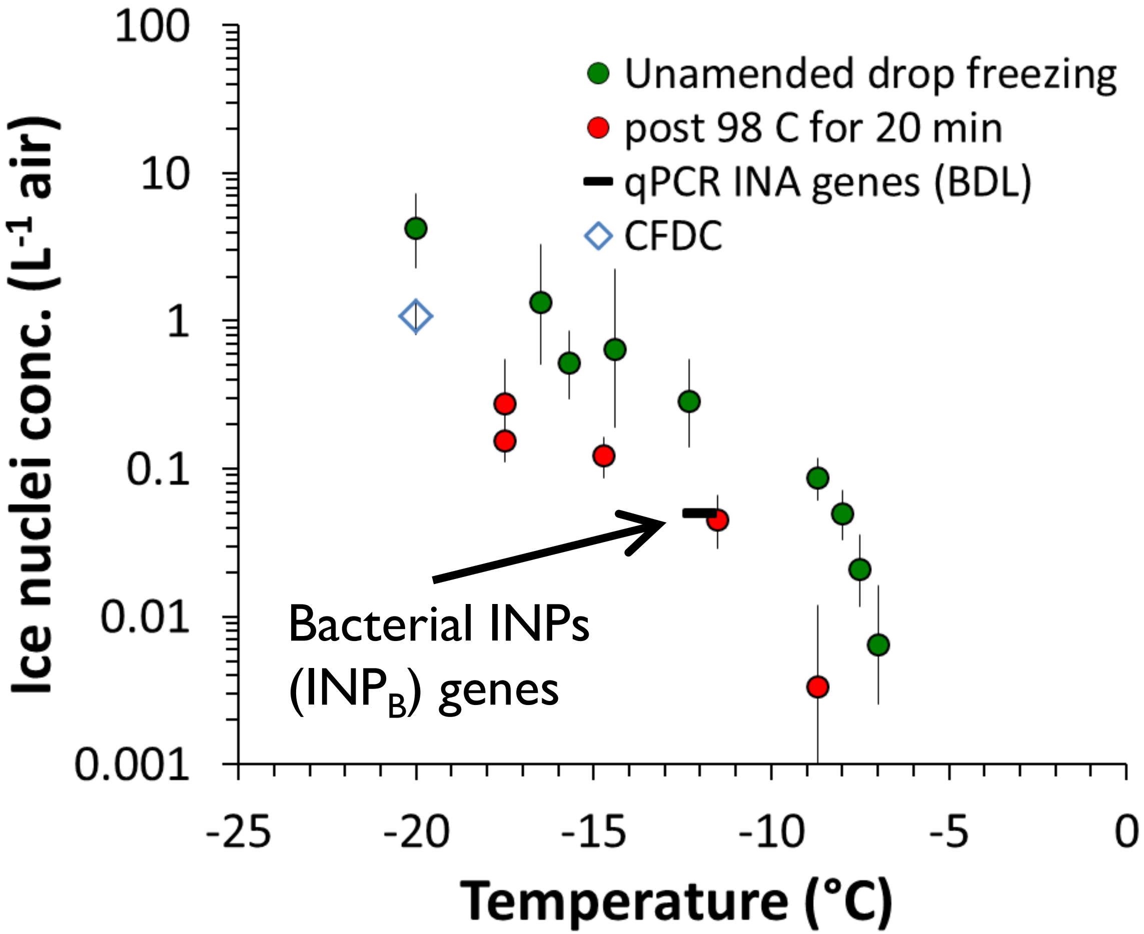
Offline treatments:







Unidentified organic (heat labile→proteinaceous) INPs are present in boundary layer air nearly always over arable regions

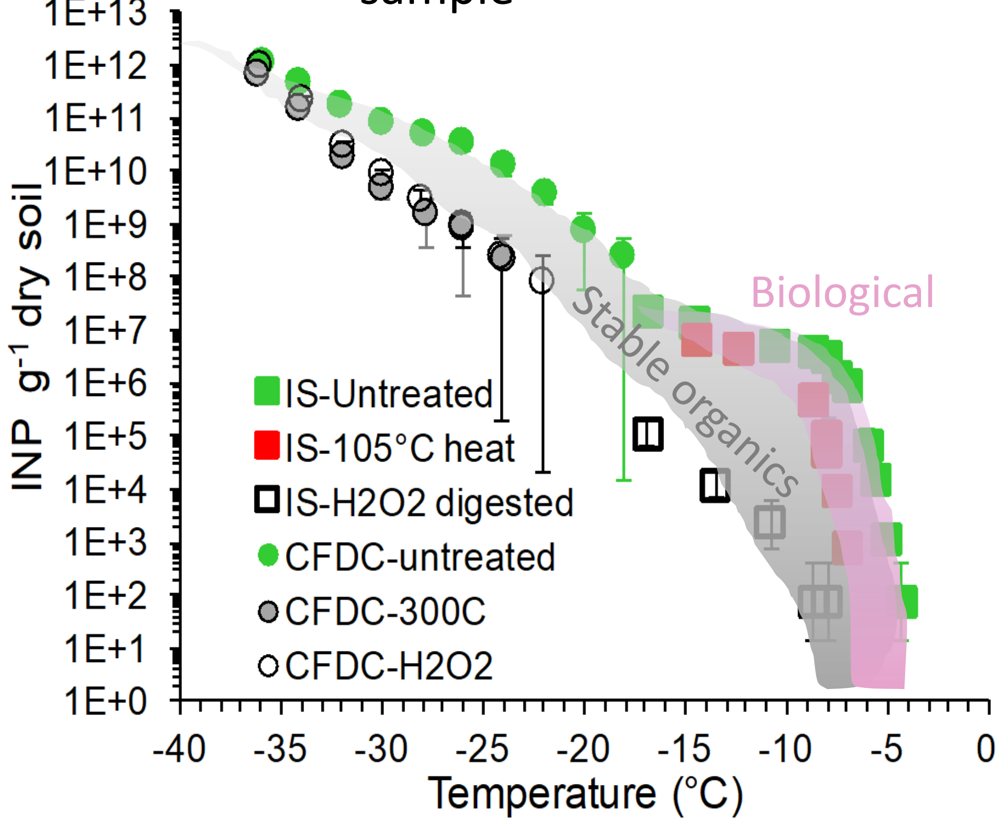


Wheat field stubble

Garcia et al., 2012: Biogenic ice nuclei in boundary layer air over two U.S. High Plains agricultural regions, J. Geophys. Res.117, D18209, doi:10.1029/2012JD018343.)

From soils or from plants?

Arable soils contain more active organic INPs compared to minerals, but heat labile contributions are only a special component



Stable soil organics are not humic and fulvic acids (Hill et al., 2016). They appear instead to be from the humin SOM fraction, (macromolecules, aggregates of peptides, aliphatics, peptidoglycan, carbohydrates, and lignin, microscopic fragments of plant roots, fungi and bacteria in various stages of decay).

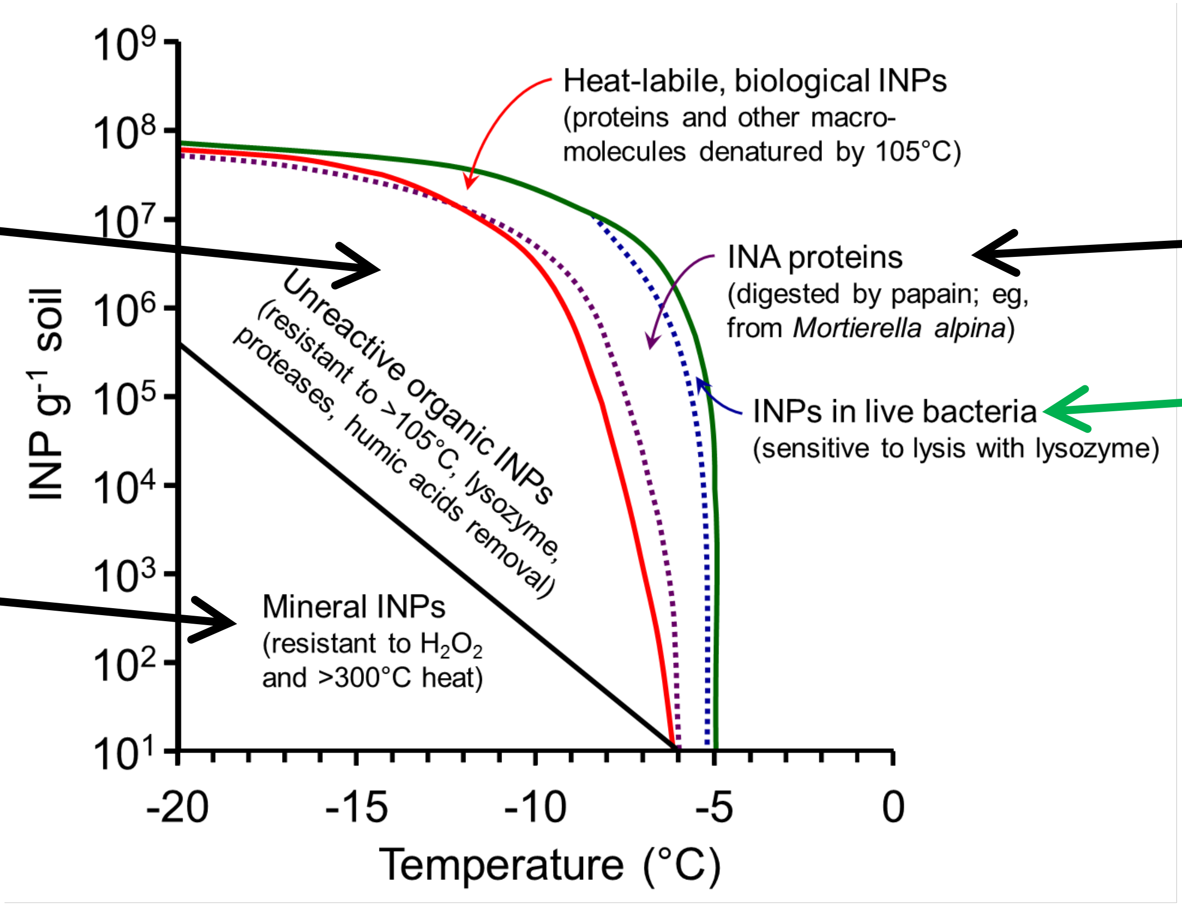
Adapted from:

Hill, T. C. J. et al., 2016: Sources of organic ice nucleating particles in soils, Atmos. Chem. Phys., 16, 7195–7211, doi:10.5194/acp-2016-1.
Tobo, Y., et al., 2014: Organic matter matters for ice nuclei of agricultural soil origin. Atmos. Chem. Phys., 14, 8521–8531.

Arable soils summary: Non-minerals are INPs, but perhaps not the primary source for biological INPs in the air

Other soil organic matter

Minerals



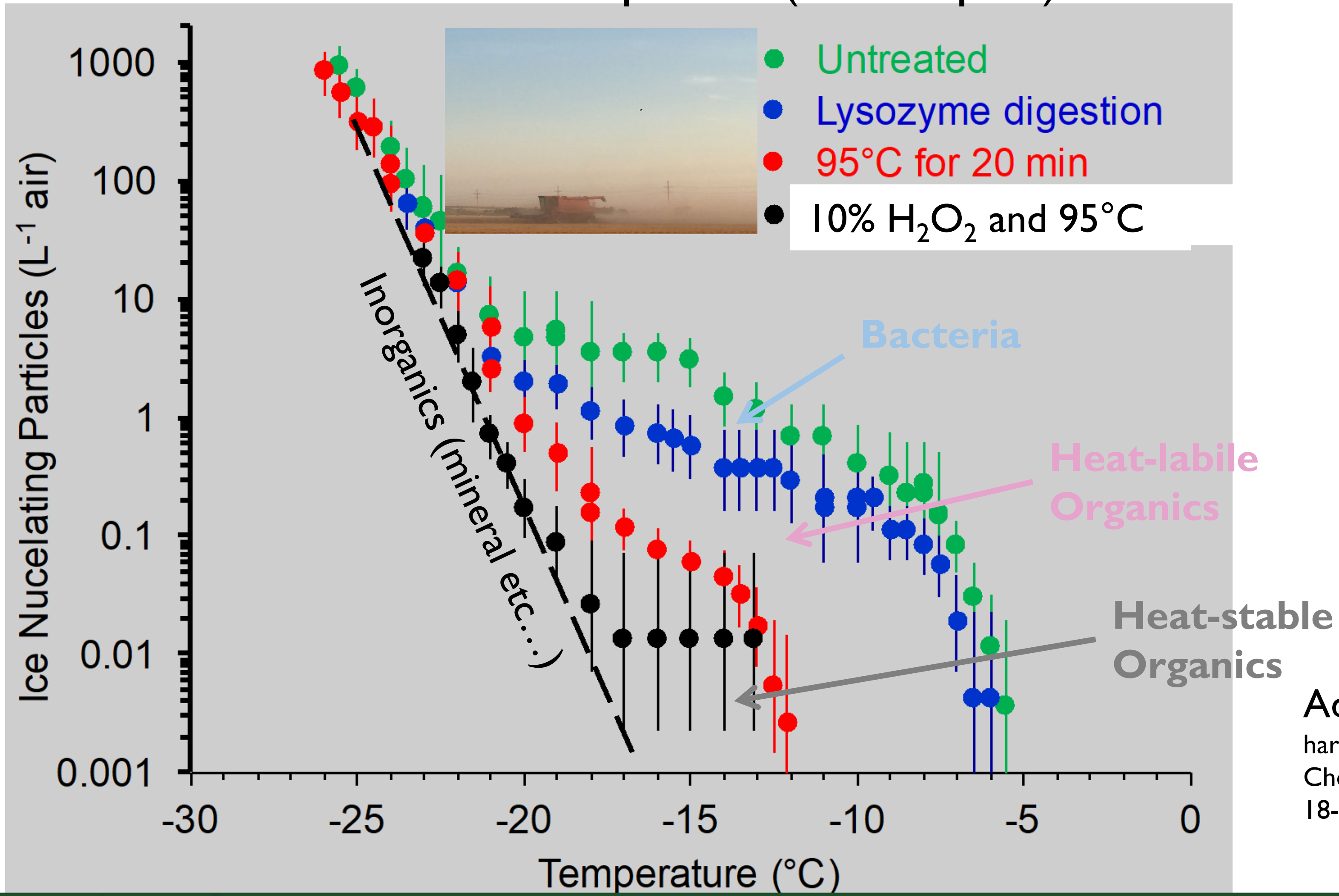
Fungi and free proteins

Bacteria

Adapted from: Hill, T. C. J. et al., 2016: Sources of organic ice nucleating particles in soils, Atmos. Chem. Phys., 16, 7195–7211, doi:10.5194/acp-2016-1.

Role of plant microbes and plant matter emissions to heat-labile organic INPs in the Central U.S.

Kansas wheat harvest period (air samples)



Adapted from: Suski, K. J. et al., 2018: Agricultural harvesting emissions of ice-nucleating particles, Atmos. Chem. Phys., 18, 13755-13771, https://doi.org/10.5194/acp-18-13755-2018.

Use for seasonal classification of all INP types using immersion freezing treatments in DOE-supported studies (CACTI; Testa et al., 2021)

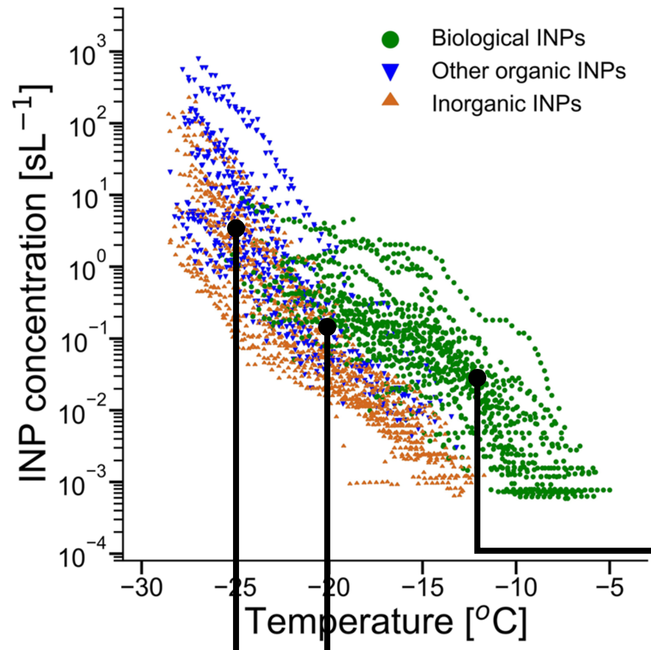


Figure 8. Ice nucleation activation temperature spectra of processed INP samples. Spectra of computed biological, other (stable) organic and inorganic INP contributions were derived from statistically significant differences between the untreated and heated spectra, significant differences between the heat-treated and the H_2O_2-digested spectra, and the residual activity remaining after H_2O_2 oxidation, respectively.

The relatively minor contributions of inorganic (mineral) INPs ("soil dust") to the total population is somewhat surprising for a mid-continental, rural site.

[From *Testa et al.*, 2021]

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At -25 ˚C, the variations in inorganic INPs can be most reliably assessed; they are relatively minor contributors at warmer temperatures

At and below -20 ˚C, stable organic INPs are major contributor to the population

At -12 ˚C, biological INPs clearly dominate the number

They remain important at -20 ˚C but number concentrations of stable organic INPs are becoming comparable

Biological INPs do not relate to other types or to size-selected aerosols in general (use of many studies)

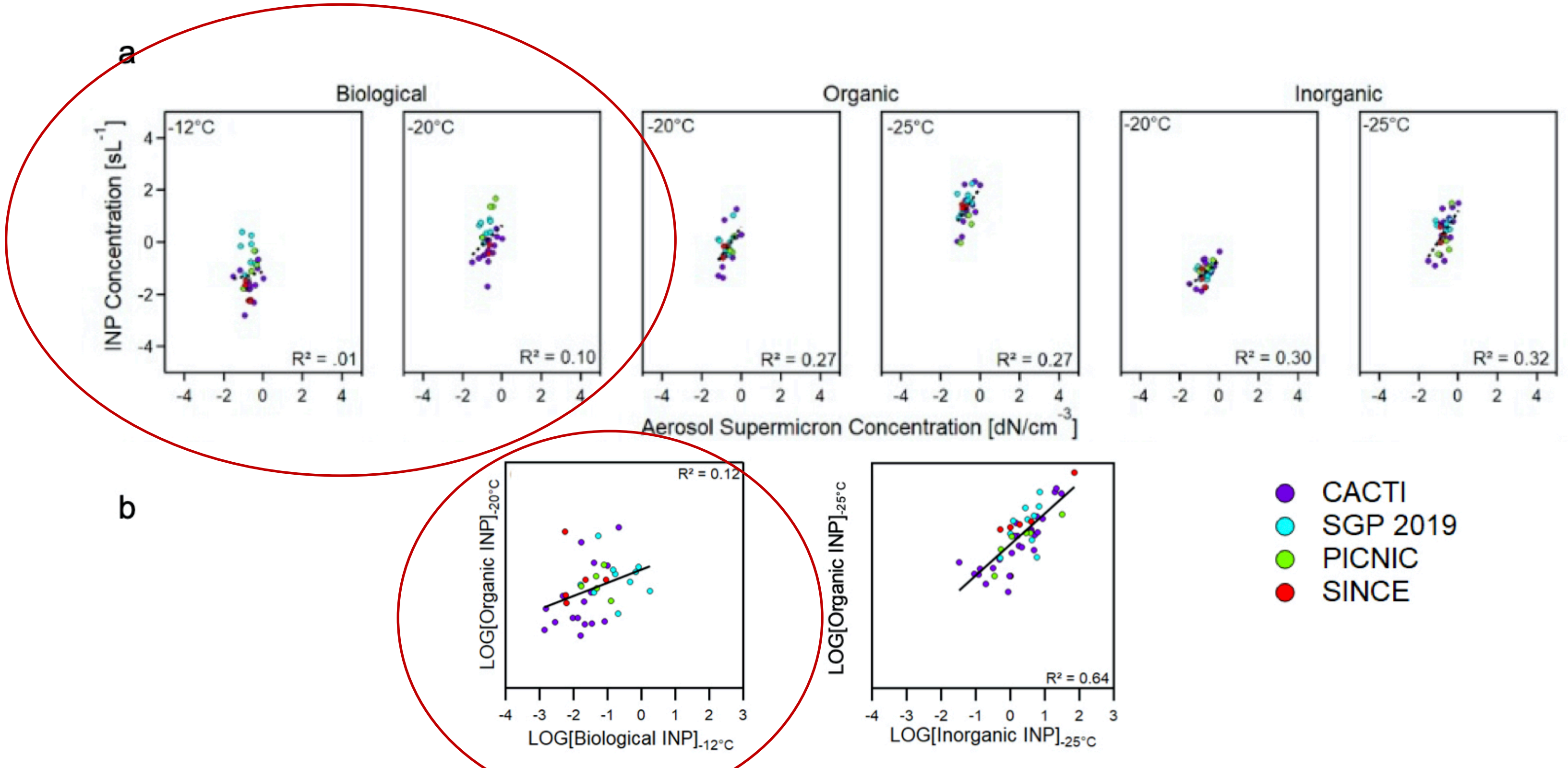
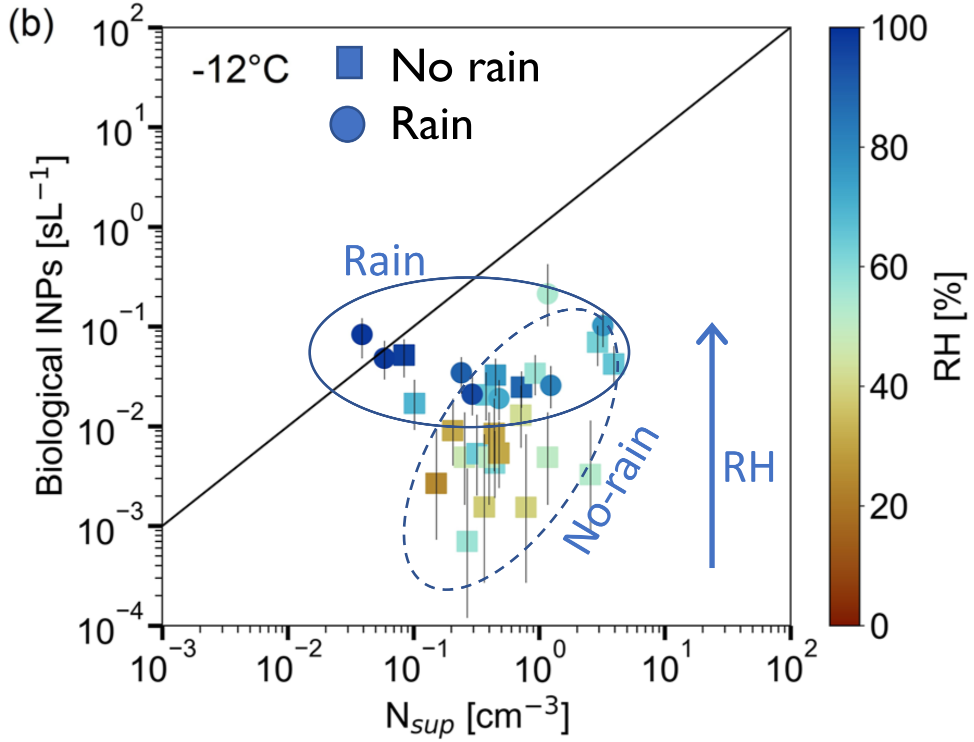


Figure 1. a) INP concentration versus supermicron aerosol concentration (cm^{-3}) for the three INP types characterized in four different regions, CACTI in the Argentinian Spring to Fall, the ARM SGP site in Fall (SGP 2019) and Spring (SINCE), and Puy de Dome, France in the Fall (PICNIC).Linear correlation coefficients are noted b) Relations between organic INPs versus biological INPs and inorganic INPs at the noted temperatures for each.

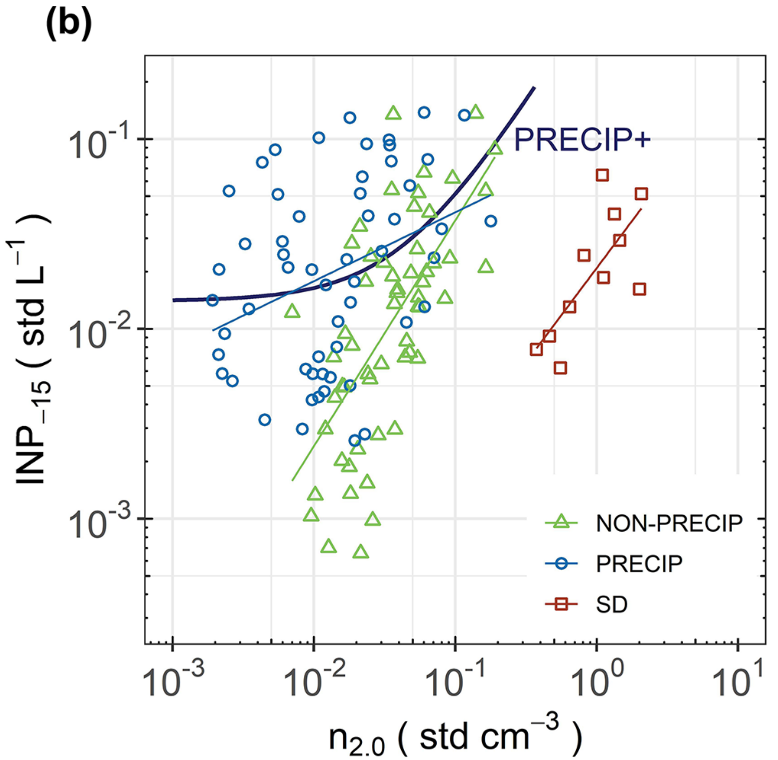
The most "efficient" bio-INPs are also affected by RH and rain, again suggesting a role of plant sources in addition to soil sources

Argentina (CACTI)



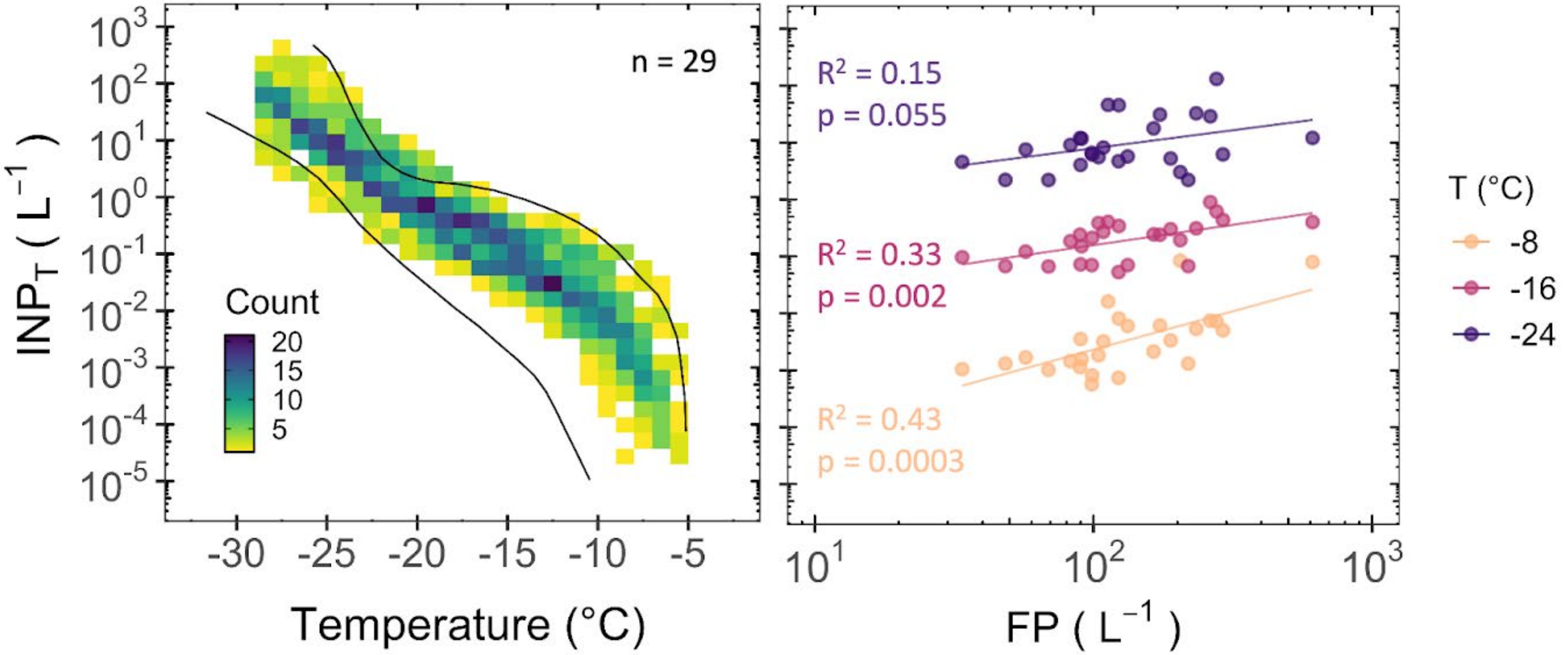
Testa, B., et al., 2021: Ice nucleating particles in the boundary layer of the Sierras de Córdoba, Argentina, during the Cloud, Aerosol, and Complex Terrain Interactions experiment, Journal of Geophysical Research: Atmospheres 126, e2021JD03518, https://doi.org/10.1029/2021JD035186.

Switzerland (late winter – assumed bio-INPs, not "typed")



Mignani, C.et al., 2021: Towards parameterising atmospheric concentrations of ice-nucleating particles active at moderate supercooling, Atmos. Chem. Phys., 21, 657–664, https://doi.org/10.5194/acp-21-657-2021, 2021.

Bio-INPs apparent in springtime relations with real-time bioaerosols (WIBS) in High Plains grassland environments



Unpublished data under support of the NSF Biology Integration Institute Regional One Health Aerobiome Discovery Network (BROADN.colostate.edu); courtesy of Claudia Mignani

What can we do better to constrain these phenomena

- Assure supermicron aerosol measurements (number, composition)
- Assure collections that address particle typing
- Ways to do single particle INPs in real-time? Pumped CVI works, but to capture higher temperature INPs requires extreme concentration of ambient aerosols.
- Deployment of real-time (many new ones recently) and offline bioaerosol collections in all studies - feasible for networks?
- Need to understand emission mechanisms at species level, via connecting metagenomics and INP measurements (opportunity to connect with JGI?)

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