



Towards the development of a baseline in ground-based **ice-nucleating particle (INP)** properties at three fixed ARM sites

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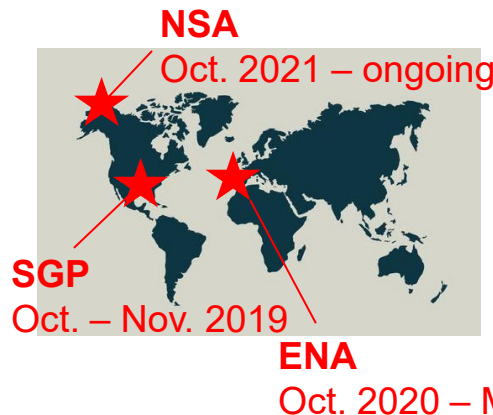


Project Goals

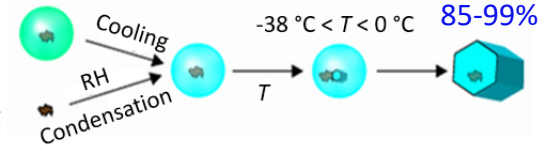


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- **Goal 1:** Adapting new INP measurement techniques for long-term ambient INP concentration (n_{INP}) monitoring at T above -33 °C at each Atmospheric Radiation Measurement (ARM) site,
- **Goal 2:** Elucidating sources, chemical composition, and ice nucleation pathways of aerosol particles at the ARM sites,
- **Goal 3:** Developing ice nucleation parameterizations that are useful for n_{INP} prediction and representative of “mixed-phase” clouds at the ARM sites.



Immersion Freezing



Condensation Freezing

Deposition Nucleation



= Ice-nucleating particle (INP)
e.g., dust, organic, and biogenic particles

'Potential' Climatic Impact of INPs

- INPs are quantitatively small but possess the substantial potential to impact climate.

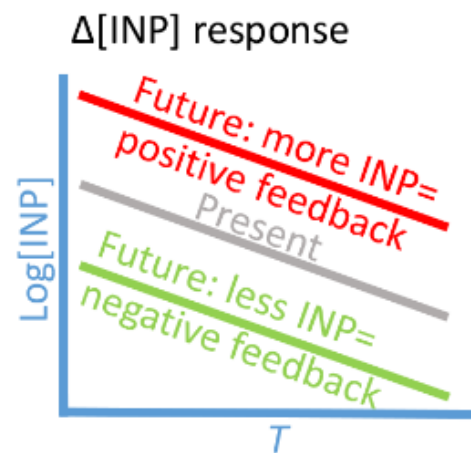
“...INPs are generally not included in CMIP6 models.”

“...previous studies have produced model estimates of opposing signs.”

- Model prediction of effective radiative forcing from aerosol-cloud interactions = $-1.0 \pm 0.8 \text{ W m}^{-2}$ → **notorious uncertainty**.

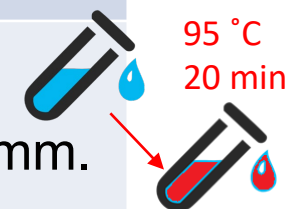
[IPCC, 2021: Ch. 7 40-41 pp. & Table 7.6]

- 1) Snow and ice coverage decreases, leading to greater INPs emissions.
- 2) Ambient INP concentration may increase in the future in response to warming.
- 3) INPs are cloud-destroying agents in mixed-phase clouds → back to (1)



Methods Summary

Technique	PINE-3* (Online)	WT-CRAFT** (Offline)
Instrument Type	Mobile Expansion Cooling Chamber	Cold Stage Freezing Assay
Ice Nucleation Mode	Immersion + Condensation + Deposition	Immersion + Heat-treated imm.
Measurable T Range	-15 to -33 °C (± 1.0 °C)	0 to -25 °C (± 0.5 °C)



*Calibrated at AIDA, **Calibrated by and co-assessed with INSEKT



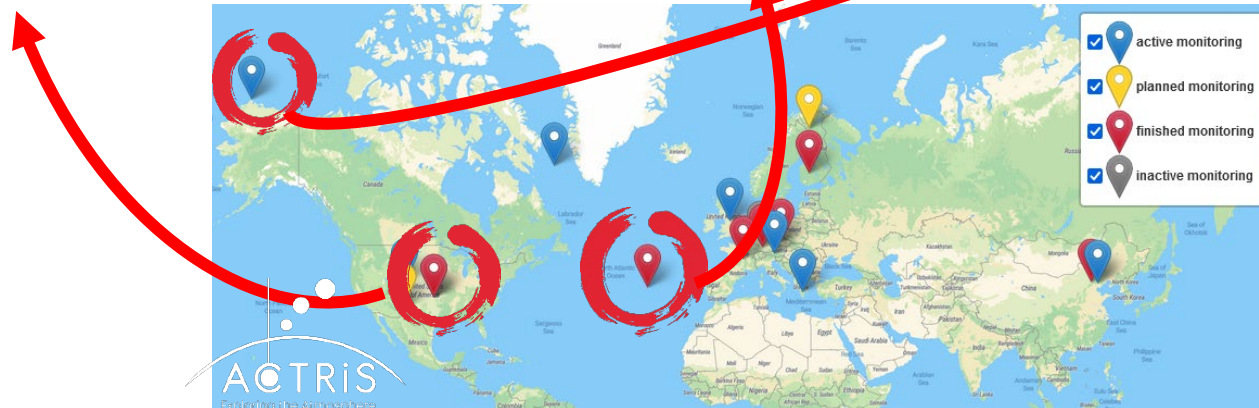
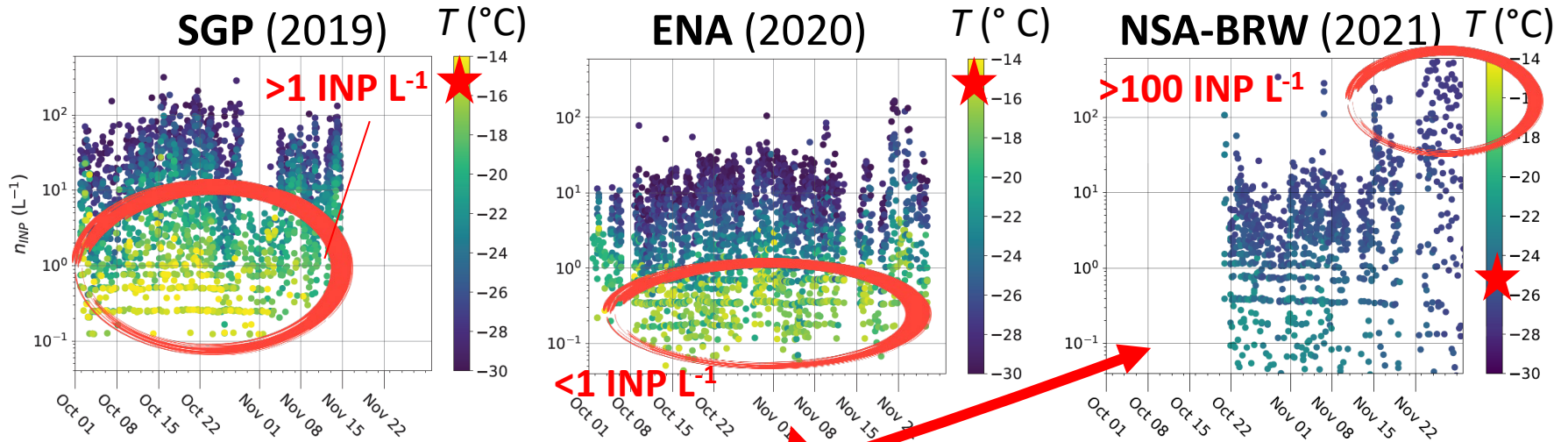
Measurements Summary

- Total aerosol concentration, $n_{\text{INP}}(T)$, and $n_{\text{CCN}}(\text{SS}\%)$ are higher @ SGP than other ARM sites.

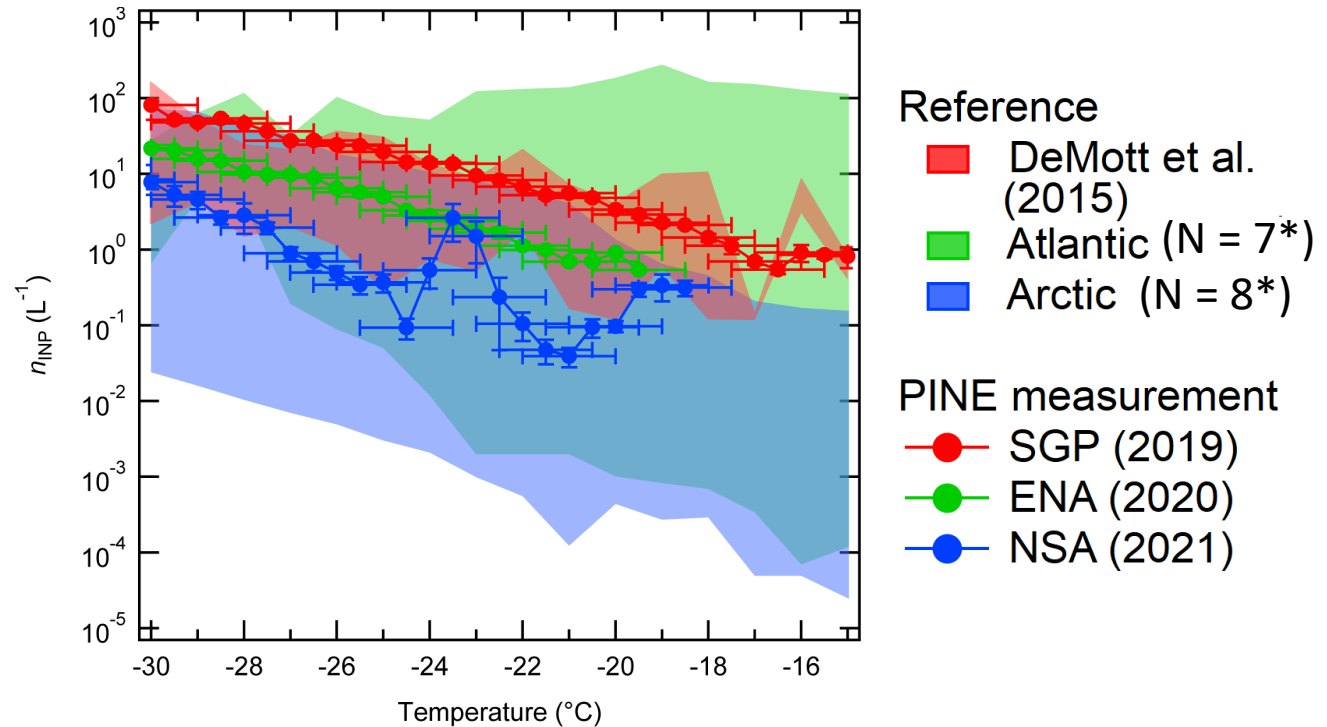
		SGP	ENA	NSA-BRW
		Oct-Nov 2019	Oct-Nov 2020	Oct-Nov 2021
Total Aerosols, $n_{\text{aer}} [\text{cm}^{-3}]$		2896.7 ± 0.9	> 339.0 ± 1.4	> 96.4 ± 2.8
Online $n_{\text{INP}}(T)$ $\times 10^{-3} [\text{cm}^{-3}]$	-15 °C	0.8 ± 0.3	-	-
	-20 °C	3.4 ± 0.4	> 0.9 ± 0.0 ₄	> 0.1 ± 0.0 ₂
	-25 °C	19.6 ± 1.5	5.0 ± 0.3	0.4 ± 0.1
	-30 °C	81.4 ± 10.7	21.8 ± 0.8	7.8 ± 1.5
$n_{\text{CCN}}(\text{SS})$ $[\text{cm}^{-3}]$	0.1 %SS	161.6 ± 7.6	> 57.7 ± 3.9	-
	0.2 %SS	510.3 ± 25.1	108.5 ± 7.6	-

$n_{\text{INP}}(-20^\circ\text{C})/n_{\text{aer}} \approx 1/\text{million}$ $\approx 1/\text{million}$ $\approx 1/\text{million}$
 $n_{\text{CCN}}(0.1\%\text{SS})/n_{\text{aer}} \approx 1/30$ $\approx 1/5$

Online $n_{INP}(T)$ Time Series (6-hour time average)



Online $n_{\text{INP}}(T)$ Spectra (T -bin = 0.5 °C)

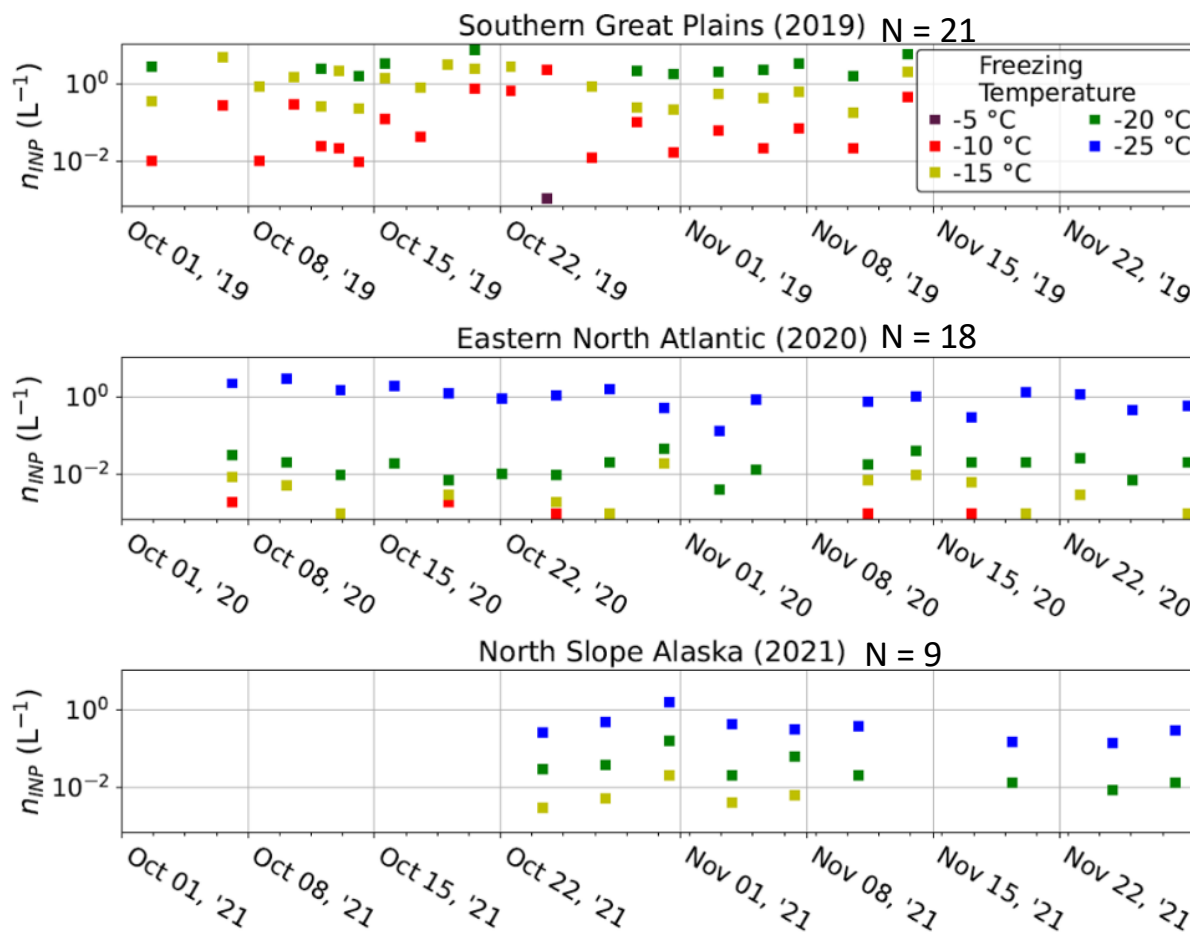


$n_{\text{INP,SGP}} > n_{\text{INP,ENA}} > n_{\text{INP,NSA}}$

NOTE: NSA High INP episodes during the Chukchi Sea storms

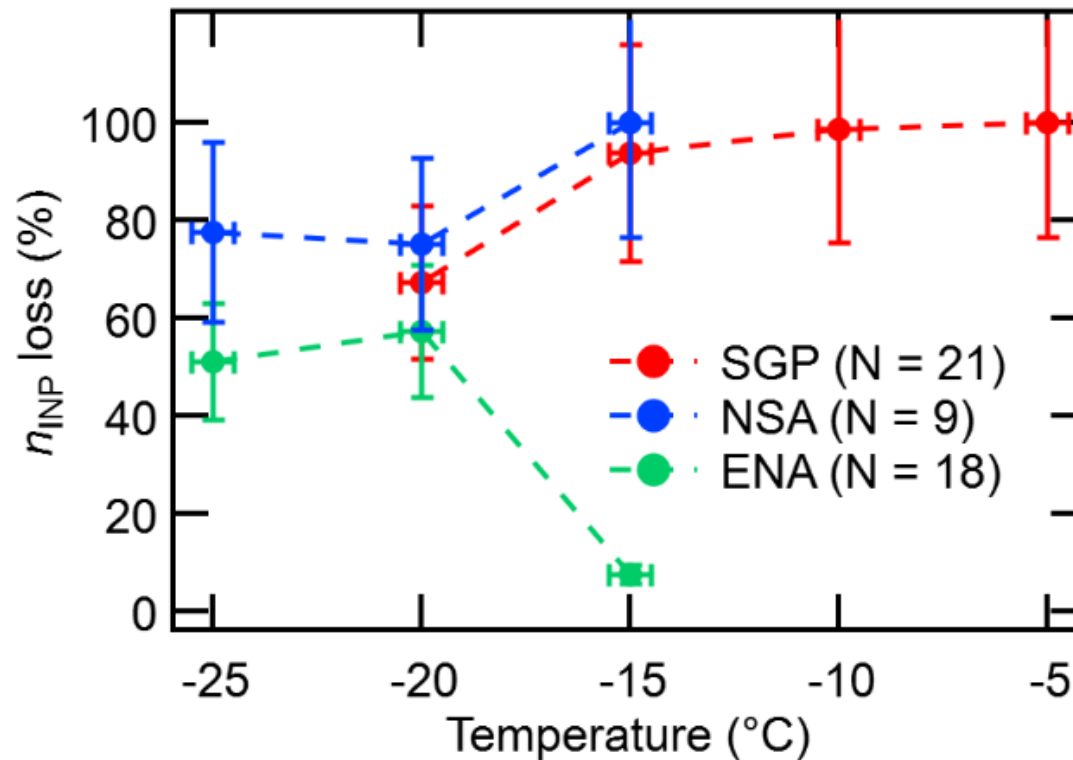
→ Breakout Session 5 [Eisenhower] @ 2 PM

Offline n_{INP} Time Series



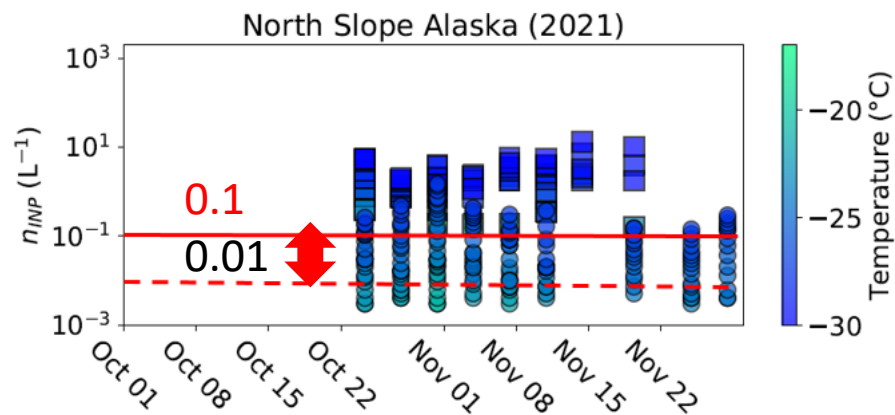
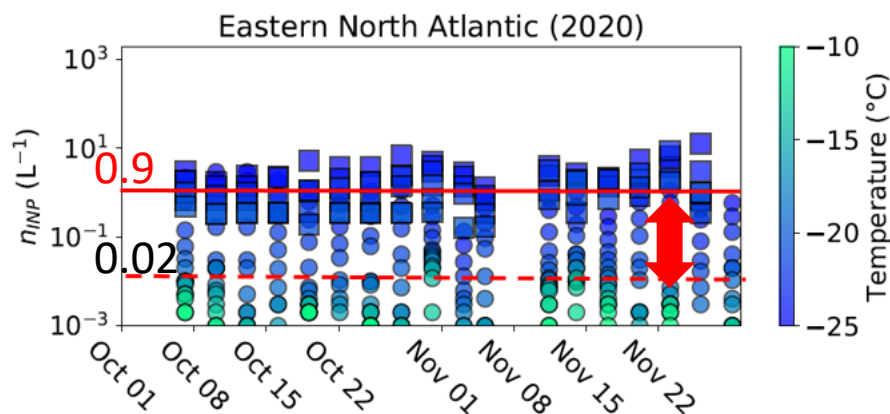
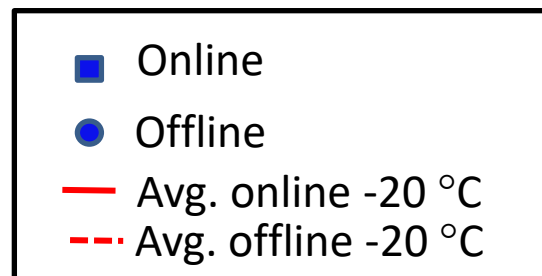
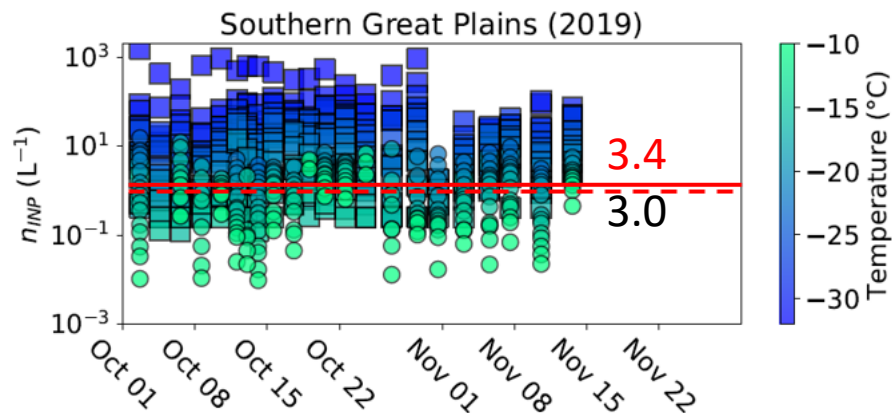
$$\square n_{\text{INP,SGP}} > n_{\text{INP,ENA}} > n_{\text{INP,NSA}}$$

Offline $n_{\text{INP}}(T)$ Heat Sensitivity



- ❑ SGP & NSA INPs are heat-sensitive (>67% loss across).
- ❑ Presence of biogenic INPs is seen at all ARM sites.

Online vs. Offline n_{INP} Comparison



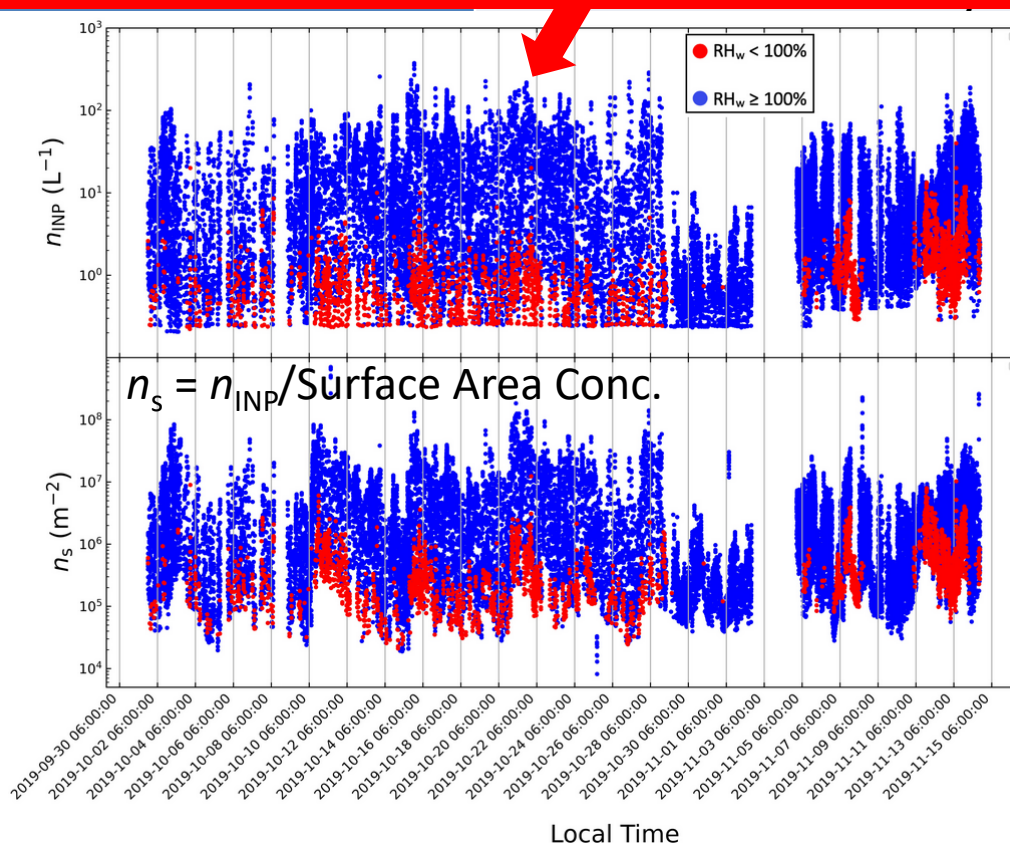
☐ Online – Offline = Condensation & Deposition \gg Immersion?*

INP Properties: SGP vs. ENA



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Site	SGP	ENA
Major IN mode	Immersion-dominant (<3% deposition)	Probable condensation freezing ($n_{INP} \propto n_{CCN}$)

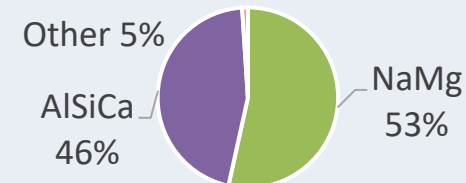


$$m_{ORG} = 0.94 \mu\text{g m}^{-3}$$

$$m_{Cl} = 0.14 \mu\text{g m}^{-3}$$

$$**m_{BC} = 0.90 \text{ ng m}^{-3}$$

Arctic air mass (54.8%)

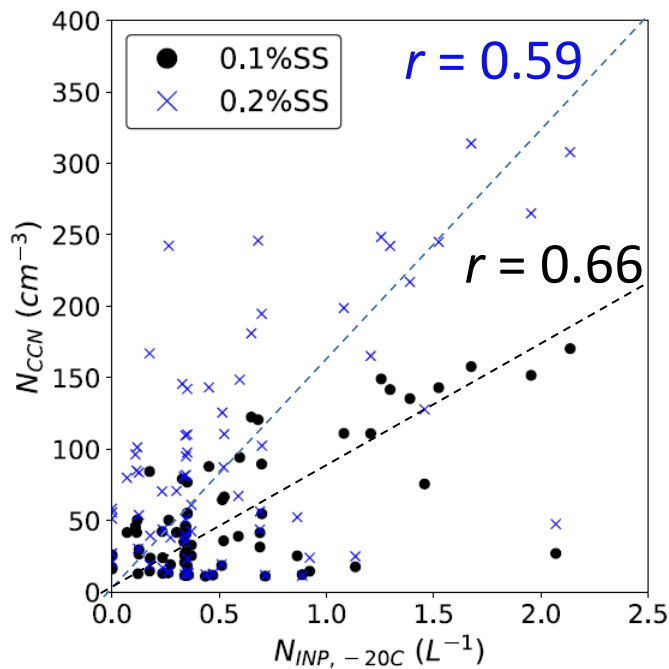


$$\text{Na/Cl} = 1.91\text{-}2.73$$

In-progress

Session 5 [Eisenhower] @ 2 PM

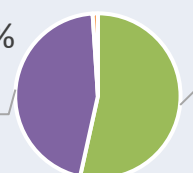
n_{INP} & n_{CCN} Correlation @ ENA



ENA Campaign			
Temperature (°C)	-30 °C	-25 °C	-20 °C
Supersaturation (%)			
0.1	-0.14	0.17	0.66
0.2	-0.06	0.14	0.59
SGP Campaign			
Temperature (°C)	-30 °C	-25 °C	-20 °C
Supersaturation (%)			
0.1	-0.03	0.05	0.001
0.2	0.12	0.001	0.06

- ❑ Probable marine contribution for both INP and CCN at ENA
- ❑ Predominance of condensation freezing at around -20 °C, explaining the gap between online and offline measurements?

INP Properties: SGP vs. ENA

Site	SGP	ENA
Major IN mode	Immersion-dominant (<3% deposition)	Probable condensation freezing ($n_{INP} \propto n_{CCN}$)
Aerosol Composition	$m_{ORG} = 1.26 \mu\text{g m}^{-3}$ $m_{Cl} = 0.02 \mu\text{g m}^{-3}$ $m_{BC} = 0.74 \text{ ng m}^{-3}$	$m_{ORG} = 0.94 \mu\text{g m}^{-3}$ $m_{Cl} = 0.14 \mu\text{g m}^{-3}$ ** $m_{BC} = 0.90 \text{ ng m}^{-3}$
Aerosol & INP Source	*Ag soil dust	Arctic air mass (54.8%)
Mixing State	In-progress (EMSL-LSR)	Other 5% AlSiCa 46%  NaMg 53% $\text{Na/Cl} = 1.91\text{-}2.73$ (N = 490)
IN Parameterization	n_s & *ABIFM	In-progress

NOTE: NSA Data → Breakout Session 5 [Eisenhower] @ 2 PM

Will further explore the mixing state and IN parameterization

Summary & Outlook



- ❑ We implemented PINE-3 & WT-CRAFT for short-term & long-term campaigns.
- ❑ Given the predominance of heat-sensitive biogenic particles at all ARM sites, **systematic measurements of biological aerosol particles** would be a great addition to the ARM program.
- ❑ **Co-located measurements of INP and CCN** might be meaningful for the INP community.
- ❑ We have many more things to look into – e.g., aerosol size distribution (esp. **supermicron range**), **particle chemical speciation** (esp. organics) etc. What is the priority for the modeling community?

ARM Archived Data

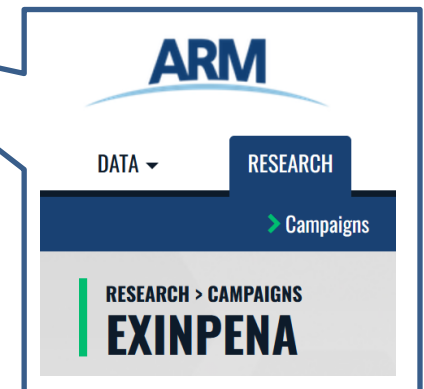
ExINP-SGP: <https://www.arm.gov/research/campaigns/sgp2019exinpsgp>

ExINP-SGP II: <https://www.arm.gov/research/campaigns/sgp2020exinpsgpii>

ExINP-ENA: <https://armweb0-stg.ornl.gov/research/campaigns/ena2020exinpena>

Publications

1. *Möhler, O.* et al.: Atmos. Meas. Tech., 14, 1143–1166, <https://doi.org/10.5194/amt-14-1143-2021>, 2021
2. *Vepuri, H. S. K.* et al.: Atmos. Chem. Phys., 21, 4503–4520, <https://doi.org/10.5194/acp-21-4503-2021>, 2021
3. *Hiranuma, N.* et al.: Atmos. Chem. Phys., 21, 14215–14234, <https://doi.org/10.5194/acp-21-14215-2021>, 2021
4. *Knopf, D. A.* et al.: Bull. Amer. Meteor., 102, E1952-E1971., 2021





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EMSL-LSR (60345) 2022 [PI: Knopf]

Supplemental Information: Data

SGP: Methods – Community Atmospheric Model 6 (CAM6), Community Earth System Model 2 (CESM2)

Runtime period: October to November 2019

Resolution: 0.9° x 1.25°, 56 vertical layers

Meteorology: Horizontal winds and

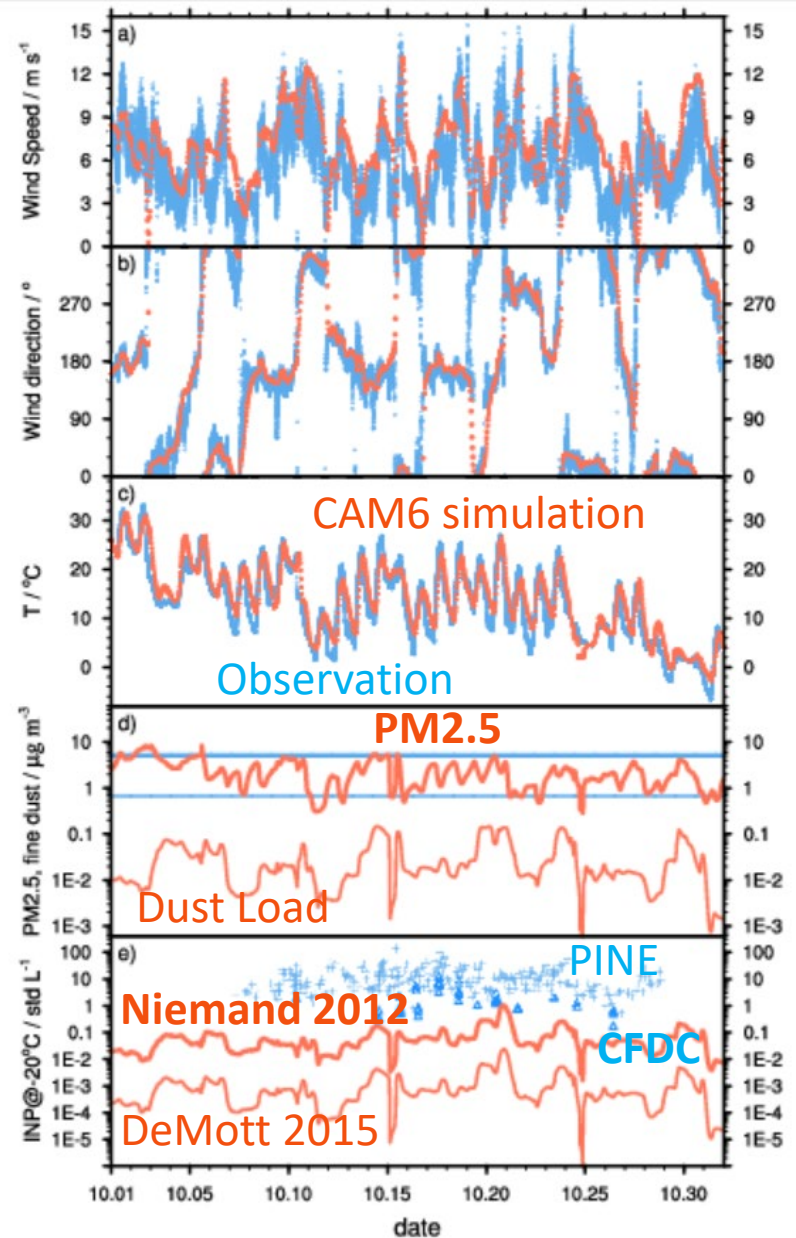
temperature nudged to MERRA2 data

Aerosol module: Four-mode version of Modal

Aerosol Module (MAM4; Liu et al., 2016)

Aerosol emissions: Shared Socioeconomic

Pathways (SSP) 2-4.5



These data are the courtesy of Xiaohong Liu and Yang Shi

ENA: Methods – Energy Exascale Earth System Model (E3SM)

Runtime period: October to November 2020

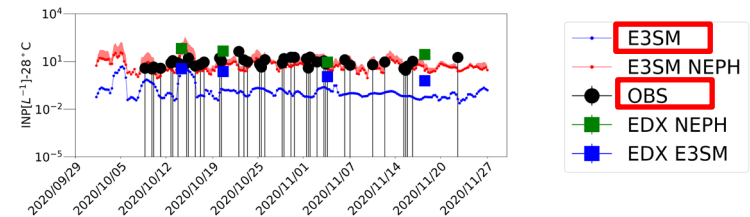
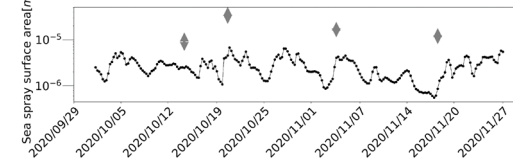
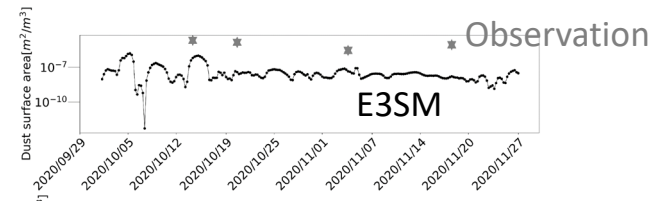
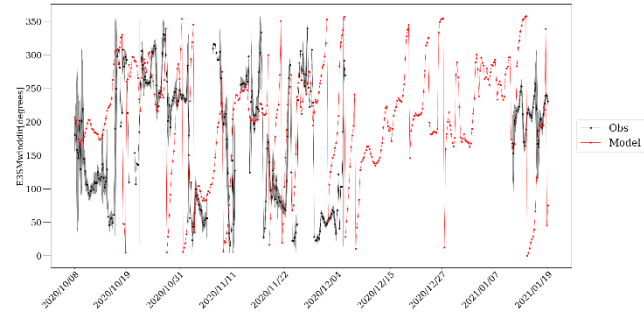
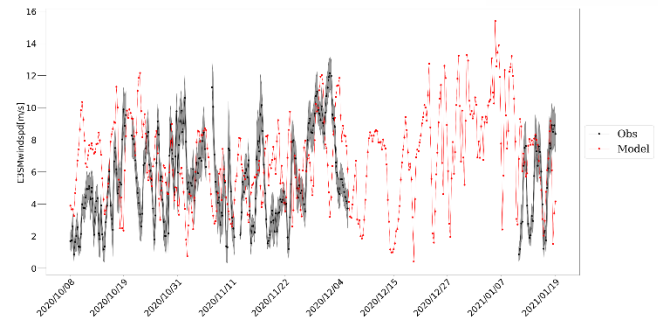
Resolution: 1°, 72 vertical layers

Meteorology: Horizontal winds and temperature nudged to MERRA2 data

Aerosol module: Four-mode version of Modal Aerosol Module (MAM4; Liu et al., 2016)

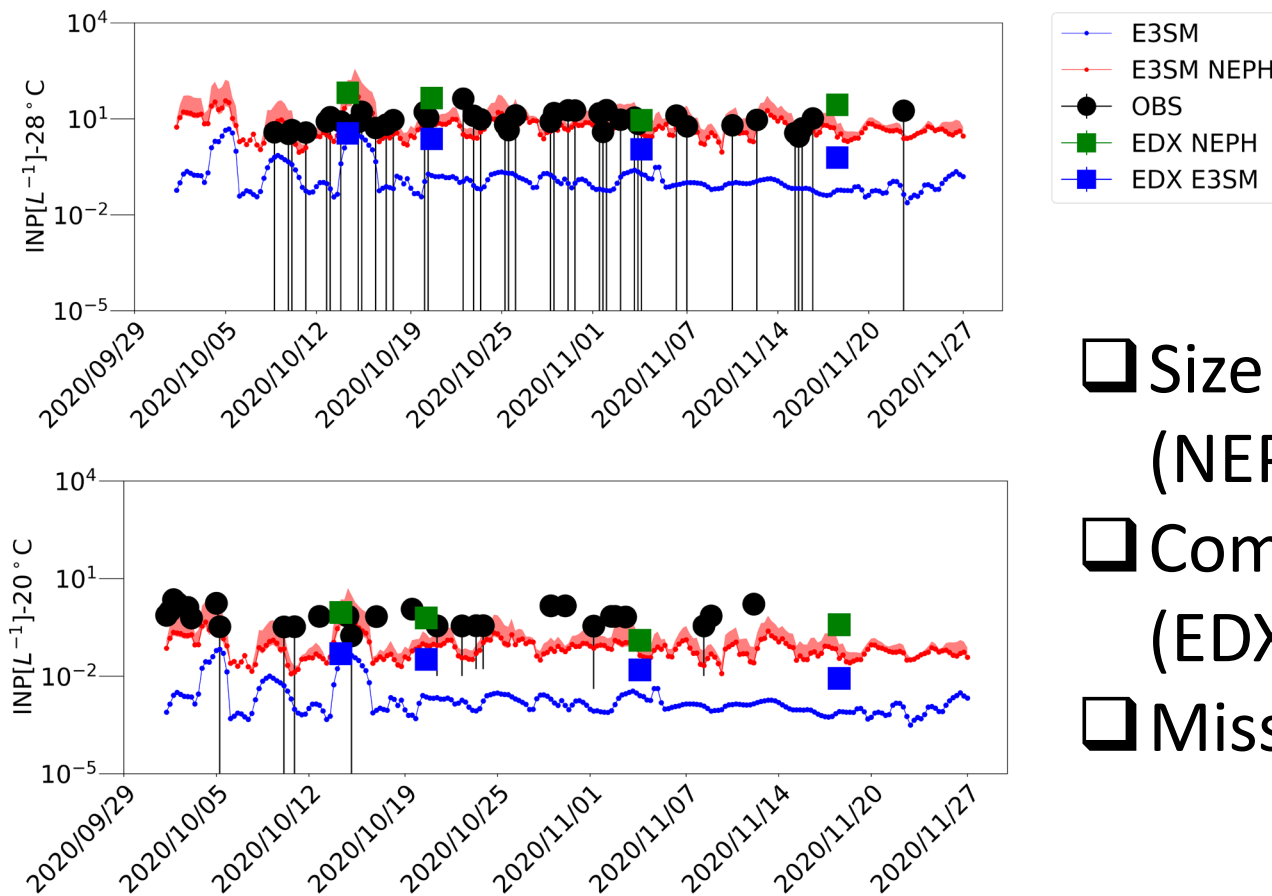
Sea salt emissions: OCEANFILMS

Dust emissions: Zender et al. (2003)



These data are the courtesy of Aish Raman and Susannah Burrows

ENA Closure Study



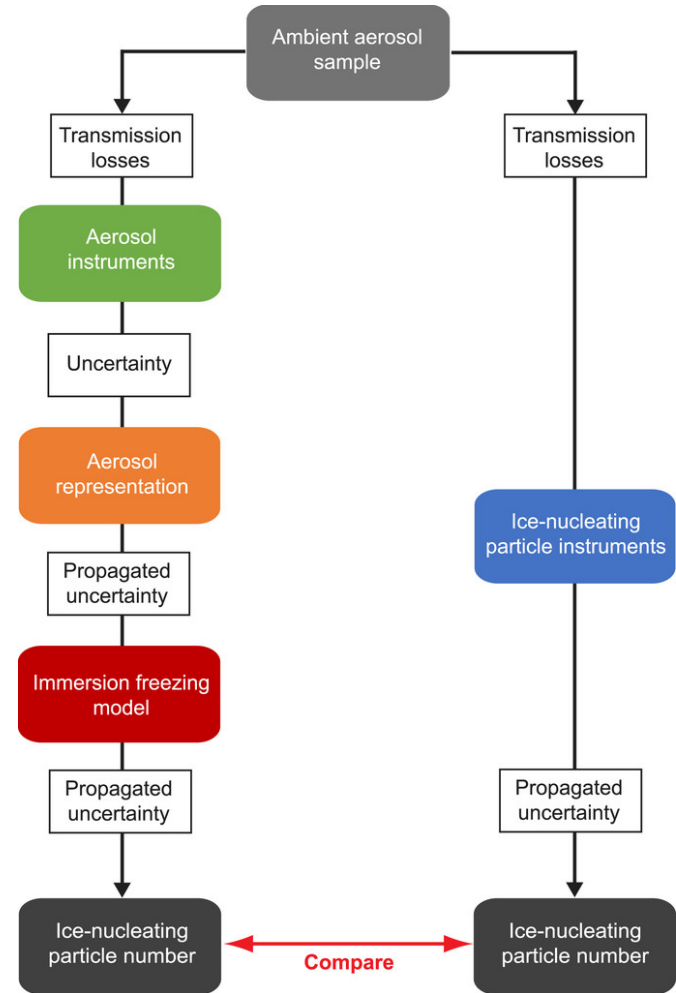
- Size Distribution (NEPH)?
- Composition (EDX)?
- Missing INPs?

The data in the courtesy of Aish Raman and Susannah Burrows

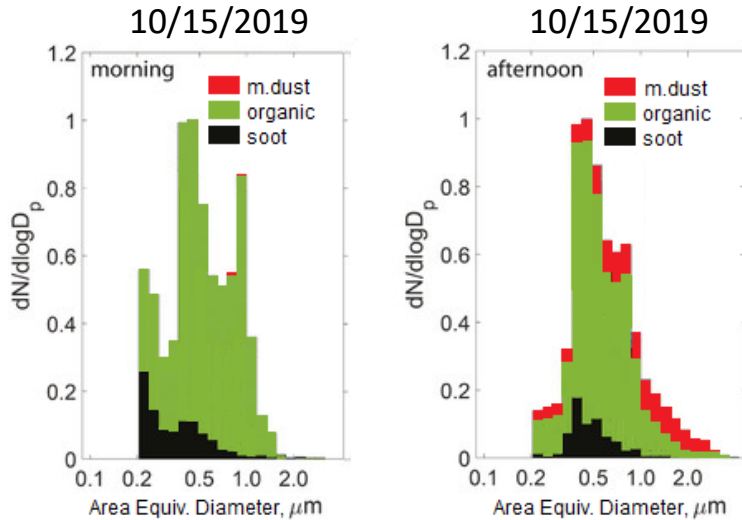
SGP Closure Study

Can we predict n_{ice} from...

- Aerosol size (i), surface area (S), and number conc. (n_{aer}) based on SMPS-APS
- Composition (mineral dust, organic, & soot) based on CCSEM-EDX
- n_s or J_{het} parameterization



SGP INAS parameterization



$$n_{ice,i} = n_{aer,i} (1 - e^{-S_i n_s(T)}) [L^{-1}]$$

Mineral Dust (N12) $-36\text{ }^\circ\text{C} < T < -12\text{ }^\circ\text{C}$

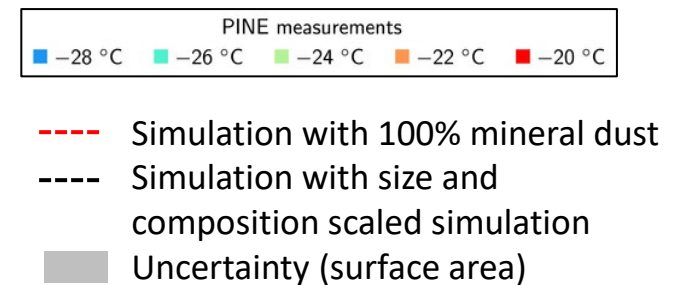
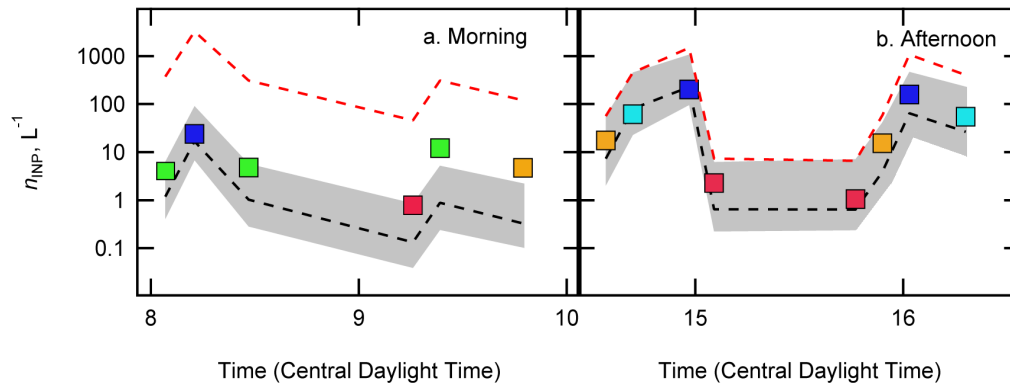
$$n_{s, \text{desert dust}}(T) = \exp[-0.517(T-273.15) + 8.934] [m^{-2}]$$

Organic (R13) $215\text{ K} < T < 273\text{ K}$

$$n_{s, \text{peat}}(T) = 2.59 \times 10^{-4} \times (T - 245.80)^2 [m^{-2}]$$

Soot (S20) $-36\text{ }^\circ\text{C} < T < -11\text{ }^\circ\text{C}$

$$n_{s, \text{soot}}(T) = \exp(1.844 - 0.687 \times T - 0.00597 \times T^2) [m^{-2}]$$



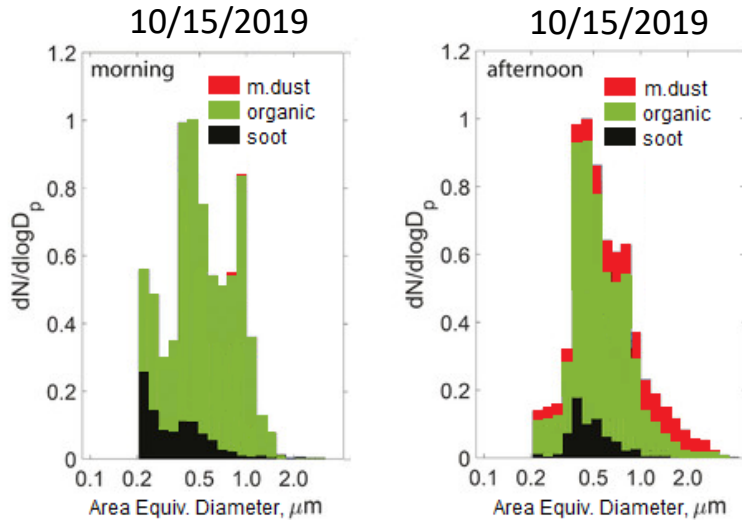
SGP ABIFM parameterization



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$$n_{ice,j} = n_{aer,j} (1 - e^{-S_j J_{het}(aw) t}) [L^{-1}]$$

(NOTE: $t = 13$ to 48 s)



Mineral Dust (A&K16)

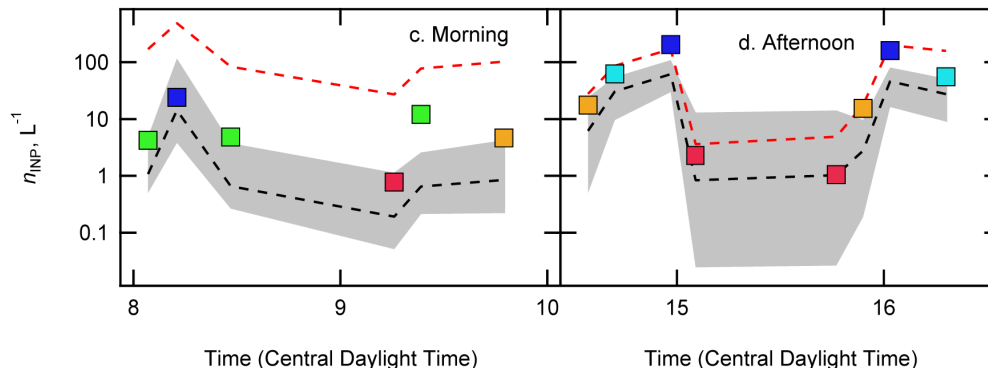
$$J_{het, illite}(\Delta aw) = 10^{-10.66873 + 54.48075 \times \Delta aw} [cm^2 s^{-1}]$$

Organic (K&A13)

$$J_{het, leonardite}(\Delta aw) = 10^{-13.40148 + 66.90259 \times \Delta aw} [cm^2 s^{-1}]$$

Soot (K21)

$$J_{het, soot}(\Delta aw) = 10^{-2.0847 + 18.0679 \times \Delta aw} [cm^2 s^{-1}]$$



- Simulation with 100% mineral dust
- Simulation with size and composition scaled simulation
- Uncertainty (surface area & stochastic error)

$BC_{SGP} \leq BC_{ENA}$
 $AE_{SGP} \gg AE_{ENA}$

Aerosol Abundance (6-hour time average)

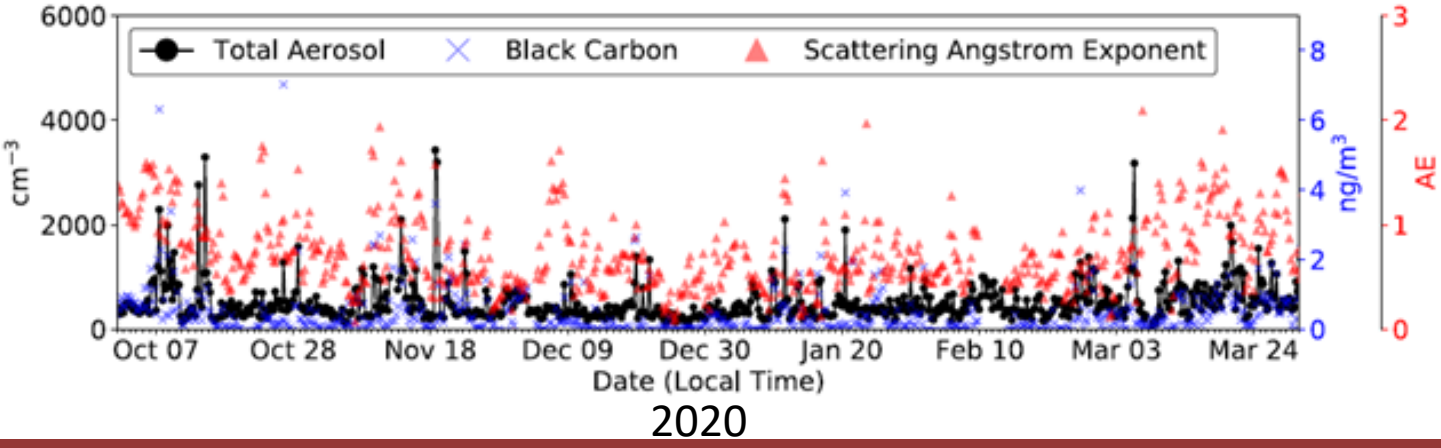
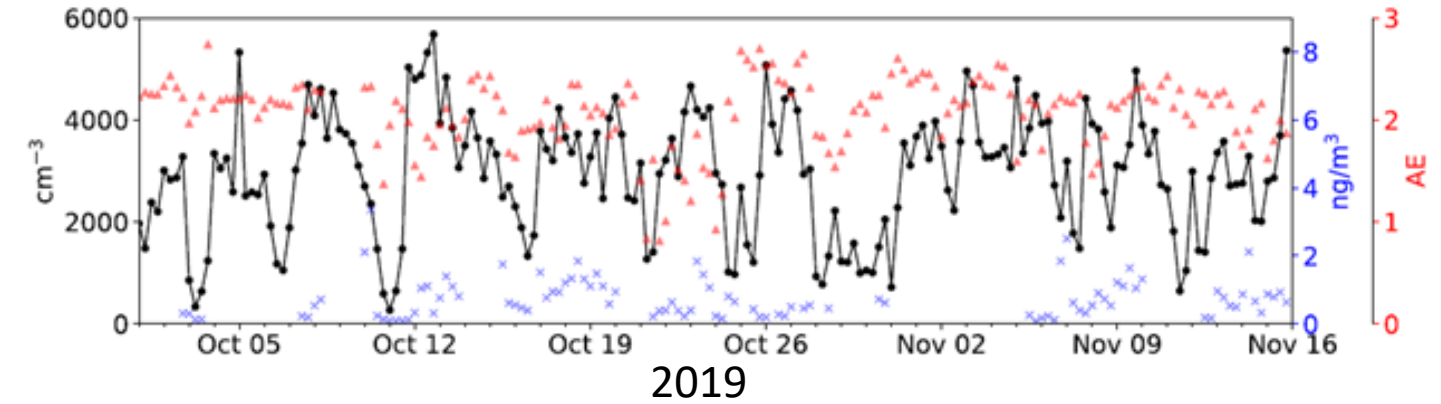


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SGP

ENA

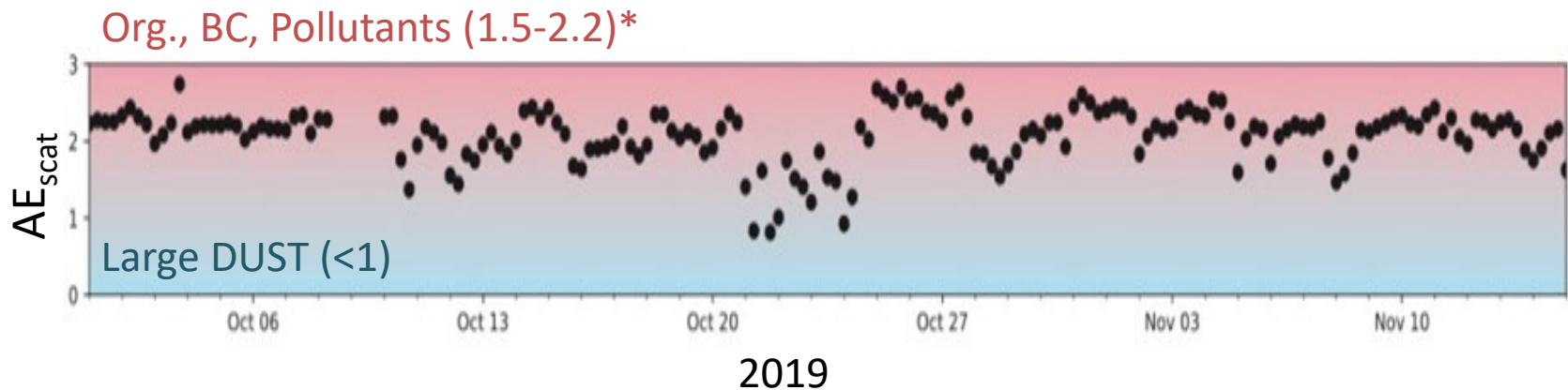
● Total Aerosol × Black Carbon ▲ Scattering Angstrom Exponent



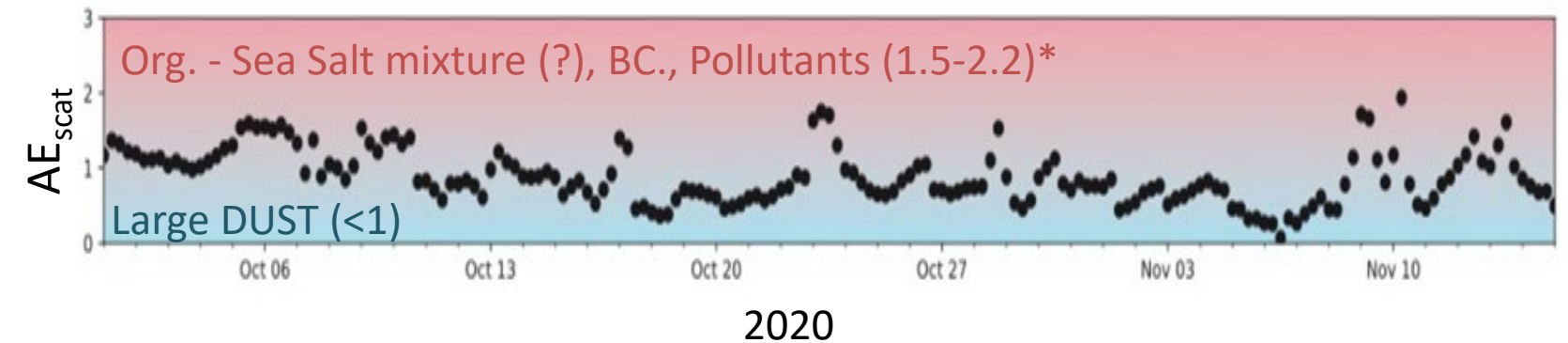
Dust Abundance

- ❖ $\text{Org}_{\text{SGP}} \gg \text{Org}_{\text{ENA}}$ (AE_{scat} of 1.5 – 2.2)
- ❖ $\text{Dust}_{\text{SGP}} \ll \text{Dust}_{\text{ENA}}$ ($\text{AE}_{\text{scat}} < 1$) with greater variability!

SGP



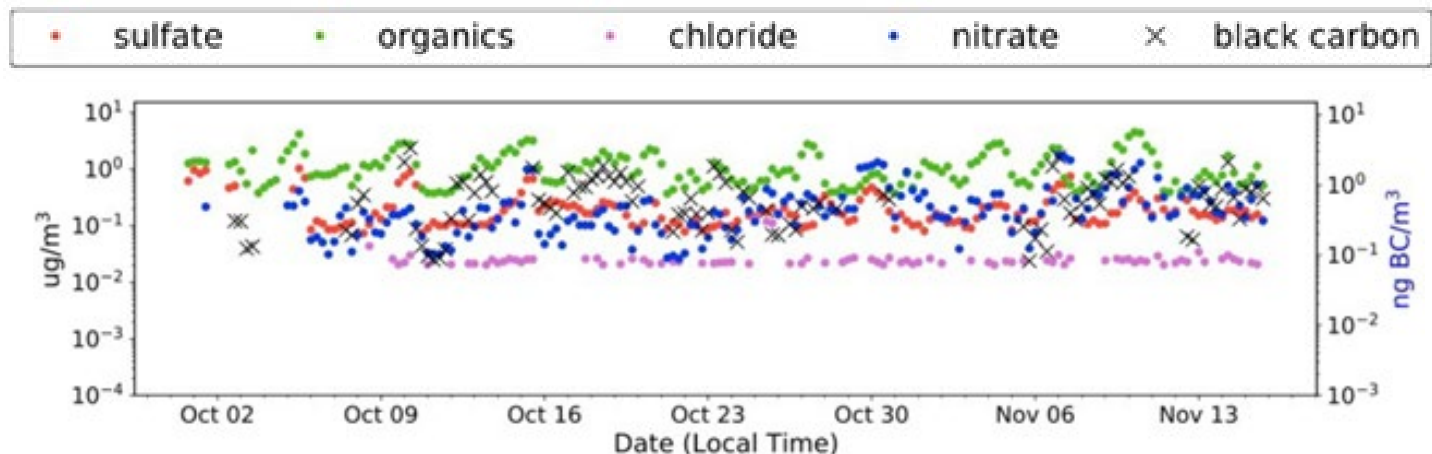
ENA



Aerosol Composition

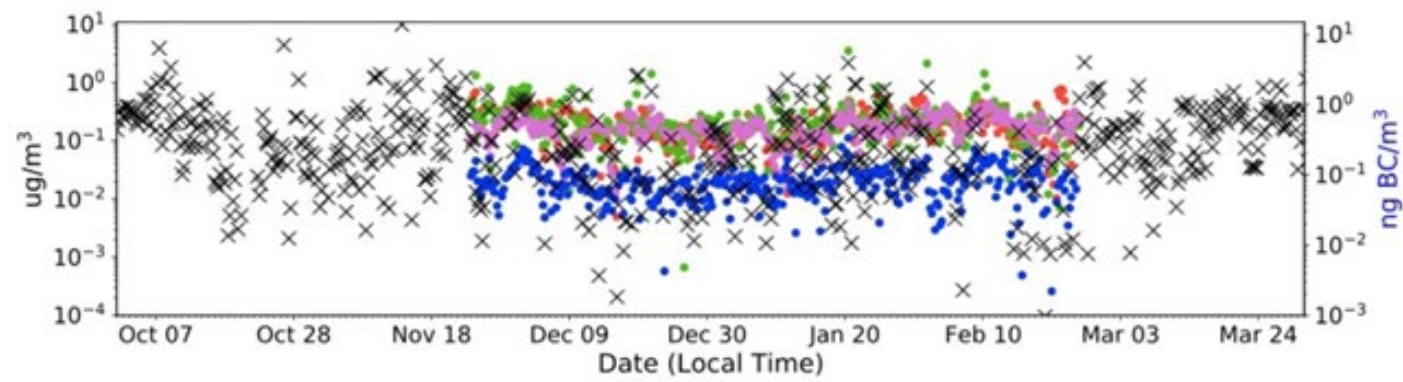
❖ $Org_{SGP} \gg Org_{ENA}$
❖ $Nitrate_{SGP} \gg Nitrate_{ENA}$

SGP



2019

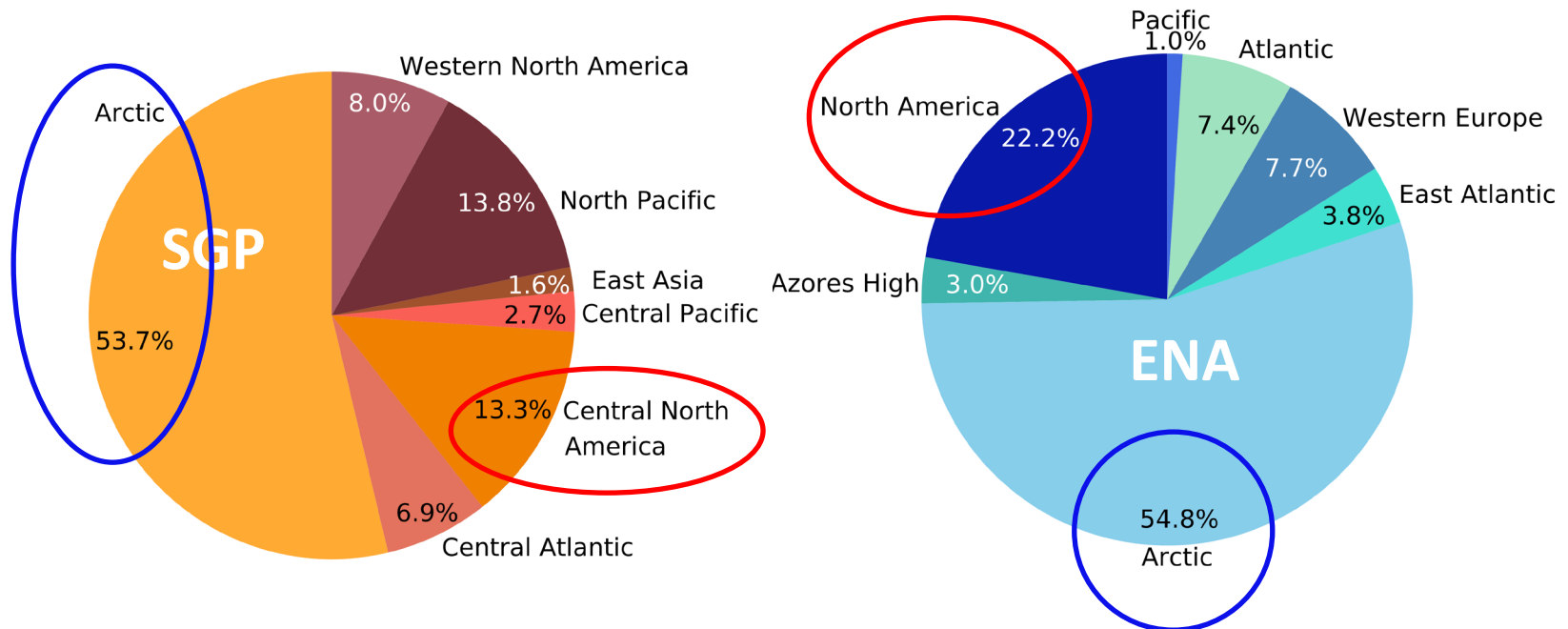
ENA



2020

10-day HY-SPLIT Back-trajectory (sampling height)

- ❖ Air mass of high INP events came from the **Arctic**.
- ❖ Air mass of high CCN (at 0.1-0.2 SS%) events came from North America (NA).
- ❖ Seasonal variability: More Arctic air mass in autumn;
More NA in winter in our study period [ENA].

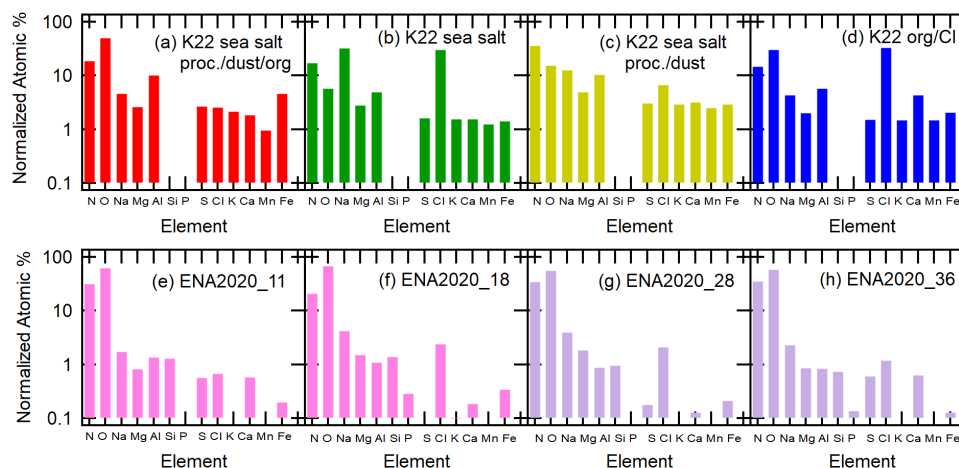


ENA Particle Composition



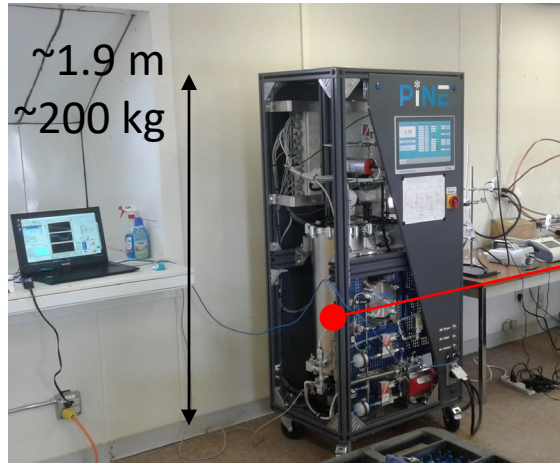
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Filter name	Start Date/Time	End Date/Time	Salt-dominant particle	Dust-dominant particle	Na-to-Cl ratio
	(UTC)	(UTC)			
Unit	mm/dd/yy hh:mm	mm/dd/yy hh:mm	% ± std. err.	% ± std. err.	n/a
ENA2020_11	10/11/20 14:24	10/14/20 15:30	29 ± 21	68 ± 14	2.73 ± 0.20
ENA2020_18	10/17/20 15:24	10/20/20 14:24	70 ± 16	30 ± 16	1.94 ± 0.08
ENA2020_28	11/1/20 13:47	11/4/20 16:03	85 ± 13	15 ± 18	1.91 ± 0.06
ENA2020_36	11/15/20 16:42	11/18/20 13:24	56 ± 16	42 ± 16	2.00 ± 0.09



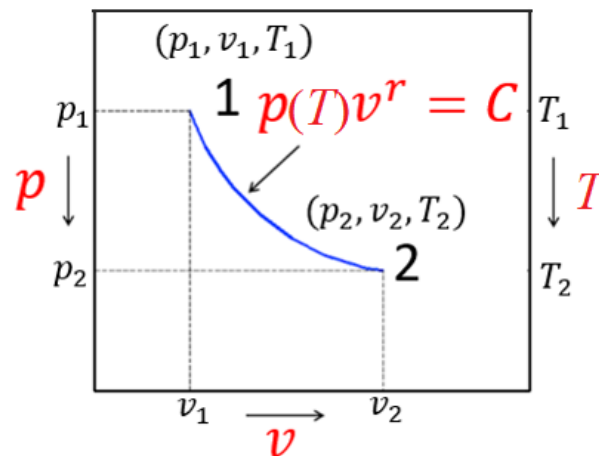
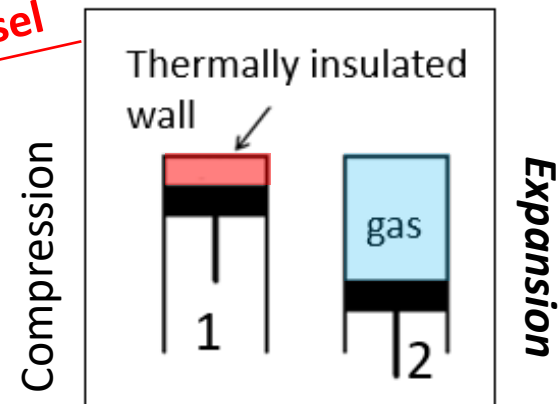
Supplemental Information: Methods

Portable Ice Nucleation Experiment (PINE-3) Chamber



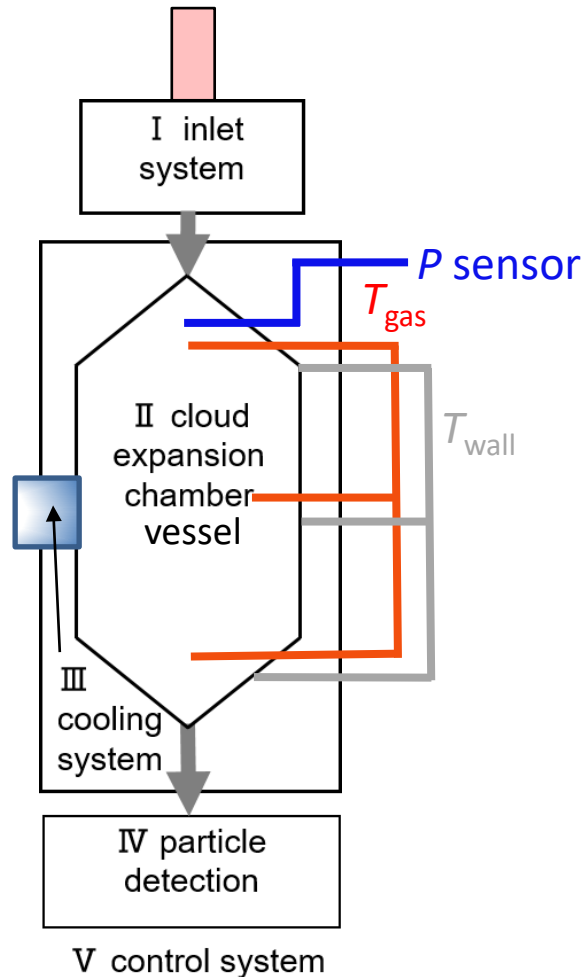
Simulated adiabatic *expansion* cooling

10 L volume vessel



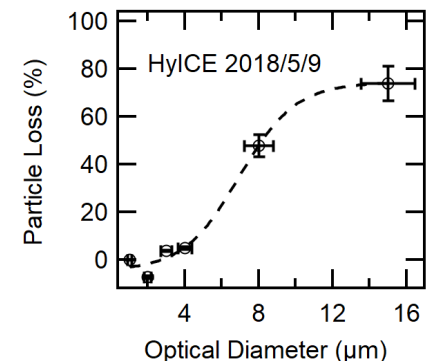
$\Delta T_{\text{gas}} \approx -4 \text{ } ^\circ\text{C}$
 $\Delta P \approx -200 \text{ to } -250 \text{ mb}$
 $\Delta v \approx +2.2 \text{ to } +2.5 \text{ L}$
 $\Delta T/\Delta t \approx 4 \text{ } ^\circ\text{C min}^{-1}$

Portable Ice Nucleation Experiment (PINE-3) Chamber



- I. **Nafion dryer:** as a part of the inlet system
- II. **Vessel with multiple sensors:** 3 T_{gas} thermocouple, 3 T_{wall} pt-100, P sensor ($\pm 1^\circ\text{C}$ accuracy)
- III. **Cryo-cooler:** controlling T_{gas} in the vessel between 0°C and -60°C (T_{wall} cooling = $0.6^\circ\text{C min}^{-1}$)
- IV. **Optical particle counter:** for n_{INP} measurement ($\pm 20\%$ accuracy) for $\approx 0.7 - 220 \mu\text{m}$ ($\approx 0.4 \text{ INP L}^{-1}$ detection limit)
- V. **LabView console:** autonomously controlling 'expansion' experiment **every ≈ 12 min** with 3 pumps, 3 mass flow controllers & 6 valves

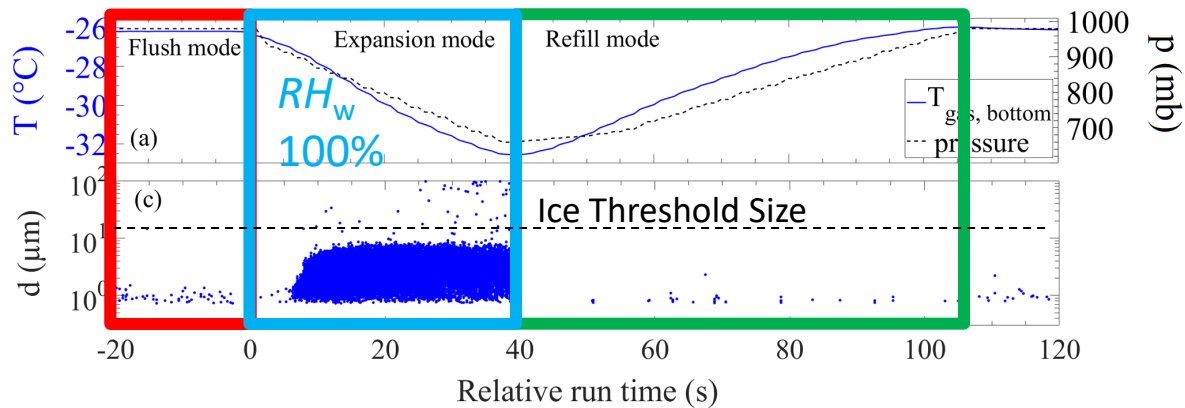
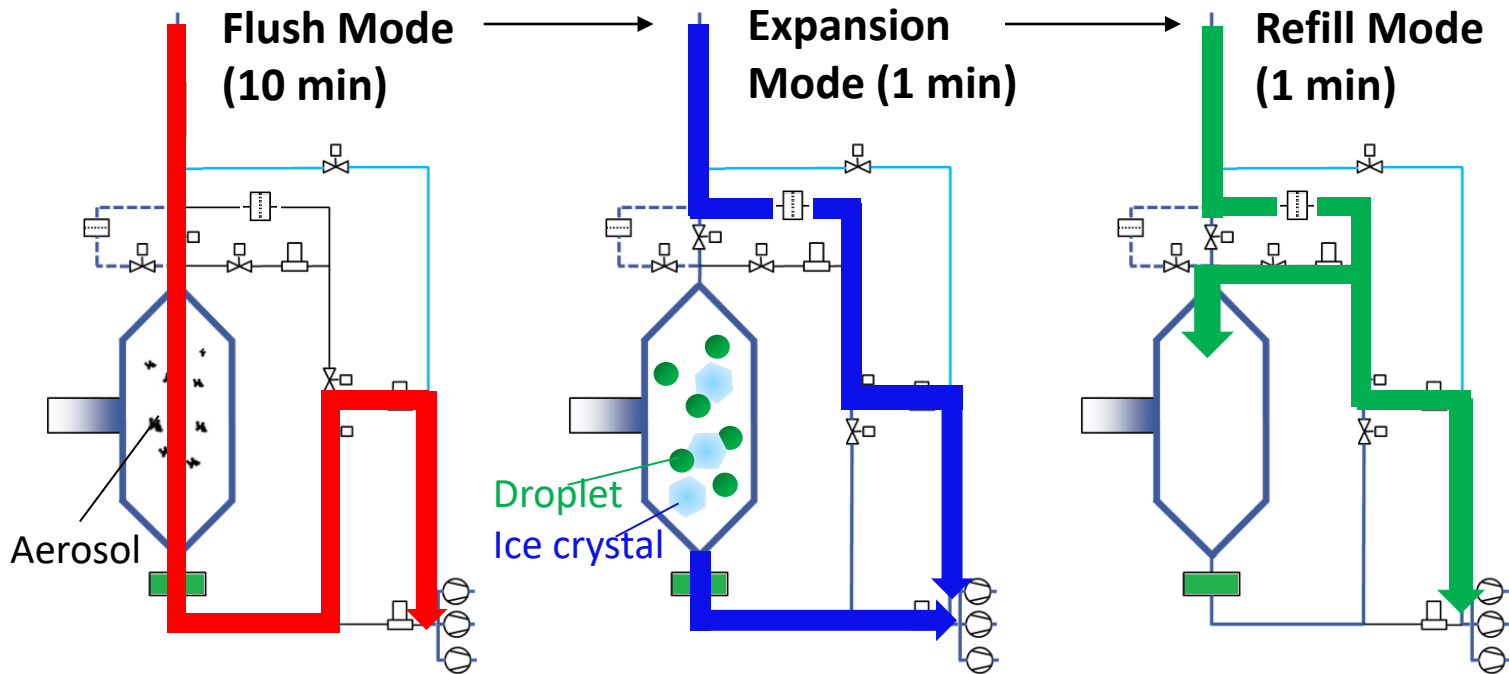
NOTE: The measured particle loss: $\leq 5\%$ at $\leq 3 \mu\text{m } D_{\text{opt}}$



PINE-3 Run-Mode

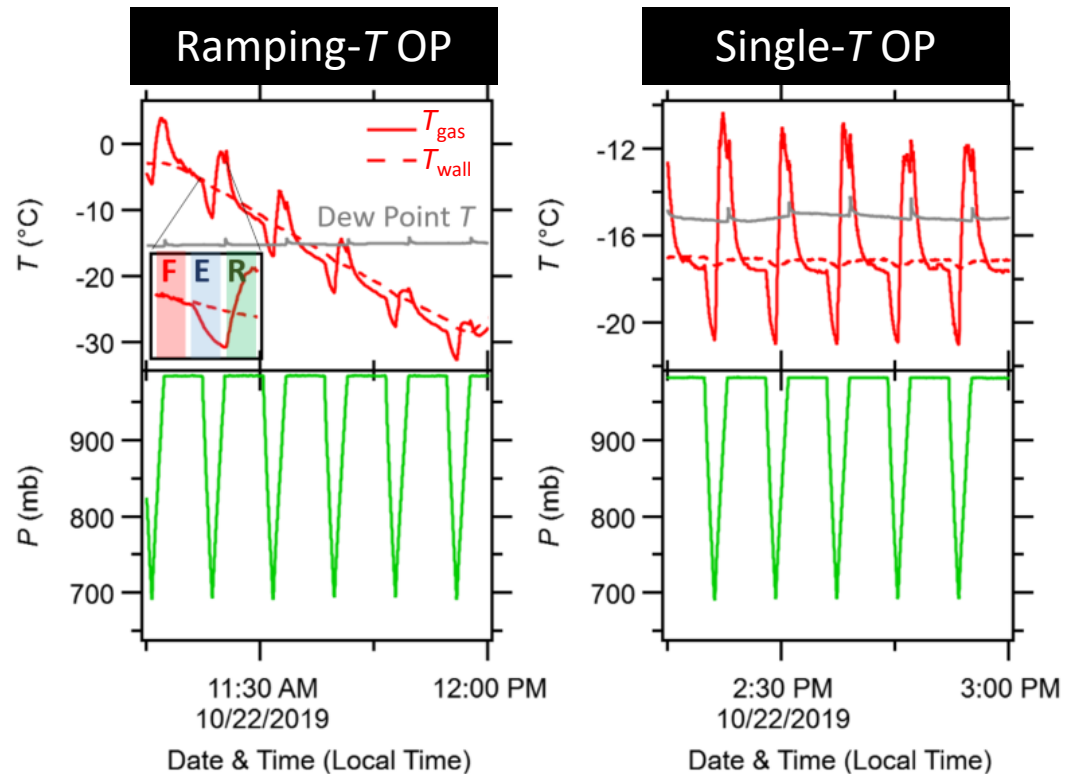


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PINE-3 Operation

- ❖ **Ramping- T Operation:**
 T_{wall} cycles of $-5\text{ }^{\circ}\text{C} \leftrightarrow -35\text{ }^{\circ}\text{C}$ every 90 min with automated sequence of **F**lush \rightarrow **E**xpand \rightarrow **R**efill
- ❖ **Single- T Operation:**
Measurements at a fixed T_{wall}
- ❖ **Background Operation:**
Expansions with filtered air are carried out daily for ~ 1 hour to ensure a zero-INP background

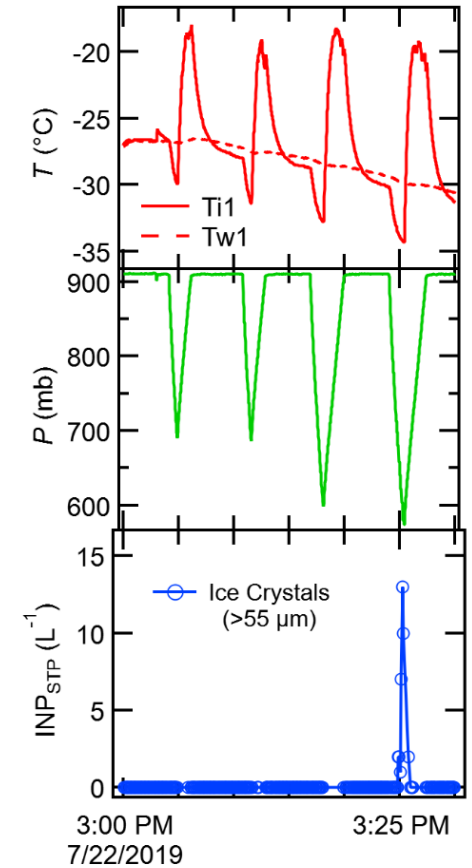
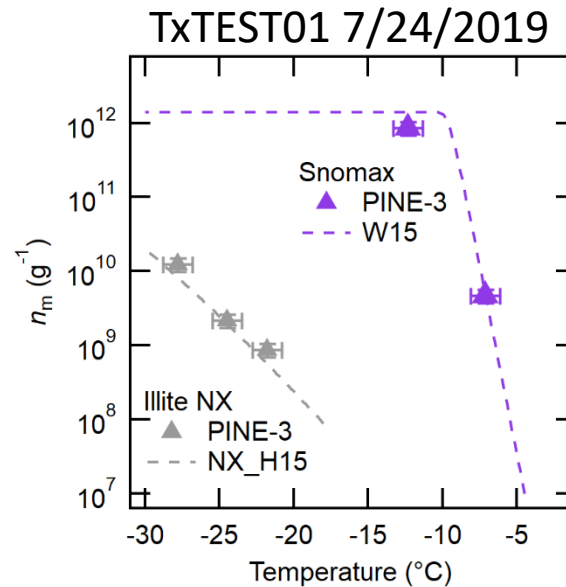
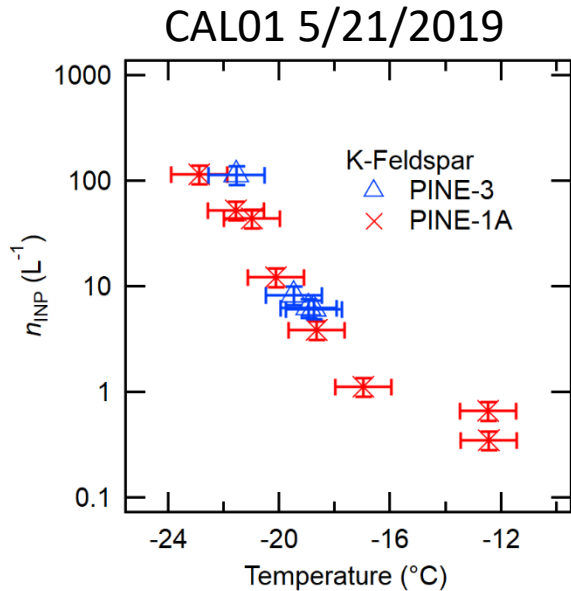


PINE-3 Calibration



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Homogeneous Freezing with Ammonium Sulfate < -33 °C



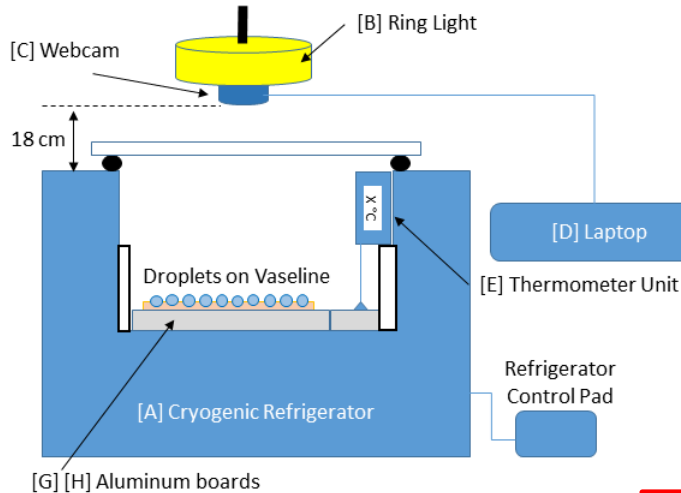
Date & Time (Local Time)

$$n_{INP}(T) = \frac{\text{Ice Crystal Counts}}{\text{Expanded Air Vol.}} [L^{-1}]$$

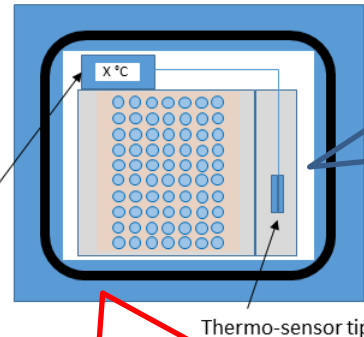
$$n_m(T) = \frac{n_{INP}(T)}{\text{Particle Mass}} [g^{-1}]$$

West Texas Cryogenic Refrigerator Applied to Freezing Test (WT-CRAFT) System

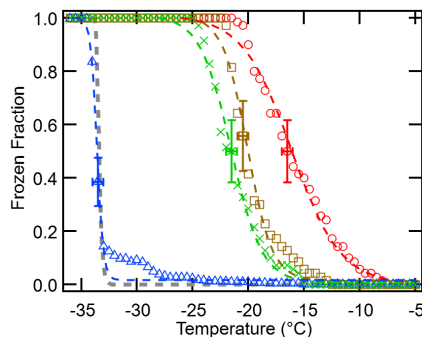
a. Side View



b. Top View

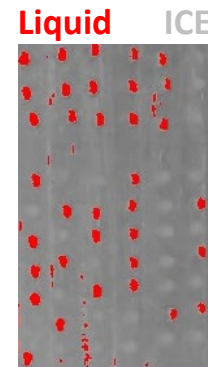


- 70 of 3 μ L droplets containing particles
- 1 °C min⁻¹ cooling for -25 °C < T < 0 °C
- Frozen fraction (f_{frozen}) at each 0.5 °C

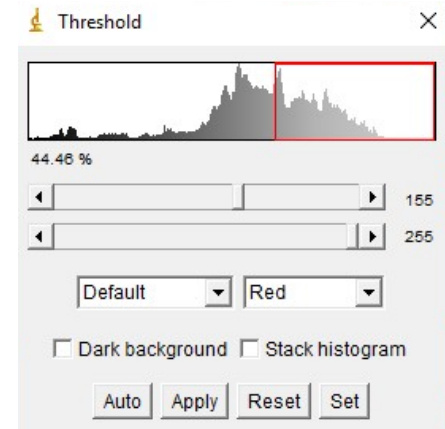


- Unfiltered-tap water
- illite NX suspension 0.1 wt%
- × Filtered-tap water
- △ HPLC-grade water
- Homogeneous Freezing of pure water droplet (theory)

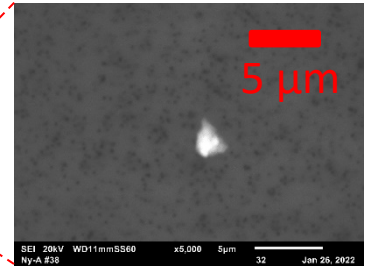
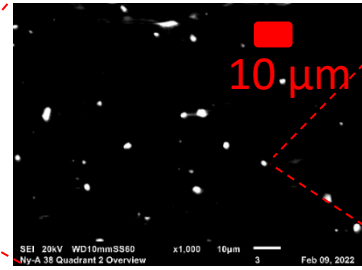
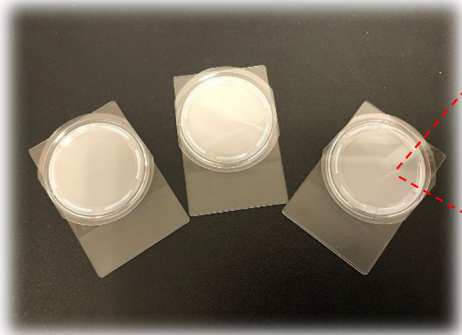
-25 °C



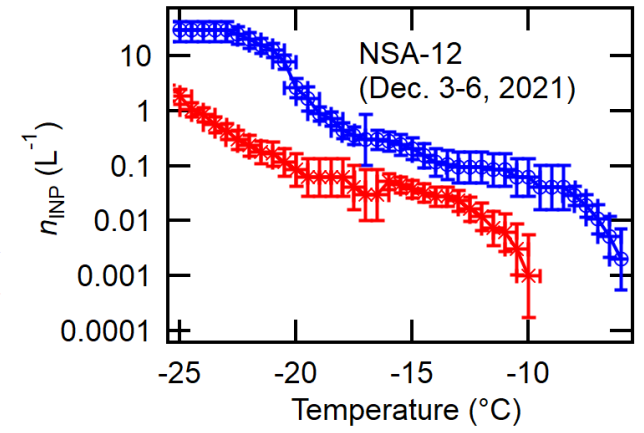
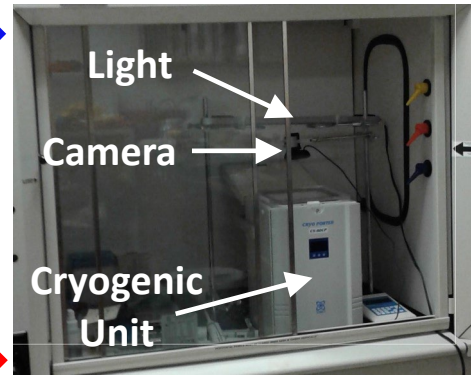
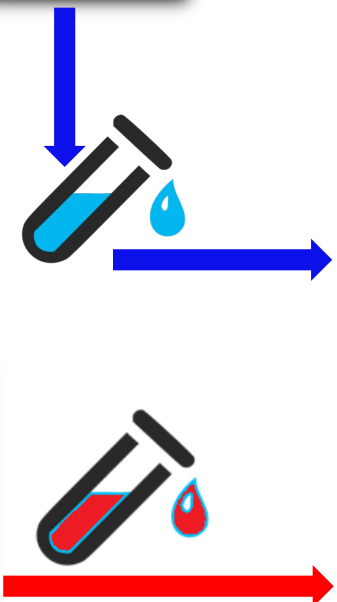
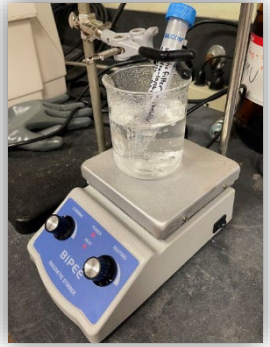
$$f_{\text{frozen}} = 39/70$$



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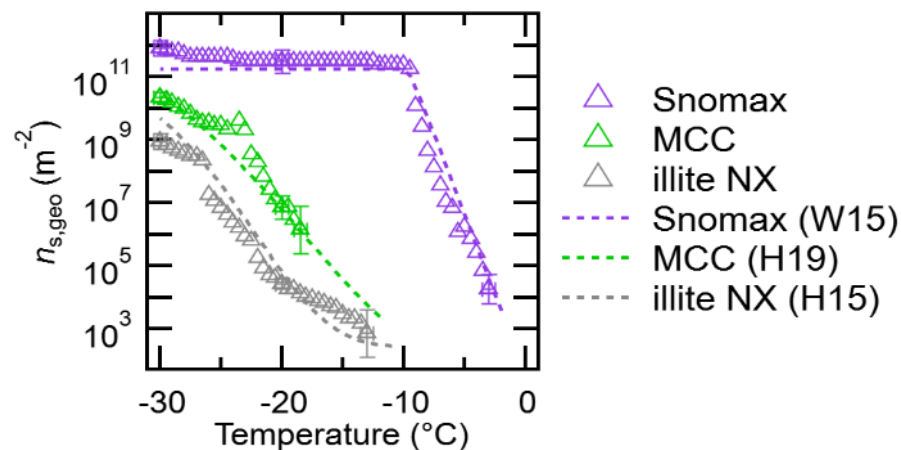
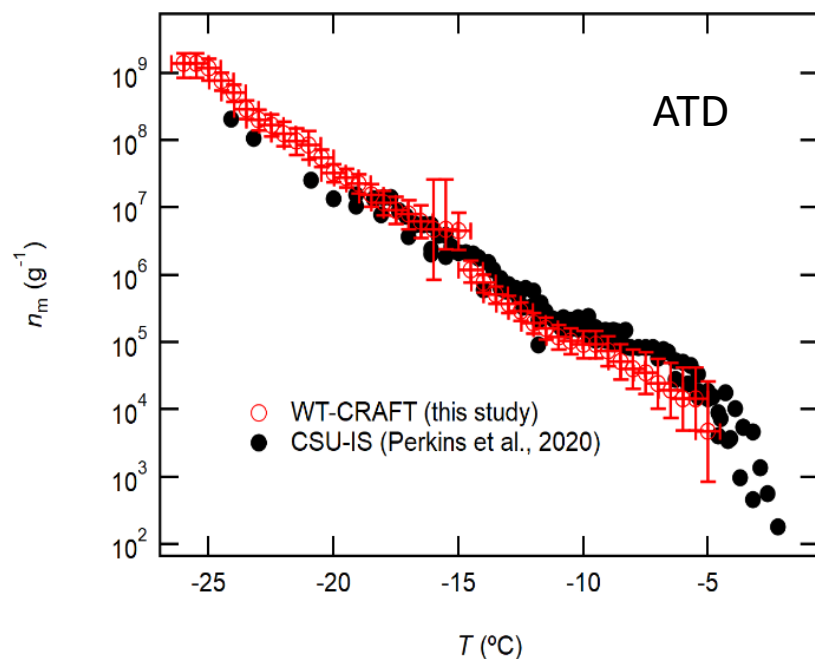
~95 °C
20 min



$$C_{INP}(T) = - \frac{\ln(1 - f_{frozen}(T))}{Droplet Vol.} [L^{-1} liq.]$$

$$n_{INP}(T) = C_{INP}(T) \times \frac{Suspension Vol.}{Sampled Air Vol.} [L^{-1} air]$$

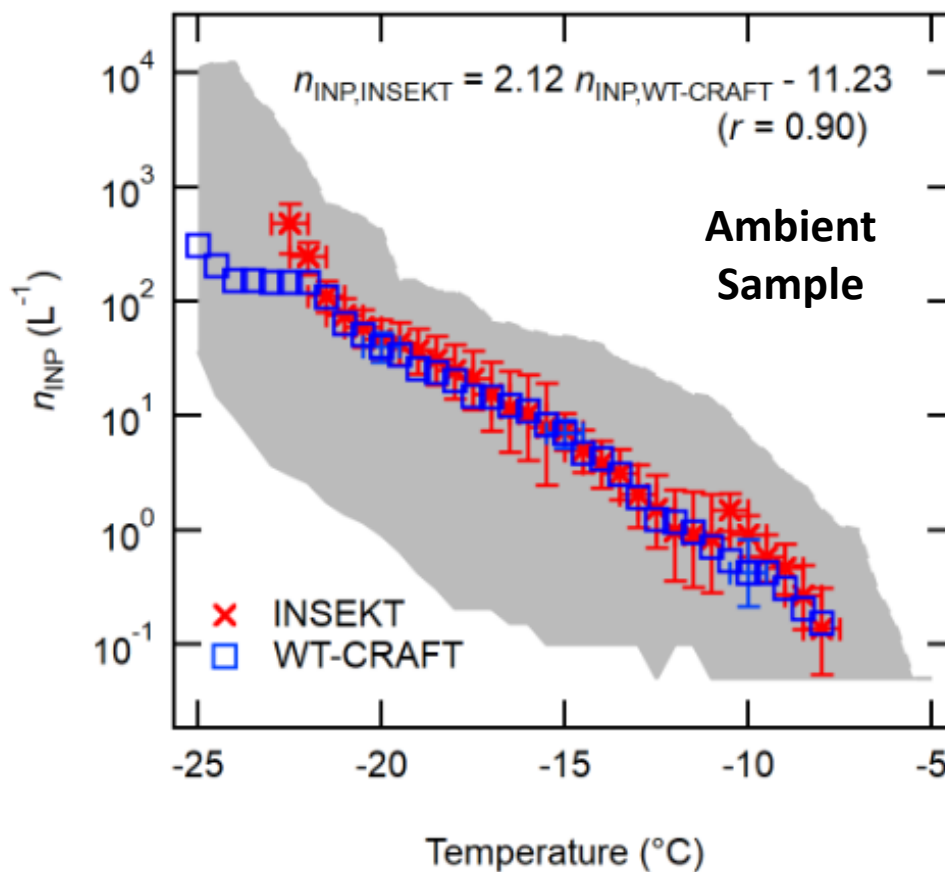
West Texas Cryogenic Refrigerator Applied to Freezing Test (WT-CRAFT) System



$$n_m(T) = \frac{n_{INP}(T)}{\text{Particle Mass}} [\text{g}^{-1}]$$

$$n_{s,geo}(T) = \frac{n_m(T)}{\text{Specific Surface}} [\text{m}^{-2}]$$

West Texas Cryogenic Refrigerator Applied to Freezing Test (WT-CRAFT) System

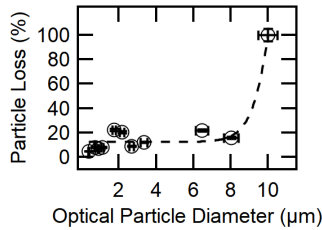
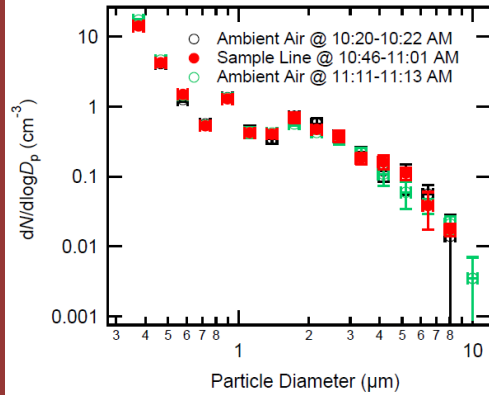


Inlet Loss

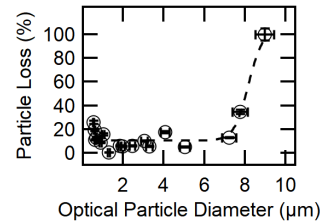
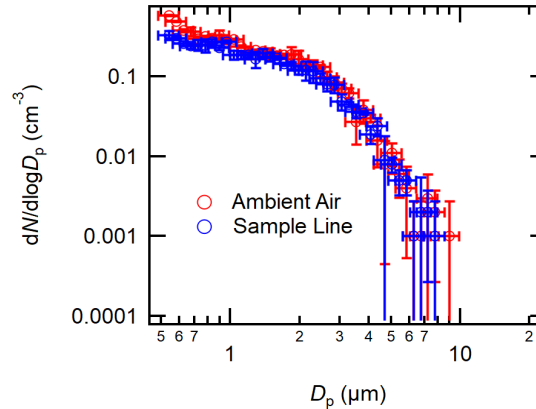


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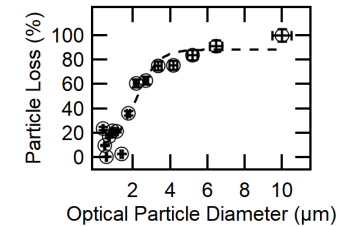
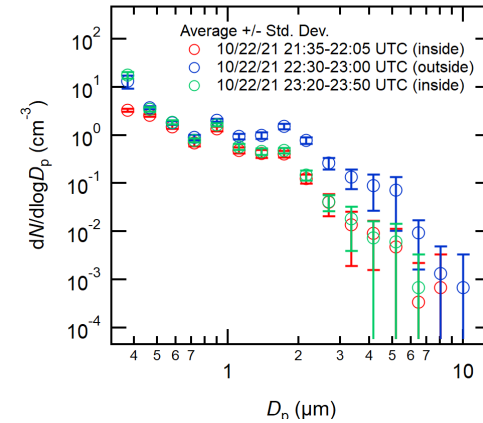
SGP



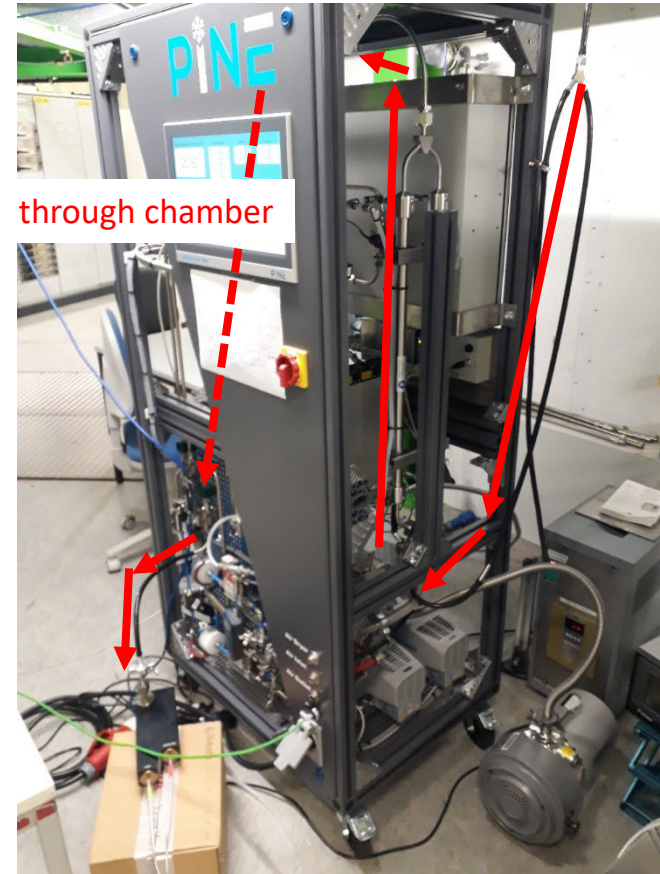
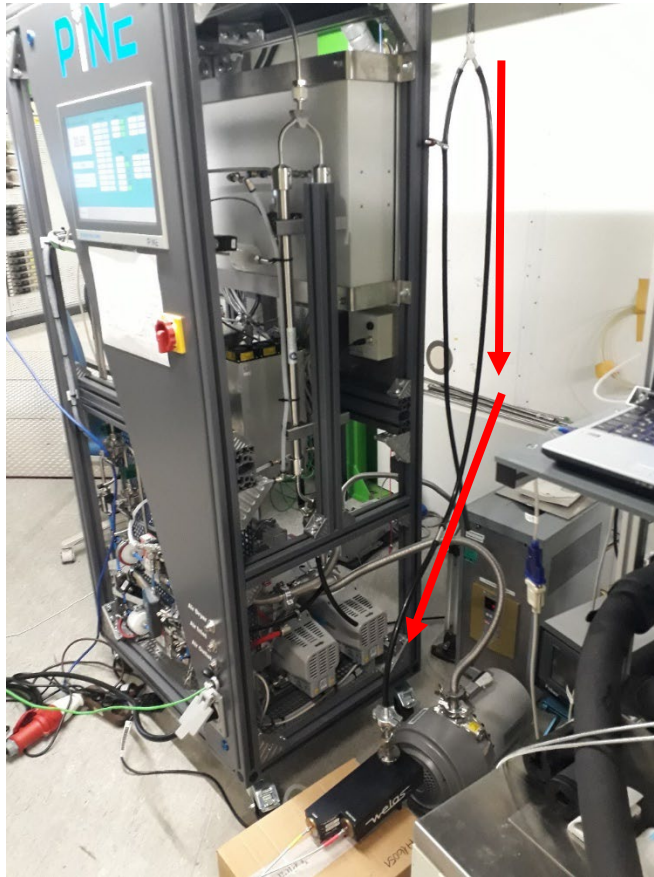
ENA



NSA-BRW



Particle Loss in PINE



“Memory Effect” Tests

- Can we clean up the chamber by introducing “filtered-air” for 300 sec or 600 sec?
- We did a series of expansion with the following sequence:
Expansion w/ filtered air (F) → Expansion w/ ambient sample air (S) → S → F
- We repeated this sequence at 4 different set point T_s : -16, -21, -26, and -31 °C

Poisson Analysis

To estimate n_{INP} errors as a function of T
 = memory effect

- Suggested by CSU – see Kathryn Moore’s M.S. thesis - <https://mountainscholar.org/handle/10217/208435>
- Based on equations from Krishnamoorthy and Lee, 2013
- Poisson mean:

- $\hat{\lambda}_s - \hat{\lambda}_f + \frac{3.84}{2} \left(\frac{1}{n_s} - \frac{1}{n_f} \right)$ where $\hat{\lambda}_s = \frac{N_s}{n_s}$ and $\hat{\lambda}_f = \frac{N_f}{n_f}$
- $N_s = n_{\text{INP, sample}}$ with ambient air
- $N_f = n_{\text{INP, filtered}}$
- $n_s =$ number of expansions with ambient air
- $n_f =$ number of expansions with filtered air

- $CI_{95\%} = \hat{\lambda}_s - \hat{\lambda}_f + \frac{1.96^2}{2} \left(\frac{1}{n_s} - \frac{1}{n_f} \right) \pm 1.96 * \sqrt{\left(\frac{\hat{\lambda}_s}{n_s} + \frac{\hat{\lambda}_f}{n_f} \right) + \frac{1.96}{4} \left(\frac{1}{n_s} - \frac{1}{n_f} \right)^2}$
- For the Poisson mean to be applicable, Z_m must be greater than 1.96 for a 95% confidence interval
- $Z_m = \frac{\hat{\lambda}_s - \hat{\lambda}_f}{\sqrt{\hat{\lambda} \left(\frac{1}{n_s} + \frac{1}{n_f} \right)}}$ where $\hat{\lambda} = \frac{N_s + N_f}{n_s + n_f}$

Preliminary “Memory Effect” Results [12/14/20]



We need an aerosol concentrator or further data processing (e.g., time average or cumulative INP counting analysis) to diagnose nINP above -16 °C

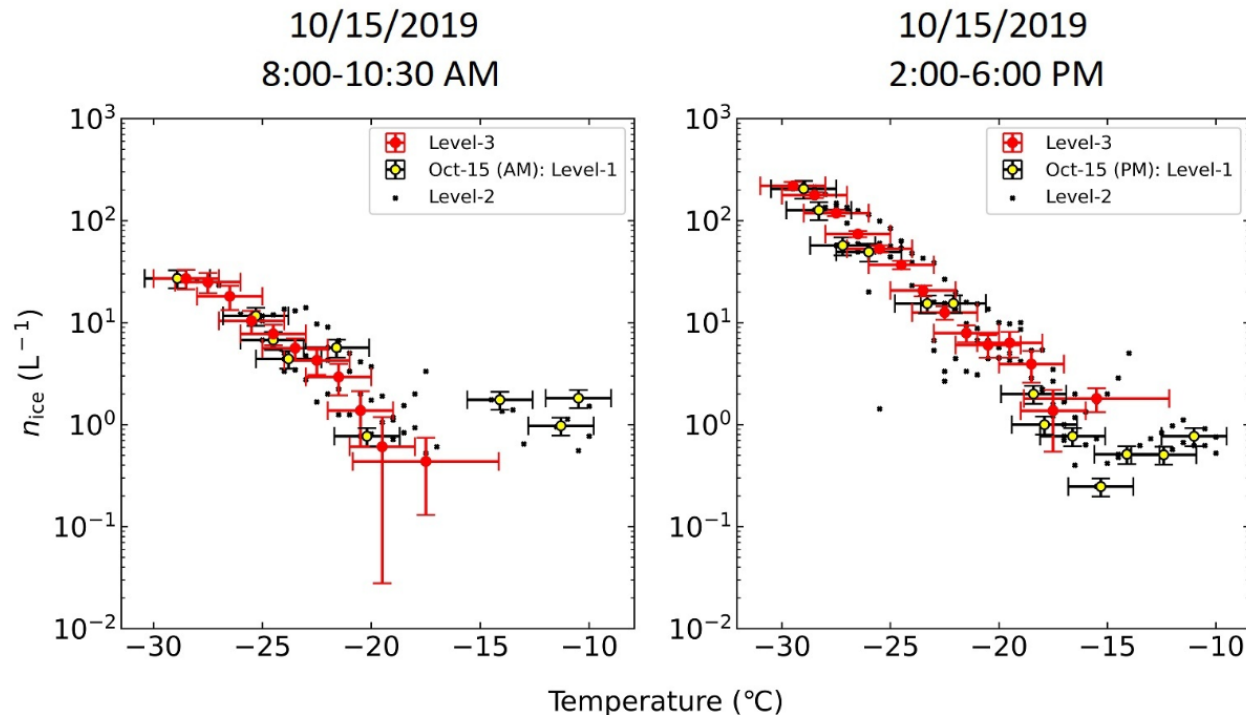
300 Second Flush Time										
Temperature (°C)	Flush Time (s)	λ_f	t_f	λ_s	t_s	λ	Mean	CI	Z_m	Mean \pm 95% CI
-16	300	0.18350168	4	0.06060606	6	0.10976431	-0.2829623	0.4905715	-0.5746607	n/a
-21	300	0.09090909	4	0.12012987	6	0.07272727	-0.1308459	0.43571273	0.1678606	n/a
-26	300	0.25423729	4	3.98964218	6	0.10844156	3.57533823	1.68054328	17.5729806	3.58 \pm 1.68
-31	300	3.90053763	4	29.5157765	6	0.17377284	25.4551722	4.76126722	95.194769	25.46 \pm 4.76
600 Second Flush Time										
Temperature (°C)	Flush Time (s)	λ_f	t_f	λ_s	t_s	λ	Mean	CI	Z_m	Mean \pm 95% CI
-16	600	0.41168629	12	0.21783849	8	0.33414717	-0.1138145	0.49275337	-0.7347044	n/a
-21	600	0.58256018	12	2.14063641	12	0.4366715	1.55807623	0.93369481	5.77546426	1.56 \pm 0.93
-26	600	0.54963303	12	6.61994079	14	1.36159829	6.04744109	1.41173315	13.2237362	6.05 \pm 1.41
-31	600	3.45739366	8	26.7392024	8	1.34513472	23.2818088	3.80793979	40.1479914	23.28 \pm 3.81

Positive aspect of Poisson



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The Poisson analysis processed data (red) **exclude high T humps & reasonable $CI_{95\%}$** as compared to the raw data (yellow).



Offline-droplet freezing results

