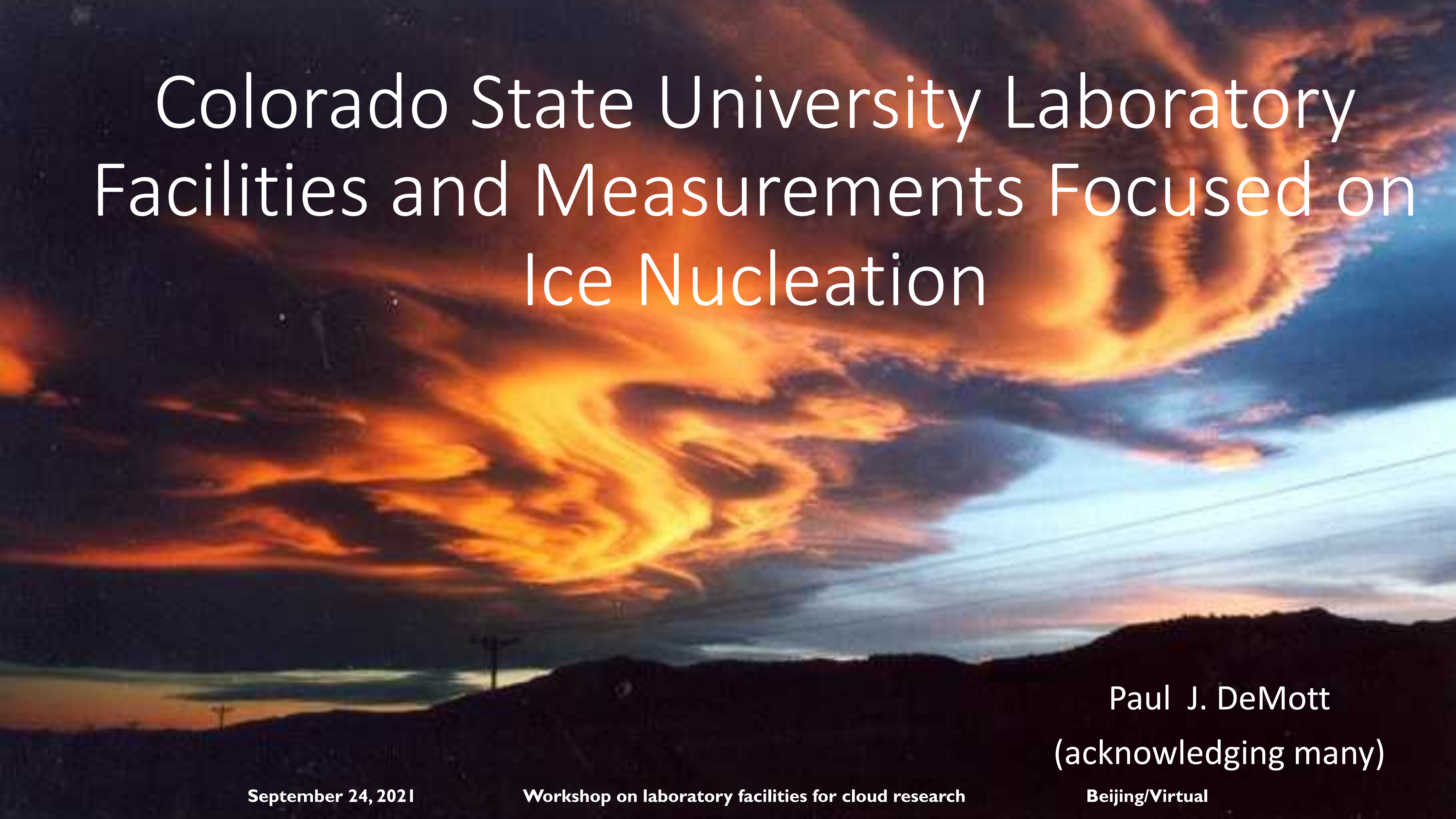


Colorado State University Laboratory Facilities and Measurements Focused on Ice Nucleation

Paul J. DeMott
(acknowledging many)

September 24, 2021 **Workshop on laboratory facilities for cloud research** **Beijing/Virtual**

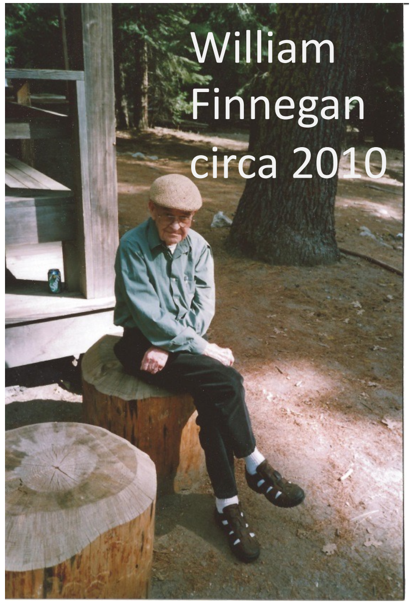
Today's talk

- <u>Former</u> fixed facilities and applications at CSU and research made possible by such facilities
  - Isothermal cloud chamber
  - Dynamic cloud chamber
  - Applications for cloud seeding and fundamental research
- Developed instruments for applications in lab or field for ice nucleation research (ongoing)
  - Continuous flow diffusion chambers
  - Ice spectrometer
- Other unique facilities for aerosol-cloud research we have used that should be considered in any overall plans for major facility capabilities

Cloud Seeding Research at the Cloud Simulation and Aerosol Laboratory


Lewis O. Grant

- Past cloud chamber studies have provided:
  - Cloud chamber activation spectral (#particles/g)
  - Rates of ice crystal formation (ice formation mechanisms and quantification)
  - New types of agents that express specific behaviors tested under operational conditions
- Calibrating in-situ ice nucleating particle counter to lab "standard"
- New methods for sampling and characterizing ice nucleating particles


William Finnegan circa 2010


Feng Daxiong - 1984

Producing "AgI-type" aerosols

Solution combustion: Vonnegut (1949)

AgI + NH_4I (solubilizing agent) + acetone (solvent) + H_2O

2% by weight typical

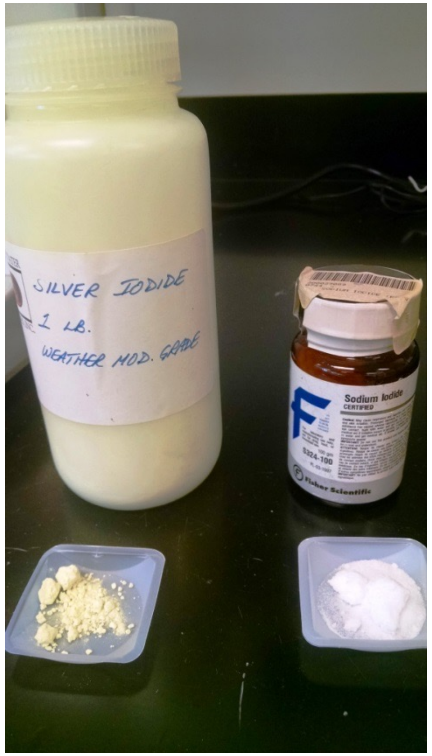
0.64% by weight

97% by weight

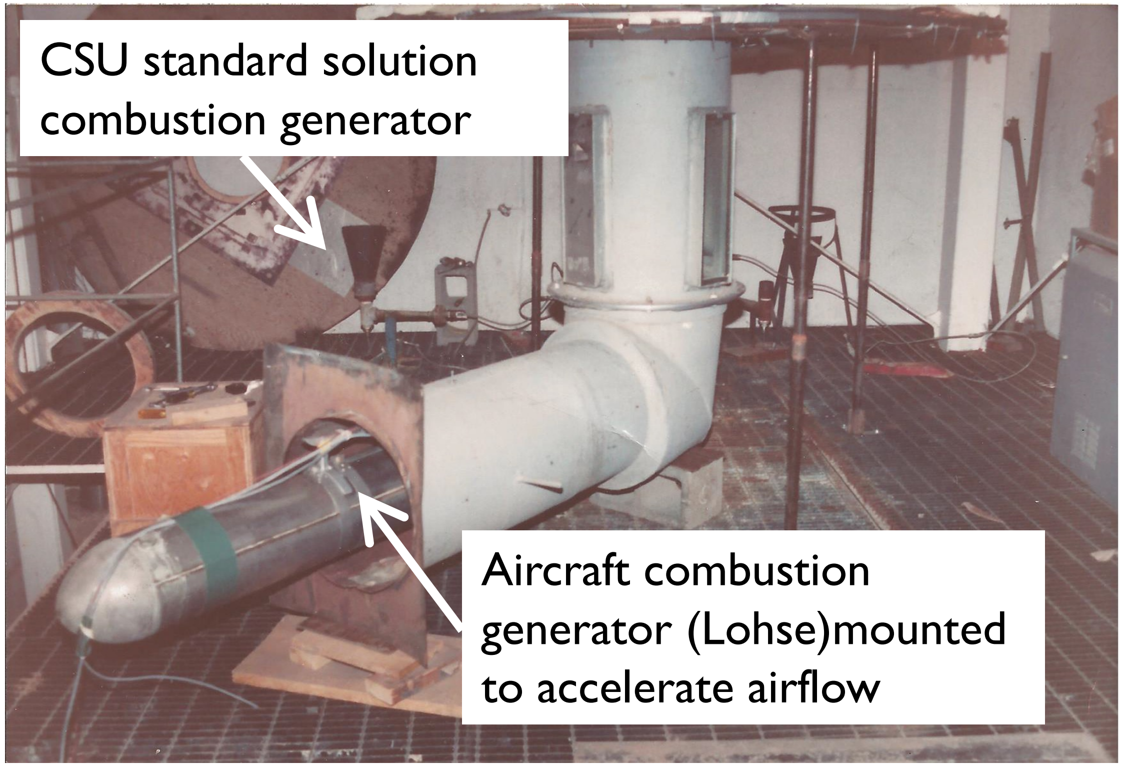
Pyrotechnic combustion: St. Amand et al. (1970)

$AgIO_3$ + 2Al (+ other metals, oxidizer such as KIO_3+ binder) → AgI + Al_2O_3 + …

Almost <u>never</u> simply AgI as a product



Varied mounting of field generators



CSU standard solution combustion generator

Aircraft combustion generator (Lohse)mounted to accelerate airflow

Wind tunnel

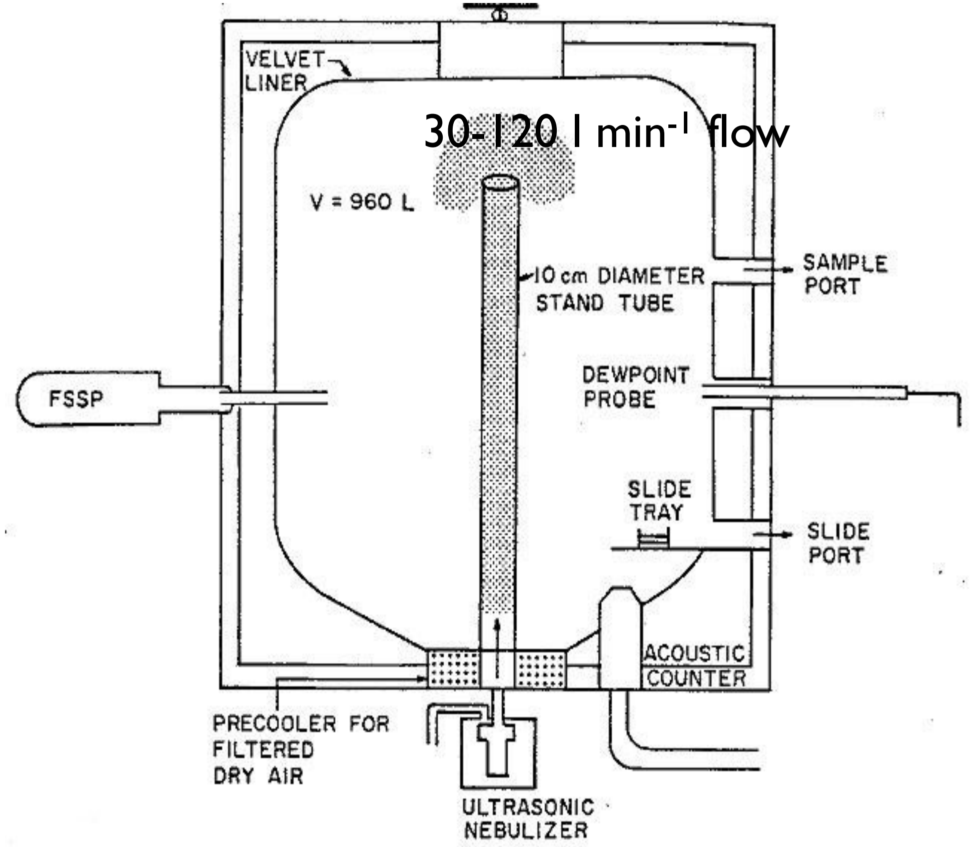
~3.2 million l min^{-1} natural air
55 m s^{-1} tunnel flow at midpoint

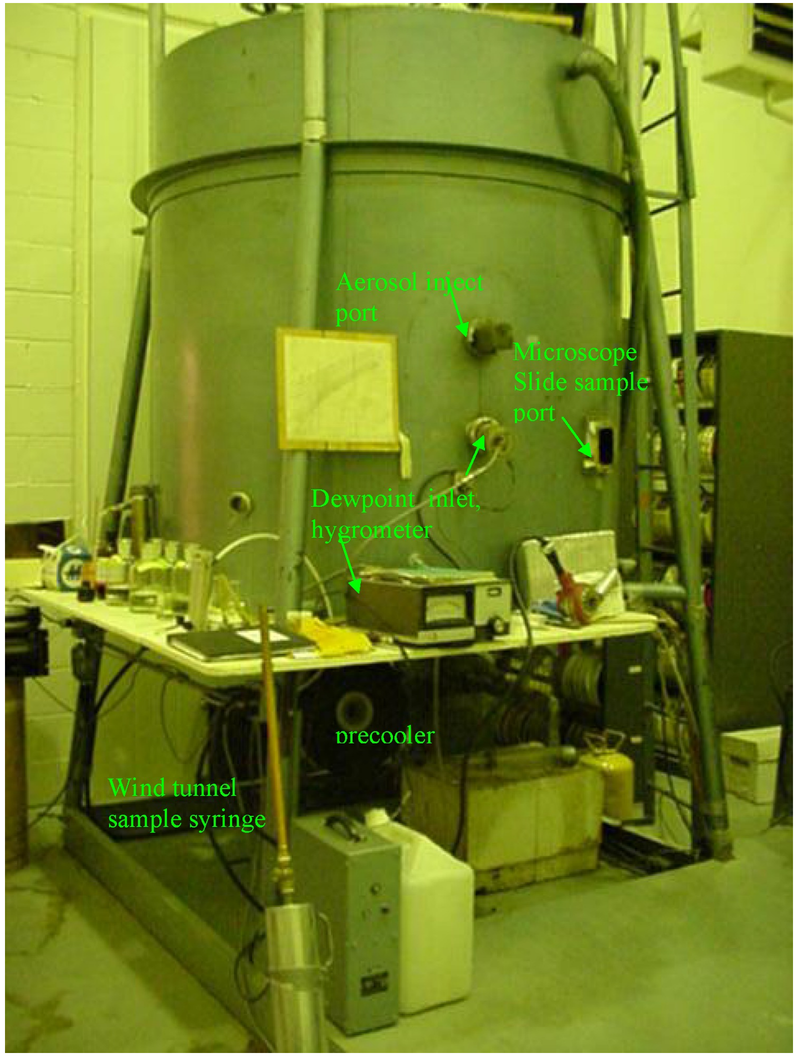
Collection and Dilution

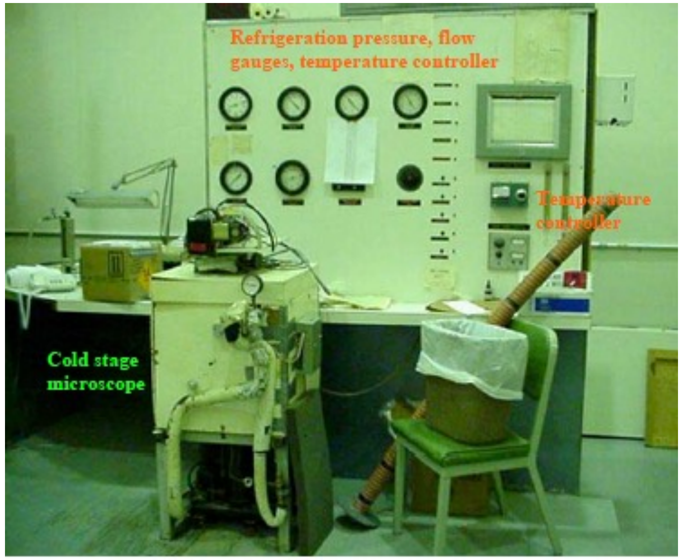
Boe, B. A. and P. J. DeMott, 1999: Comparisons of Lohse Wing-tip nuclei generators and burn-in-place pyrotechnics in the North Dakota Cloud Modification Project. *Journal of Weather Modification*, **31**, 109-118.

CSU Cloud Simulation Laboratory

Static, isothermal chamber
(Garvey et al. 1975;
DeMott et al. ,1982)


30-120 l min^{-1} flow





Calibration of seeding aerosols (Yield, Effectiveness, Activity)

Yield was the key factor determining decisions on seeding materials.

$$\text{Yield} = N_{ic} * (A_c/A_v) * (R_d/R_g) * (D_s/V_s) = \# \ g^{-1}$$

N_{ic}: total # ice crystals collected per microscope slide viewing area

A_c: chamber cross-sectional area(cm^2)

A_v: microscope viewing area(cm^2)

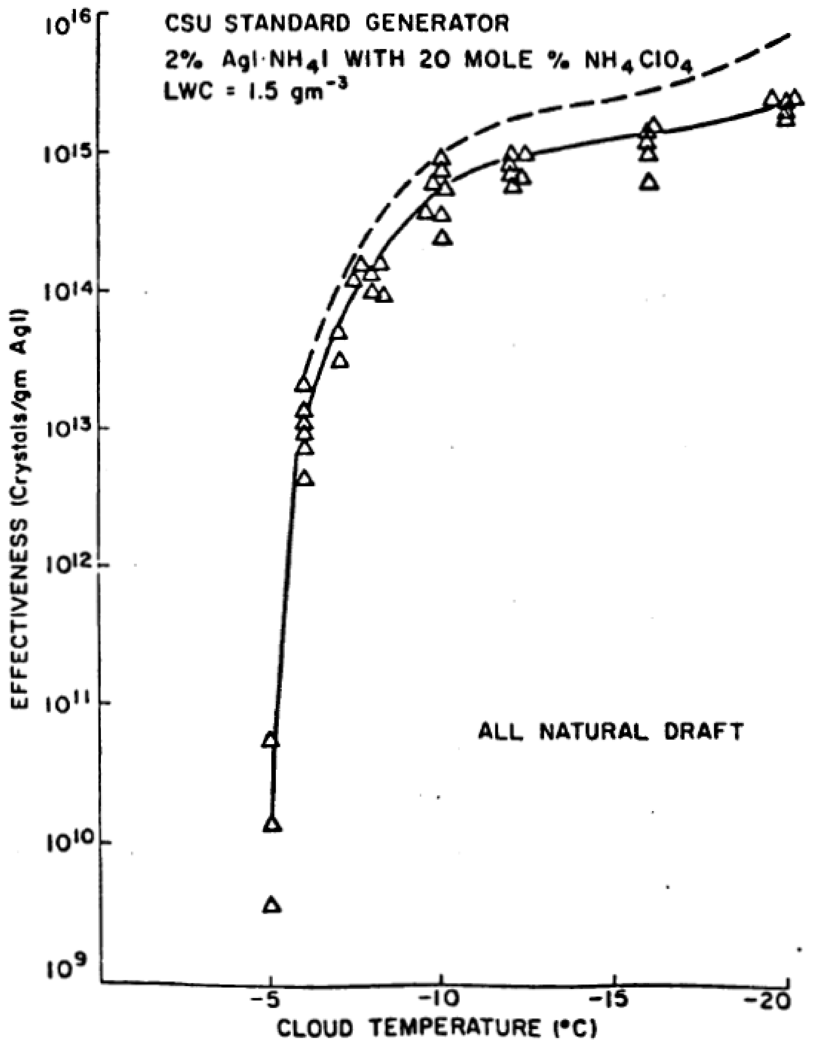
R_d: wind tunnel dilution rate($l \ min^{-1}$)

R_g: AgI generation rate($g \ min^{-1}$)

V_s: volume of collected sample (l)

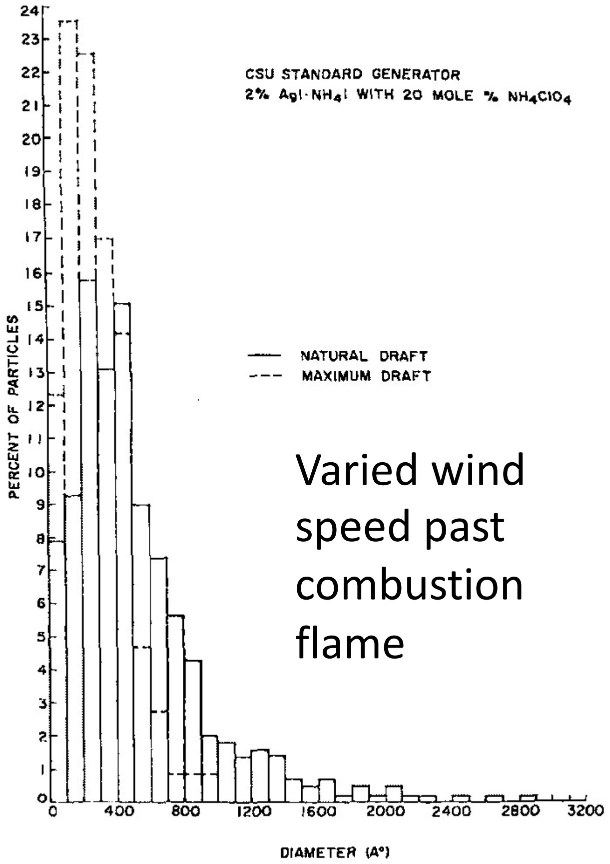
D: sample dilution factor

- In all cases, there was an attempt not to "overseed"

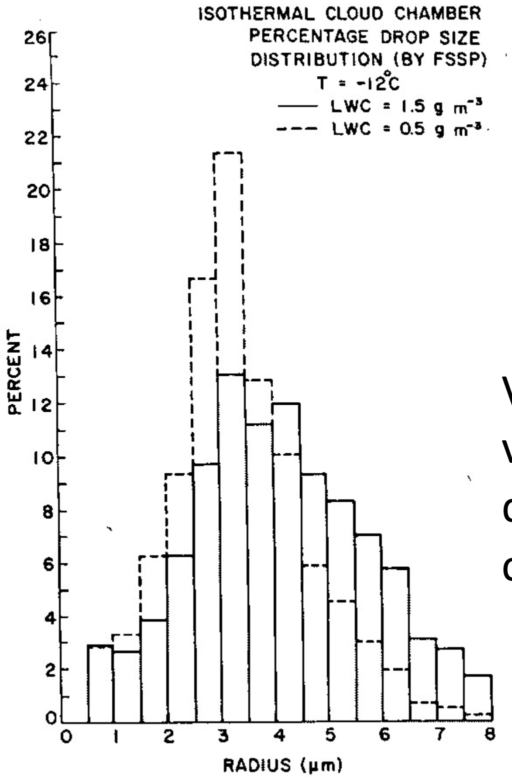


Able to explore aerosol size and droplet concentration impacts – to provide hints at nucleation mechanisms

DeMott (1982 – thesis)
DeMott et al. (1983, J. Clim. Appl. Meteor.).



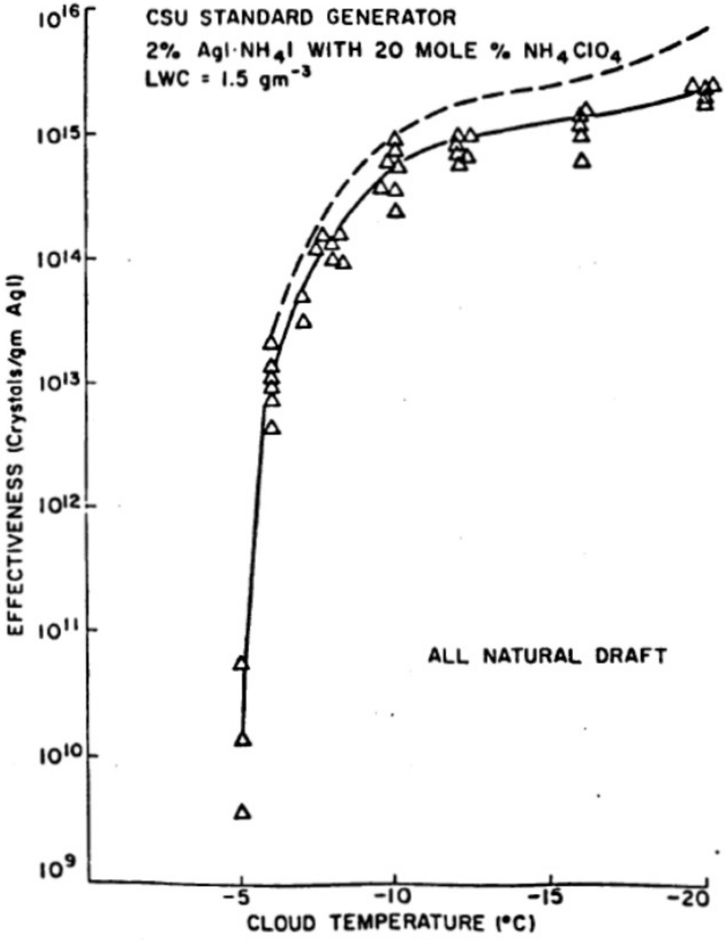
Varied wind speed past combustion flame

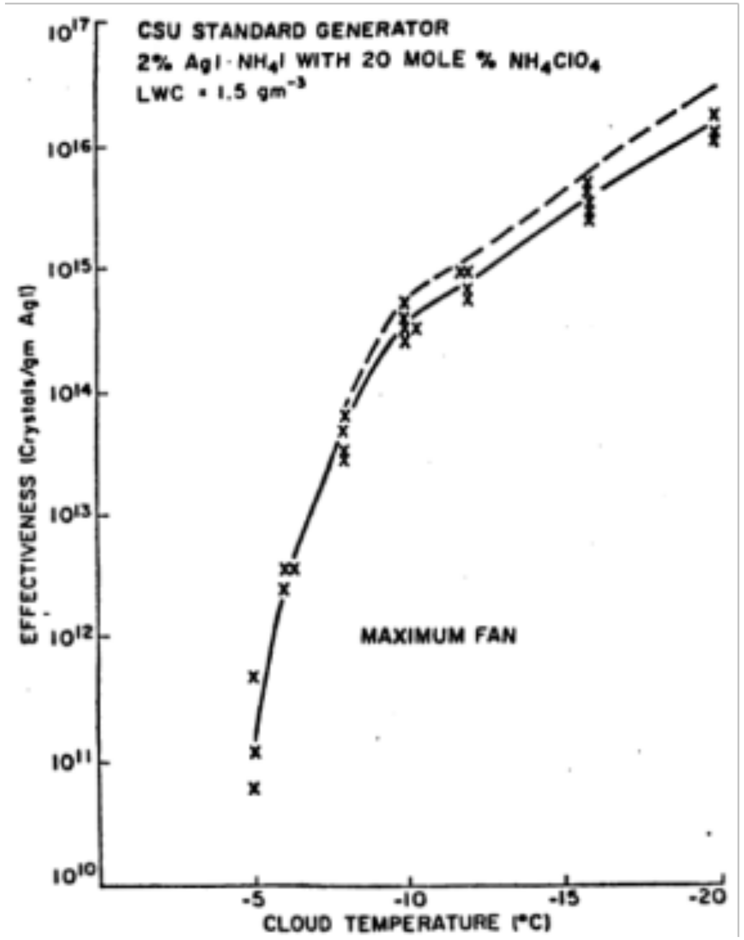


4300 cm^{-3}
2100 cm^{-3}

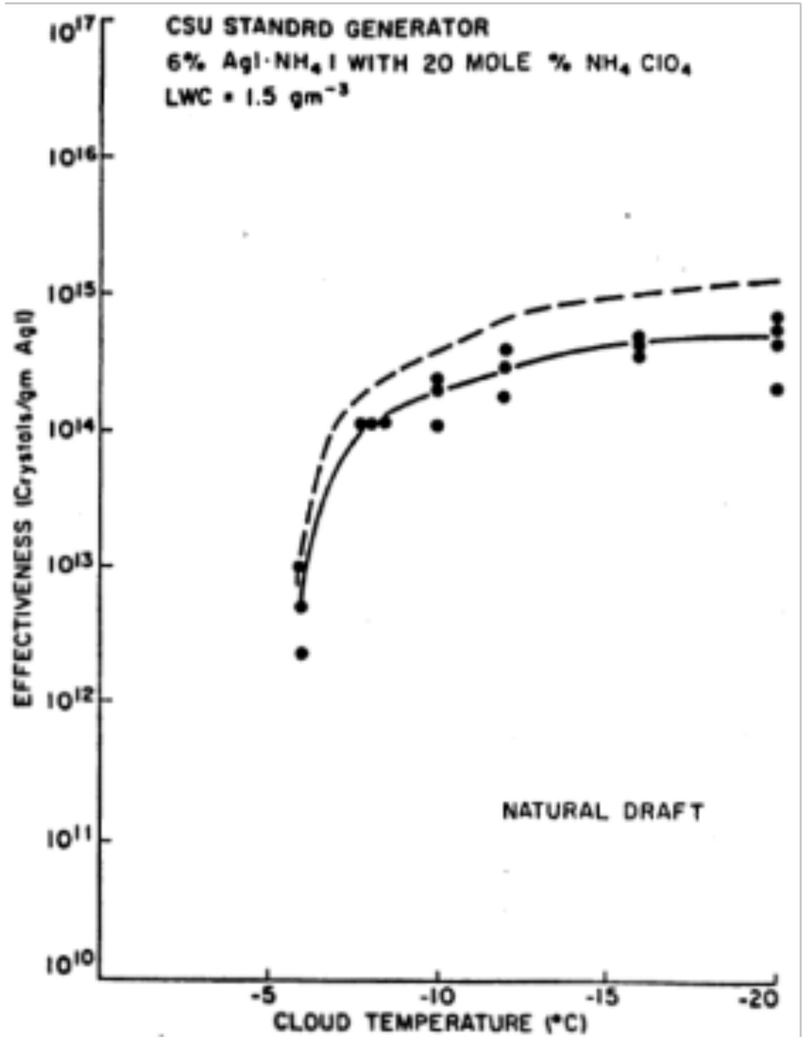
Varied LWC via varied cloud droplet concentration

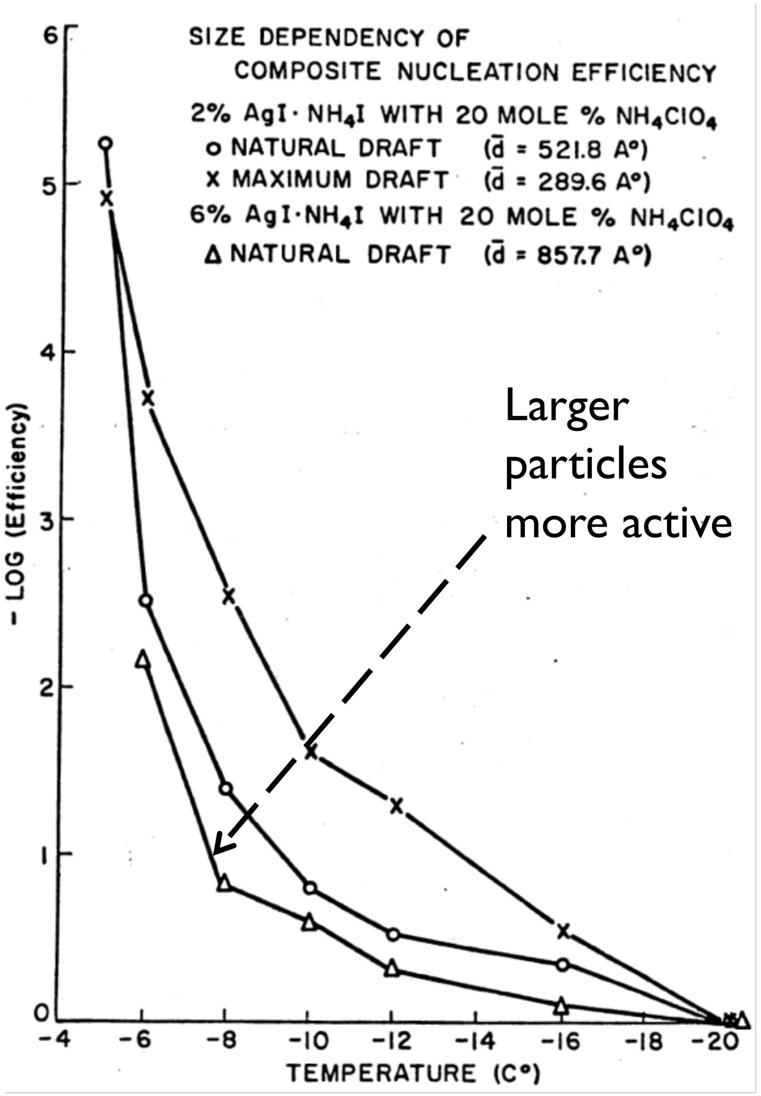
Size distribution impacts Yield for most ice nuclei from combustion – flame quenching impact





Size distribution from combustion can also be controlled by AgI mass → size effects on ice nucleation

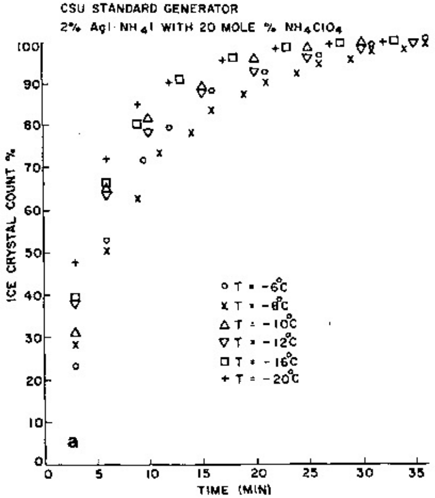


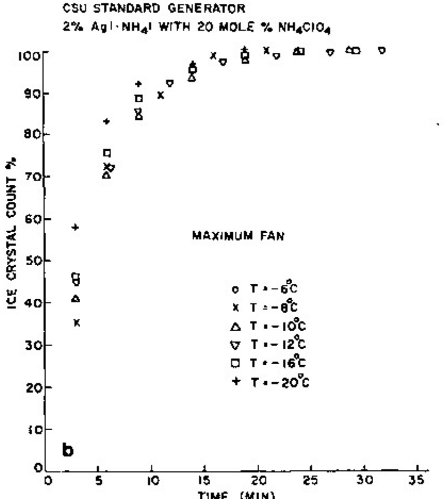


Larger particles more active

Rates and mechanisms of ice formation reflect changes in particle sizes and cloud droplet concentration (LWC)

DeMott et al. (J. Clim. Appl. Meteor., 1983)





Ice formation is faster for smaller particles!

$N_{ICt} = N_{IC\infty}\,(1-\exp(-k_{app}t))$

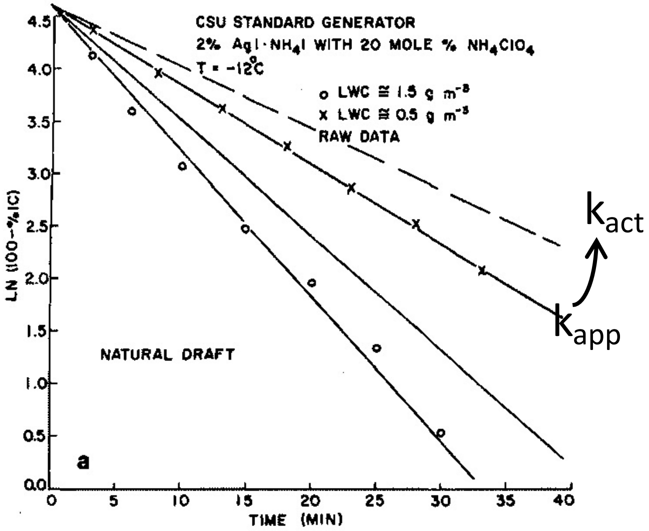
N_{ICt}: cumulative ice crystal count at time t

$N_{IC\infty}$: total count at time infinity

$k_{app} = k_{act} + k_{dilution}$

$\mathrm{Ln}\,(100-\%IC) = -k_{app}\,t$

Ice formation is psuedo-first order process. Faster for higher droplet concentrations!

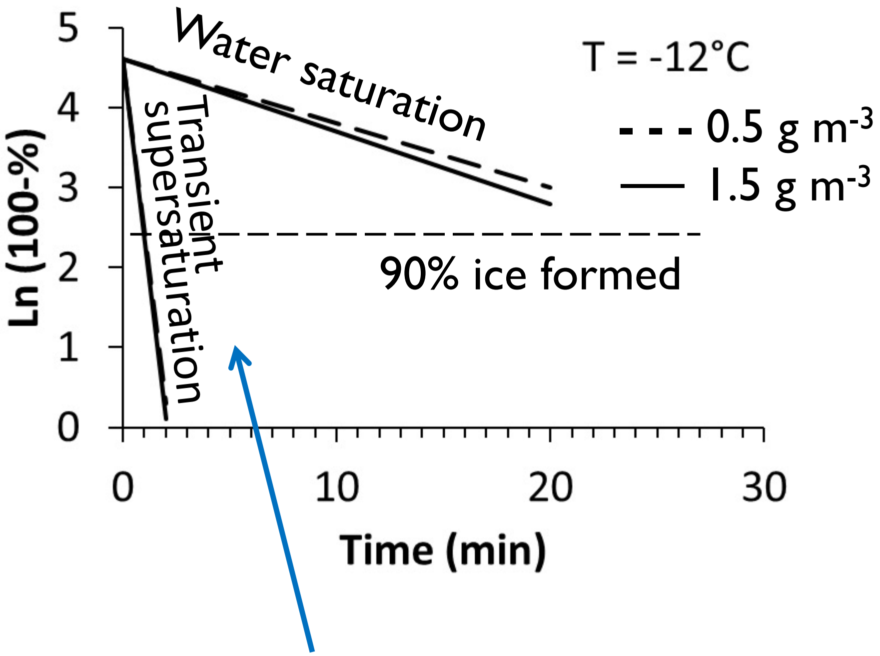


→ Contact freezing by AgI and AgI_xCl_y systems

“Complexed, AgI-alkali halide nuclei systems were “slow” for condensation freezing, motivating designing “fast” hygroscopic AgI_xCl_y-zNaCl system

Slow freezing at water saturation

AgI·NaI complex (Blumenstein et al. 1987)



However…

No sensitivity to droplet concentration → condensation freezing nucleation

Fast freezing at water saturation

AgI-AgCl-4NaCl (Feng Daxiong and W.G. Finnegan, JWMA, 1984)

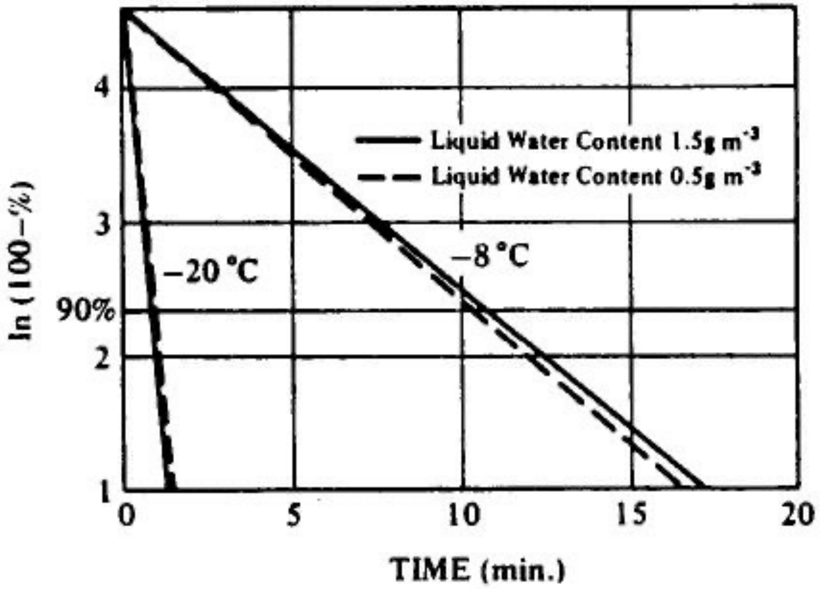
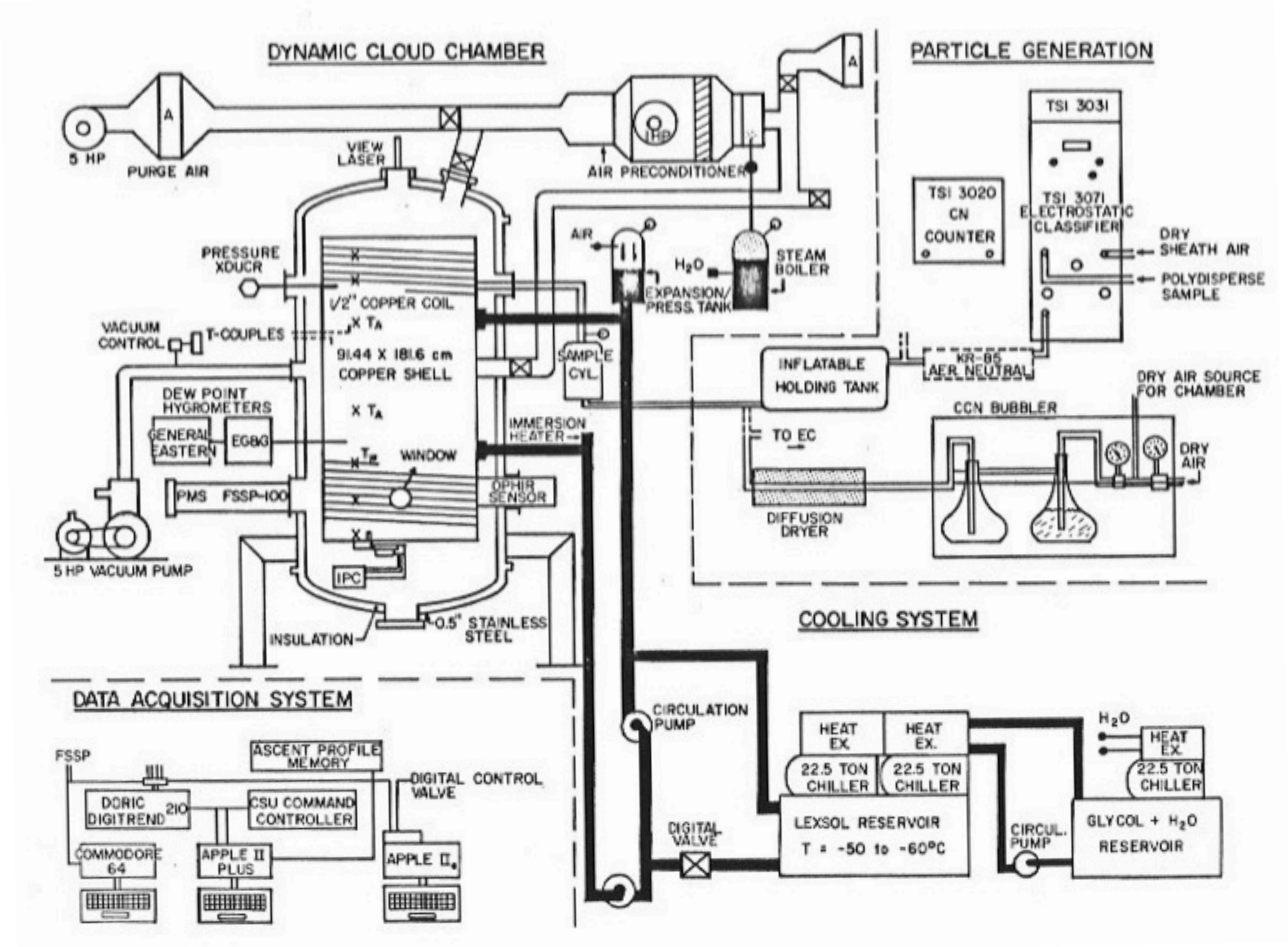
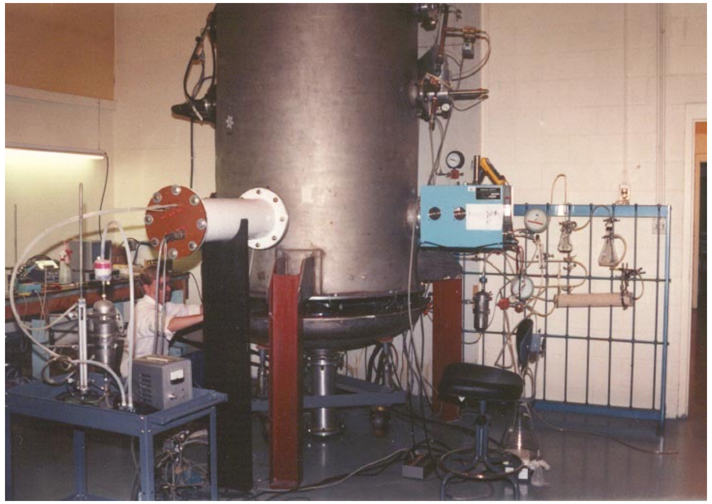


Fig. 3 Rates of depletion of AgI.AgCl-4 NaCl composite nuclei after introduction into the cloud chamber at different liquid water contents.

CSU Dynamic Cloud Chamber

Dynamic cloud chamber (Garvey, 1975; DeMott and Rogers, 1990)

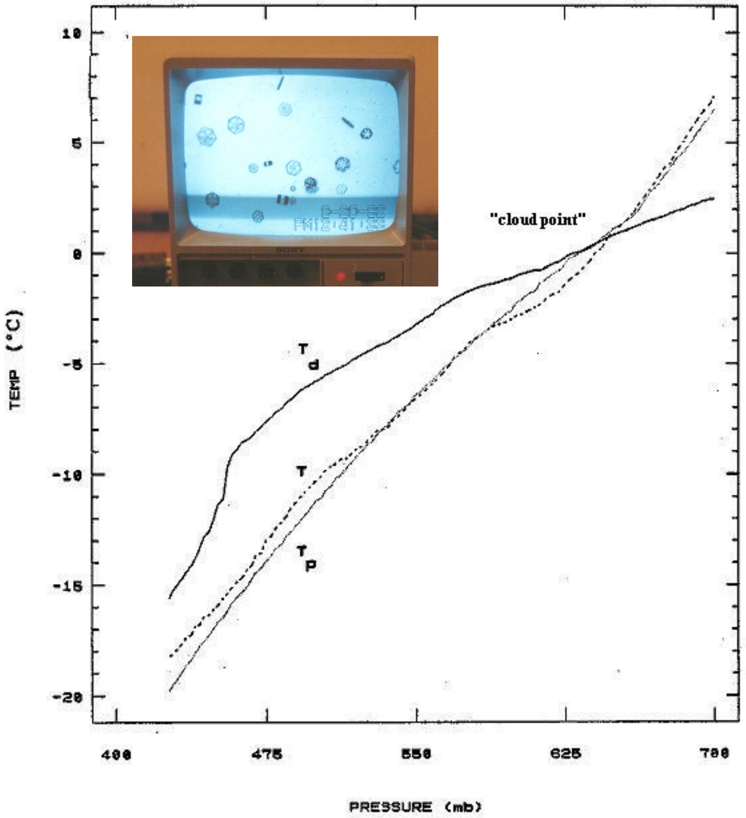


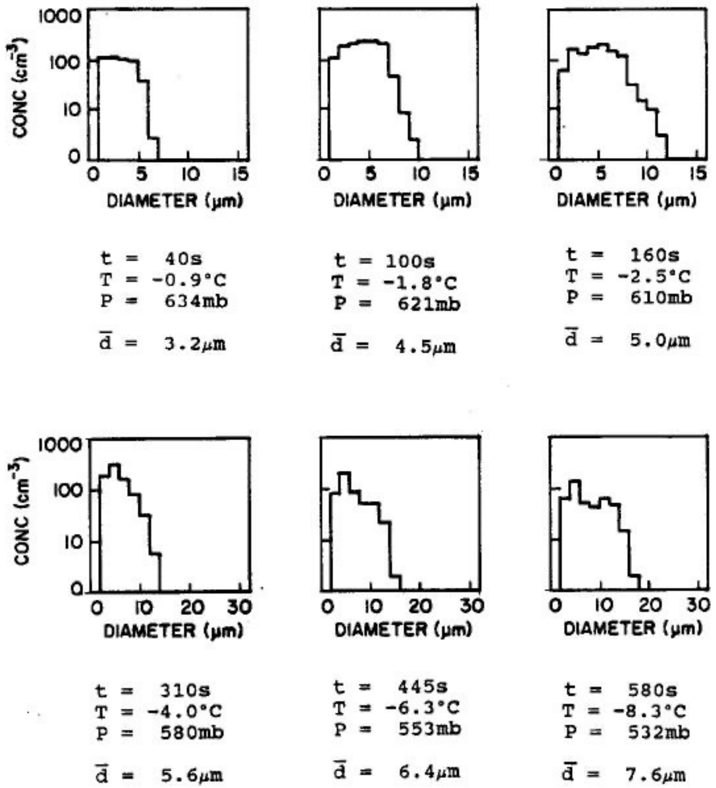


NCAR Electra air sample delivered to laboratory (circa 1992)

CSU Dynamic Cloud Chamber

Could reproduce adiabatic profile through cooling rate to form clouds

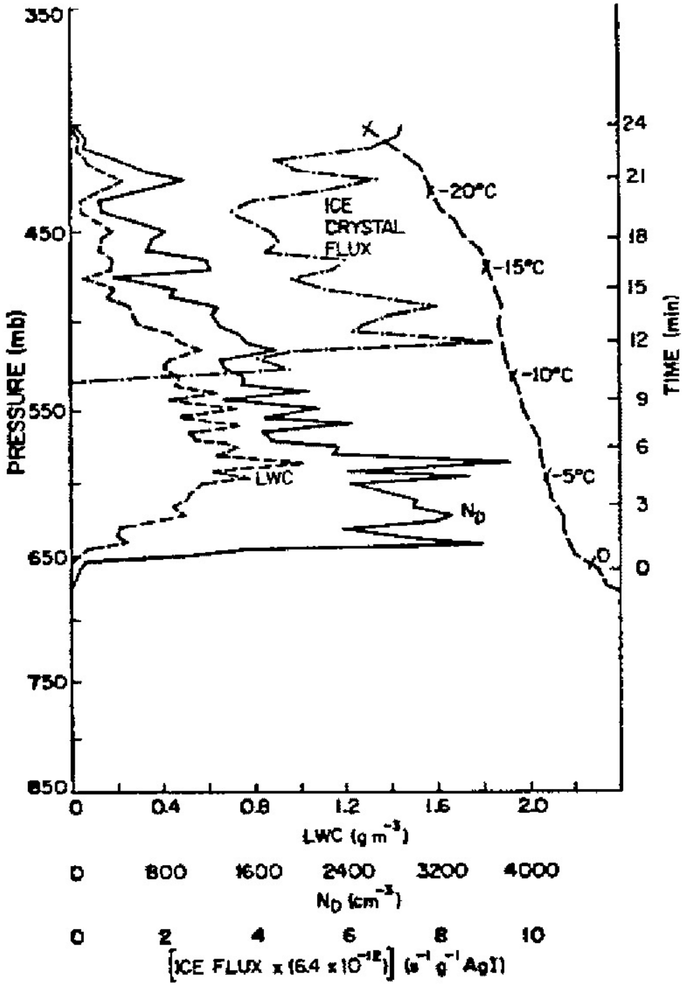




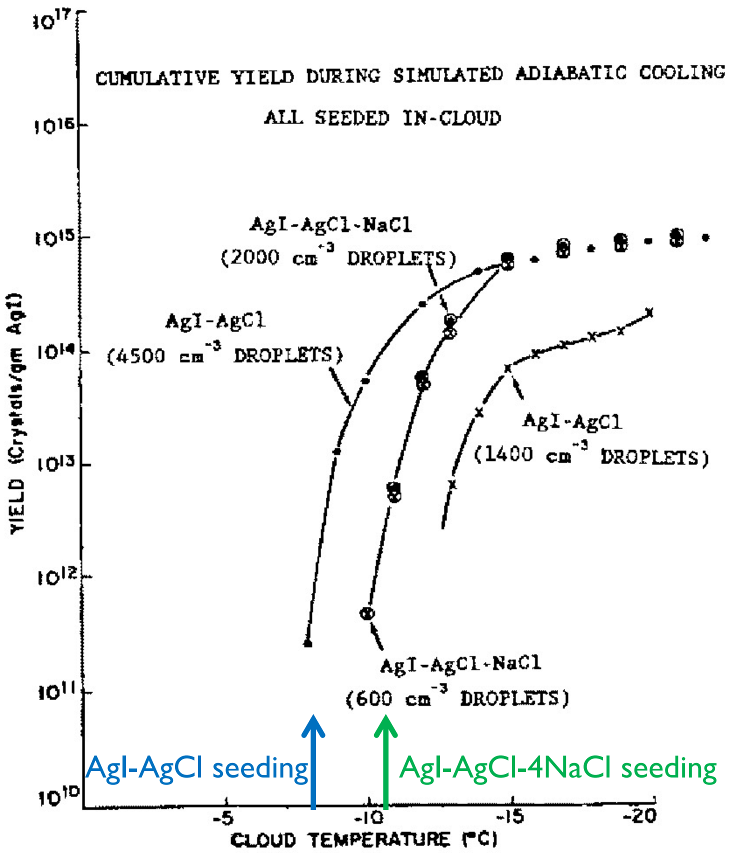
Mechanistic behaviors preserved when AgI-AgCl and AgI-AgCl-NaCl aerosols are seeded into supercooled clouds

Inject
Into
cloud
→

Sulfate
CCN
added
→



DeMott et al. (JWMA1988)



Corroborates primary action by contact freezing versus condensation freezing of the hydrophobic versus hygroscopic systems seeded into a cloud

Supercooled cloud base supersaturations can switch AgI-AgCl mechanism from contact-freezing to condensation/immersion freezing

74 P.J. DeMott / Atmospheric Research 38 (1995) 63–99

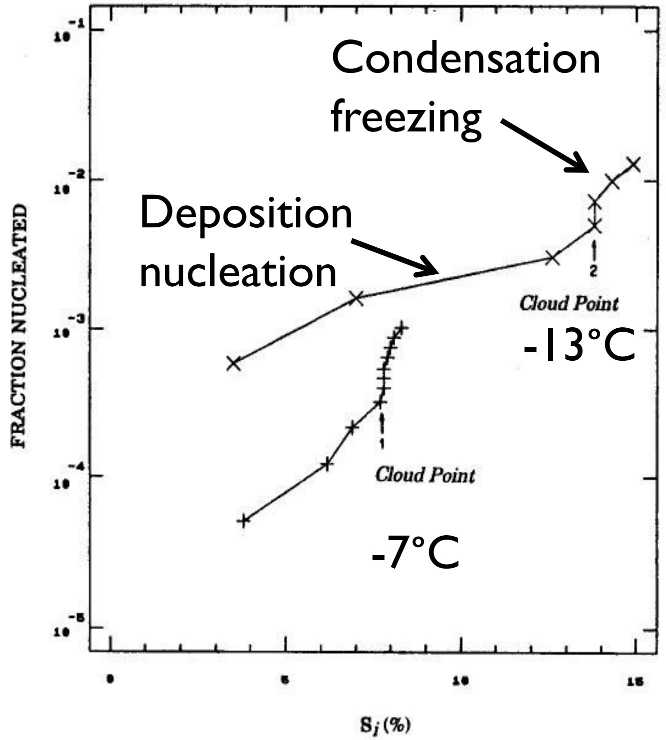
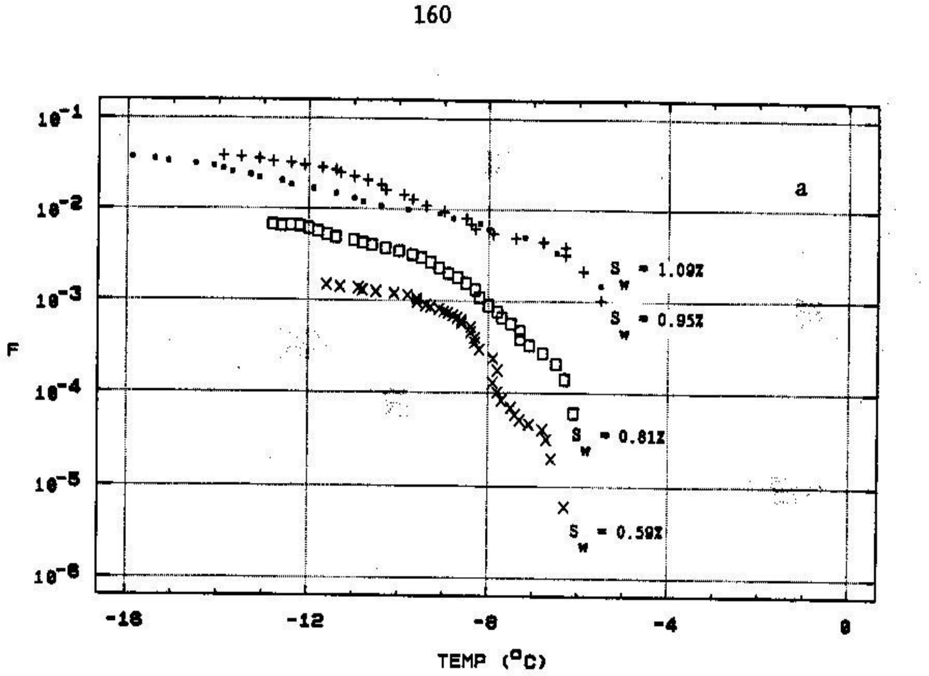


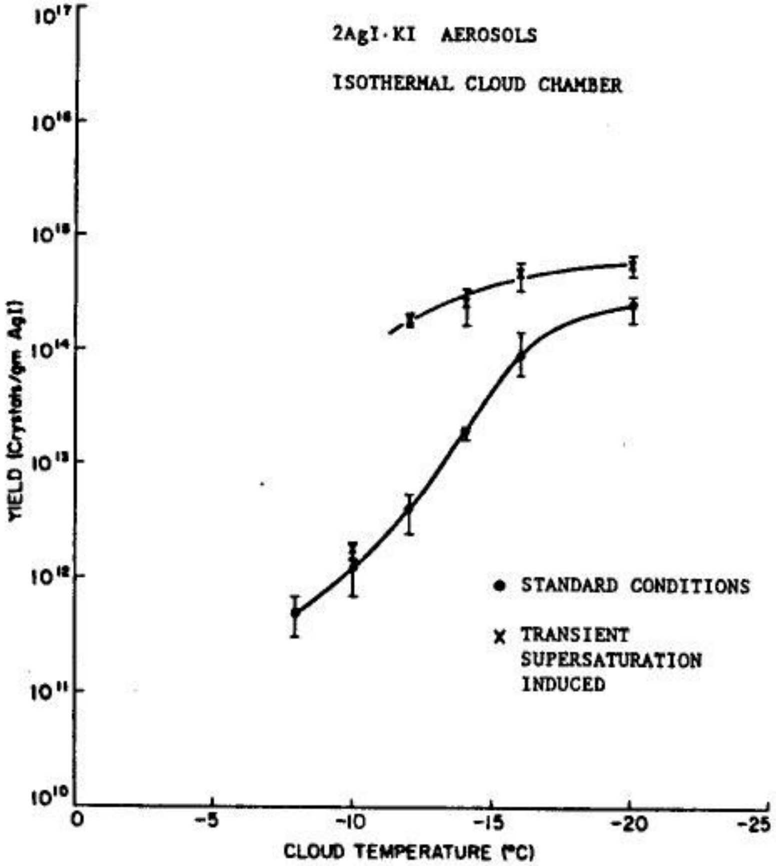
Fig. 2. Fraction of monodisperse AgI–AgCl aerosols nucleated versus ice supersaturation in two continuous expansions. Cloud formation point is indicated by the numbered arrows at $T = -7°C$ (1) and $T = -13°C$ (2). Experiments proceed left to right.



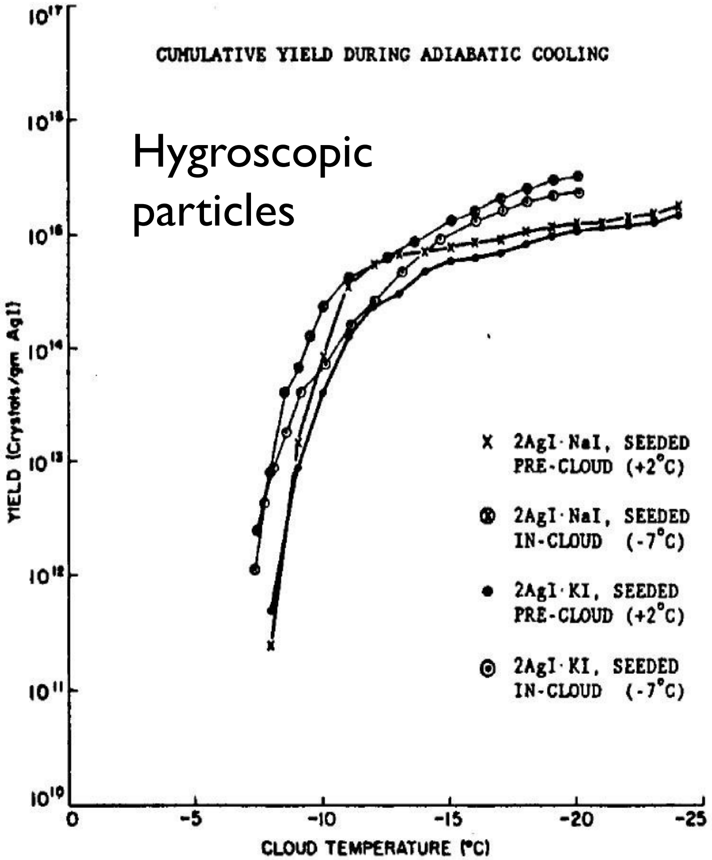
AgI-AgCl are contact freezing nuclei but also express condensation freezing behavior in response to water vapor supersaturations at supercooled cloud base (calculated for varied cooling rate in dynamic chamber).

How seeding agents are “tested” can affect estimate of their utility, as can their conditions of exposure after seeding

Transient water vapor supersaturation enhances yield of “complexed” AgI



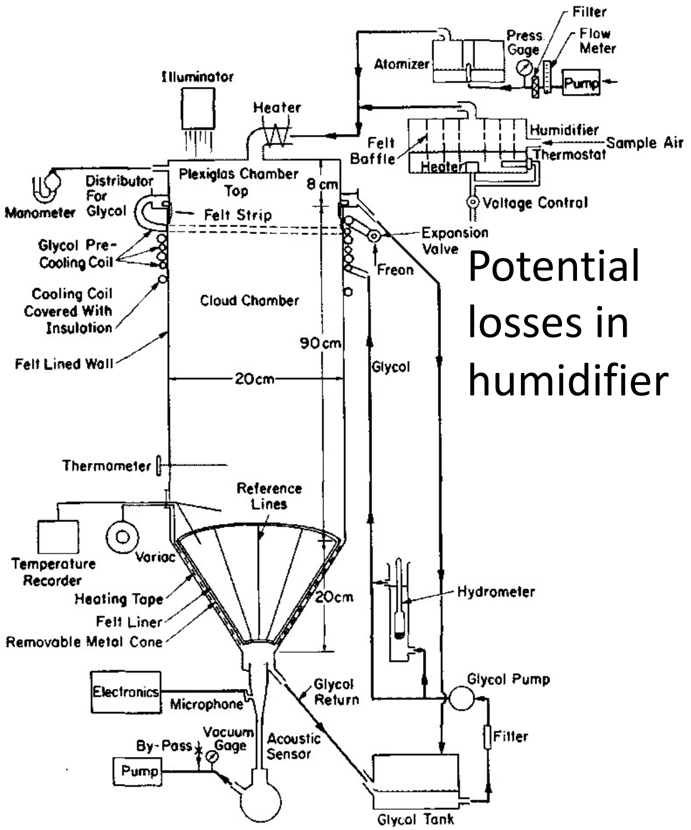
As does low steady state supersaturation in dynamic cloud chamber


Hygroscopic particles

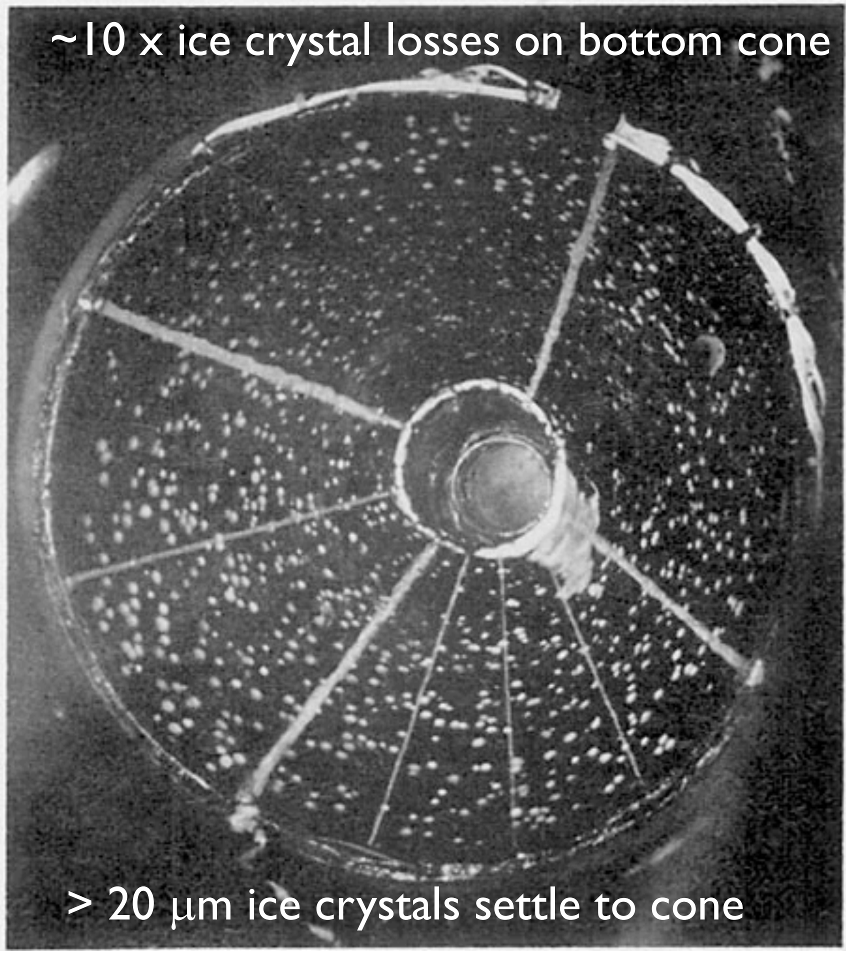
See Blumentstein et al. (JCAM, 1987) for demonstration of impact in 2D orographic cloud scenario.

Special studies at Cloud Simulation and Aerosol Laboratory – comparison of portable instruments to fixed devices (a paradigm carried forward to present day)

Portable INP counters (Langer et al., 1973)



Potential losses in humidifier



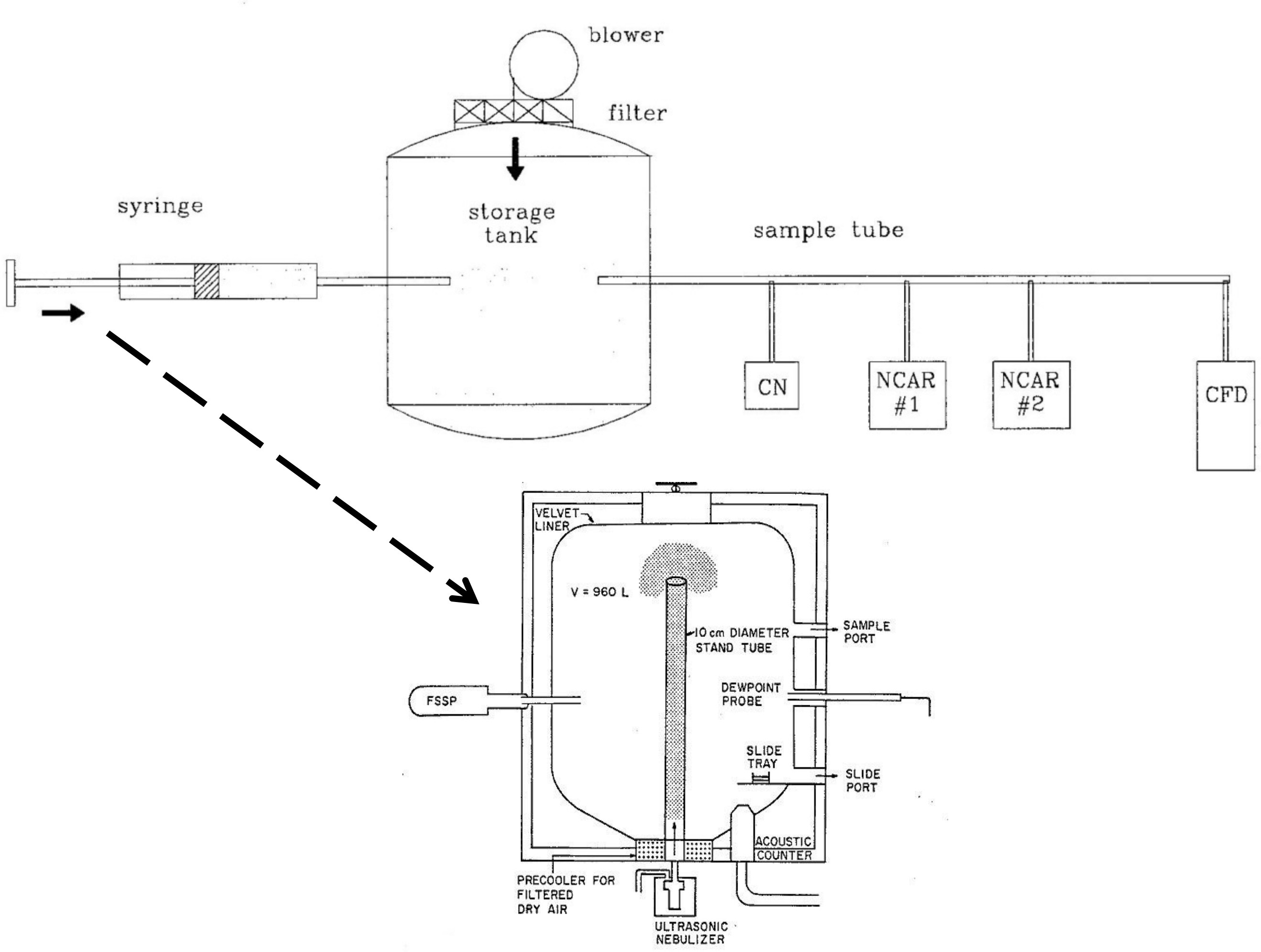
~10 x ice crystal losses on bottom cone

> 20 μm ice crystals settle to cone

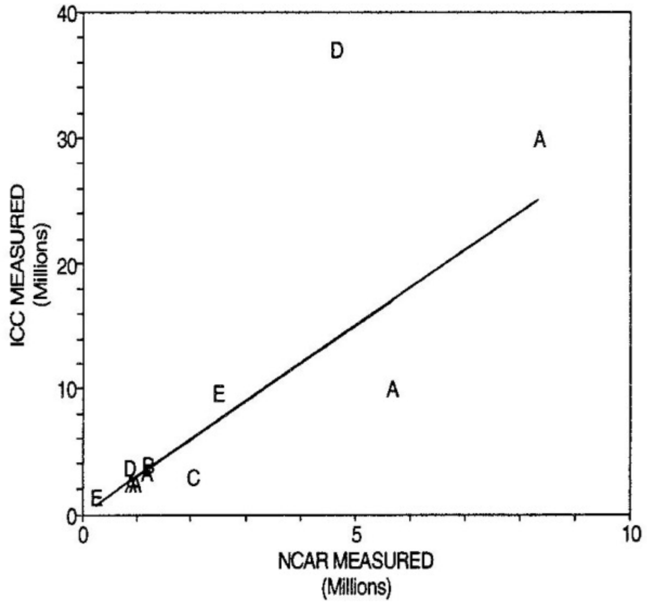
Super et al. (2007)

Jim Heimbach in photo, built these

Comparisons between portable INC and Isothermal cloud chamber gives confidence and calibration (DeMott et al. ,JWMA, 1995)

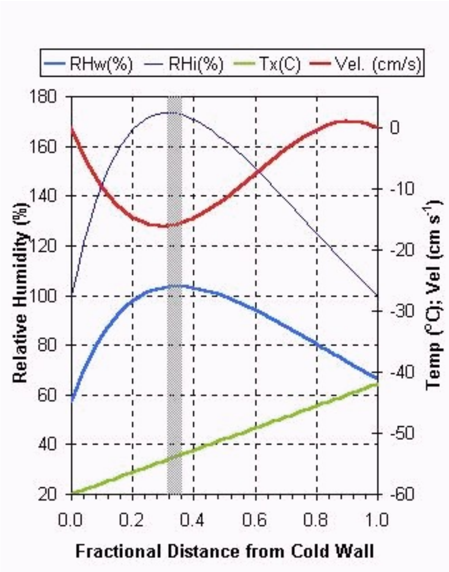


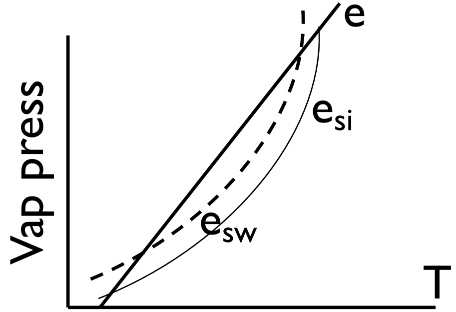
NCAR = ICC * 0.34



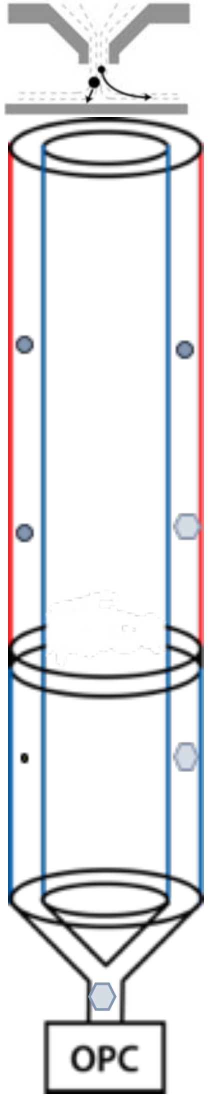
DeMott, P. J., A. B. Super, G. Langer, D. C. Rogers, and J. T. McPartland, 1995: Comparative characterizations of the ice nucleus ability of AgI aerosols by three methods. *J. Wea. Mod.*, **27**, 1-16.

Continuous Flow Diffusion Chamber





See *Rogers et al.* (1988; 2001) for fundamentals



refrigerated walls set processing temperature

temperature gradient controls ice and water supersaturations

warm wall temperature matched to cold wall

ice saturated, water subsaturated

inertial impactor removes particles larger than 1.5 µm

Varied cut-point

↓

particles encounter controlled ice supersaturation

↓

Conditions may also be water supersaturated to activate particles into droplets

↓

a fraction of particles freeze

↓

evaporation section reduces any liquid droplets back to haze sizes

↓

optical counting of ice crystals

IN measurement by CFDC as function of supersaturation. These instruments are now well understood and mature

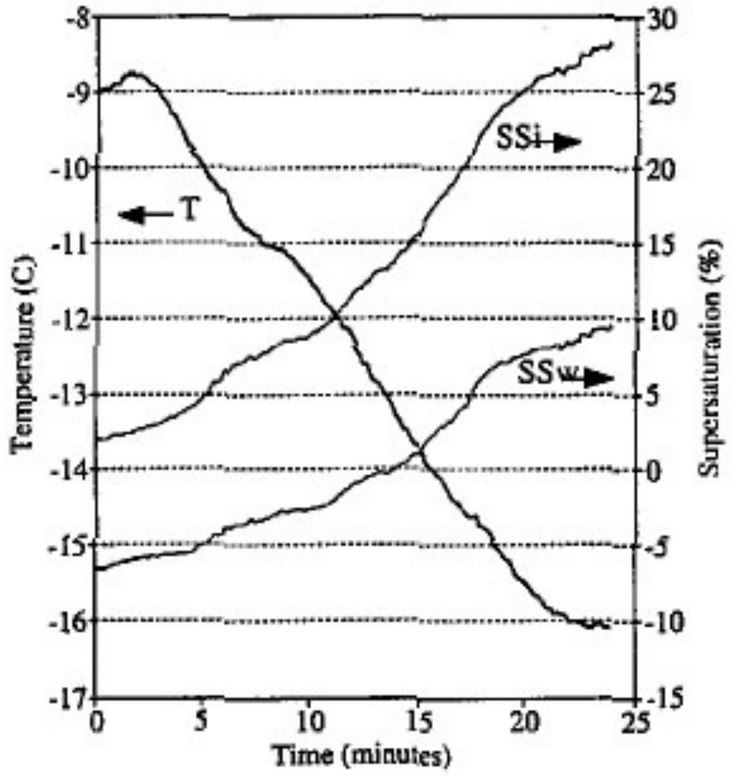


Figure 9. History of temperature, SSw, and SSi at the location of the AgI aerosol in the CFD.

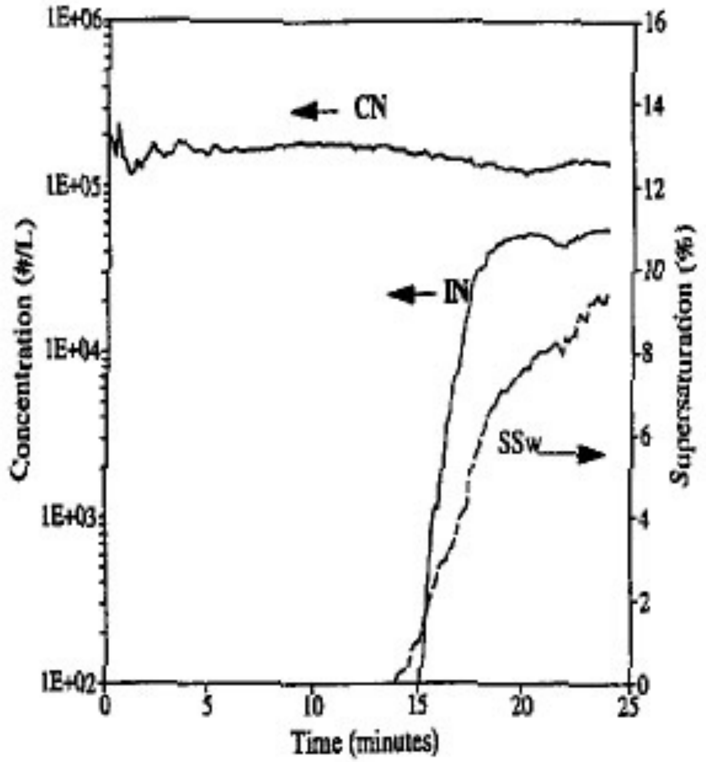


Figure 10. History of particle concentrations and water supersaturation in CFD experiment. Note that IN and SSw traces are in phase.

40% active fraction within factor 2 of ICC values

Future Needs, Possibilities, Recommendations for cloud seeding laboratory studies applications

- Need to understand natural ice nucleation processes in order to place seeding in context
- More realistic testing of INP behavior should be done
    - Lab tests designed to mimic generation conditions as well as considering all IN mechanisms
    - Detailed modeling of dynamics of aerosol generation like what has been done for aircraft engines?
- Major facilities for research and testing seeding agents are limited or no longer active to quality control product
    - AgI solution combustion and pyrotechnic generation methods have probably been exploited to their maximum extent, though not fundamentally understood (opinion)

Future Needs, Possibilities, Recommendations for cloud seeding laboratory studies applications

  - Specialized new INPs with high ice formation efficiency and rates should be investigated (coatings, biological)
- Models should include detailed information on nucleation processes, so that seeding hypotheses can be tested
- In-situ INP detection as part of field research programs would be beneficial and the technology is ready

Non-weather modification related studies at CSAL cloud chamber: Homogeneous and heterogeneous freezing

Drop diam.
Freezes in 1 s

DeMott and Rogers (J. Atmos. Sci., 1990)

5 µm →
20 µm →
35 µm →

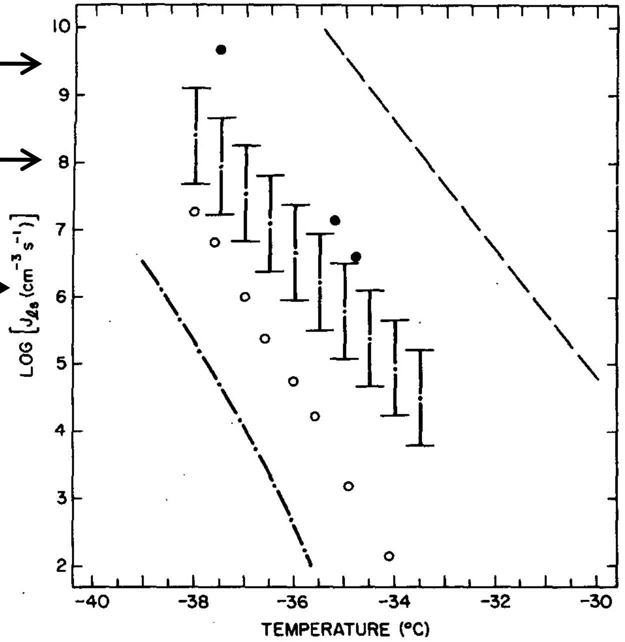
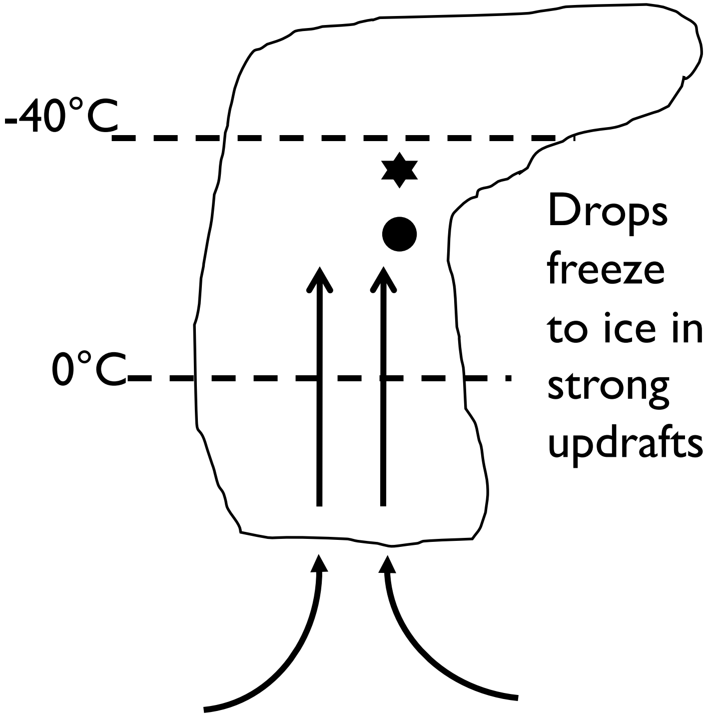


FIG. 9. Average J_{ls} versus temperature from this study is plotted (with uncertainties given by bars) for comparison with results extrapolated from Hagen et al. (1981) (– – –), from Sassen and Dodd (1988) (●), from Butorin and Skripov (1972) (○), and from theoretical homogeneous-freezing nucleation (Pruppacher and Klett 1978) (—●—).



Drops freeze to ice in strong updrafts

Acetylene soot freezing as a function of updraft

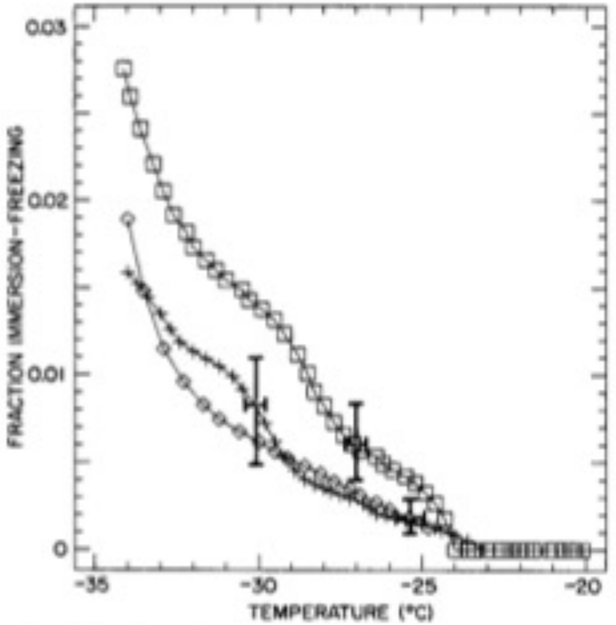


FIG. 5. Fractions of soot nucleated as ice with respect to the numbers of aerosol immersed in cloud droplets for 0.08 µm soot in 1°C min^{-1} (—+—) and 2°C min^{-1} (—◇—) cooling rate expansions, and for 0.12 µm (actual diameter, not mobility diameter) soot in 1°C min^{-1} cooling rate expansion (—□—). Uncertainties are noted by horizontal and vertical bars at one data point in each experiment.

CSU laboratory studies (natural aerosol activation and freezing; instrument testing)

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Petters, M. D., M. T. Parsons, A. J. Prenni, P. J. DeMott, S. M. Kreidenweis, C. M. Carrico, A. P. Sullivan, G. R. McMeeking, E. Levin, C. E. Wold, J. L. Collett, Jr., and H. Moosmüller, 2009: Ice nuclei emissions from biomass burning. J. Geophys. Res., 114, D07209, doi: 10.1029/2008JD011532.

Prenni, A. J., M. D. Petters, A. Faulhaber, C. M. Carrico, P. J. Ziemann, S. M. Kreidenweis and P. J. DeMott, 2009: Heterogeneous ice nucleation measurements of secondary organic aerosol generated from ozonolysis of alkenes, Geophys. Res. Lett., 36, L06808, doi:10.1029/2008GL036957.

Popovicheva O. B., Persiantseva N. M., Shonija N. K., P. J. DeMott, K. Koehler, M. D. Petters, S. M. Kreidenweis, V. Tishkova, B. Demirdjian, J. Suzanne, 2008: Water interaction with hydrophobic and hydrophilic soot particles. Phys. Chem. Chem. Phys., 10, 2332 – 2344 (DOI:10.1039/B718944N).

Sullivan, R. C., M. D. Petters, P. J. DeMott, S. M. Kreidenweis, H. Wex, D. Niedermeier, F. Stratmann, P. Reitz, and J. Schneider, 2010: Irreversible loss of ice nucleation surface sites in mineral dust particles induced by sulphuric acid condensation. Atmos. Chem. Phys., 10, 11471-11487.

Sullivan, R. C., L. Miñambres, P. J. DeMott, A. J. Prenni, C. M. Carrico, E. J.T. Levin, and S. M. Kreidenweis, 2010: Chemical processing does not always impair heterogeneous ice nucleation of mineral dust particles. Geophys. Res. Lett., 37, L24805, doi:10.1029/2010GL045540.

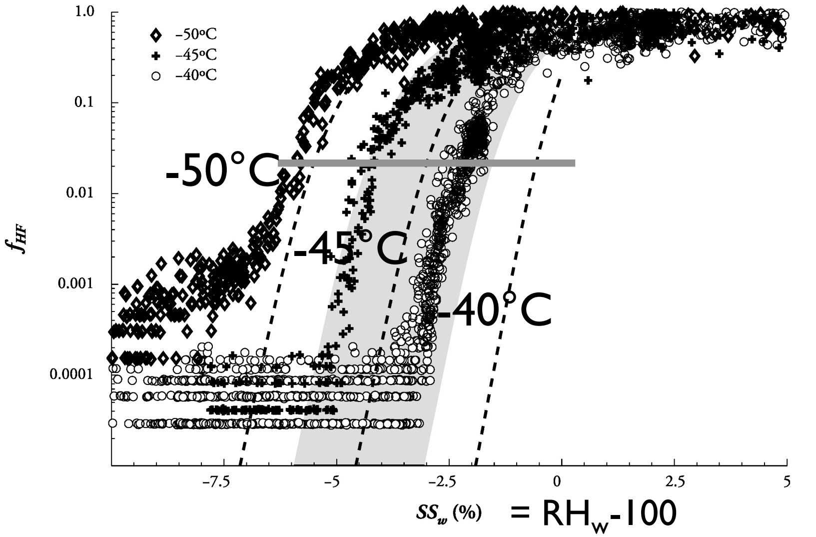
Laboratory-field interface for ice nucleation studies

There is a continuing need to understand our ability to measure ice nucleation and capture INP properties via online and offline methods at a variety of tropospheric conditions, with different ways of capturing INPs, different activation times, capturing different aerosol systems etc…

Low temperature (e.g., cirrus) CFDC applications for a variety of relevant aerosol systems – as for any large cloud chamber

Natural air from outside of home laboratory

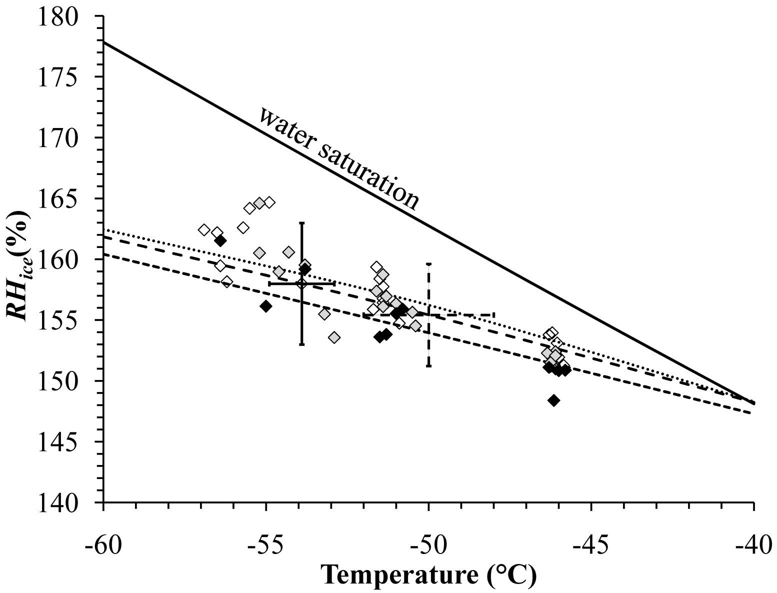
- *Richardson* et al. (2010) [Copyright [2010] American Geophysical Union. Reproduced by permission of American Geophysical Union.]



Freezing fraction of ambient particles at indicated T versus water activity prediction of homogeneous freezing (dashed).

USFS Fire Sciences Laboratory

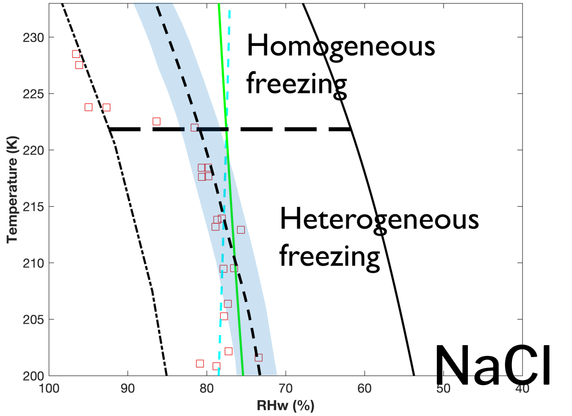
DeMott, P. J., et al., Ice nucleation behavior of biomass combustion particles at cirrus temperatures, J. Geophys. Res. 114: D16205, doi:10.1029/2009JD012036, 2009. Copyright [2009] American Geophysical Union. Reproduced by permission of American Geophysical Union.

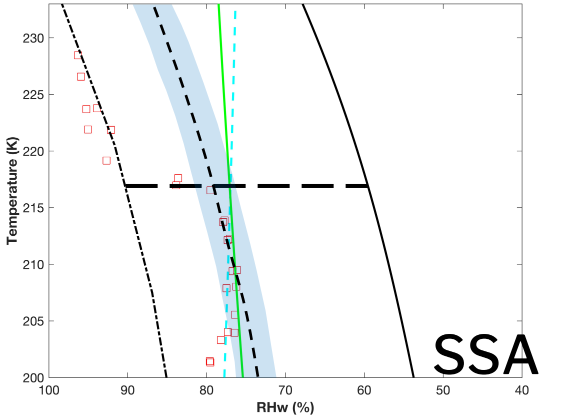


Conditions of 1% particles freezing for biomass combustion particles versus water activity prediction of homogeneous freezing (dashed lines)

Seawater sea spray particles

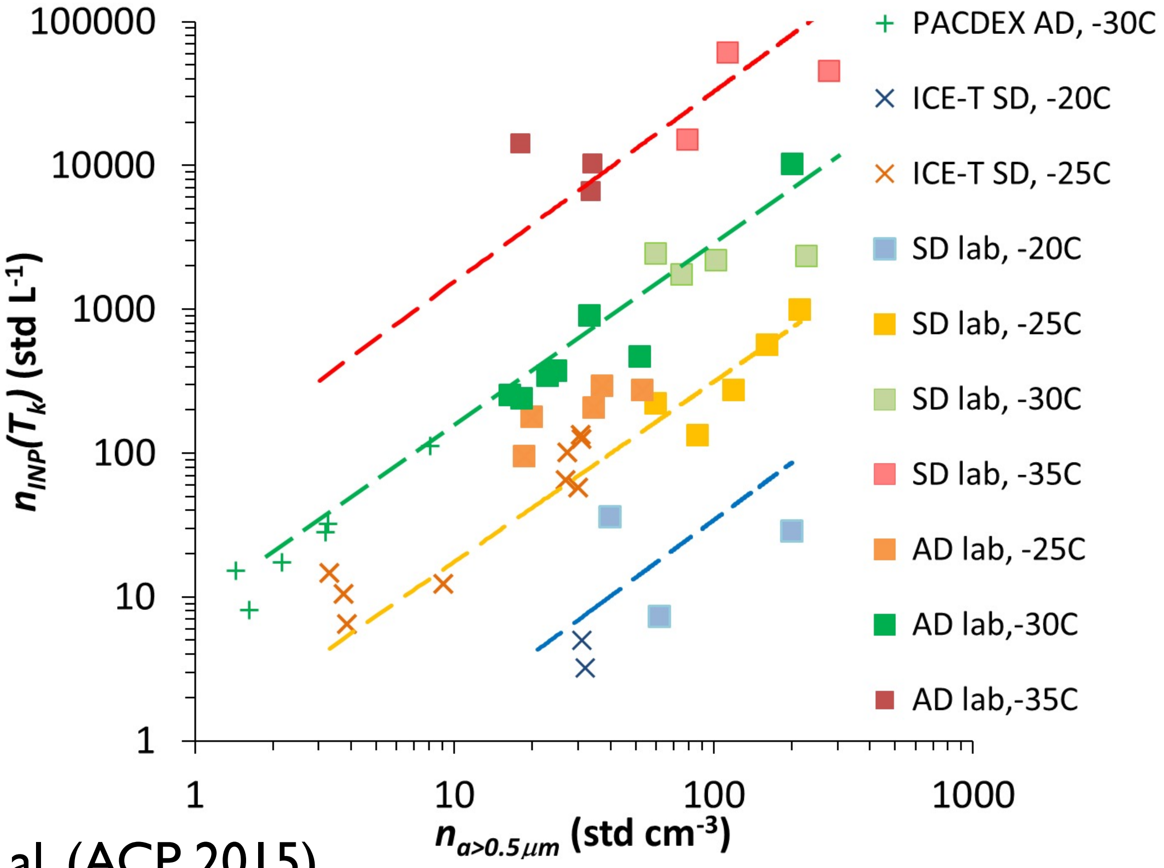
Patnaude, R., et al. (2021), Is ice formation by sea spray particles at cirrus temperatures controlled by crystalline salts? *ACS Earth and Space Chemistry*, Article ASAP, https://doi.org/10.1021/acsearthspacechem.1c00228





Development of empirical parameterizations for mixed-phase clouds through combining lab and field measurements

$$n_{INP,T_k} = (cf)\,(n_{a>0.5\,\mu m})^{(\alpha(273.16-T_k)+\beta)}\exp(\gamma(273.16-T_k)+\delta)$$

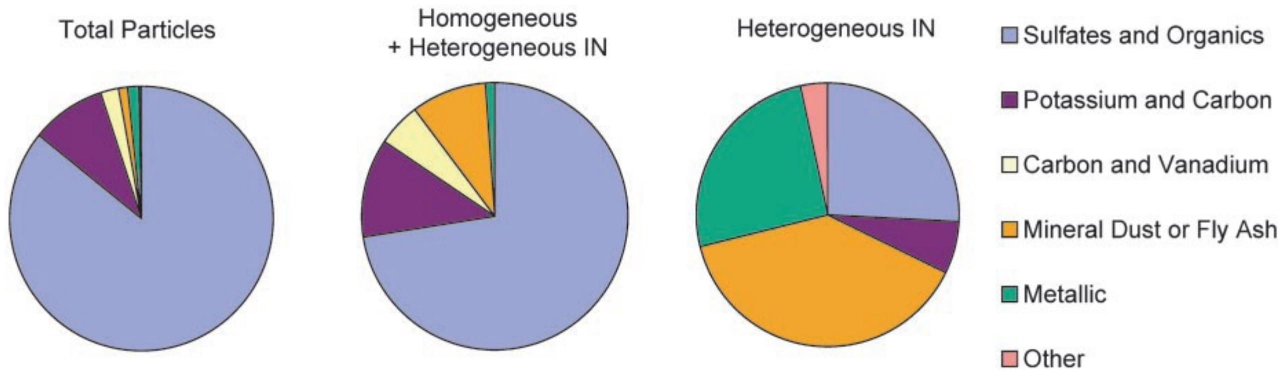


CFDC measurements at mixed phase cloud temperatures of regional mineral dust in the laboratory at KIT compared to field data in aerosol layers from Saharan and Asian deserts

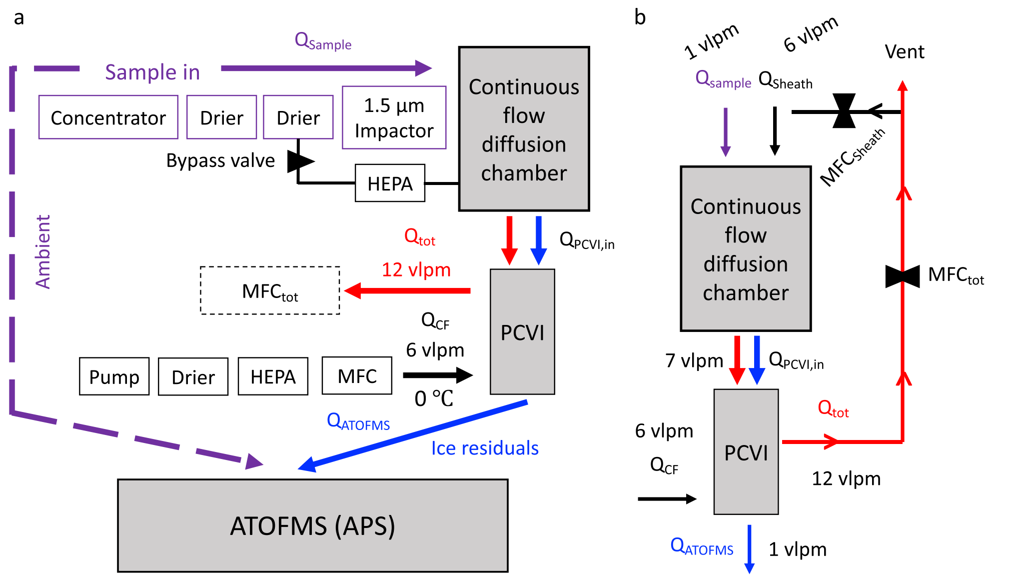
DeMott et al. (ACP, 2015)

Development and testing of systems for characterizing INP composition (indoor/outdoor laboratories)

Single particle laser ablation mass spectrometry of INPs using pumped CVI (*DeMott et al. 2003b*)



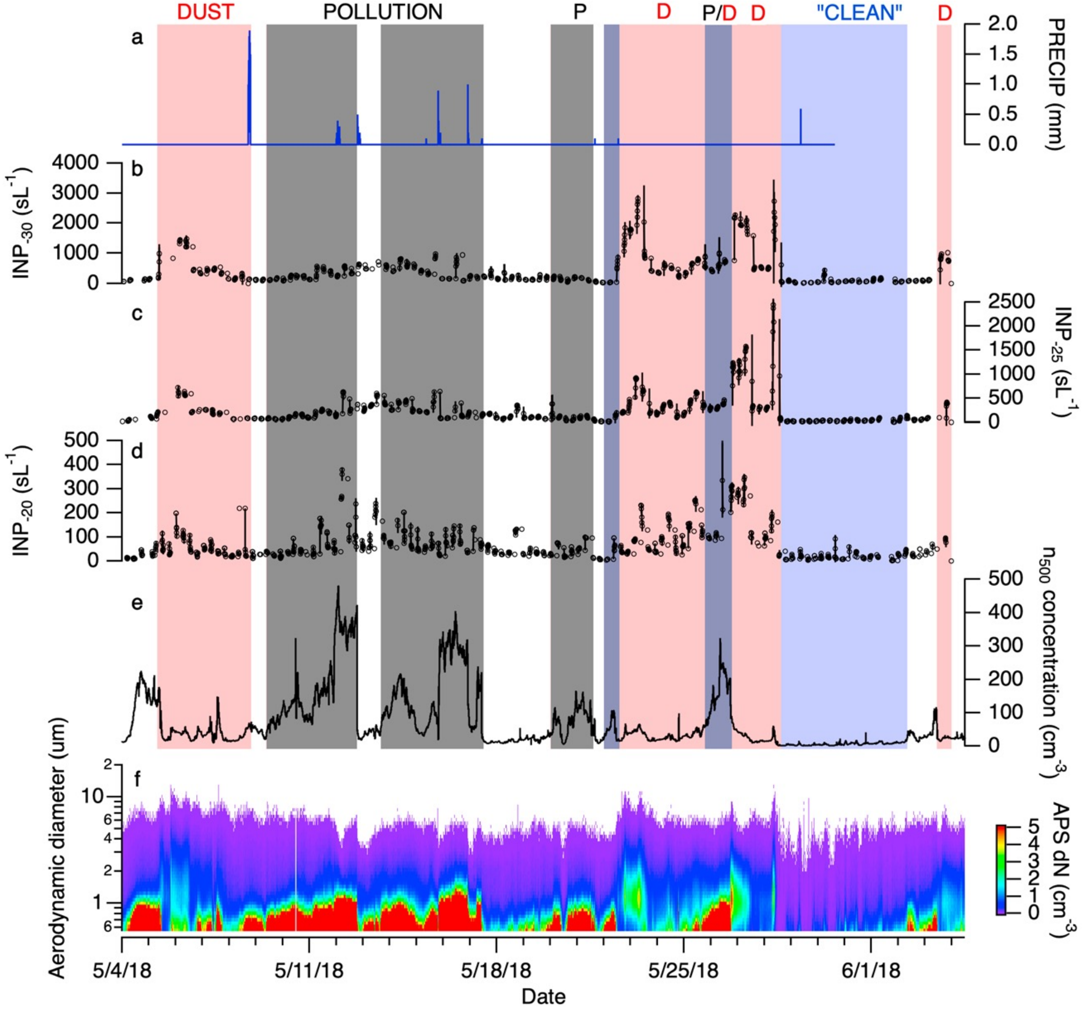
Cornwell, G. C., McCluskey, C. S., Levin, E. J. T., Suski, K. J., DeMott, P. J., Kreidenweis, S. M., & Prather, K. A. (2019). Direct online mass spectrometry measurements of ice nucleating particles at a California coastal site. Journal of Geophysical Research: Atmospheres, 124, 12,157–12,172. https://doi.org/10.1029/2019JD030466



Impaction collection for TEM analyses

Kreidenweis, S. M., Y. Chen, P. J. DeMott, and D. C. Rogers, 1998: Isolating and identifying atmospheric ice-nucleating aerosols: A new technique. *Atmospheric Research*, **46**, 263-278.

New CFDC developments (automation and commercialization)



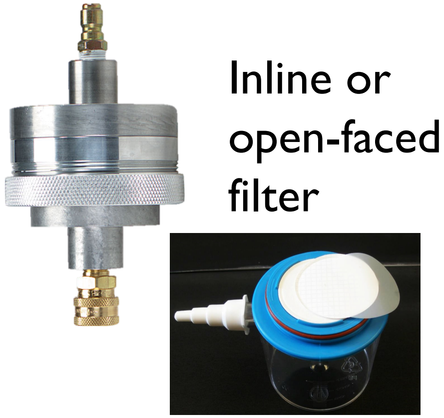
New aircraft CFDCs:

- For mixed phase conditions
- For cirrus conditions

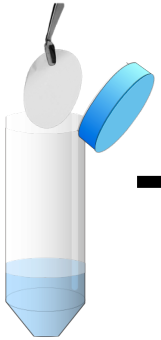
Bi, K et al., 2019: Measurements of ice nucleating particles in Beijing, China. Journal of Geophysical Research: Atmospheres, 124, 8065–8075, https://doi.org/10.1029/2019JD030609.

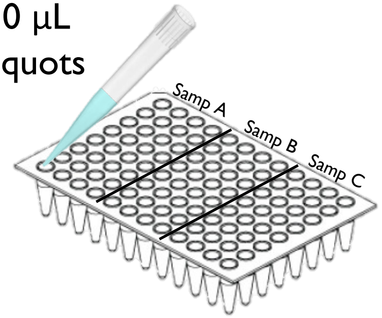
Offline (post processing of filters) immersion freezing INP measurement with the CSU Ice Spectrometer (IS) and similar devices

Methods: Tom Hill (CSU)



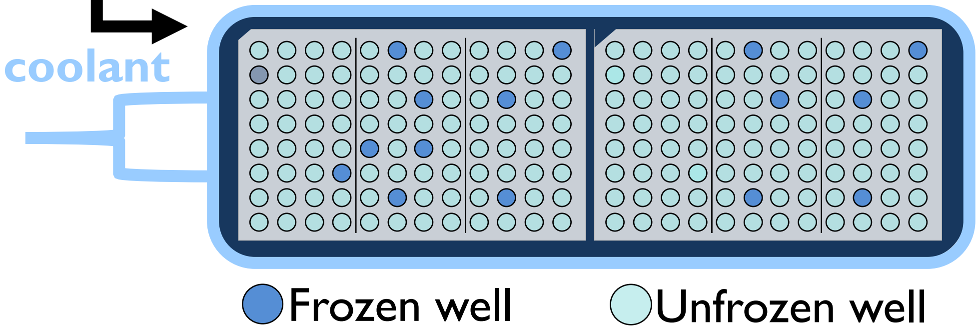
Inline or
open-faced
filter





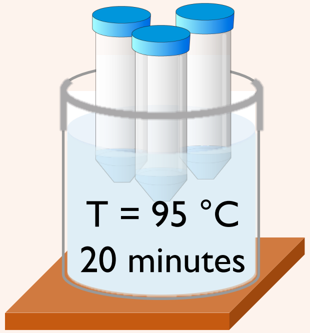
50 µL
aliquots

Samples cooled at 0.33°C min^{-1} and frozen wells recorded until ~ -28 °C



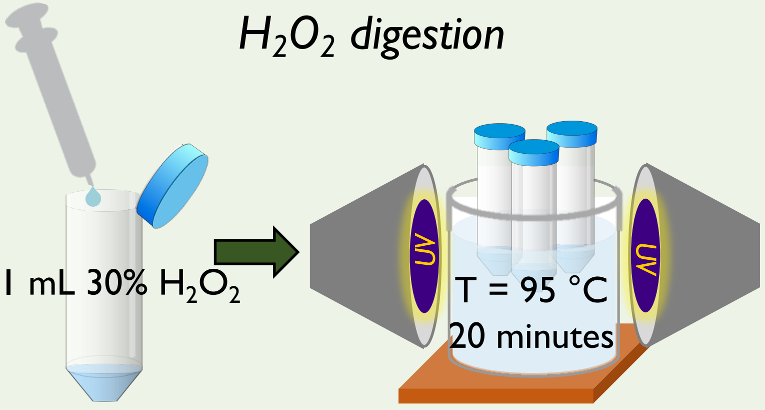
Offline treatments:

Heat



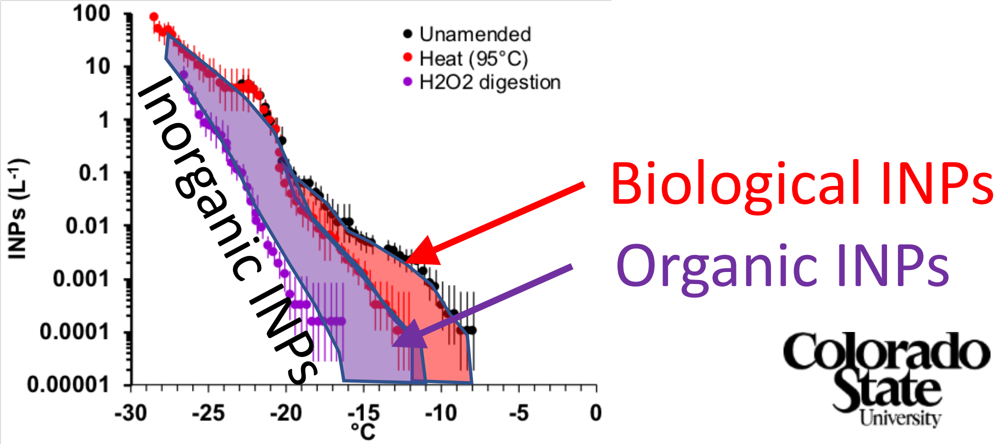
T = 95 °C
20 minutes

H_2O_2 digestion



+ 1 mL 30% H_2O_2

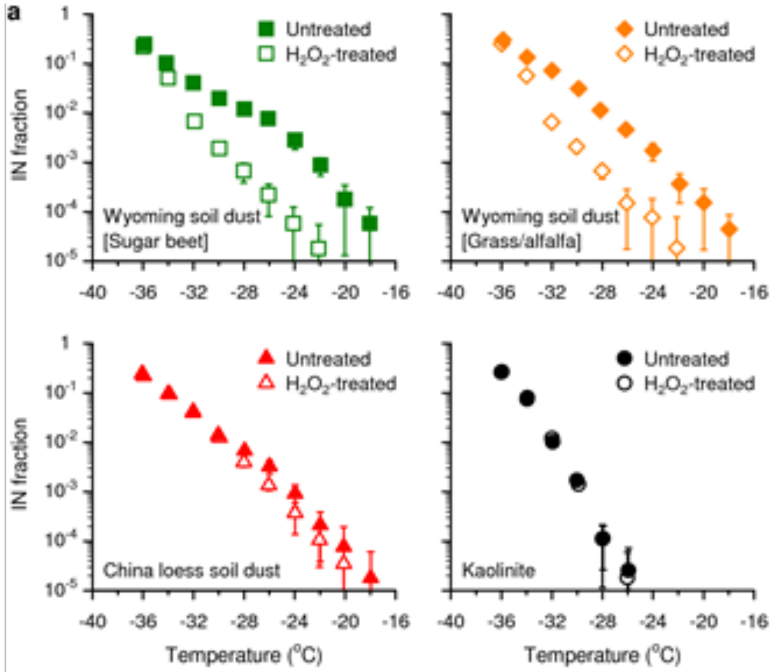
T = 95 °C
20 minutes

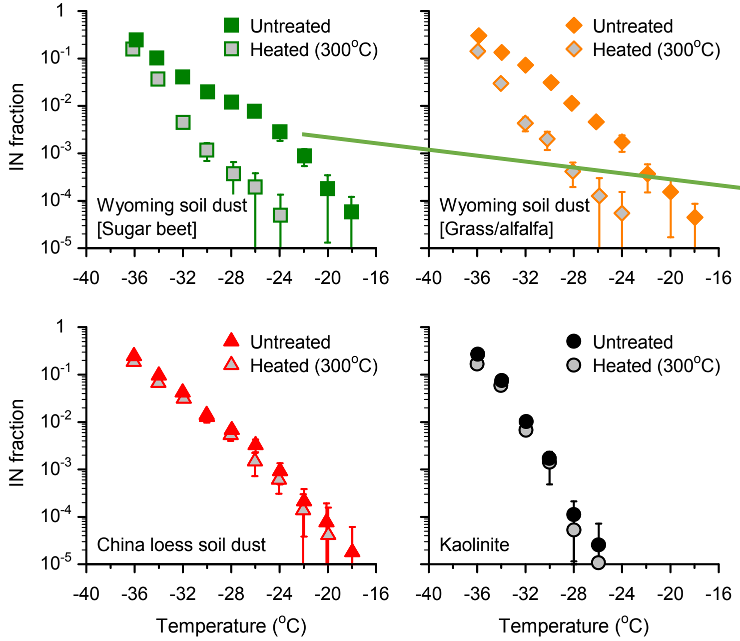


Biological INPs

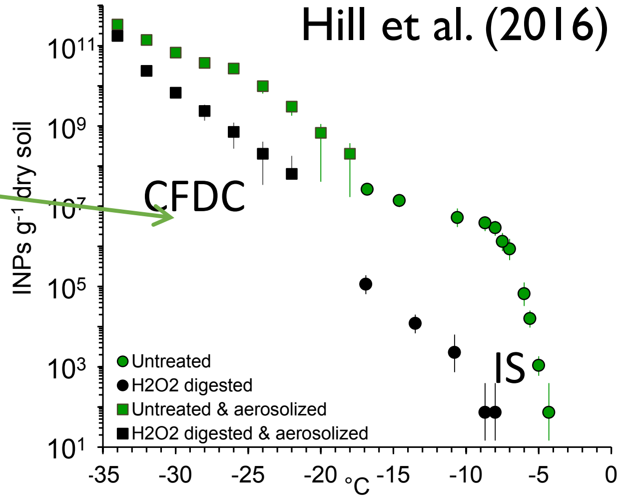
Organic INPs

Soil particles tested in laboratory examining the ability H2O2 digestions and high heat to target organic C-containing INPs

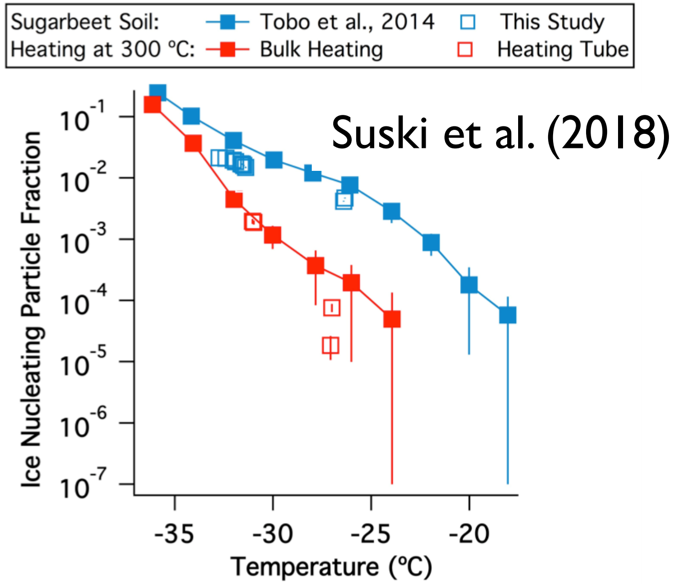




Tobo et al. (2014): CFDC tests of equivalence of heat and peroxide impact on INPs from bulk soil particles and desert dusts



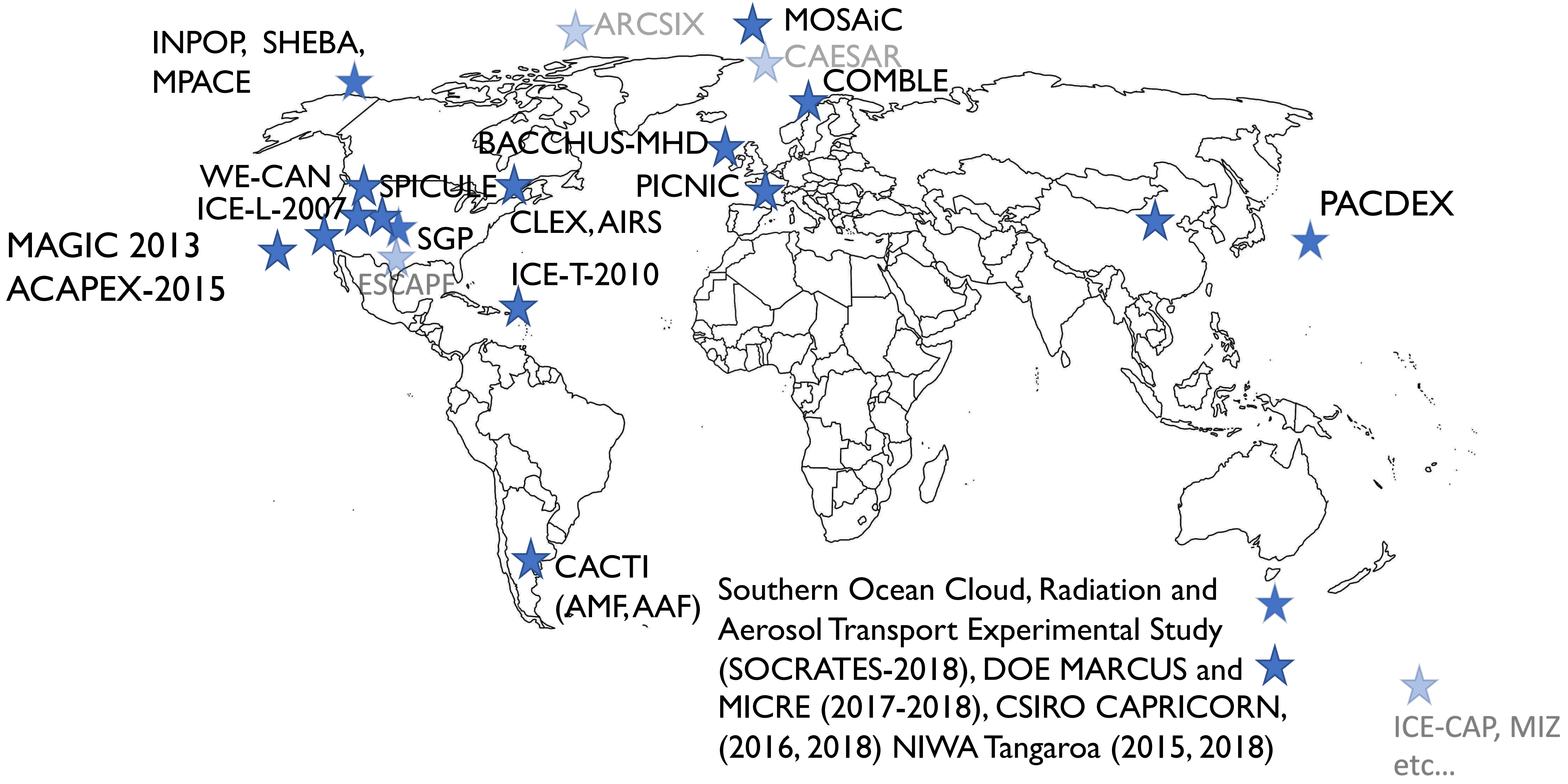
Hill et al. (2016)



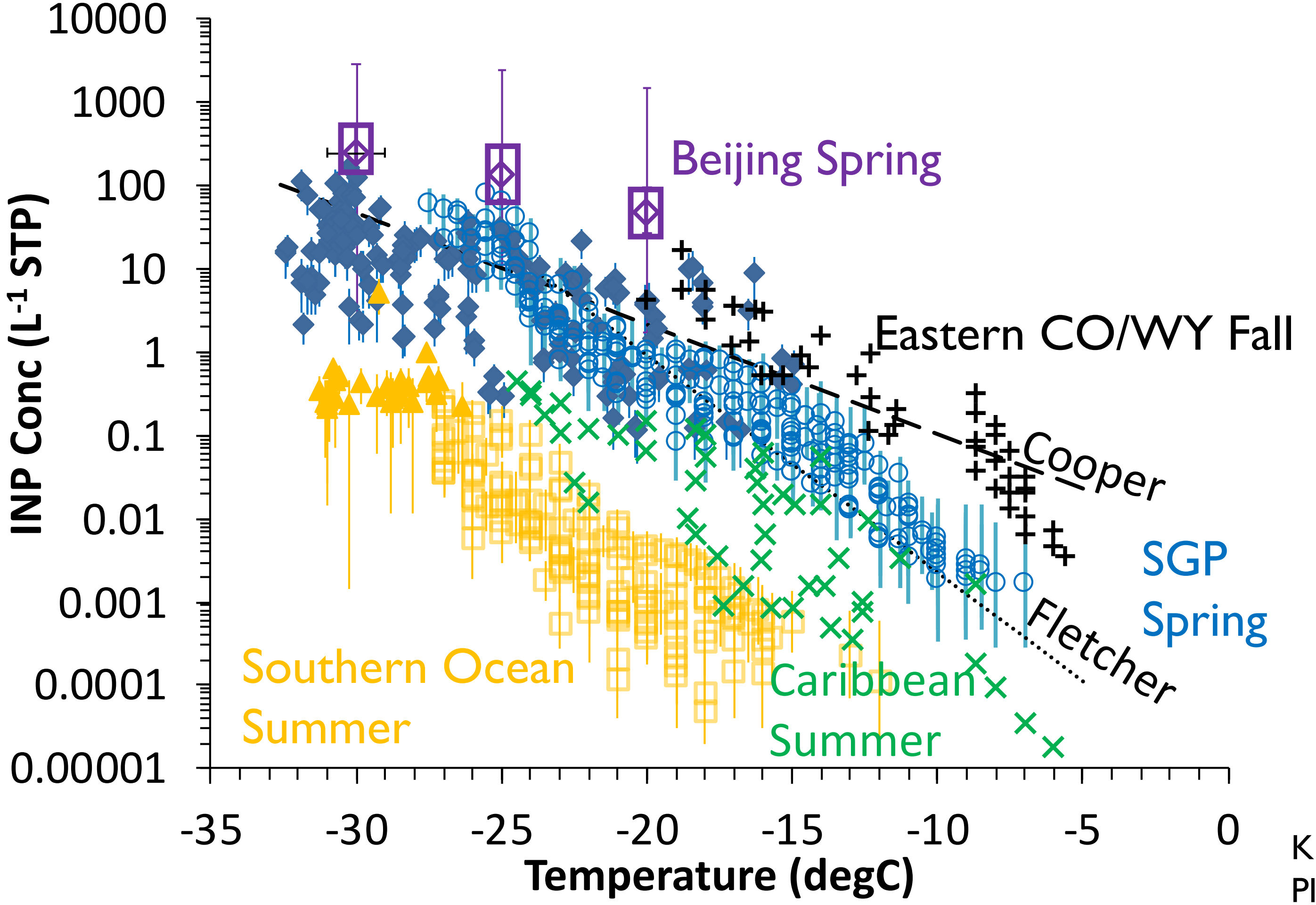
Suski et al. (2018)

CFDC single heated particles = bulk heated particles resuspended

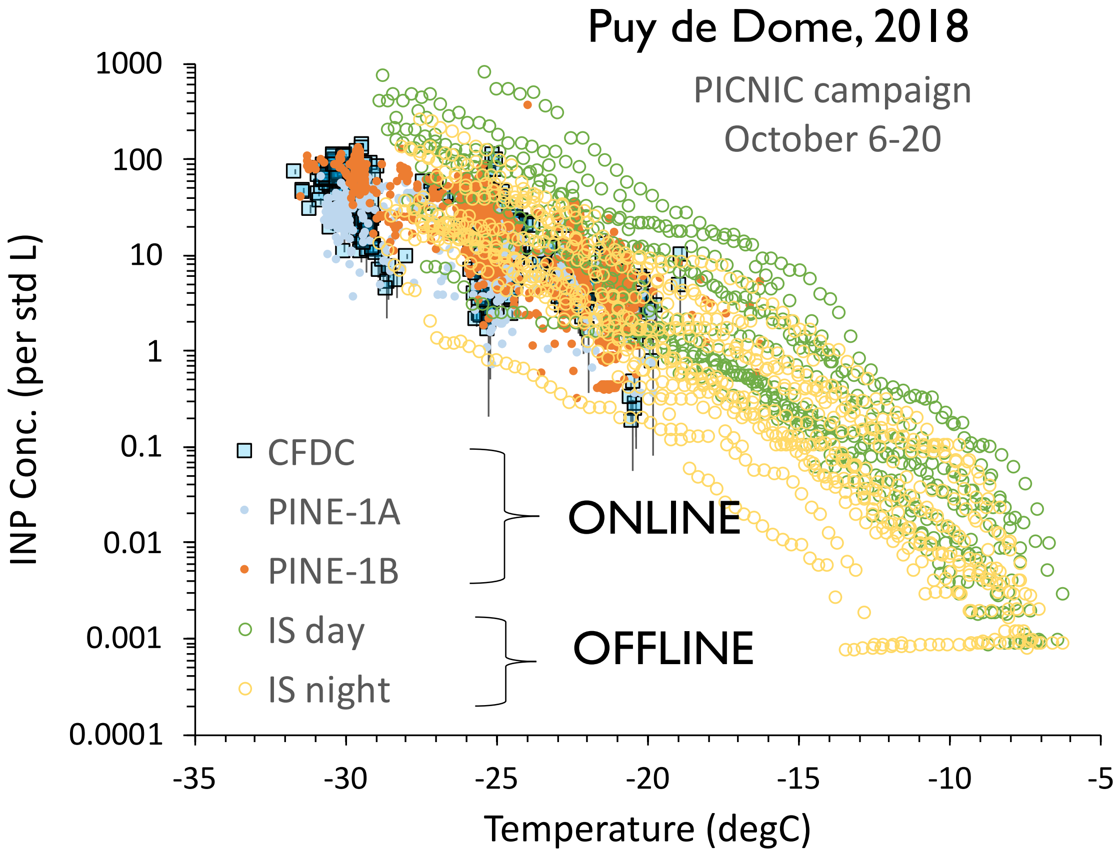
INPs sampled in a variety of marine and continental locations in both hemispheres with the CSU CFDC and IS instruments



Application of online and offline measurements in field studies and international intercomparison efforts (lab studies – See Möhler presentation)



Lacher et al., in preparation

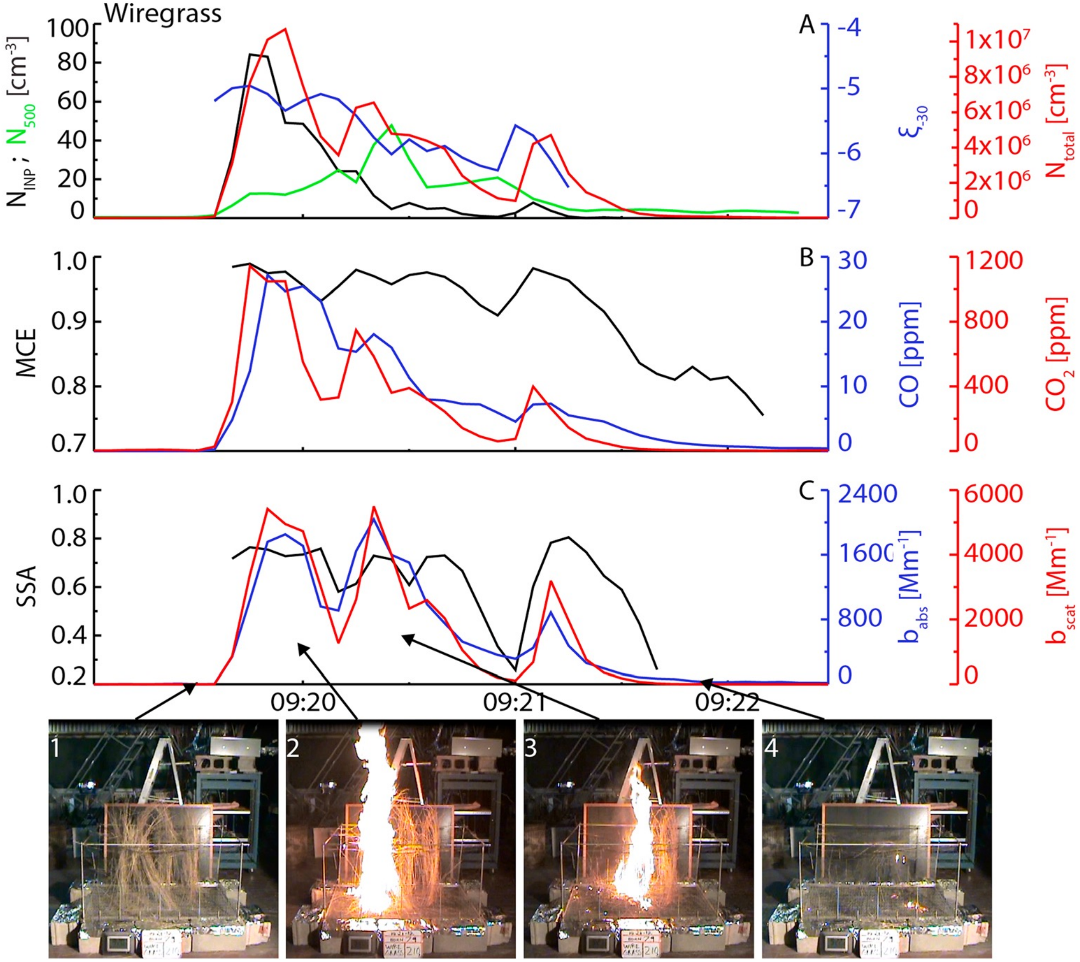


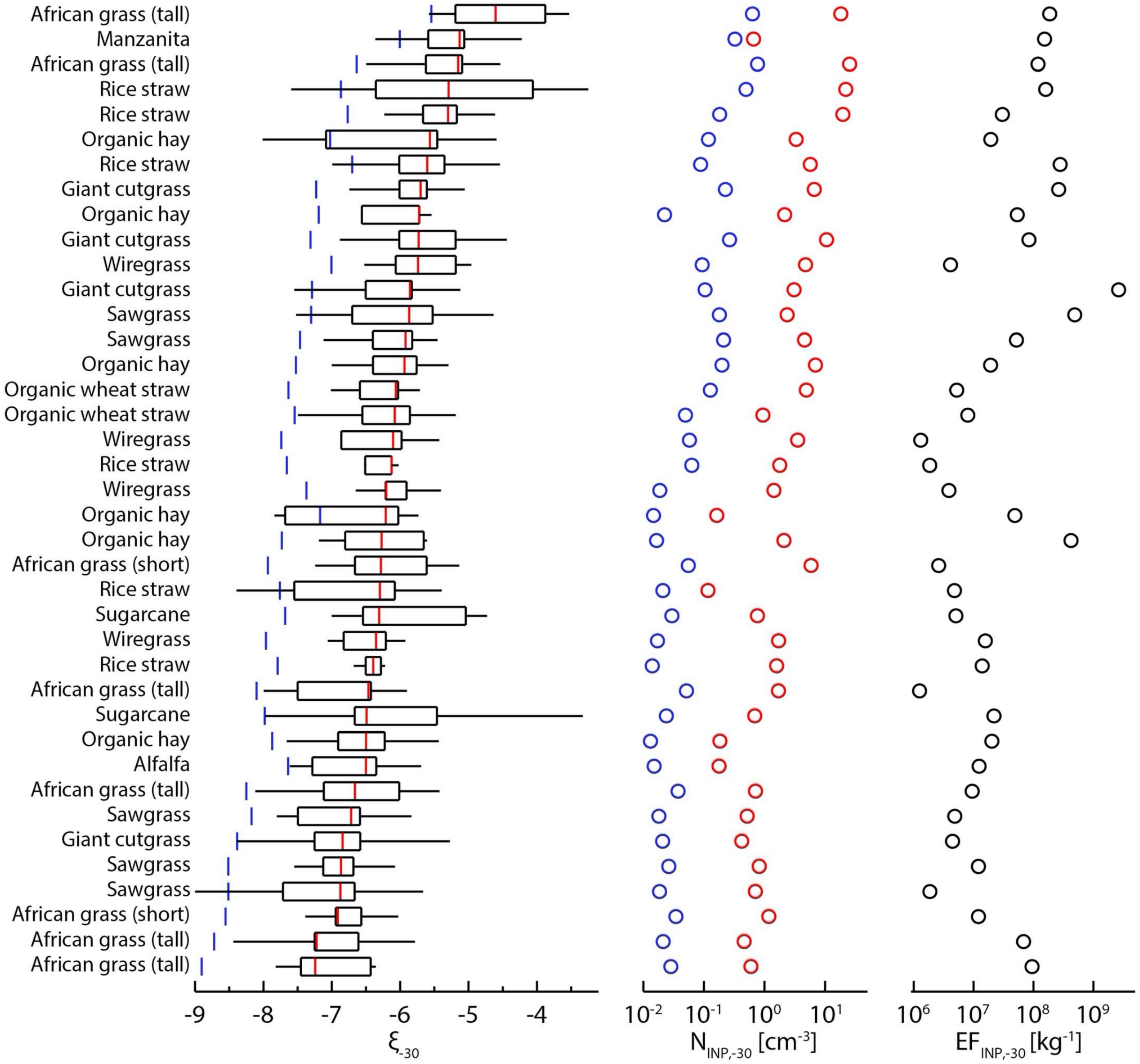
Knopf et al. (2021). Aerosol–Ice Formation Closure: A Southern Great Plains Field Campaign, Bulletin of the American Meteorological Society (published online ahead of print 2021), https://doi.org/10.1175/BAMS-D-20-0151.1

Associating with other laboratory facilities for ice nucleation studies

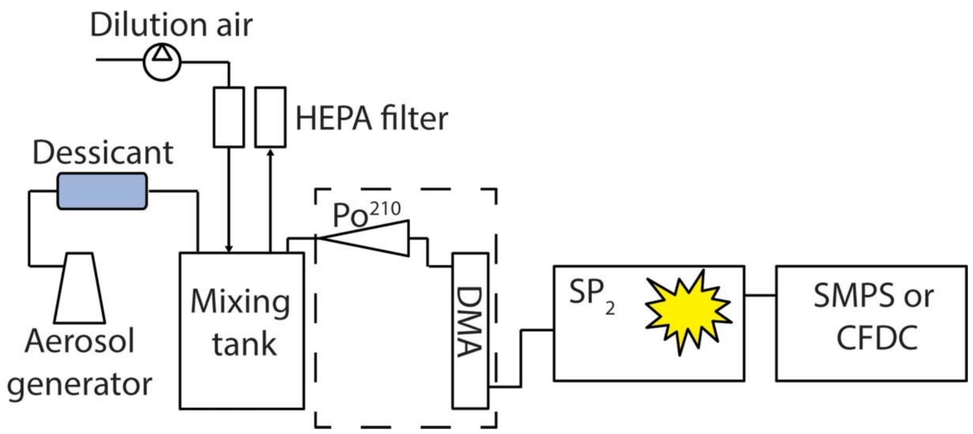
USFS Fire Sciences Laboratory (e.g., Levin et al., 2016)

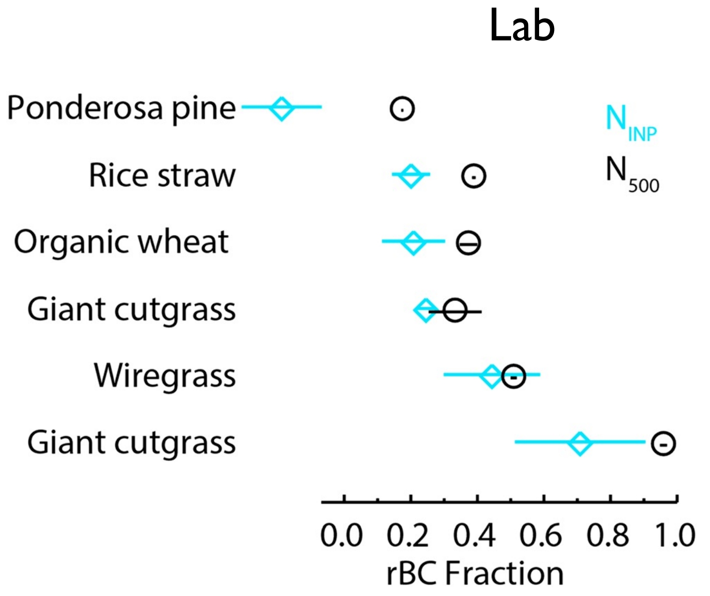
$$\xi_{-30°C} = \log (n_{in}/n_{cn})_{RHw \sim 105\%}$$

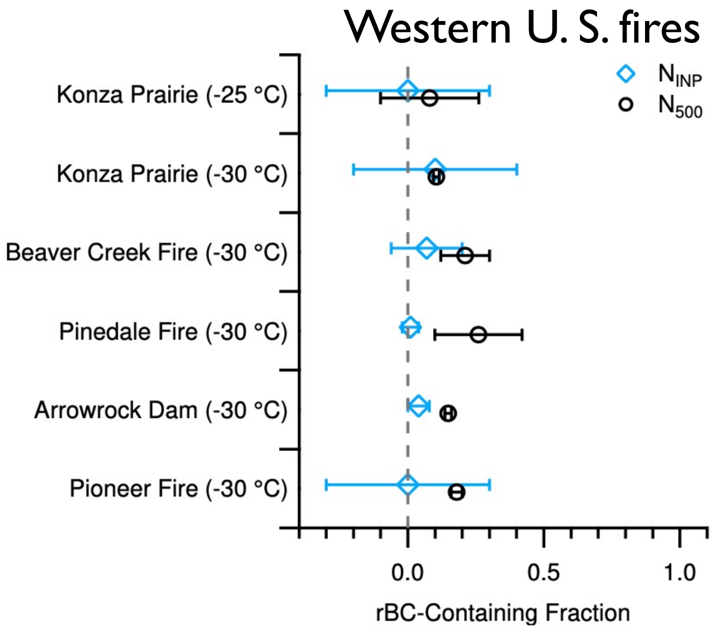




Development of integrated methods in same setting (e.g., SP2-CFDC) and subsequent application in field







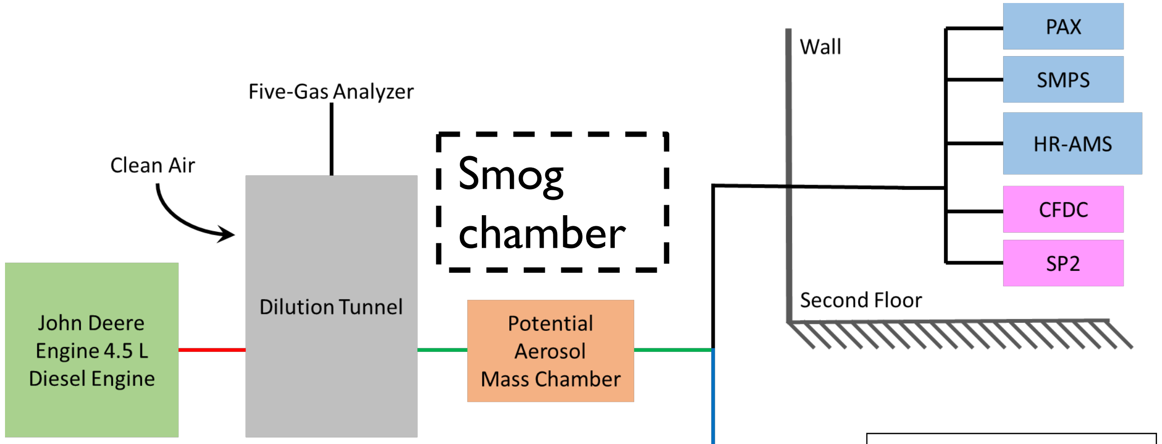
Levin, E. J.T et al., 2014: A New Method to Determine the Number Concentrations of Refractory Black Carbon Ice Nucleating Particles, Aerosol Science and Technology, 48:12, 1264-1275, DOI: 10.1080/02786826.2014.977843.

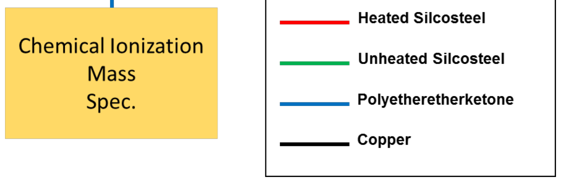
Levin, E. J. T. et al., 2016: Ice-nucleating particle emissions from biomass combustion and the potential importance of soot aerosol, J. Geophys. Res. Atmos., 121 (10), 5888-5903, doi:10.1002/2016JD024879.

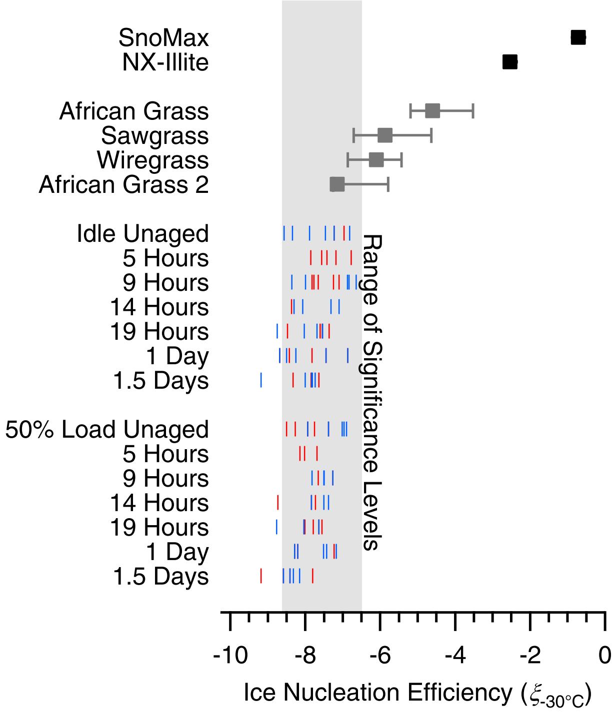
Schill, G. P., P. J. DeMott, E. J. T. Levin, and S. M. Kreidenweis, 2018: Use of the Single Particle Soot Photometer (SP2) as a pre-filter for ice nucleation measurements: Effect of particle mixing state and determination of SP2 conditions to fully vaporize refractory black carbon, Atmos. Meas. Tech., 11, 3007-3020, https://doi.org/10.5194/amt-11-3007-2018.

Schill, G. P. et al., 2020: The contribution of black carbon to global ice nucleating particle concentrations relevant to mixed-phase clouds. Proceedings of the National Academy of Sciences, 117 (37), 22705–22711, DOI: 10.1073/pnas.2001674117

Use of specialized labs: CSU Engines Laboratory (e.g., Schill et al., 2016)





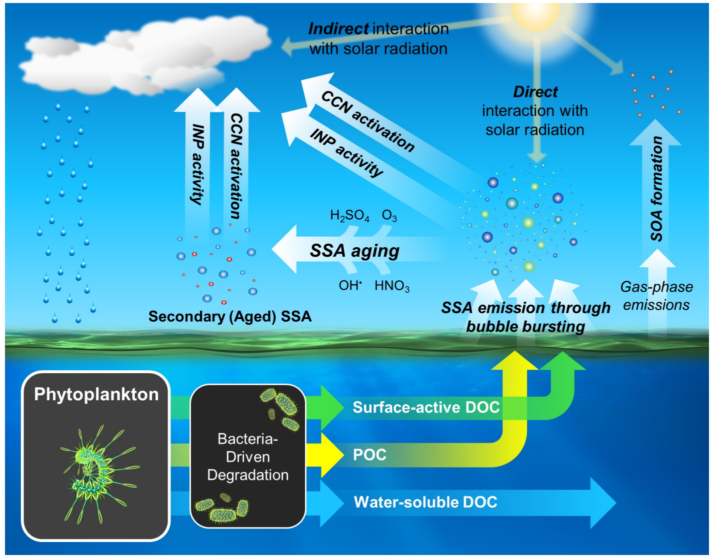


Schill, G. P., S. H. Jathar, J. K. Krodos, E. J. T. Levin, A. M. Galang, B. Friedman, D. K. Farmer, J. R. Pierce, S. M. Kreidenweis, and P. J. DeMott, 2016: Ice nucleating particle emissions from photo-chemically-aged diesel and biodiesel exhaust, *Geophys. Res. Lett.*, **43**, 5524-5531, doi:10.1002/2016GL069529.

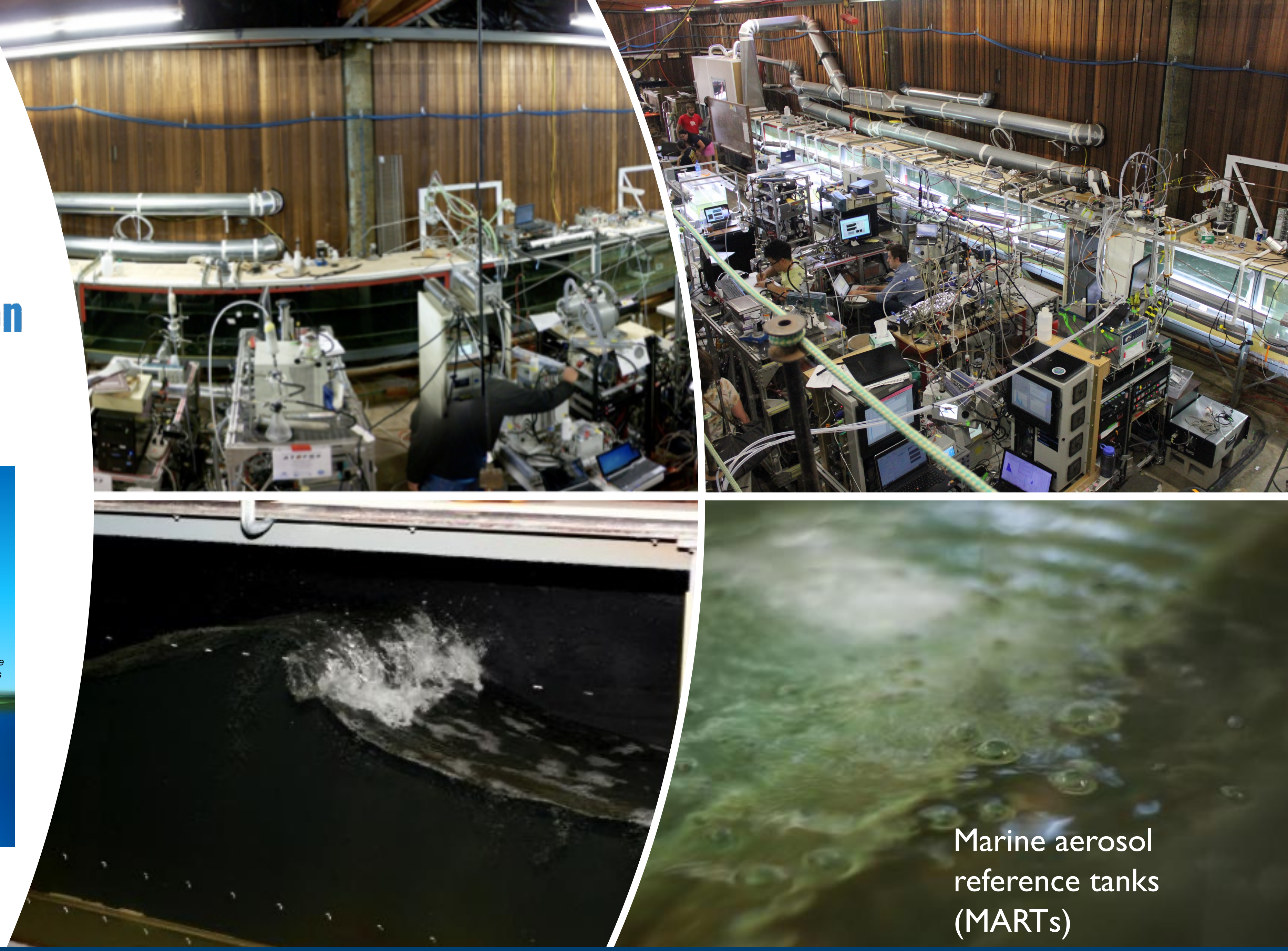
Isolated Ocean-Atmosphere Studies in the Laboratory

Centers for Chemical Innovation

CAICE NSF center for aerosol impacts™ on chemistry of the environment



Prather *et al.* ***PNAS*** **2013**

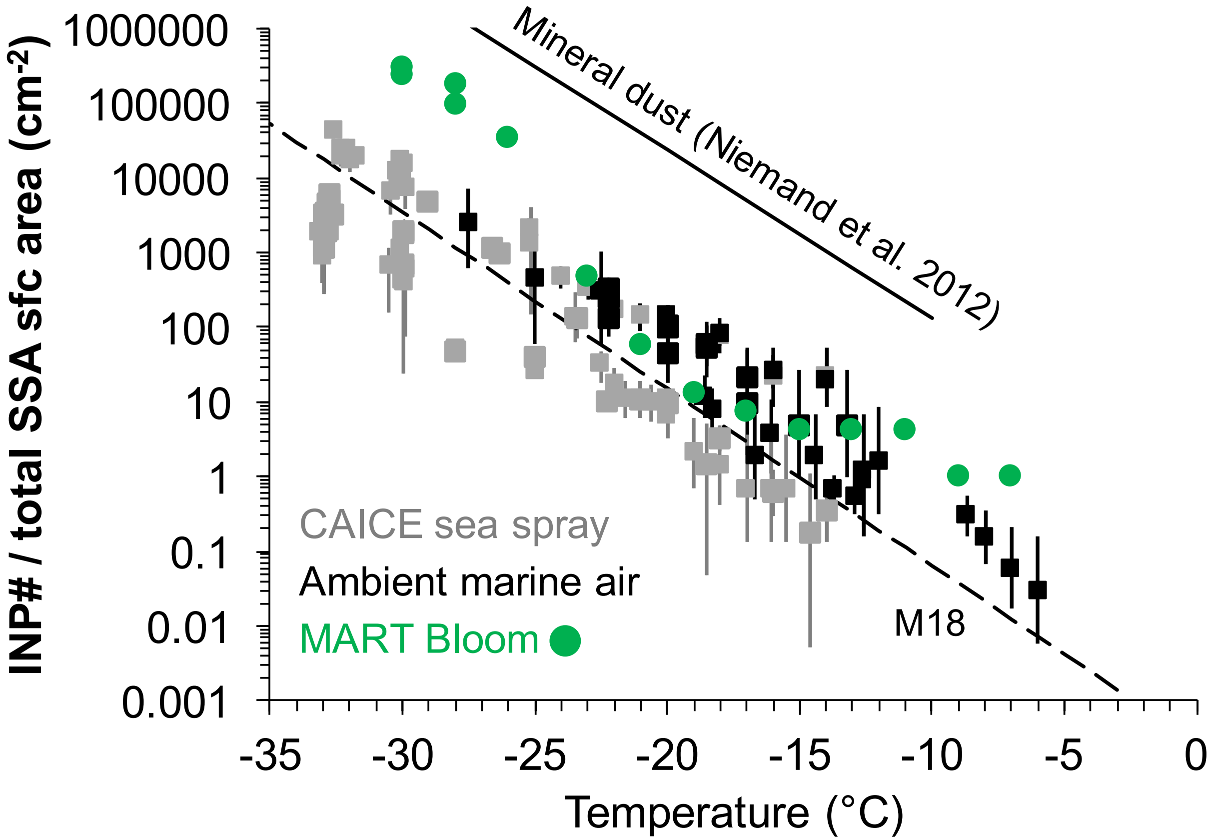


Marine aerosol reference tanks (MARTs)

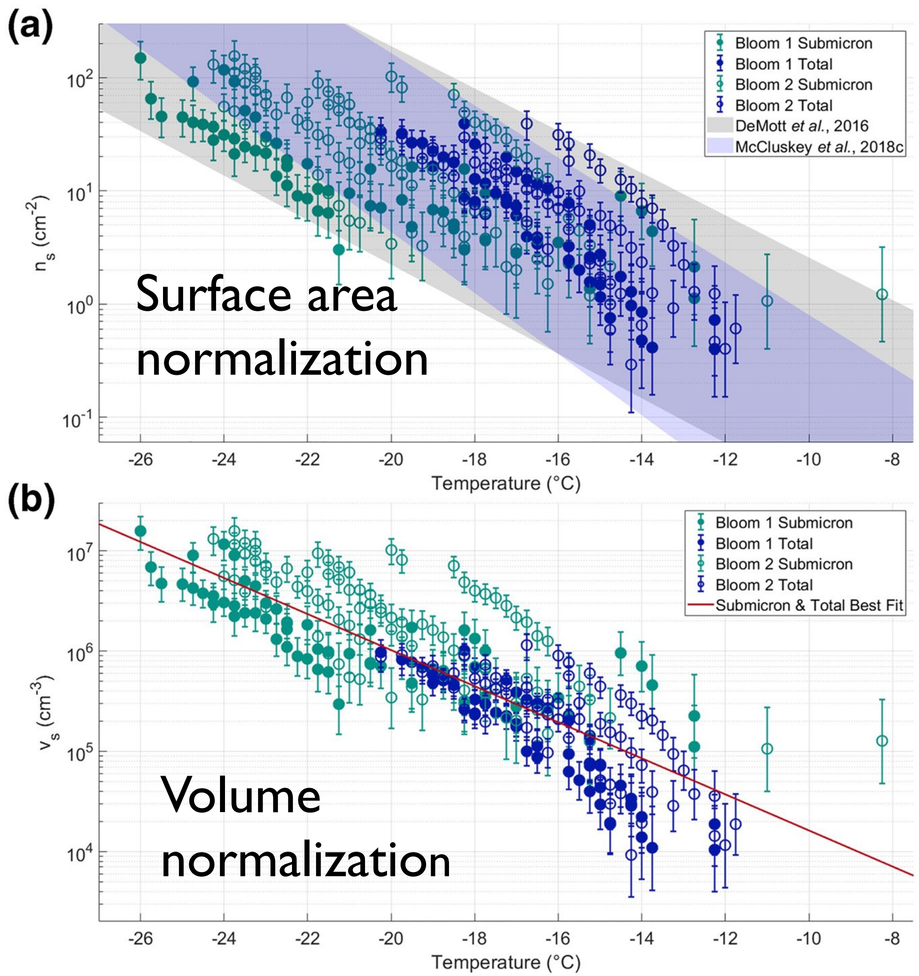
CFDC and IS studies using wave flume and portable sea spray devices for ice nucleation studies

CAICE NSF center for aerosol impacts on chemistry of the environment

DeMott et al., 2016: Sea spray aerosol as a unique source of ice nucleating particles. *Proc. Natnl. Acad. Sci.*, 113 (21), 5797-5803, doi:10.1073/pnas.1514034112.



Mitts, B. A., Wang, X., Lucero, D. D., Beall, C. M., Deane, G. B., DeMott, P. J., & Prather, K. A. (2021), Geophysical Research Letters, https://doi.org/10.1029/2020GL089633



SeaSCAPE Overview

SeaSCAPE (2019) was designed to replicate the chemical complexity of the marine atmosphere and surface ocean in the laboratory.

Properties of Nascent Sea Spray Aerosol (SSA)

Emission of Reactive Gases

Photochemical oxidation of SSA and formation of secondary marine aerosol (SMA)

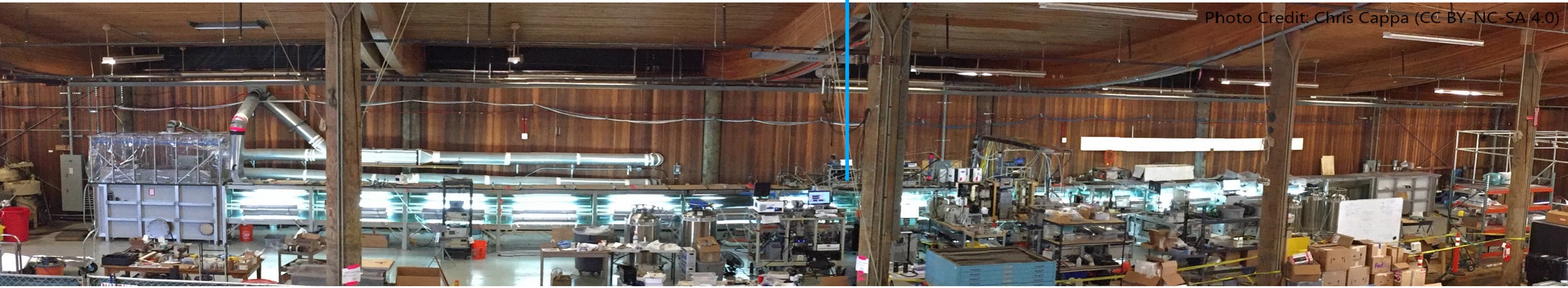
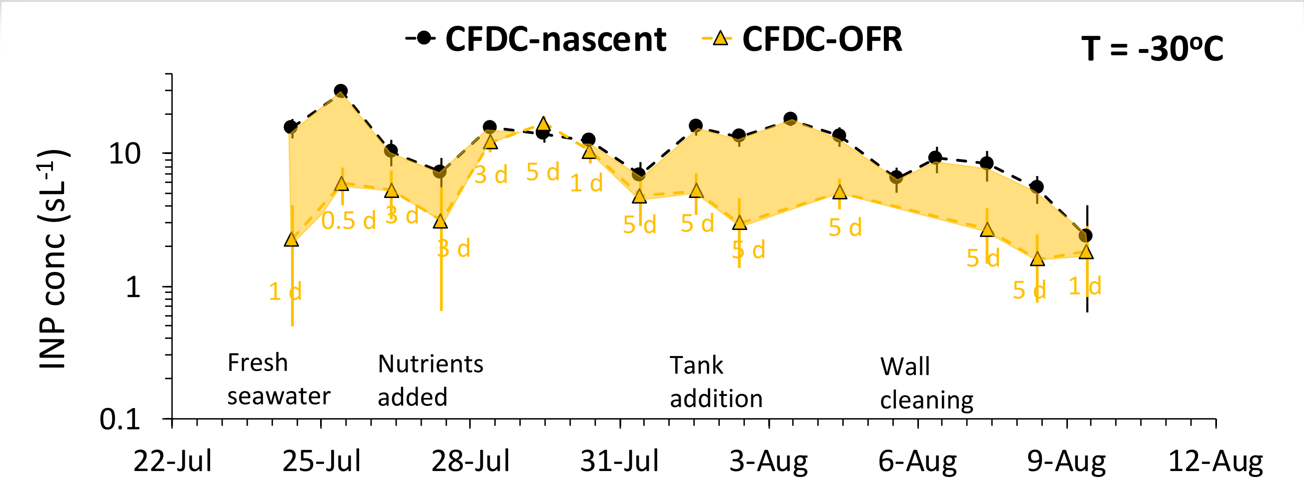


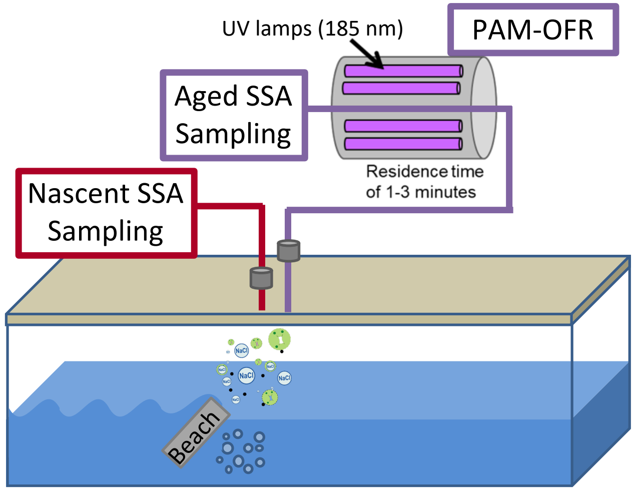
Photo Credit: Chris Cappa (CC BY-NC-SA 4.0)

Timeline of Oxidation Impact on Ice Nucleating Particles in SeaSCAPE 2019 Studies

CAICE NSF center for aerosol impacts on chemistry of the environment



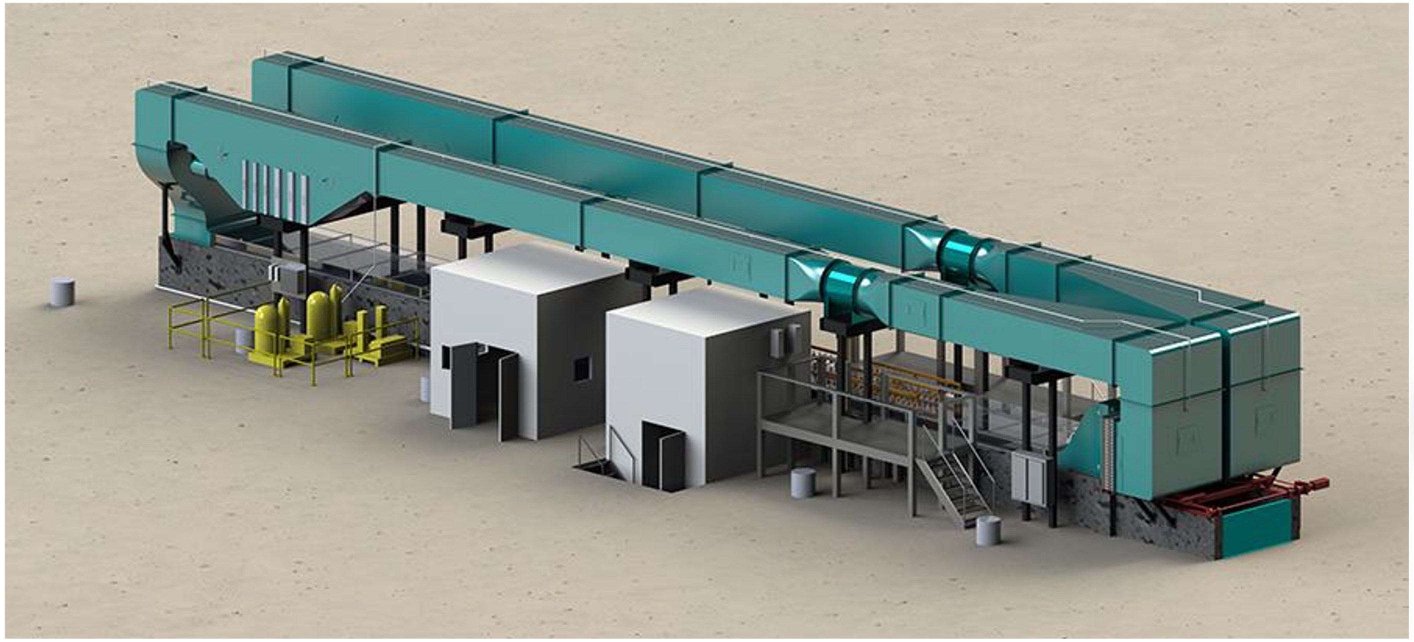
Oxidation imposes **up to 5x loss** of INPs following emissions, equivalent to up to **~2°C degradation** on basis of IS temperature spectra



SIO Waveflume
(DeMott et al. In Prep)

Scripps Ocean-Atmosphere Research Simulator (SOARS)

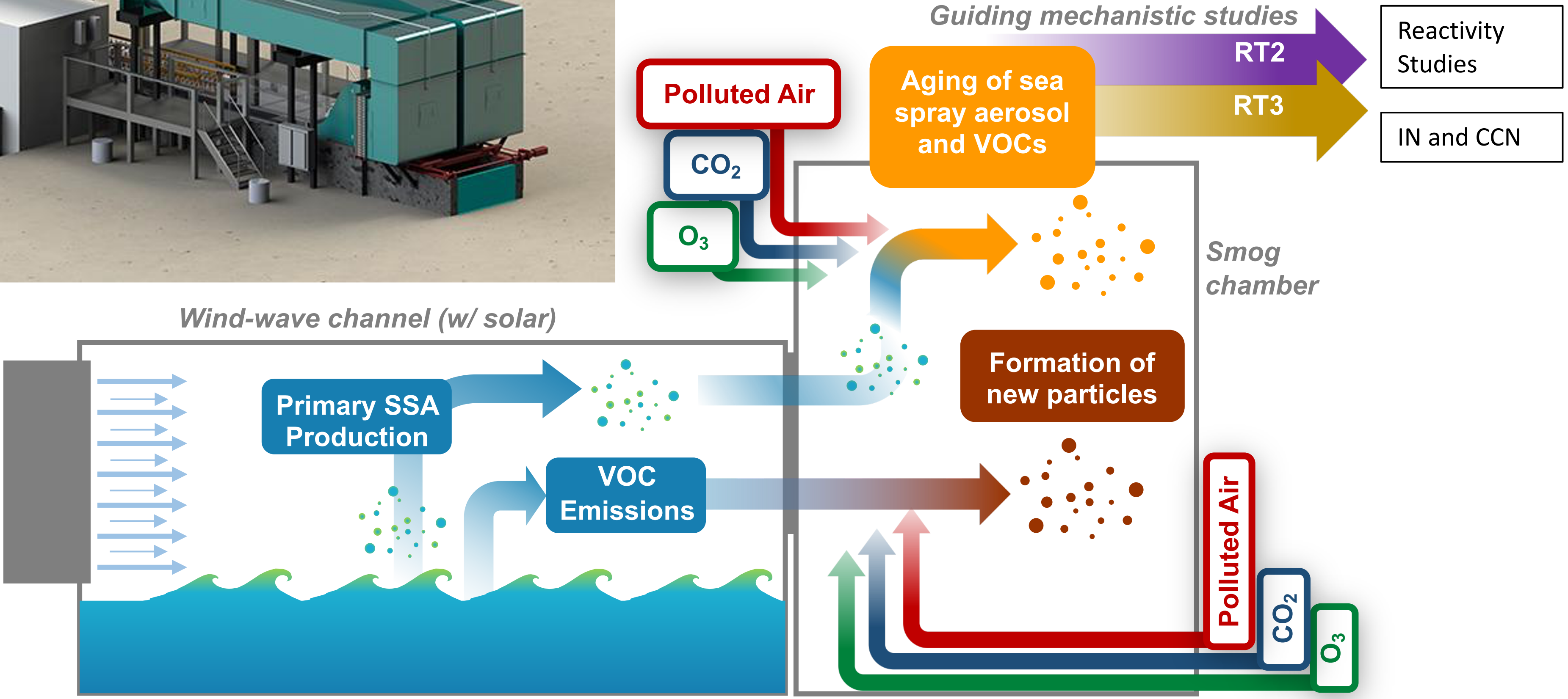
CAICE NSF center for aerosol impacts on chemistry of the environment



UC San Diego

Simulate future (and past) climate scenarios with full environmental control (T, RH, winds)

Wind speeds up to 19 m/s
T = -15 to 25°C (ocean)




UC San Diego
The Scripps Ocean Atmosphere Research Simulator (SOARS)
SPRING 2021
An interdisciplinary research instrument to mimic the current and future oceans from polar to tropical regions and study their role in human health, weather, and climate.
UC Regents
Executive Architects
AeroLab/CRB
Contractor
Align Builders
In an emergency, CALL CAMPUS POLICE:
9-1-1
858-534-HELP
24 Hour Construction Emergency: 619.494.6354
student centered
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