Single-particle Observations of Year-round Sea Spray Aerosol in the Coastal and High Arctic

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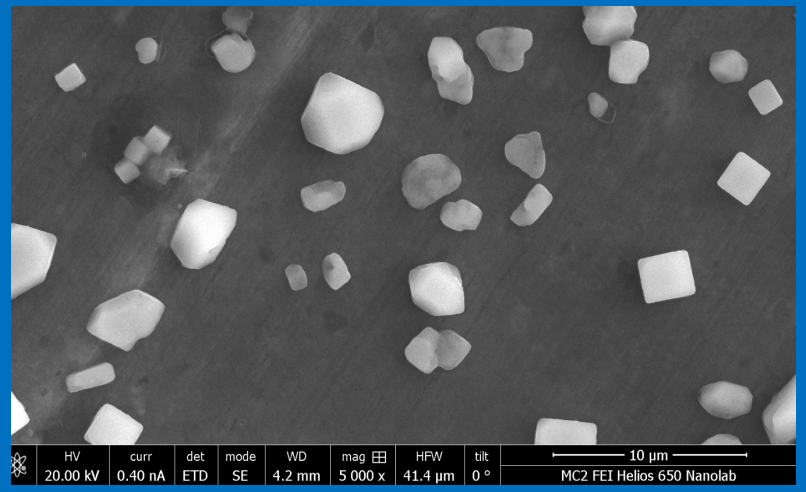
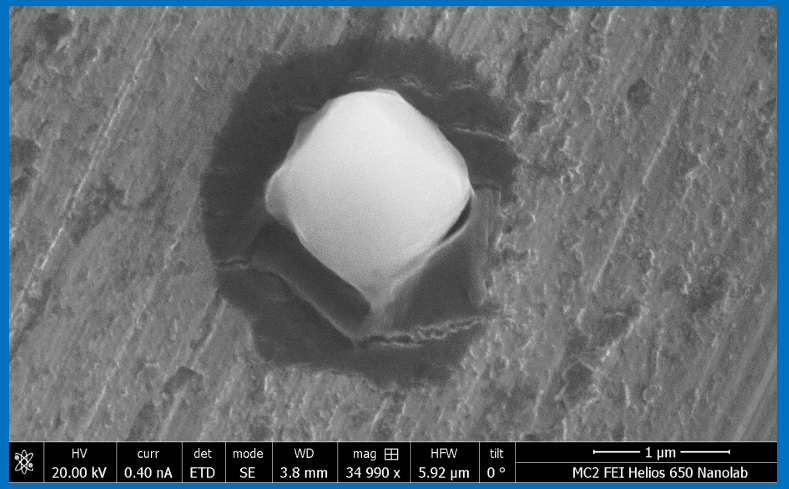
[1]University of Michigan, [2]Colorado State University, [3]EPFL, Switzerland, [4]EMSL, Pacific Northwest National Laboratory





UNIVERSITY OF MICHIGAN

EMSL

DEPARTMENT OF ENERGY · UNITED STATES OF AMERICA

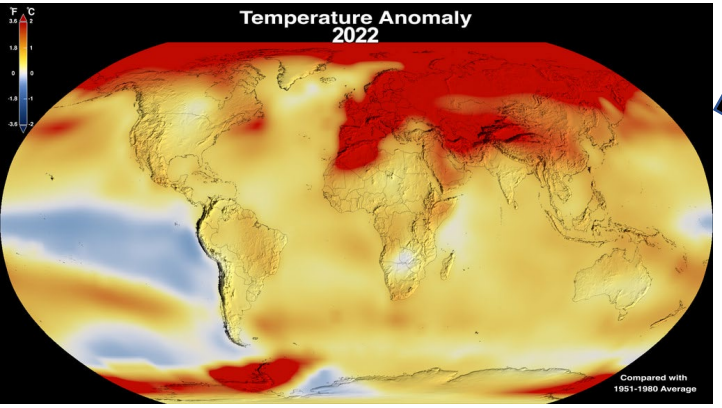
ASR Atmospheric System Research

ARM CLIMATE RESEARCH FACILITY

MOSAiC International Arctic Drift Expedition

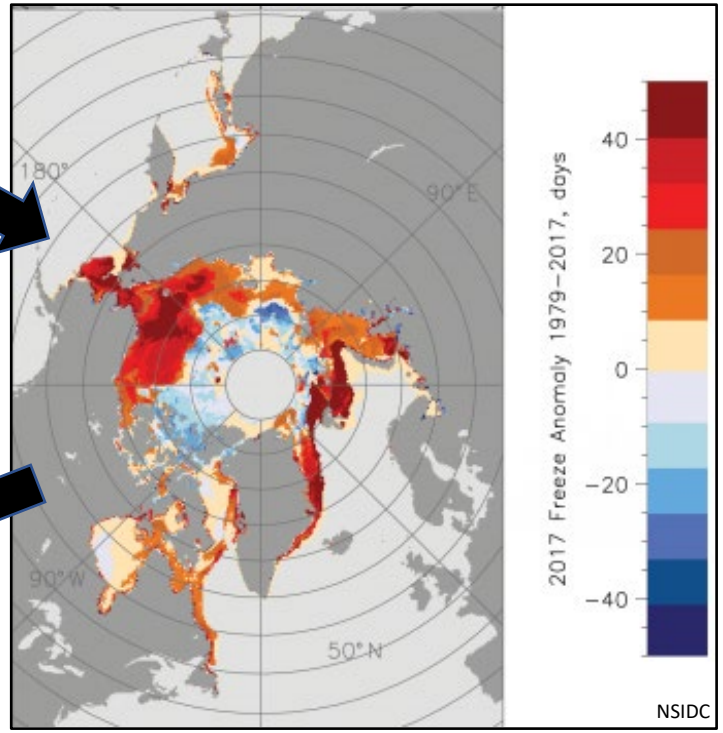
Warming Arctic = Increasing Open Water

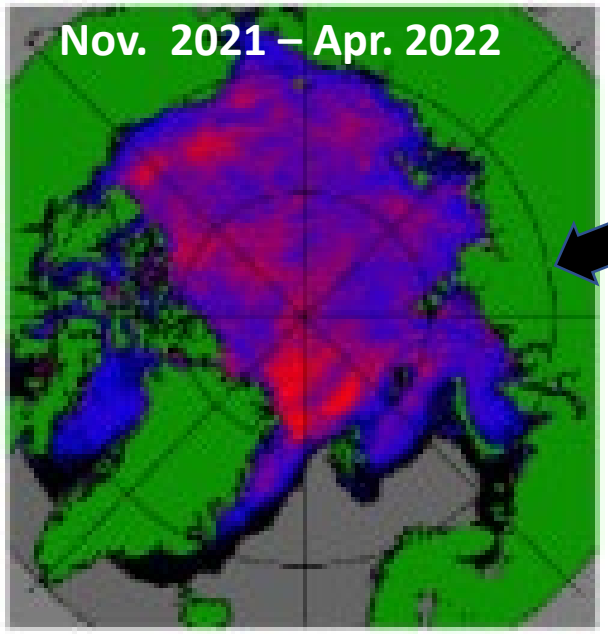
Arctic warming

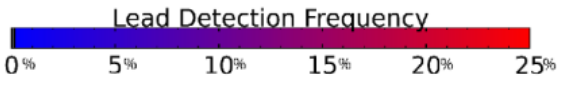


https://svs.gsfc.nasa.gov/5060

Delayed Sea Ice Freeze-up

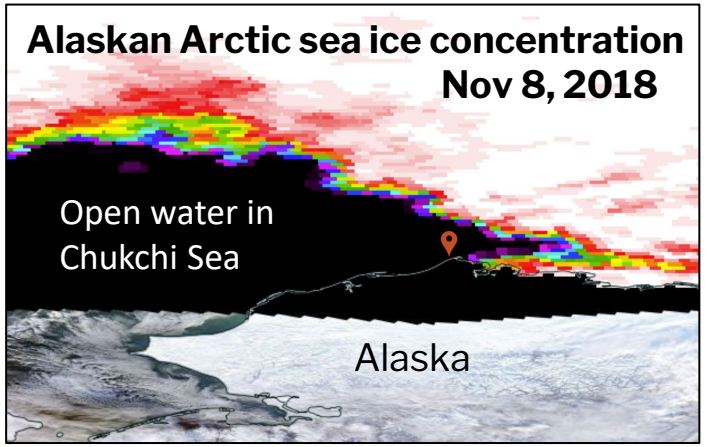


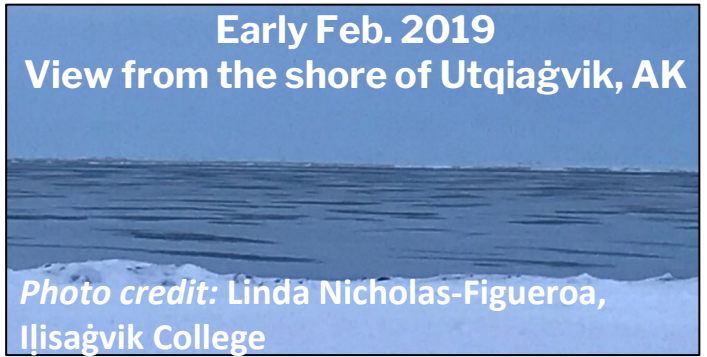




Thinning Sea Ice = Increasing Ice Fracturing (Lead Formation)

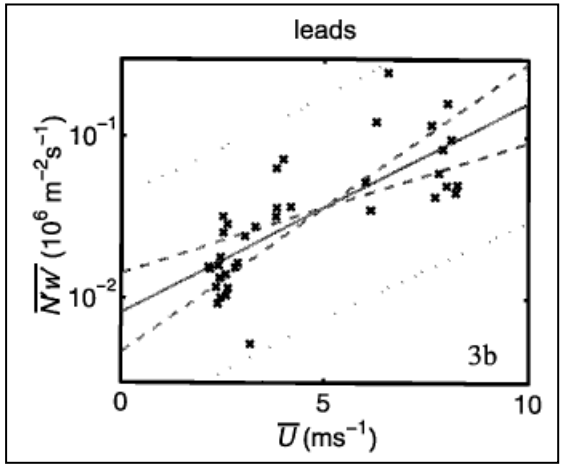
Hoffman et al 2022, *Remote Sens.*



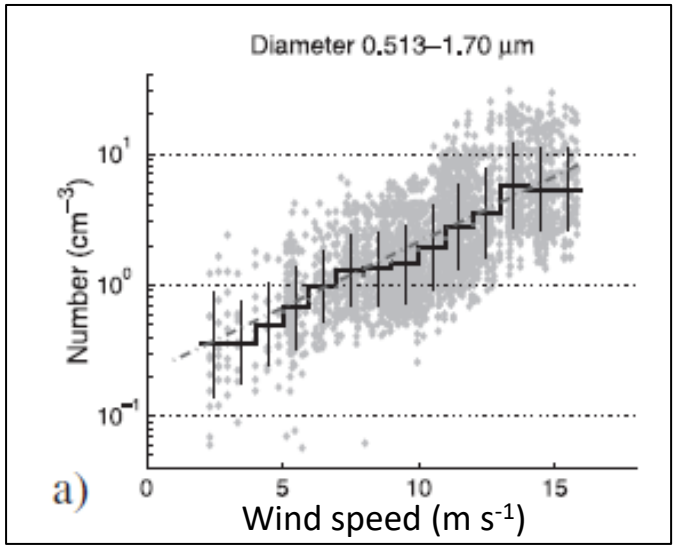


Sea Ice Leads → Sea Spray Aerosol & Clouds

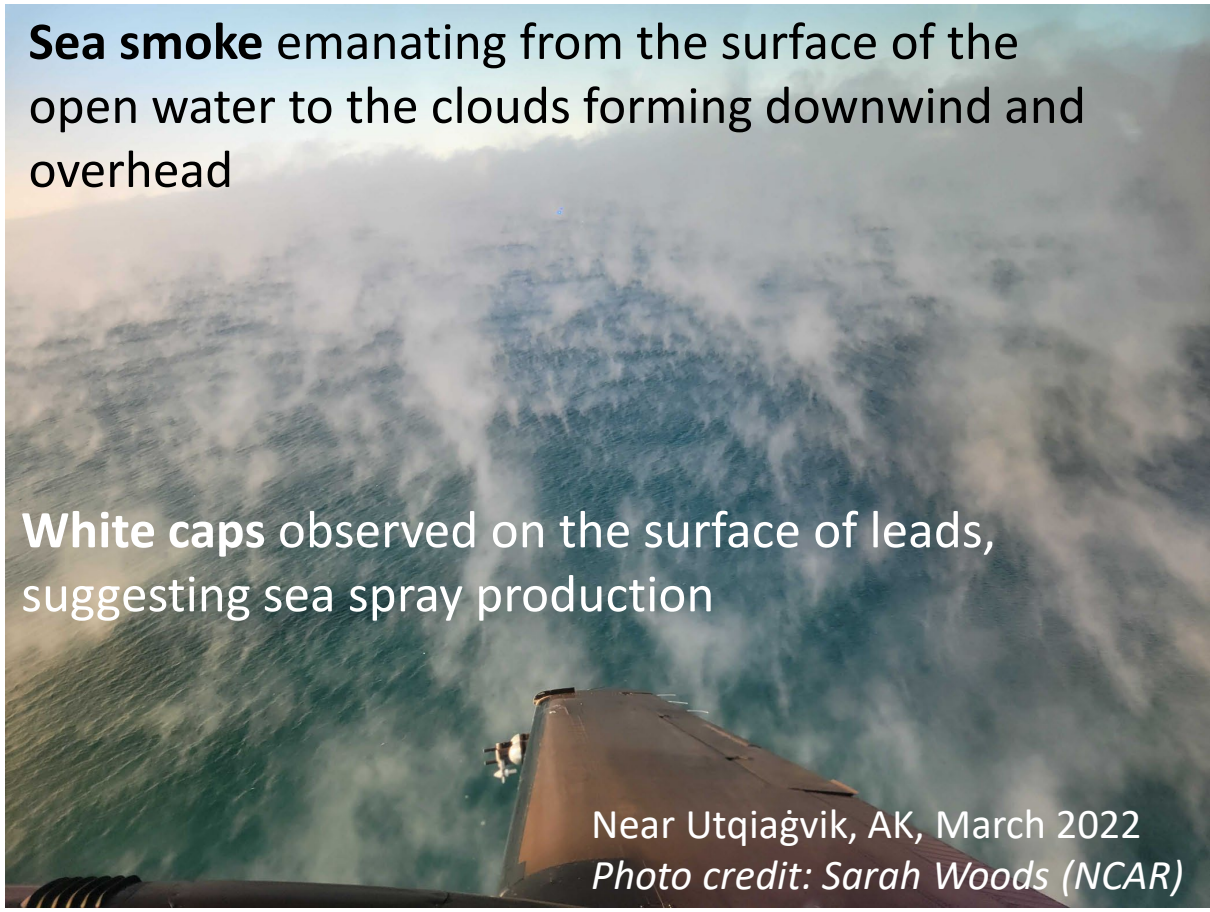
Increasing sea spray aerosol emissions from leads with increasing wind speed



Nilsson et al. 2001, *JGR*



Leck et al. 2002, *JGR*



Geophysical Research Letters

Wintertime Airborne Measurements of Ice Nucleating Particles in the High Arctic: A Hint to a Marine, Biogenic Source for Ice Nucleating Particles

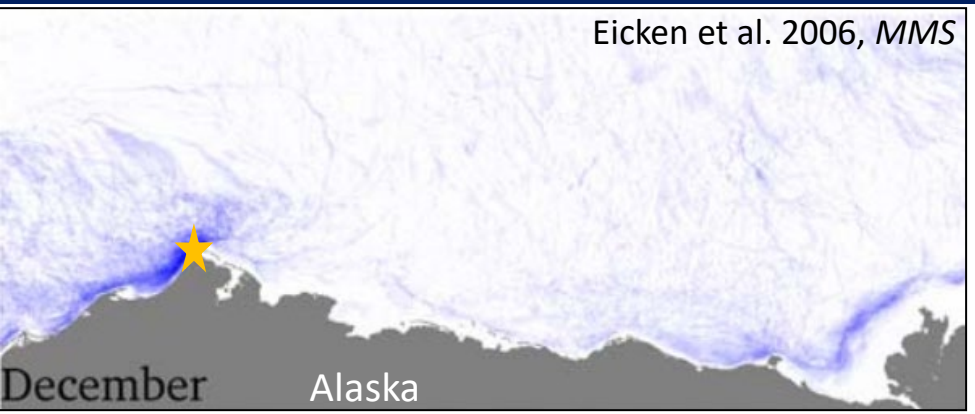
M. Hartmann[1], K. Adachi[2], O. Eppers[3,4], C. Haas[5], A. Herber[6], R. Holzinger[7], A. Hünerbein[8], E. Jäkel[9], C. Jentzsch[1], M. van Pinxteren[10], H. Wex[1], S. Willmes[11], and F. Stratmann[1]

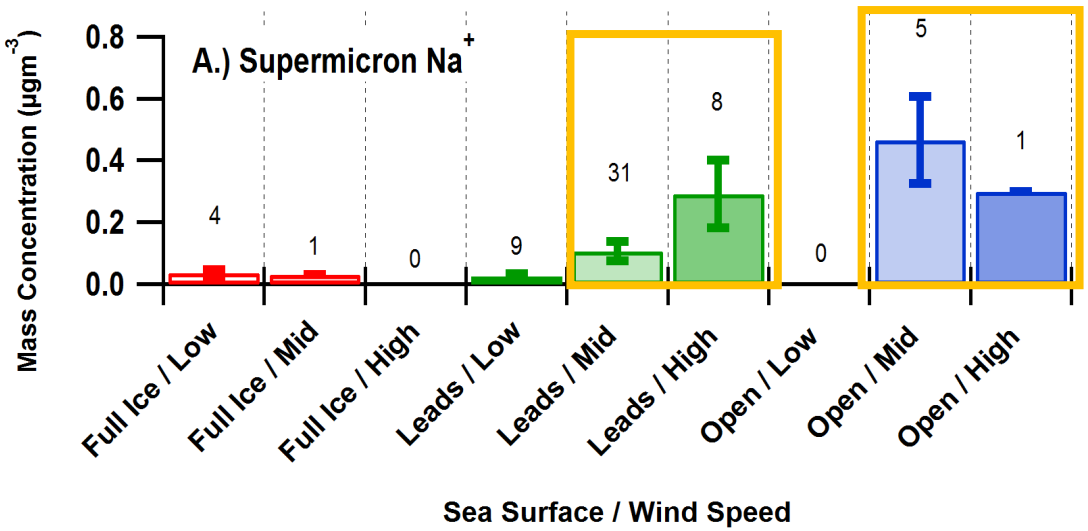
Midwinter Arctic leads form and dissipate low clouds

Xia Li, Steven K. Krueger, Courtenay Strong, Gerald G. Mace & Sally Benson

Nature Communications **11**, Article number: 206 (2020) | Cite this article

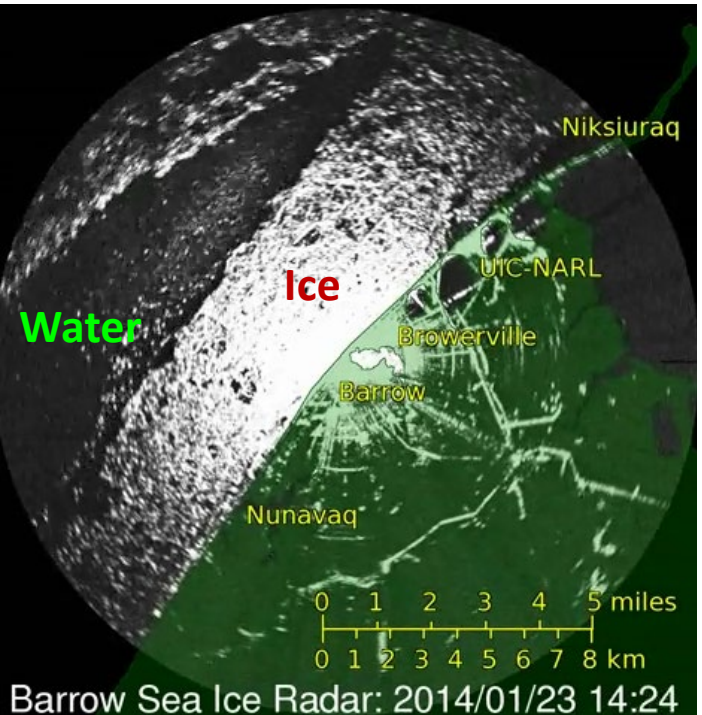
Year-Round Coastal Arctic Sea Salt Aerosol

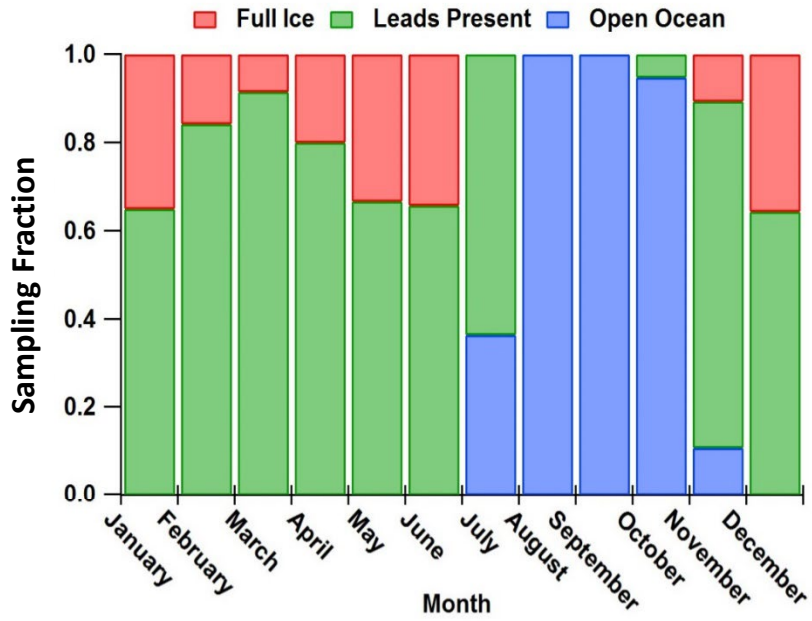




Utqiaġvik, AK
2006-2009

Low < 4 m/s
Mid 4-7 m/s
High >7 m/s



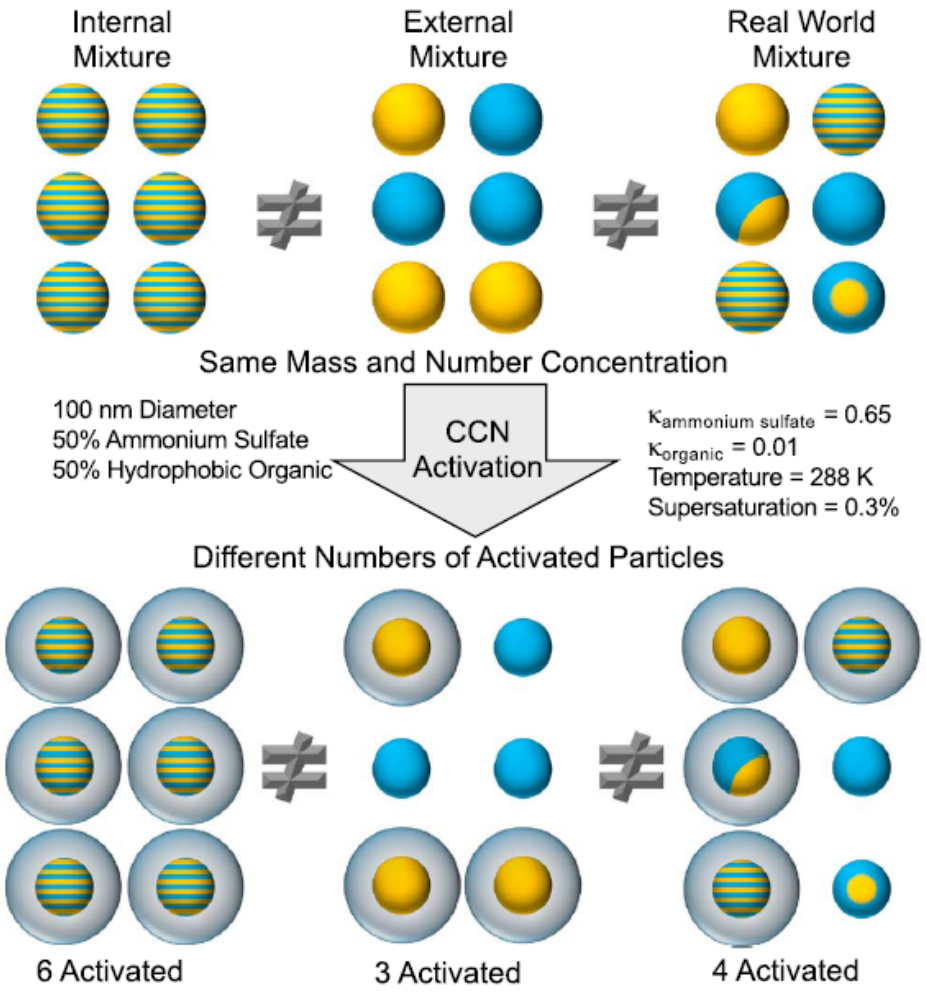


- **Sea salt aerosol emitted locally year-round from open water and leads at elevated wind speeds**
- Full ice and low winds dominated by chloride-depleted (transported) sea salt aerosol

May and Pratt et al. 2016, *JGR – Atmos.* doi:10.1002/2016/JD025273

Single-Particle Measurements are Important!

Assumptions Impact Cloud Activation

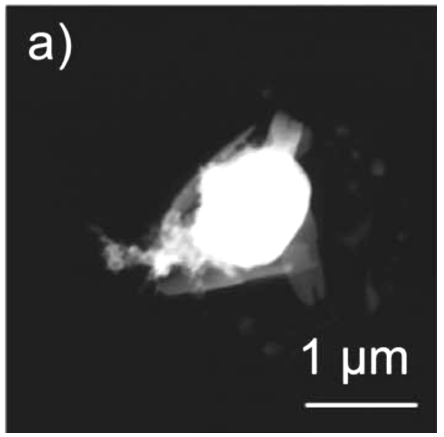


Riemer et al. 2016, *Rev. Geophys.*

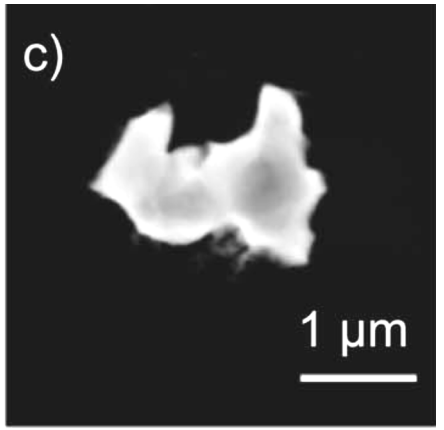
Chemical measurements of **individual particles** enable evaluation of modeling of:

- cloud droplet activation
- ice nucleation
- light scattering
- multiphase reactions

Chukchi Sea, Aug. 2016



Sea Spray Aerosol (CCN)



Mineral Dust (Potential INP)

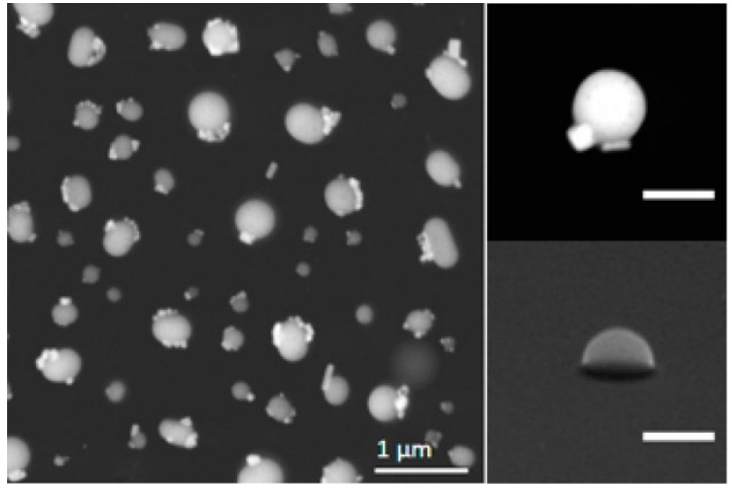
Kirpes & Pratt et al. 2020, *ESPI,* doi:10.1039/c9em00495e

Single-Particle Source Identification

Single-particle measurements enable **identification of primary aerosol source (e.g., dust, sea spray, soot, organic-sulfate)**, **aging (transport) processes**, & **direct determination of mixing state**

Utqiaġvik, AK (NSA), Aug.-Sep. 2015

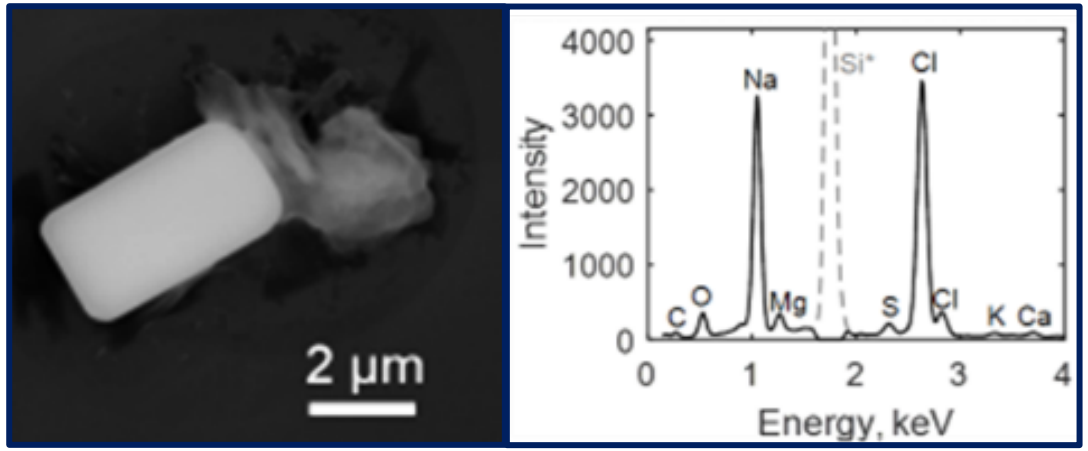
Solid organic-coated ammonium sulfate particles from new particle formation & growth



Kirpes, Pratt, & Ault et al. 2022, *PNAS.* doi:10.1073/pnas.2104496119

Central Arctic pack ice, Aug.-Sep. 2018

Sea spray aerosol generated (using a MART, marine aerosol reference tank) from surface water collected from leads



- Consistent with mid-latitude sea spray aerosol composition (Prather et al. 2013, *PNAS*)
- Collaboration for INPs (Ben Murray, Leeds)

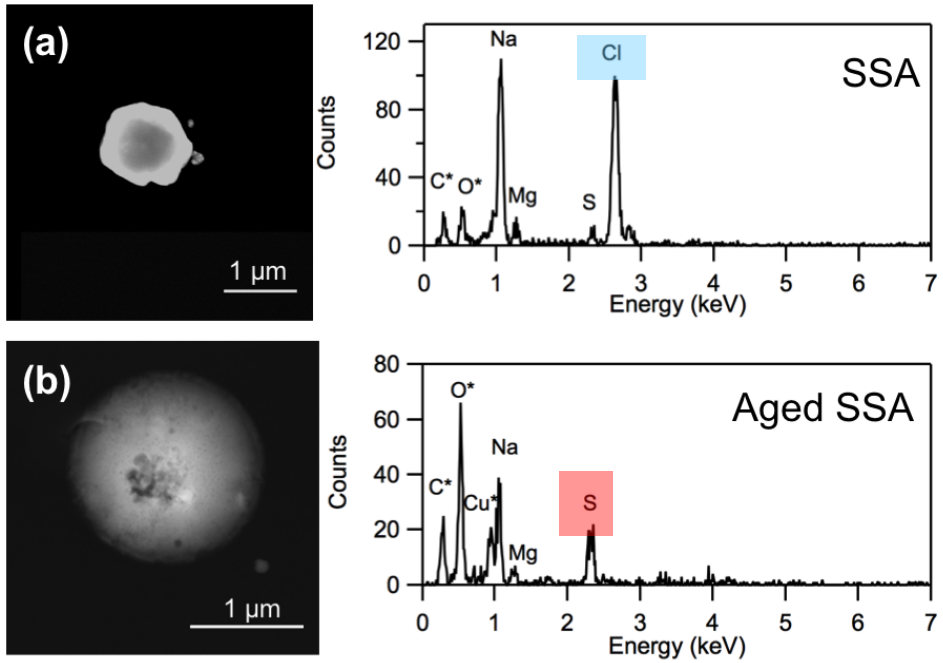
Connection to CAPE-K breakout questions!

In Progress

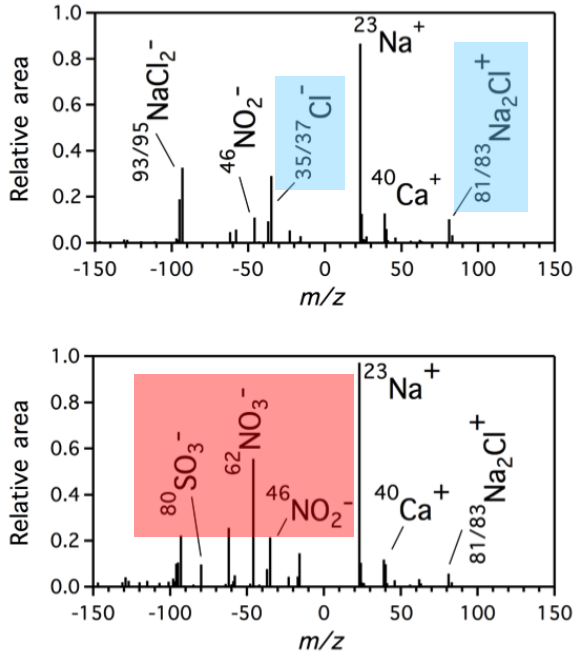
Late Summer (Aug.-Sep.) Sea Spray Aerosol

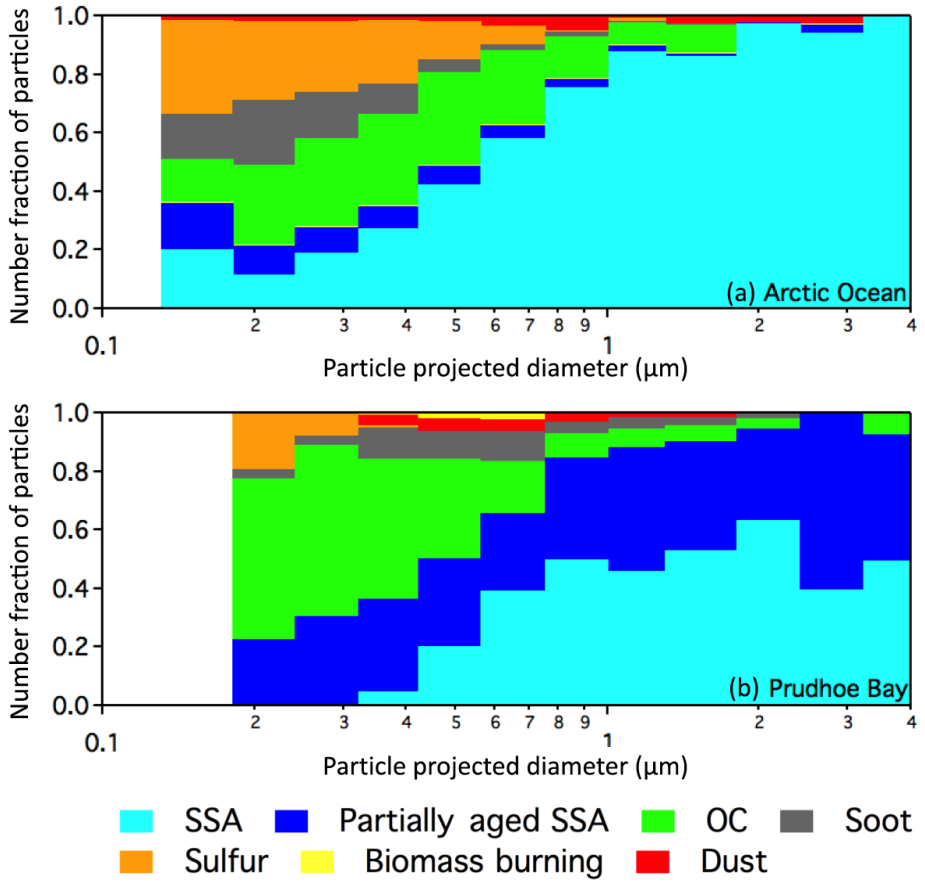
Utqiaġvik, AK (NSA), Aug.-Sep. 2015
Oliktok Point, AK (AMF3), Aug.-Sep. 2016

SEM-EDX



Single-particle mass spectrometry (A-ATOFMS)



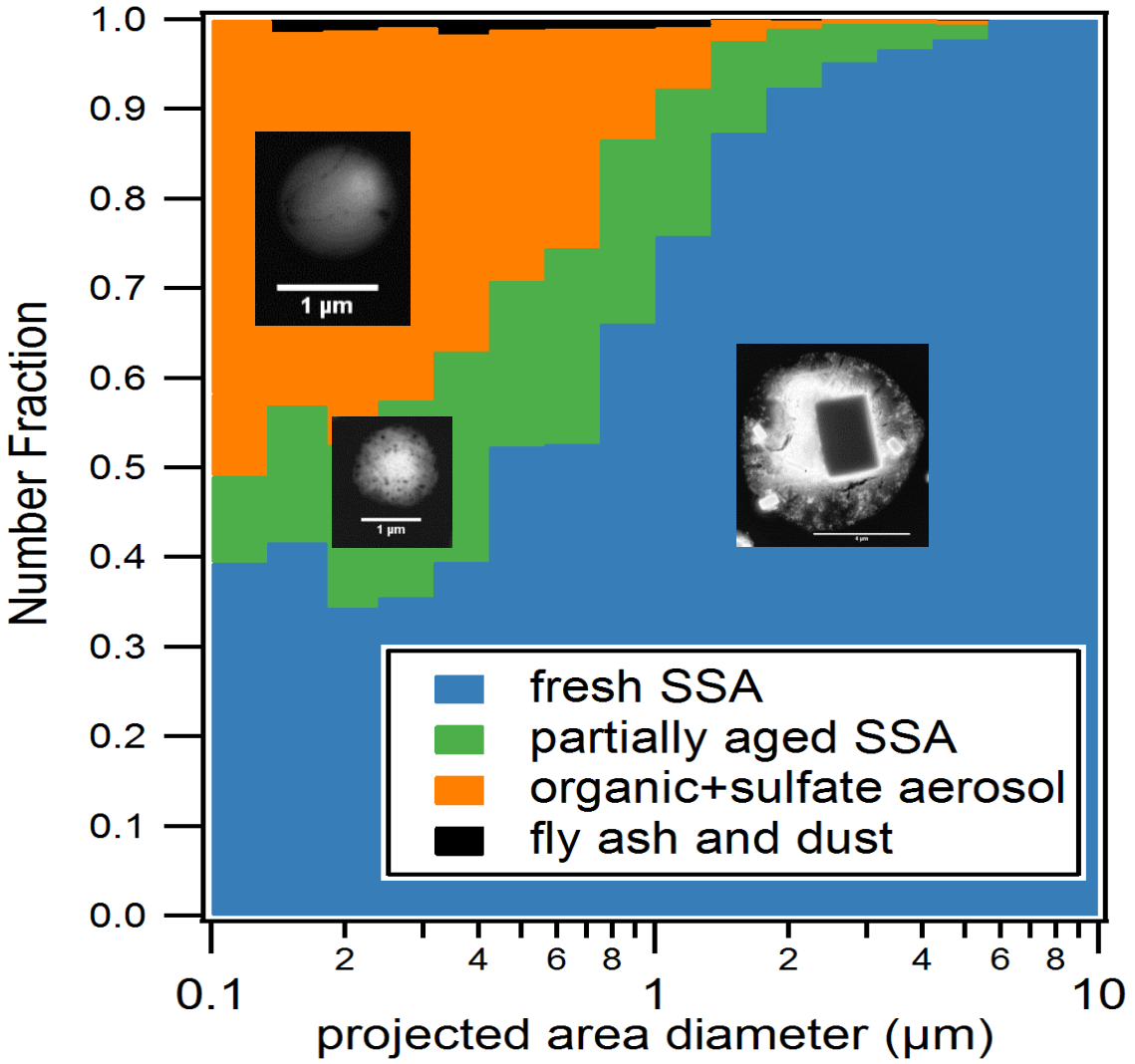


Sea spray aerosol (SSA) chloride depletion and sulfate and nitrate enrichment from multiphase reactions (Gard et al. 1998, *Science*) due to Arctic oil field emissions of NO_x and SO_2

Gunsch and Pratt et al. 2017, *ACP,* doi:10.5194/acp-17-10879-2017
Gunsch, Liu, and Pratt et al. 2020, *ES&T*, doi:10.1021.acs.est.9b04825

Late Winter (Jan.-Feb.) Sea Spray Aerosol

Utqiaġvik, AK (NSA), Jan. – Feb. 2014



- **Sea spray aerosols (SSA) were >50% of aerosol number above 0.1 µm**
- Sulfate internally mixed with 1) organic aerosol and 2) partially aged sea spray aerosol

Computer-controlled Scanning Electron Microscopy with Energy Dispersive X-ray Spectroscopy (CCSEM-EDX) of 24,847 individual particles on TEM grids

Kirpes, Ault, and Pratt et al. 2018, *ACP.* doi:10.5194/acp-18-3937-2018

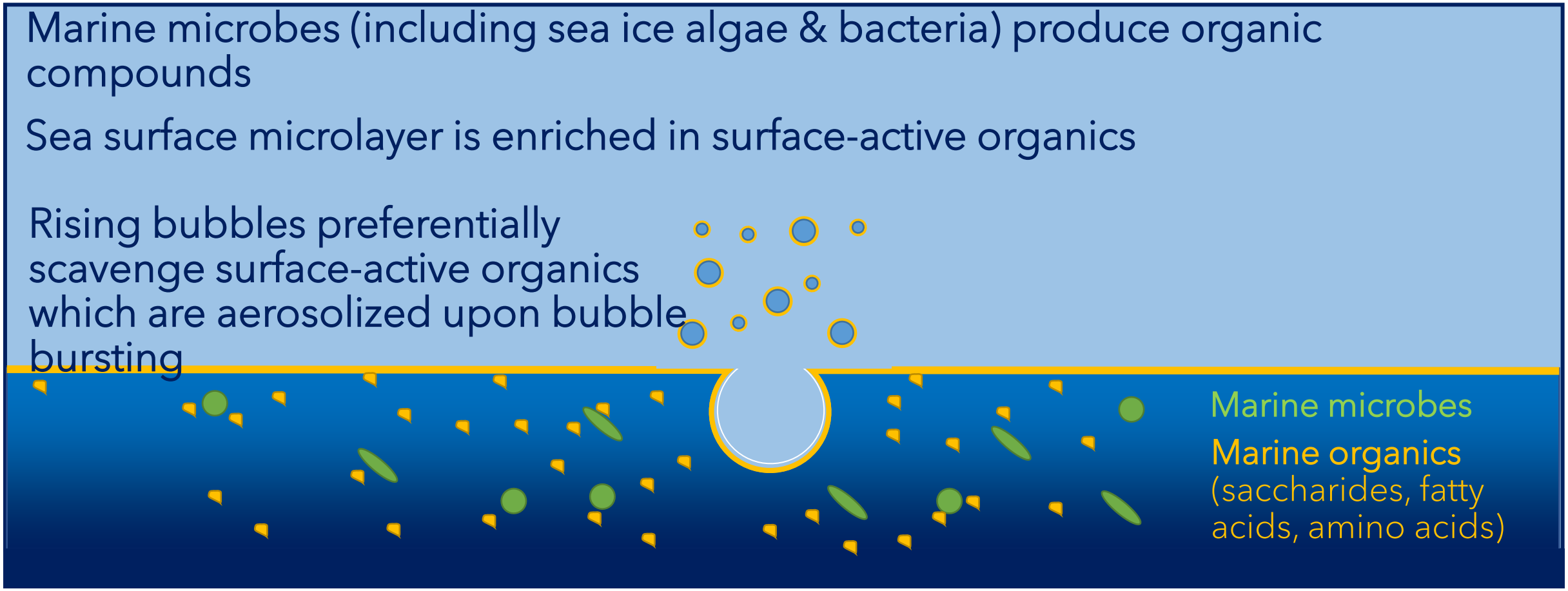
Not Just Sea Salt – Coatings of Marine-Derived Organics

Surface-active organic compounds are scavenged by rising air bubbles

Marine microbes (including sea ice algae & bacteria) produce organic compounds

Sea surface microlayer is enriched in surface-active organics

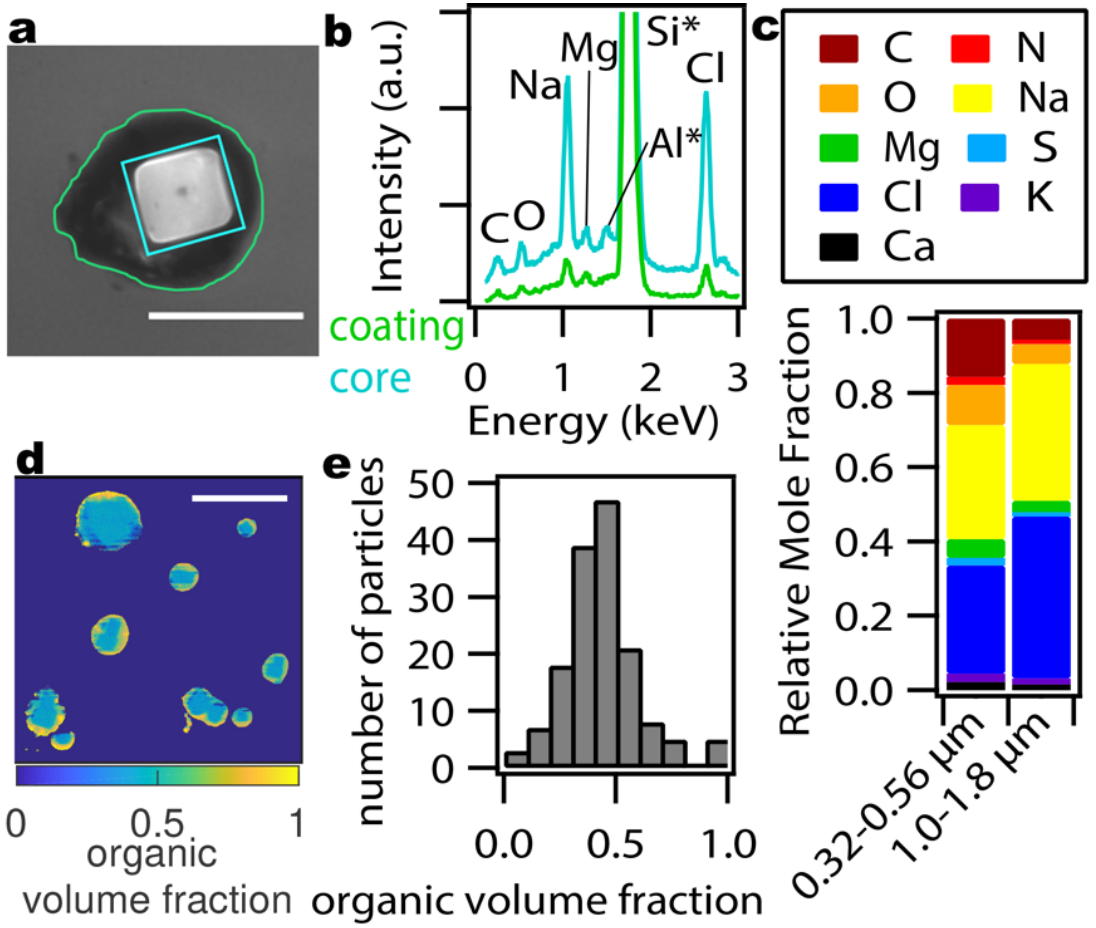
Rising bubbles preferentially scavenge surface-active organics which are aerosolized upon bubble bursting



Decho 1990 Oceanography and Marine Biology
Blanchard 1975 Applied Chemistry at Protein Interfaces, Chapter 18 Bubble Scavenging and the Water-to-Air Transfer of Organic Material in the Sea

Organic Coatings on Sea Spray Aerosol

Utqiaġvik, AK (NSA)
Jan. – Feb. 2014



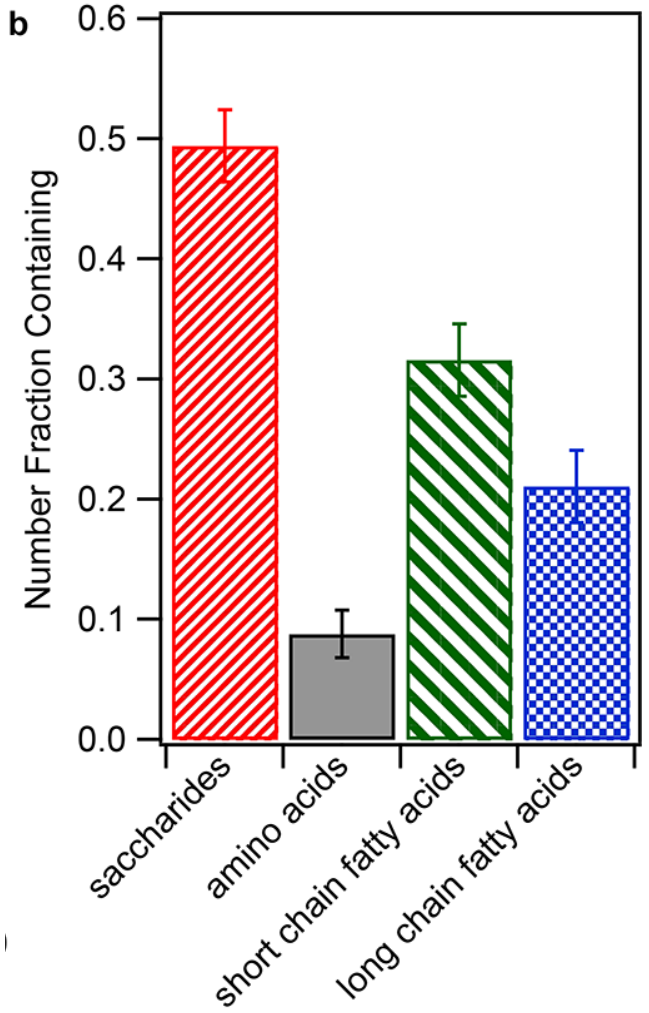
CCSEM-EDX of 1,691 individual particles on silicon, complemented by STEM-NEXAFS

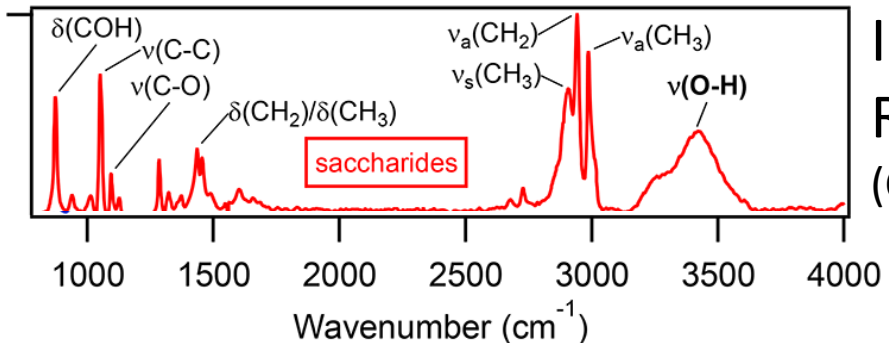
- **71 % of sea spray aerosol particles contained organic volume fractions between 0.3 – 0.5**
  - Similar to previous midlatitude algal bloom mesocosm experiments (0.2 – 0.5) (Collins et al. 2013, *JGR*; Pham et al. 2017, *ACS Earth & Space Chem)*

Kirpes, Ault, and Pratt et al. 2019, *ACS Central Science.* doi:10.1021/acscentsci.9b00541

Marine Saccharides, Fatty Acids, & Amino Acids Coating Sea Spray Aerosol

Utqiaġvik, AK, Jan. – Feb. 2014





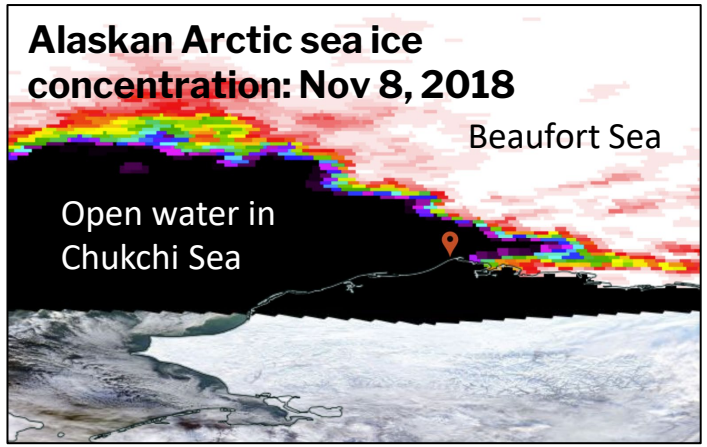
Individual particle Raman microspectroscopy (Cochran et al. 2017, *Chem*)

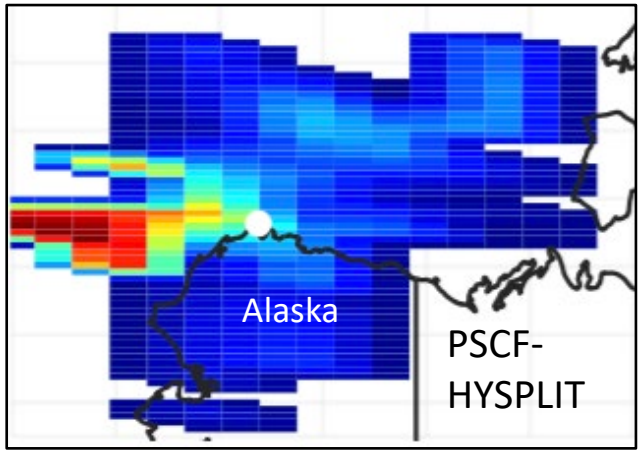
- **Marine saccharides, fatty acids, and amino acids observed**
- Previously observed in Arctic aerosol (e.g., Russell et al. 2010, *PNAS*)
- **Marine exopolymer substances** (EPS, e.g., Orellana et al. 2011, *PNAS*)**, complex with Ca^{2+}, which explains observed C and Ca enrichments**
- Sea ice algae and bacteria produce EPS as a cryoprotectant
- Similar individual particle Mg/Na, S/Na, and Cl/Na ratios to seawater (not consistent with measured snow over nearby sea ice)
- Consistent with bubble bursting aerosol from open leads

Kirpes, Ault, and Pratt et al. 2019, *ACS Central Science.* doi:10.1021/acscentsci.9b00541

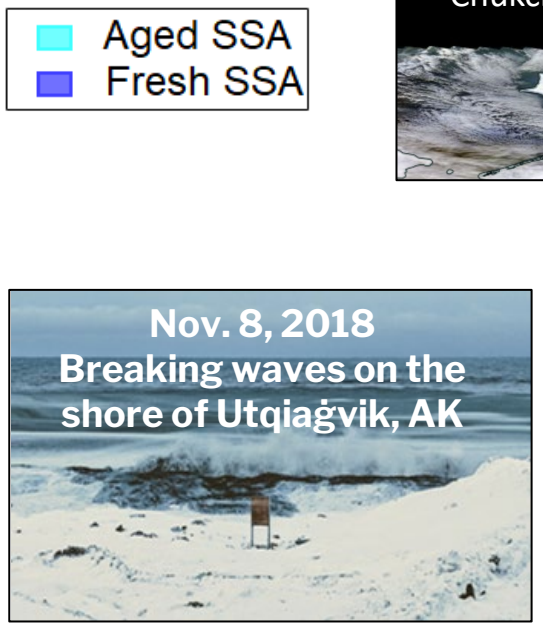
Early Winter (Nov.-Dec.) Sea Spray Aerosol

Utqiaġvik, AK (NSA)
Nov.-Dec. 2018





Fresh sea spray aerosol (SSA) from open Chukchi Sea and leads in Beaufort Sea



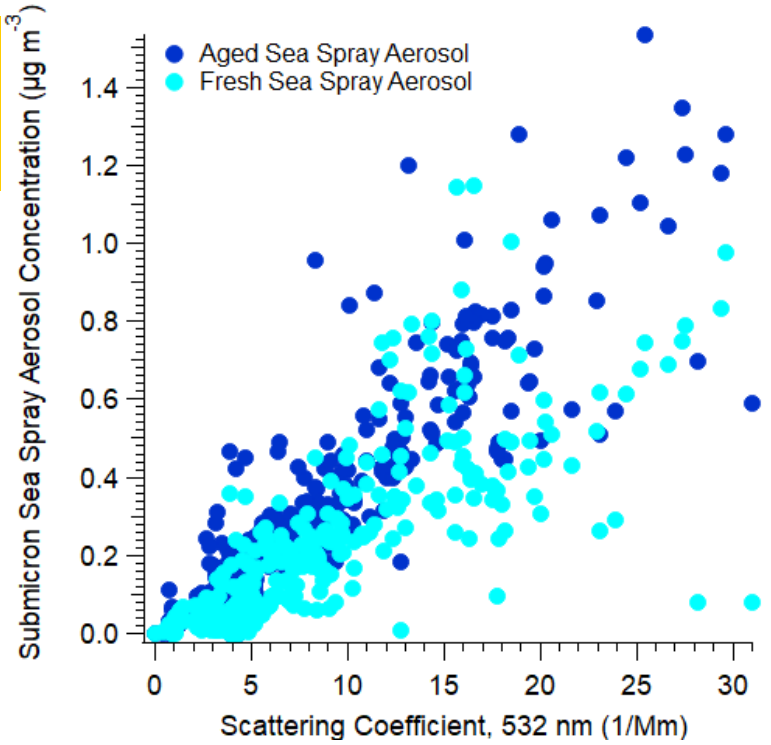
In Progress

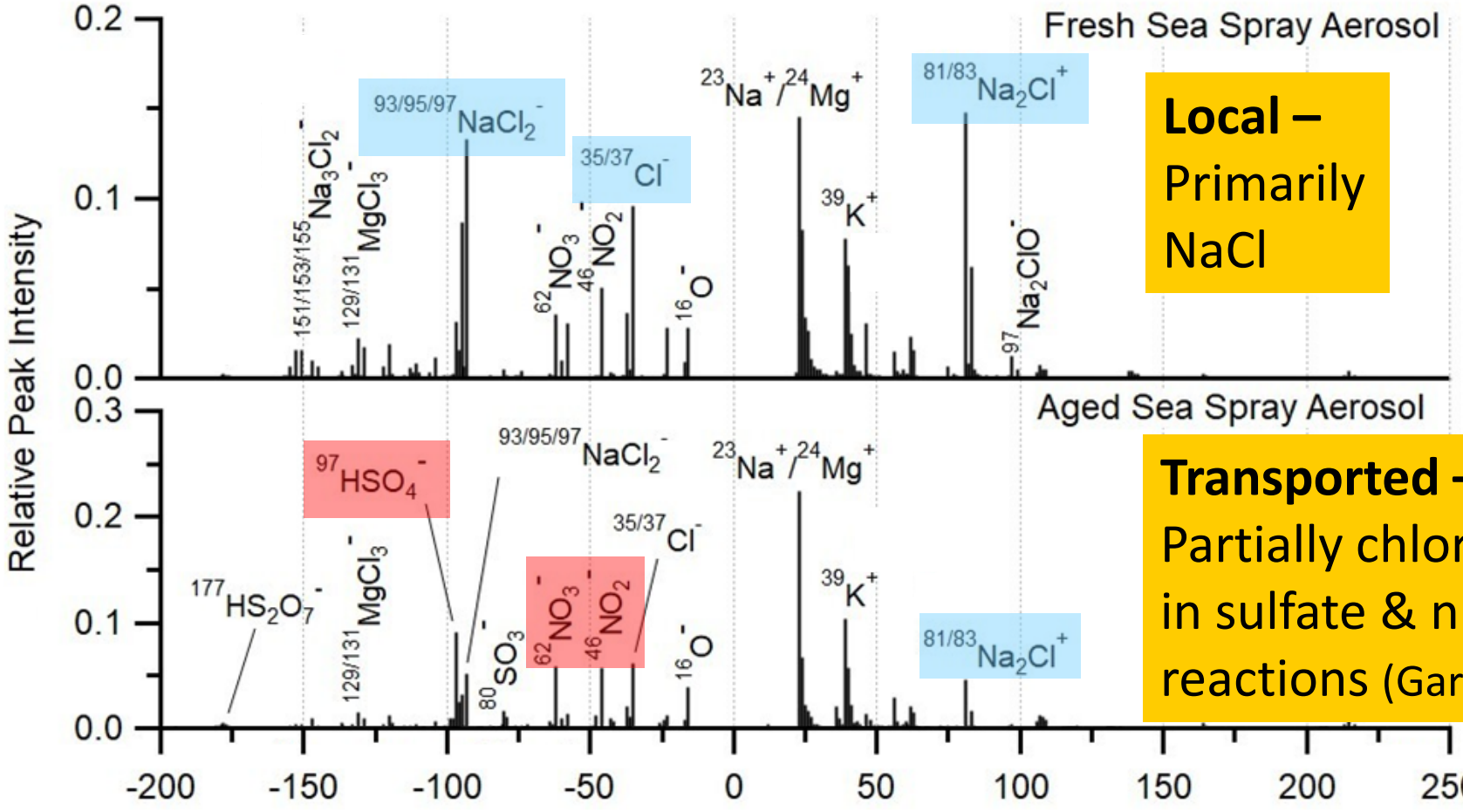
Single-Particle Mass Spectrometry (A-ATOFMS)

Early Winter Local & Transported Sea Spray Aerosol

Utqiaġvik, AK (NSA)
Nov.-Dec. 2018

Sea spray aerosol conc. is correlated (r^2>0.6) with scattering





Local – Primarily NaCl

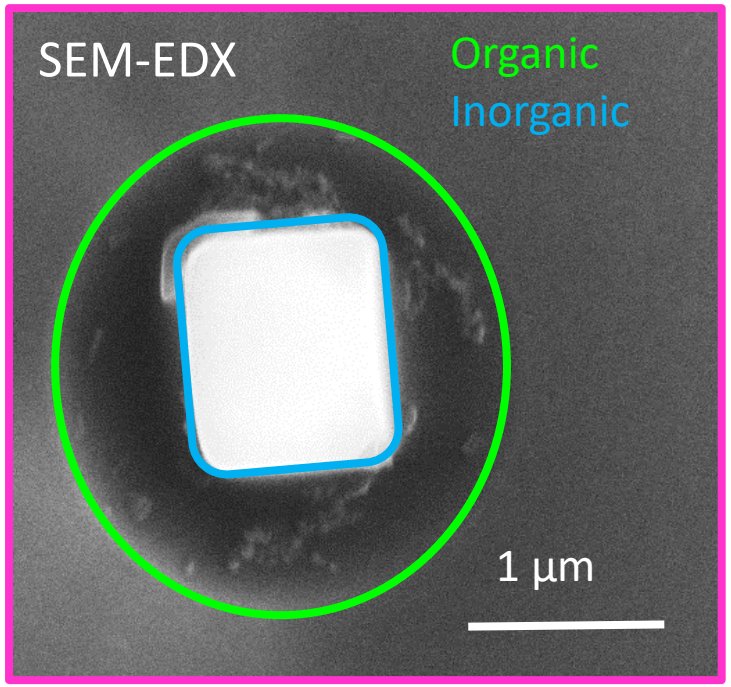
Transported – Partially chloride depleted; Enriched in sulfate & nitrate from multiphase reactions (Gard et al. 1998, *Science*)

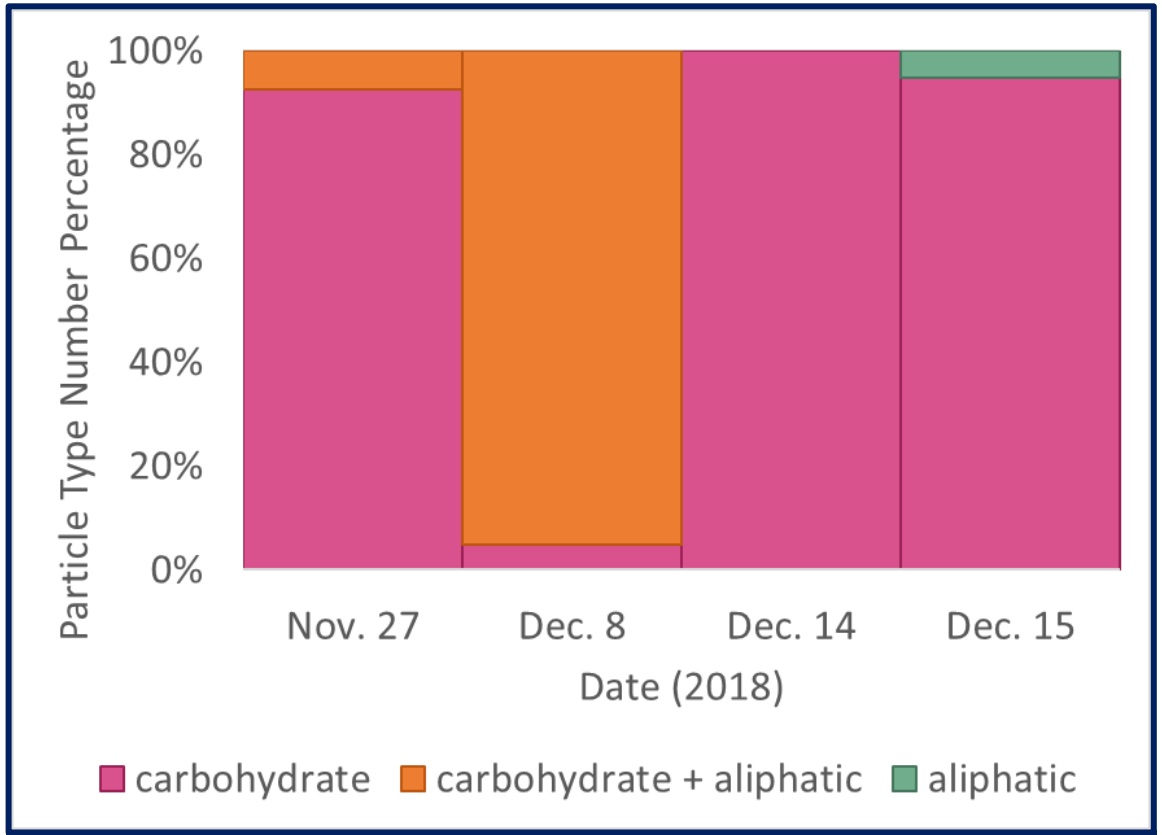
In Progress

Single-Particle Mass Spectrometry (A-ATOFMS)

Early Winter Sea Spray Marine Organic Coatings

Utqiaġvik, AK (NSA)
Nov.-Dec. 2018





Sea spray aerosol particle (1.0-1.8 µm) organic composition, by number percentage. Fluorescent

- **Marine carbohydrates (saccharides)** were the dominant sea spray aerosol organic compound class, followed by **marine carbohydrates and aliphatics (i.e. saccharides + fatty acids)**.
- Similar to Jan.-Feb. NSA sea spray aerosol results

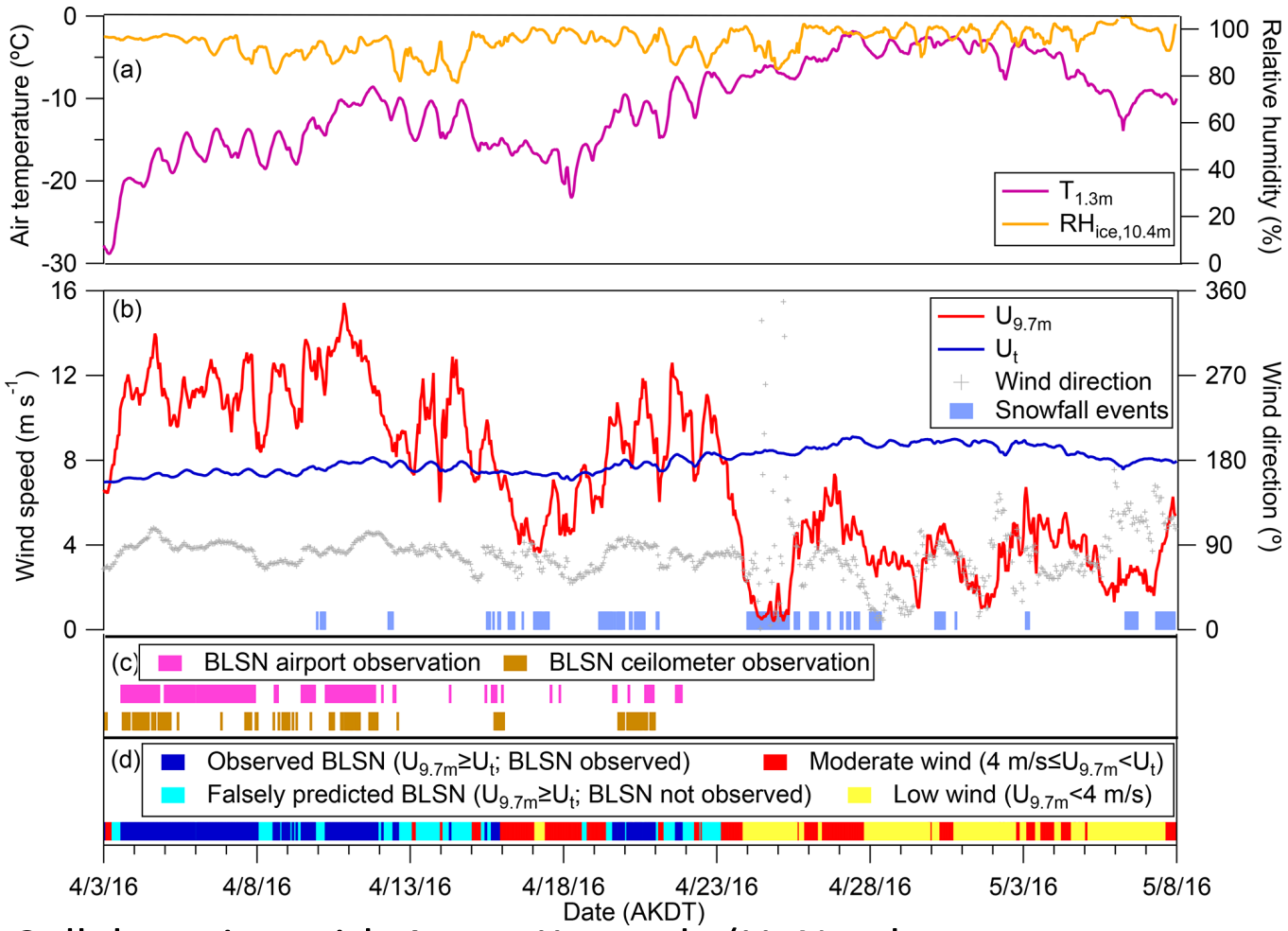
Individual sea spray aerosol particle Raman microspectroscopy

In Progress

Springtime (Apr.-May) Sea Spray Aerosol

Utqiaġvik, AK (NSA)
Apr.-May 2016

- Detailed investigation of blowing snow as a possible sea salt aerosol source via meteorological & single-particle chemical analysis



- Occurrence of blowing snow over-predicted based on wind speed & temperature
- Supermicron particles scavenged by blowing snow
- Individual sea salt particle composition (CCSEM-EDX) consistent with sea spray aerosol from upwind, local leads

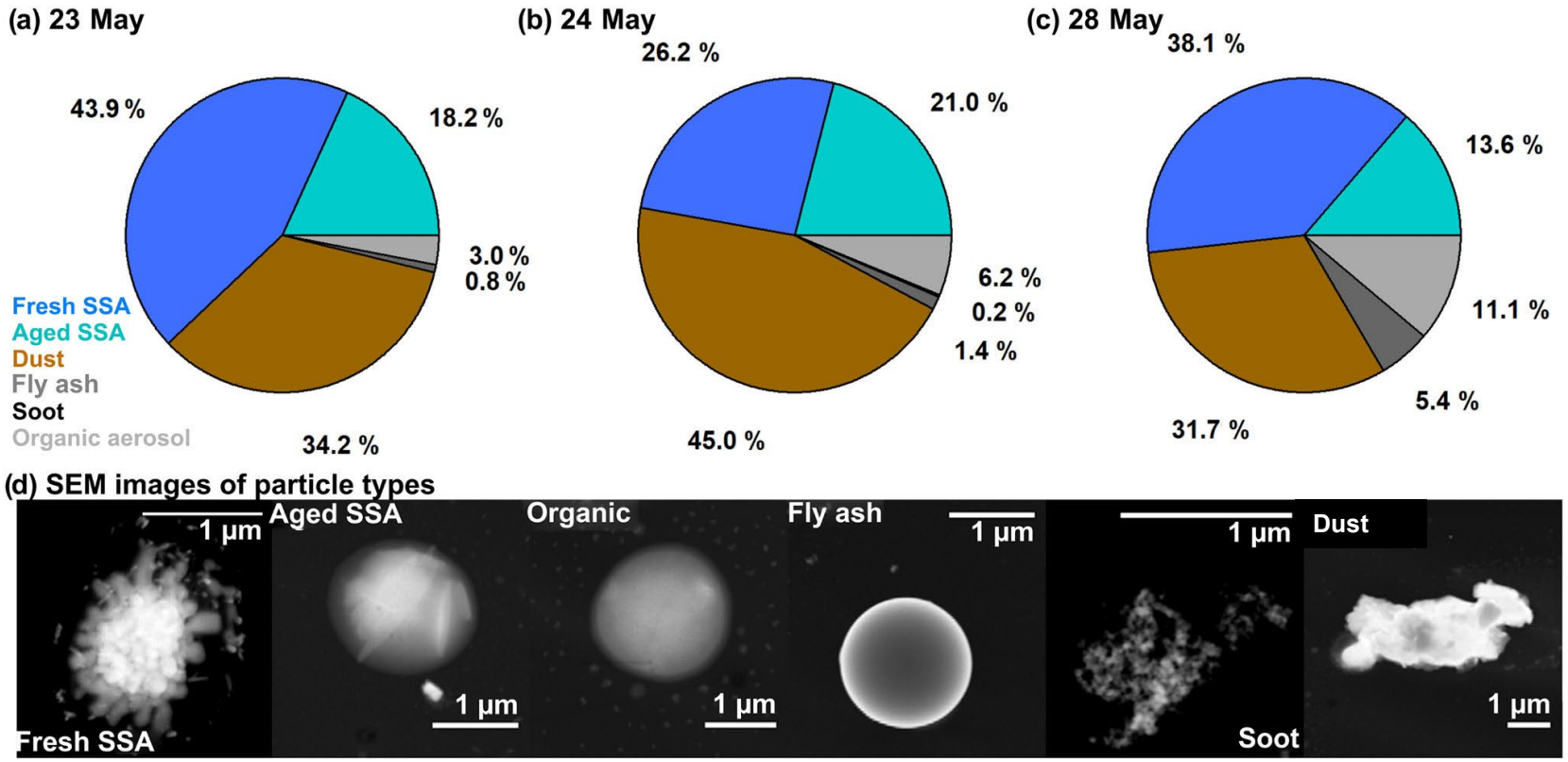
Collaboration with Aaron Kennedy (U. North Dakota) for blowing snow (BLSN) evaluation

Chen and Pratt et al. 2022, *ACP.* doi:10.5194/acp-22-15263-2022

Springtime (Mar.-May) Sea Spray Aerosol

Oliktok Point, AK (AMF3)
Mar.-May 2017

SEM-EDX



- Fresh sea spray aerosol (SSA), dust, and aged SSA are major contributors to coarse mode aerosol (>1.15 µm)
- Fresh sea spray aerosol size and composition consistent with local production
- Mineral & marine sources can explain measured INPs (Jessie Creamean, CSU)

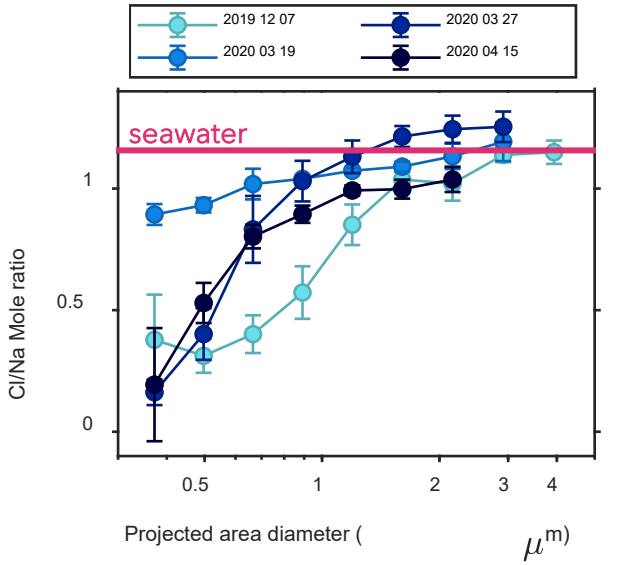
Creamean et al. 2018, *ACP.* doi:10.5194/acp-18-18023-2018

MOSAiC: Sea Spray Aerosol Observed Year-round

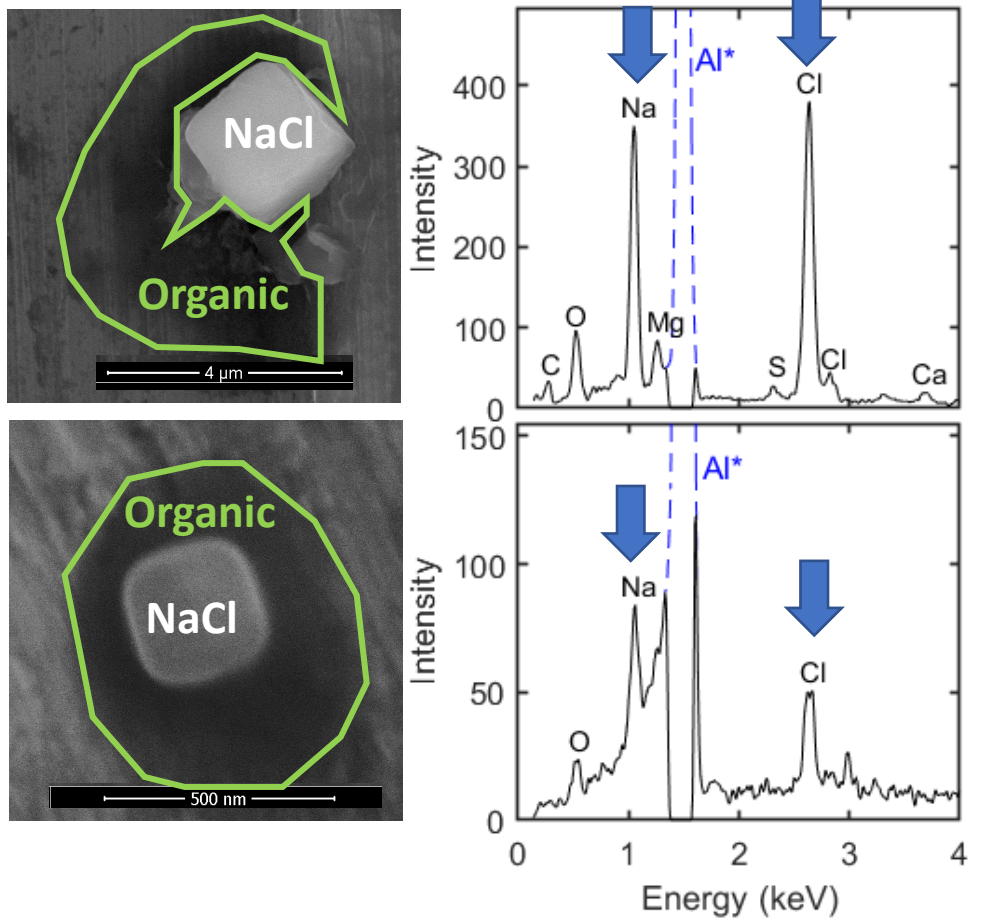
High Arctic, Oct. 2019 – Sep. 2020

- Sea salt particles often consisted of salt cores with organic coatings, similar to previous NSA observations

- Cl/Na ratios for > 1 µm particles similar to seawater indicating locally-produced sea salt aerosol
- Submicron (< 1 µm) sea salt aerosol depleted in Cl, likely from reactions during long-range transport



Cl/Na ratios calculated for 100-1000 individual sea salt particles per sample



Sea spray aerosol particles with organic coatings

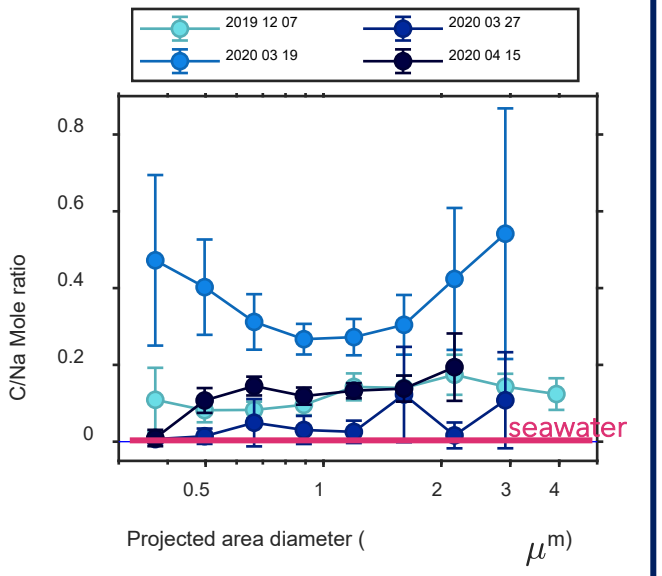
Note: Additional sea salt morphologies currently under investigation

In Progress

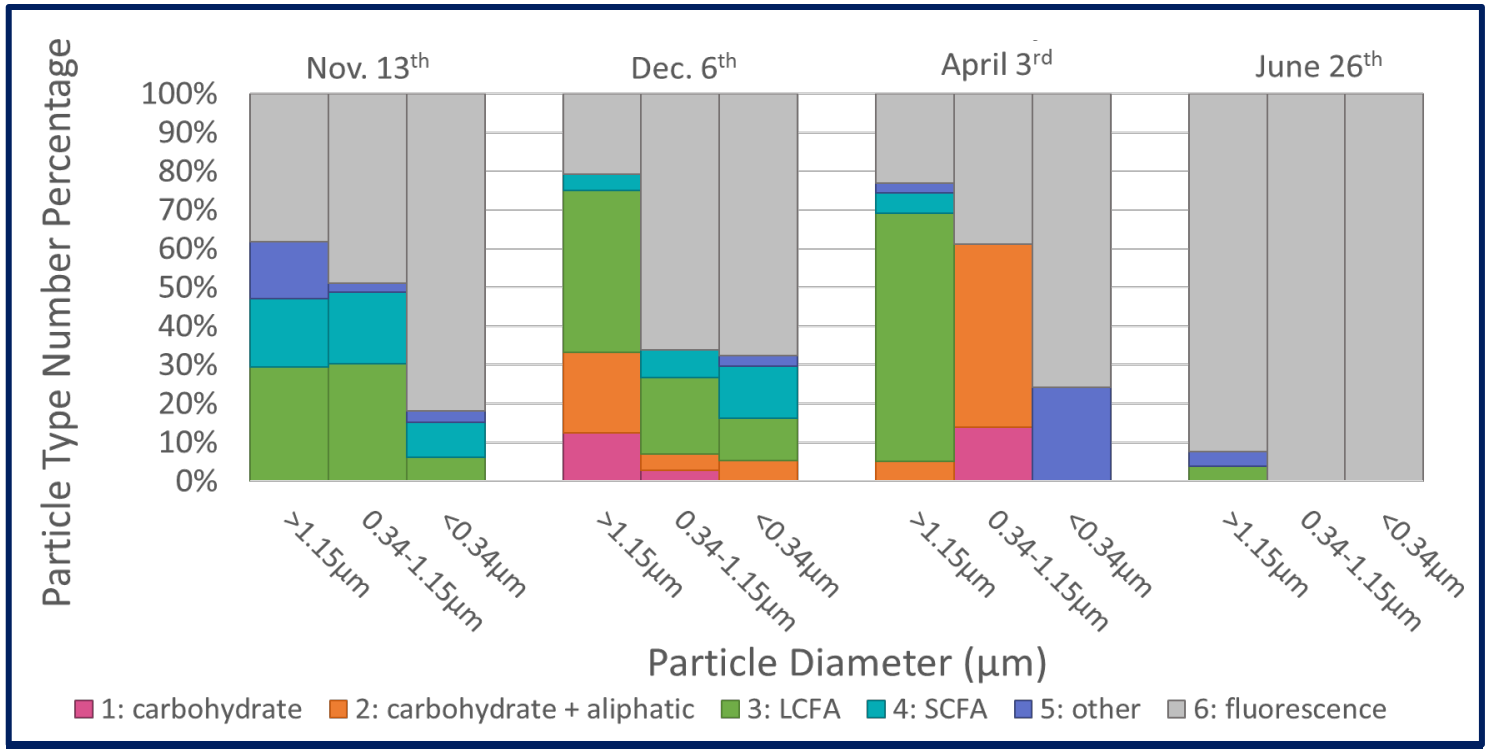
MOSAiC: Marine Organic Coatings & Fluorescence

High Arctic, Oct. 2019 – Sep. 2020

- **Large range in C/Na ratios** consistent with mid-latitude sea spray aerosol (e.g. Pham et al. 2017, *ACS Earth Space Chem*; Mirrielees et al. 2022, *ACS Meas. Sci. Au*)



C/Na ratios calculated for 100-1000 individual sea salt particles per sample (CCSEM-EDX)



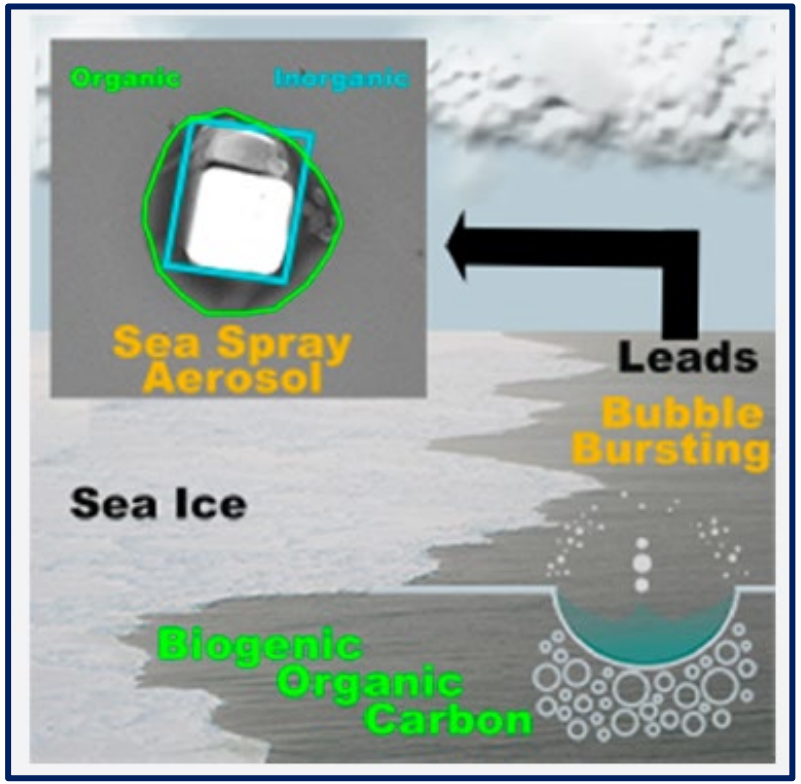
Particle organic composition, by percentage (Raman microspectroscopy)

- **Fatty acids** (long chain and short chain) were the dominant non-fluorescent organic class identified, followed by carbohydrates (saccharides).
- Fluorescence was observed in 60% of the particles analyzed, supporting the presence of biological material.

In Progress

Summary and Ongoing Work

- **Sea spray aerosols were observed year-round in the coastal Arctic and High Arctic**
  - Local production of sea spray aerosol (all sizes) and transport of aged sea spray aerosol (primarily <1.0 um)
  - Many sea spray aerosol particles have organic coatings, dominated by marine saccharides and fatty acids
- *Ongoing work & next steps:*
  - Identify potential sea salt aerosol from blowing snow sublimation (morphological & chemical analysis)
  - Continued analysis of sea spray aerosol organic composition & fluorescence
  - Connections to CCN and INP data (comparisons to Creamean et al. 2022, Nat. Comm.)



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