EMSL

Chemical Composition and Mixing State of Wintertime Aerosol from the European Arctic Site of Gruvebadet, Svalbard

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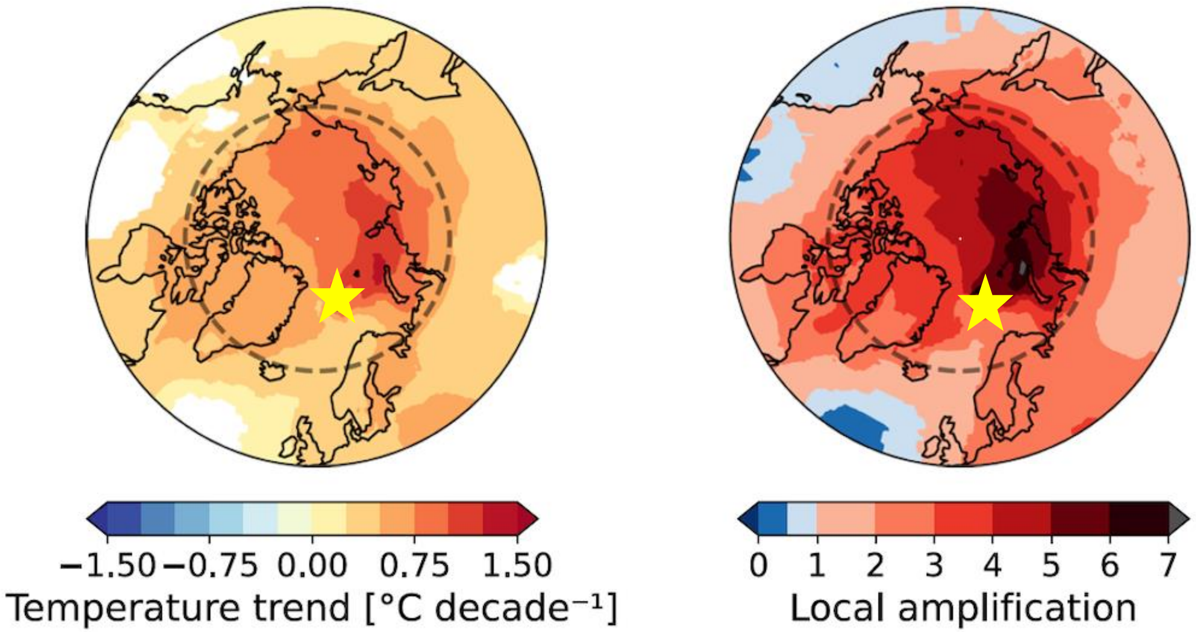
Post Masters RA

A rapid warming Arctic climate

The Arctic has warmed ‘nearly four times faster’ than the global average since 1979

Model simulation shows faster warming rate at Arctic



Rantanen et al., 2022 nature commsev.

- Arctic Ocean was warming faster than **0.75°C decade^{-1}**, with a maximum warming near **Svalbard**.

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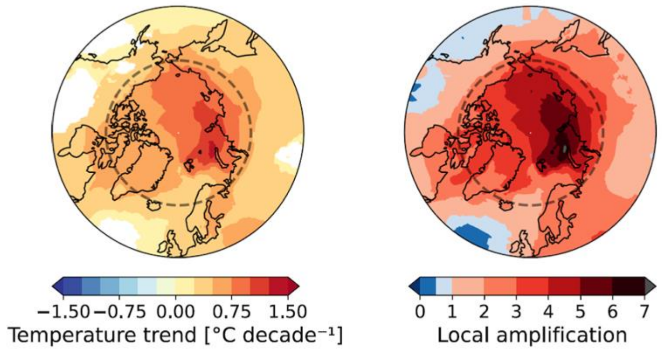
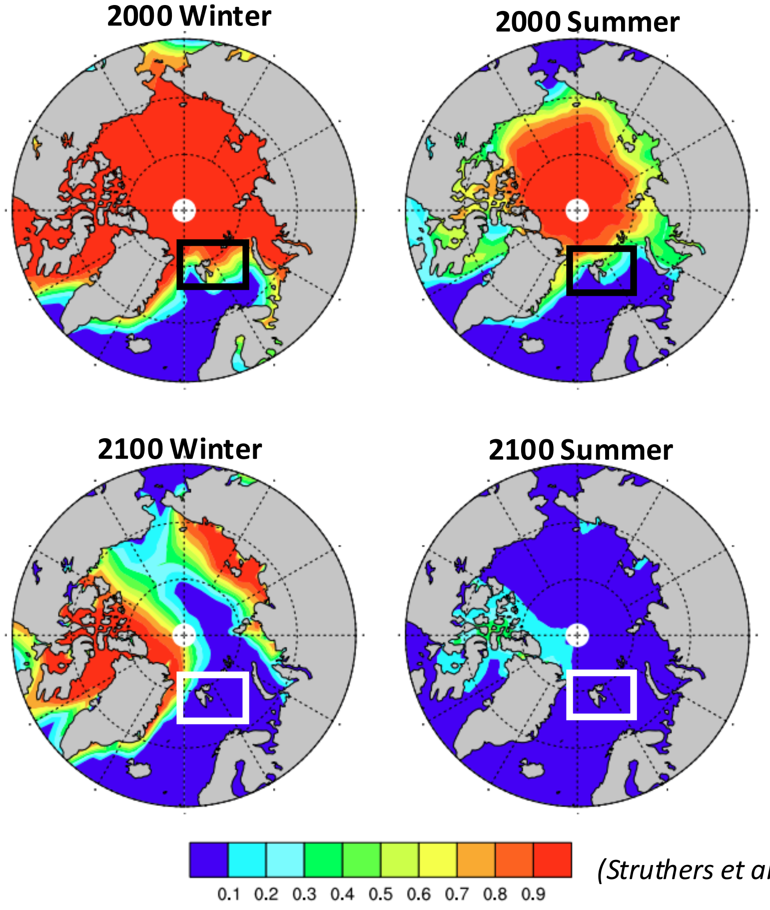


Rantanen et al., 2022 nature commsev.

Modeled Sea Ice Fraction

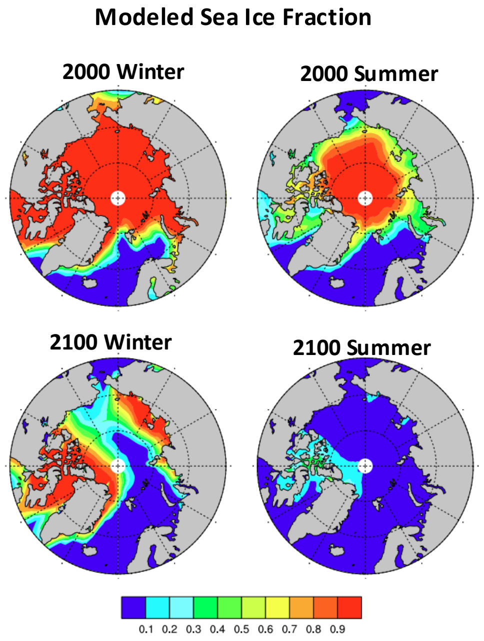


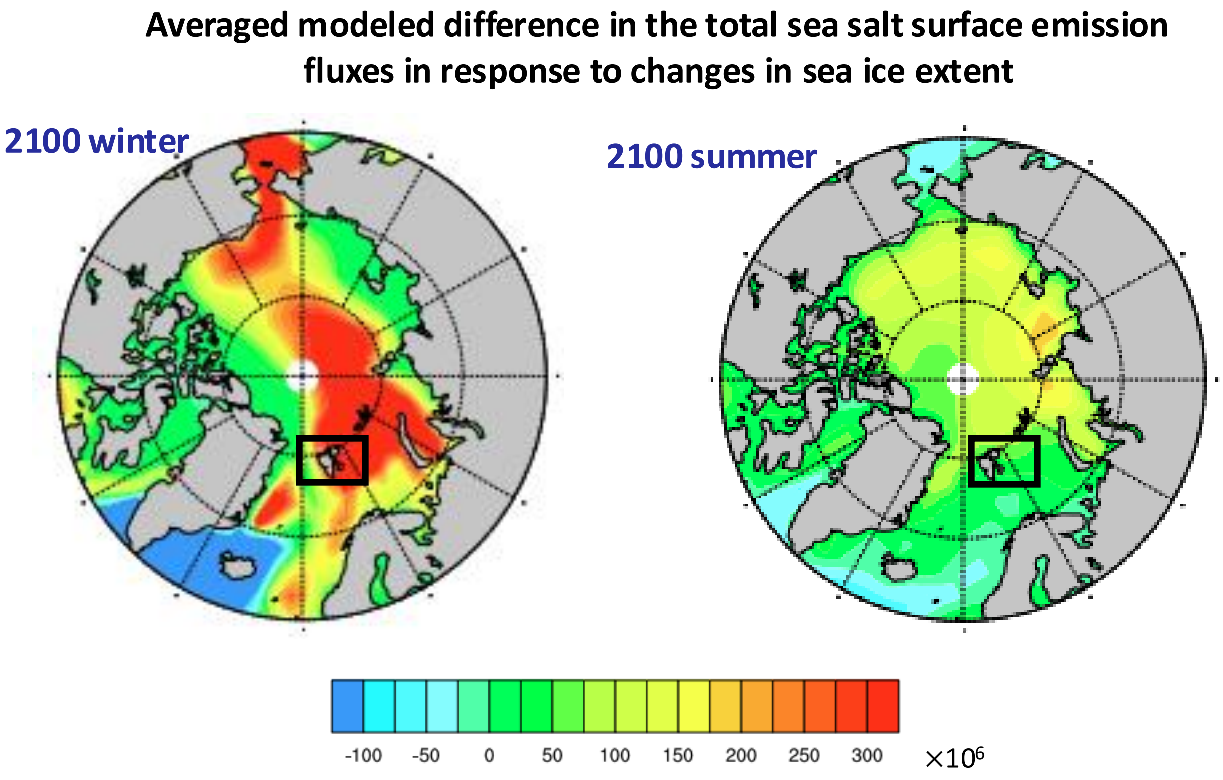
(Struthers et al., ACP, 2011)

- Summer Arctic sea ice may **completely vanish** by the end of the 21st century or earlier.

The Arctic has warmed 'nearly four times faster' than the global average since 1979

A rapid warming Arctic climate





(Struthers et al., ACP, 2011)

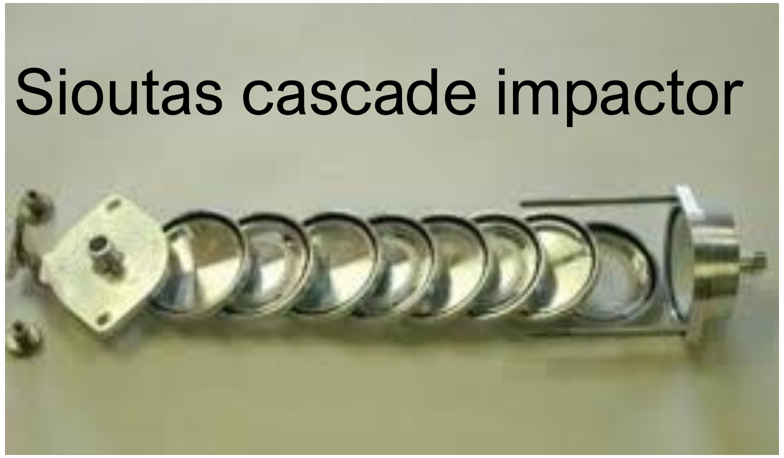
- **Sea salt aerosol (SSA) emissions increase** in response to a decrease in sea ice.

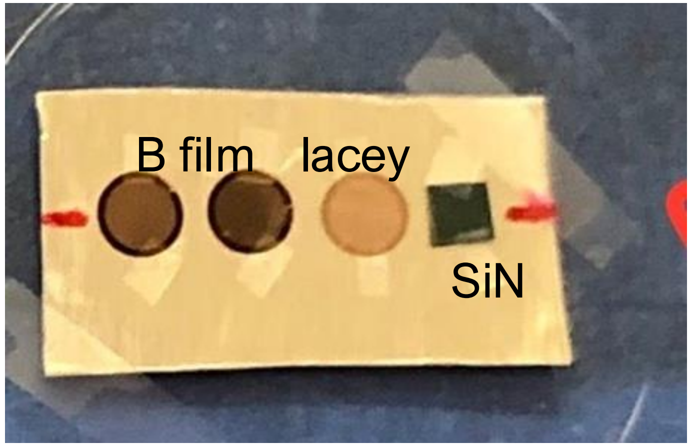
Sampling site and collection

- Observations were carried out from **November to December 2020** at **Gruvebadet Atmospheric Laboratory (GAL).**
- **9 days** of samples were collected.
- Gruvebadet Atmospheric Laboratory is dedicated to the study of **aerosol and has continuous long-term observations since 2010.**

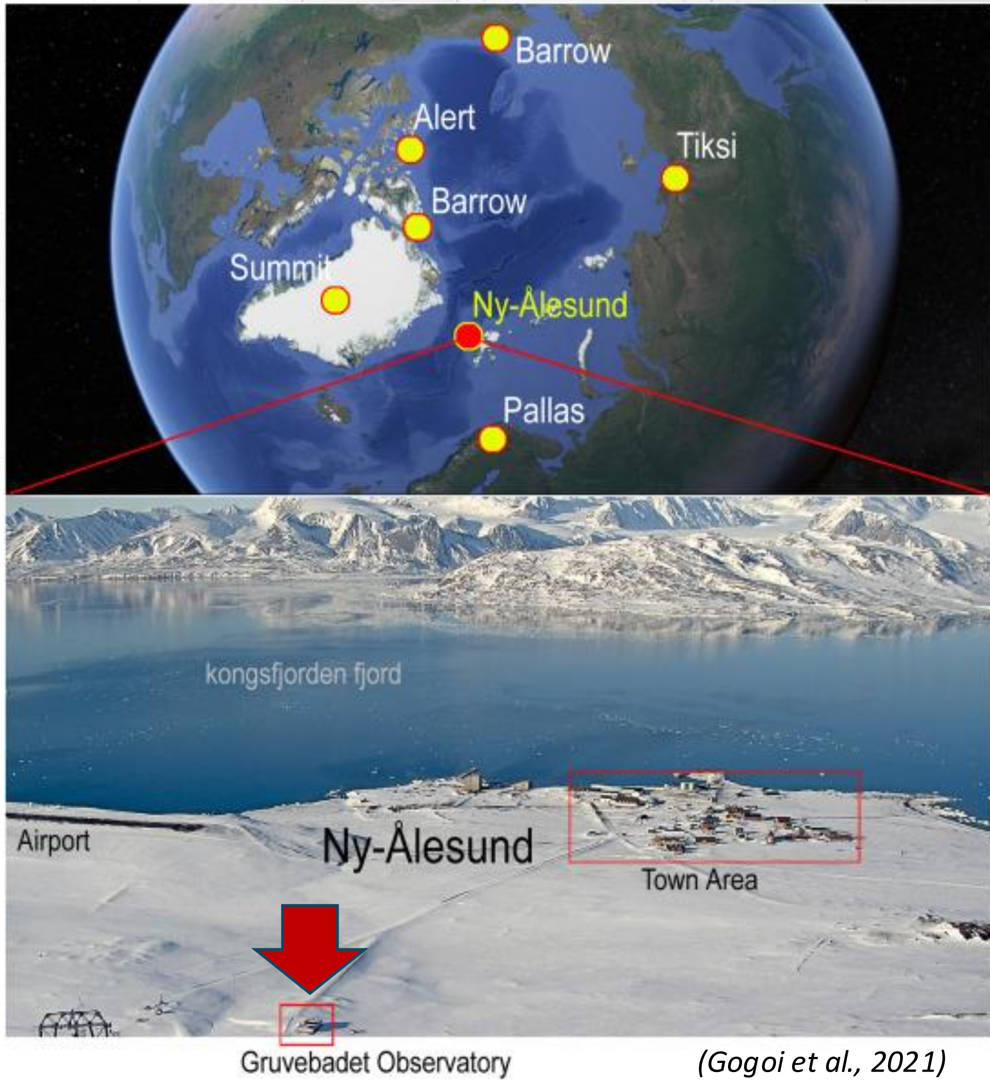
Sample collection:

Impactor: Four-stage Sioutas Cascade impactor (SKC. 9L/min)
Substrate: TEM grids (B-film and lacey)
Size range: 0.25 μm to 10 μm





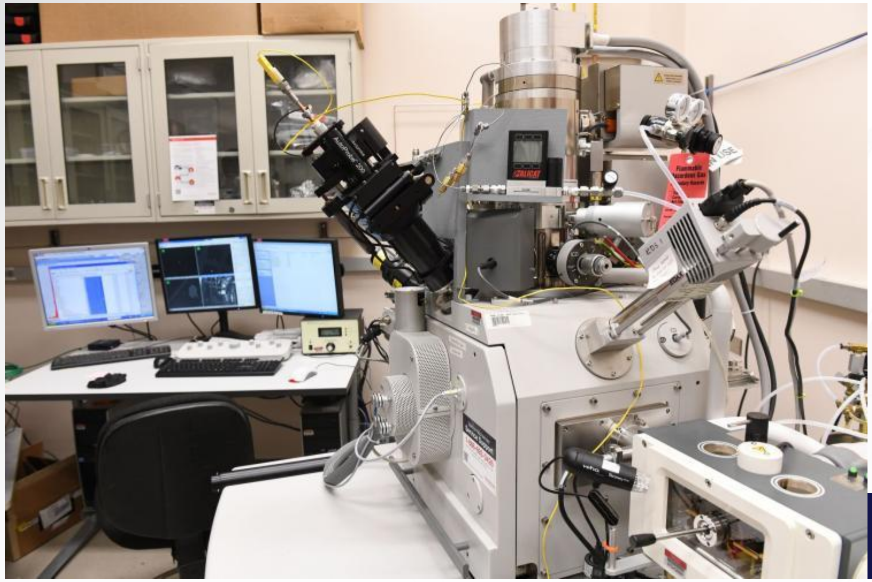
Geographical location of GAL



(Gogoi et al., 2021)

Analysis Overview

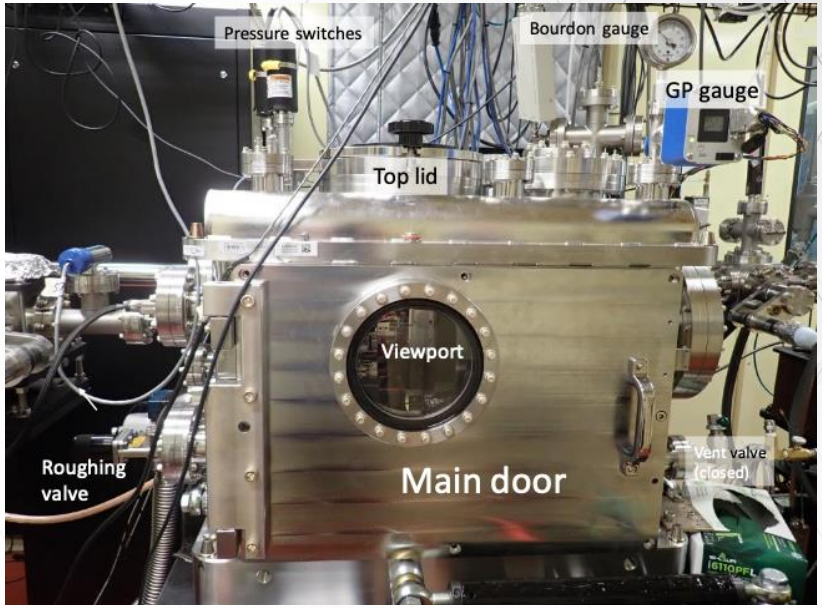
CCSEM-EDX



EMSL

(Susan Mathai, Zezhen Cheng, Nurun Nahar Lata., Quanta, EMSL)

STXM-NEXAFS



ALS
ADVANCED LIGHT SOURCE

(Zezhen Cheng, Nurun Nahar Lata, Mathew Marcus, STXM, ALS)

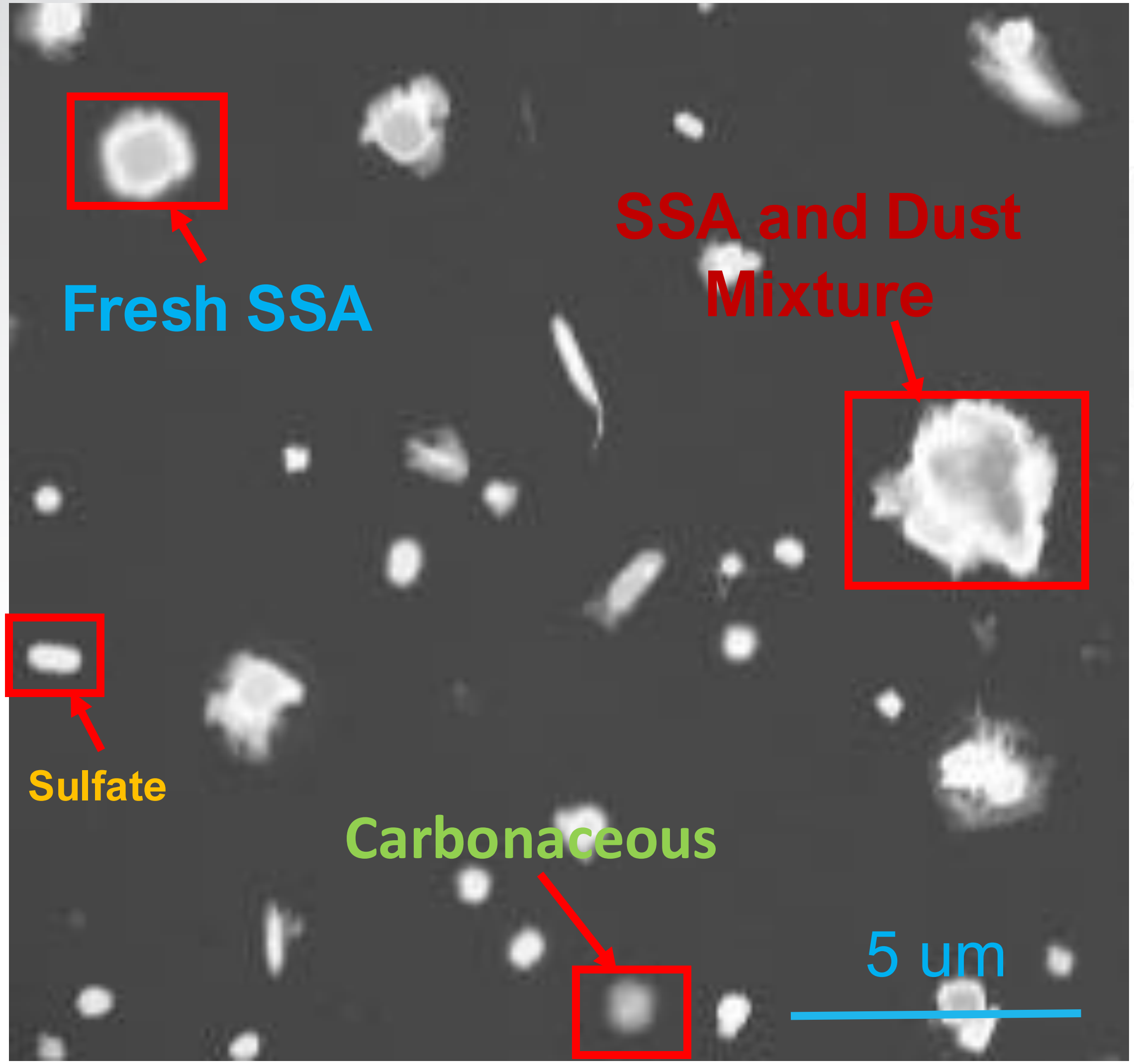
Offline analysis:

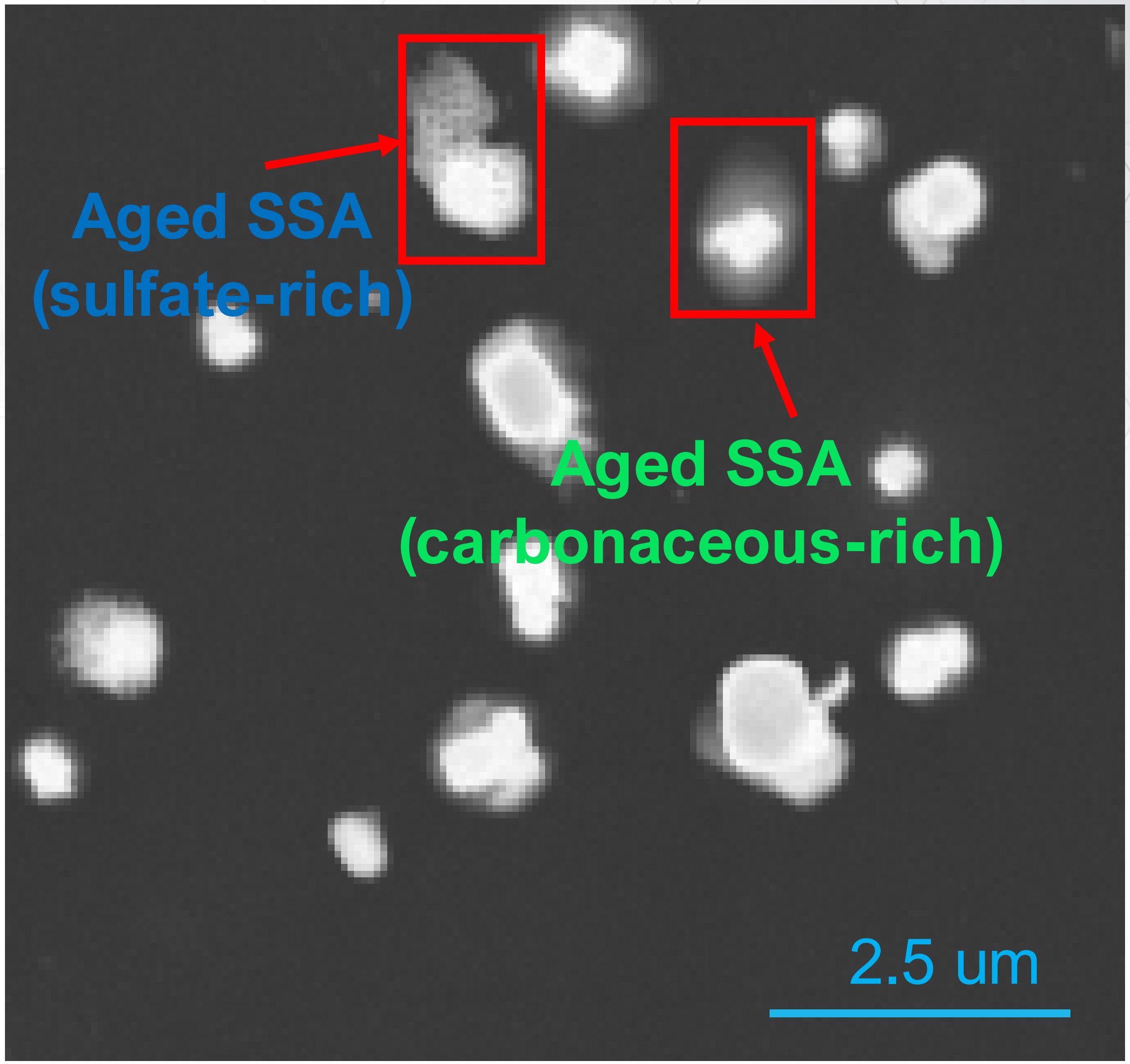
Elemental composition and Morphology: computer-controlled scanning electron microscopy, coupled with energy-dispersive X-ray spectroscopy (CCSEM-EDX)

Chemical composition and mixing state: Scanning transmission X-ray microscopy (STXM) and near edge X-ray absorption fine structure spectroscopy (NEXAFS).

What are the chemical species that contribute to Arctic aerosol population?

Morphologies of Arctic aerosol

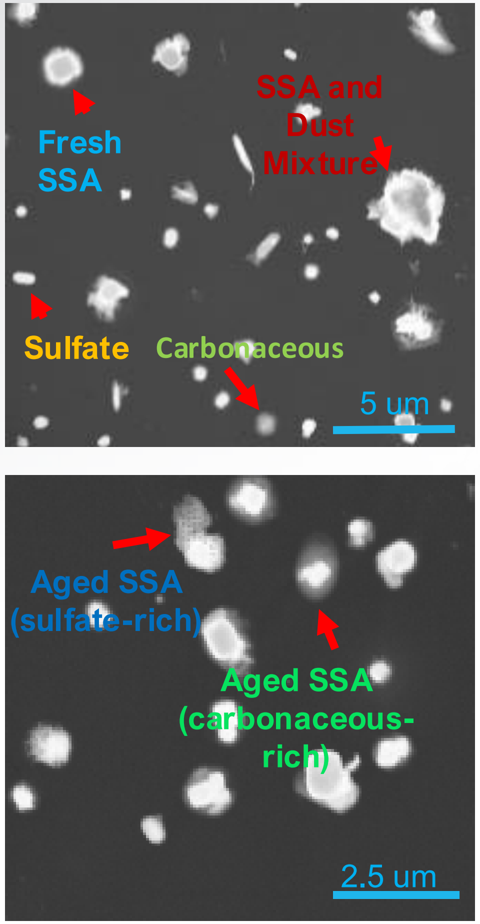


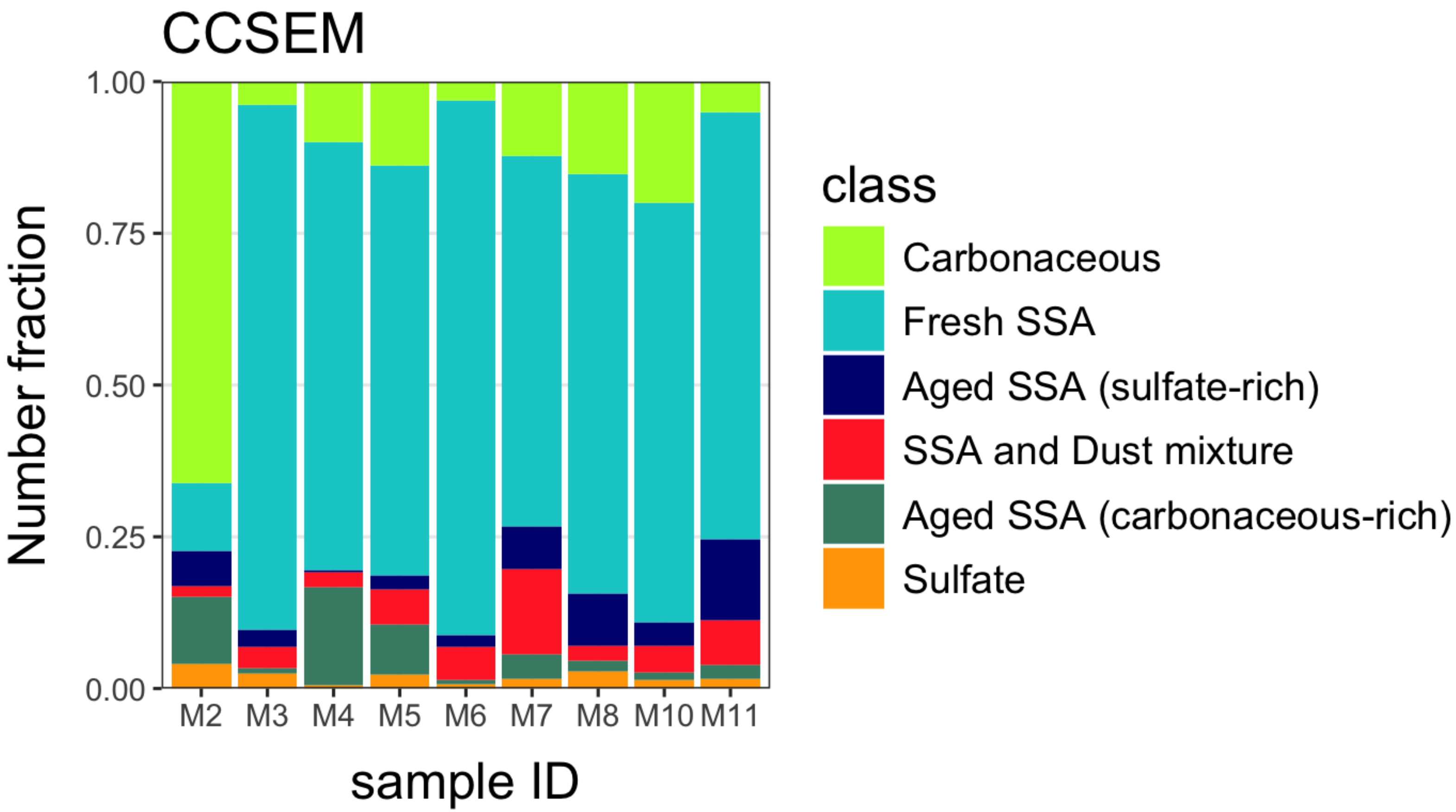


1. Six species were identified.

What are the chemical species that contribute to Arctic aerosol population?

Morphologies of Arctic aerosol





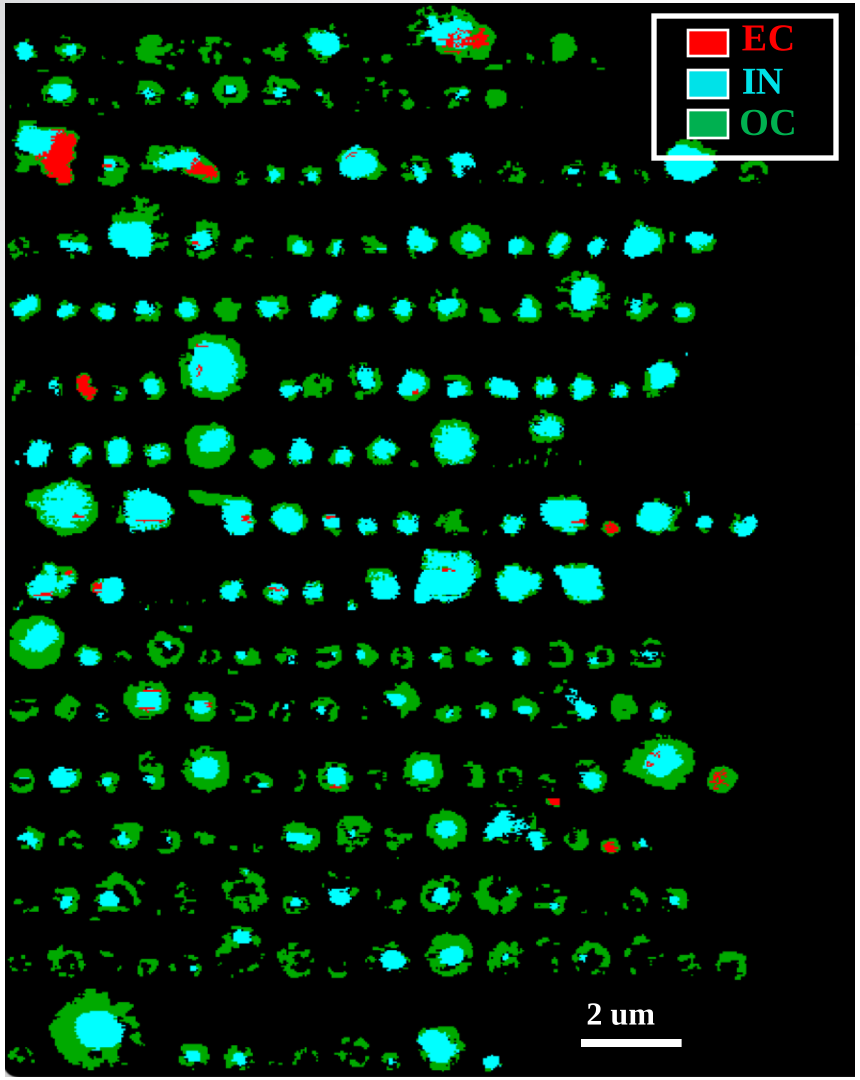
2. **Fresh SSA** is most abundant in Arctic aerosol.

3. Aged SSA: **enriched in sulfur** or **enriched in carbon and oxygen,** indicating different aging mechanisms.

4. Non-negligible number of **SSA** were **mixed with dust particles**.

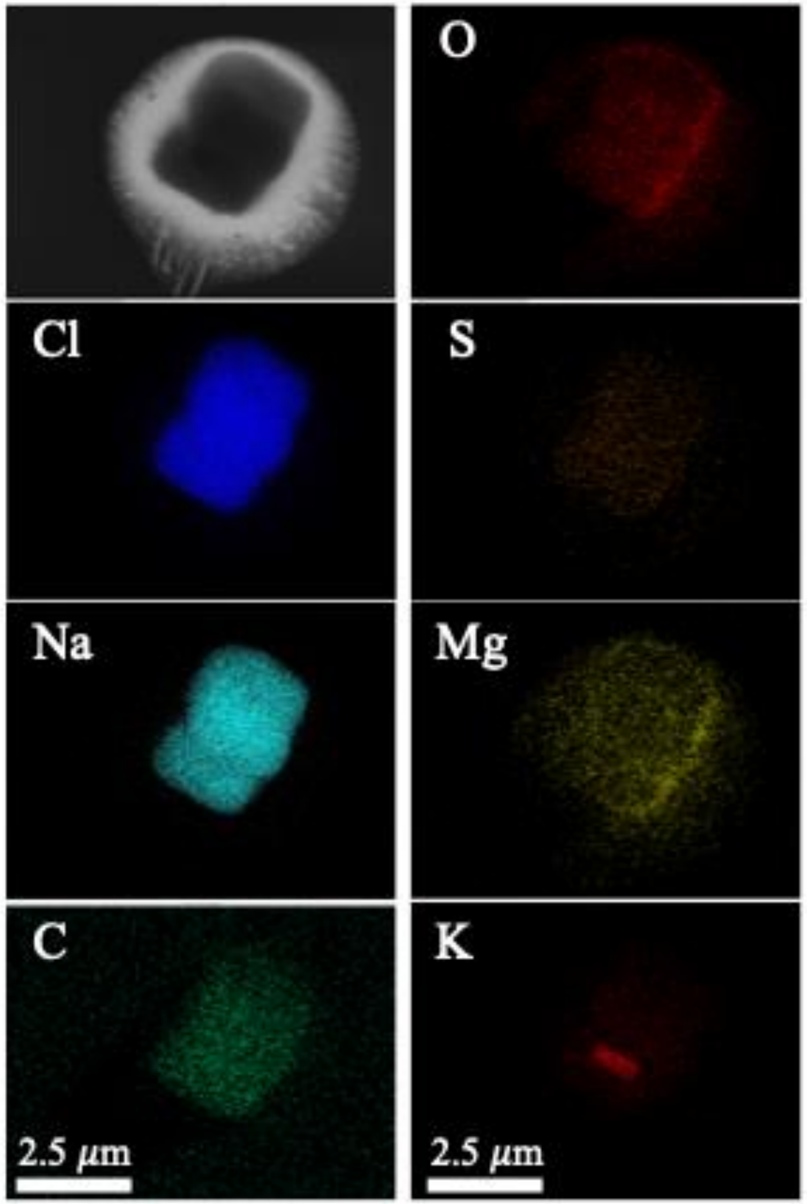
Morphology and internal mixing of sea salt aerosols (SSA)

STXM carbon map shows mixing states



Core-shell morphology is commonly found in both **fresh and aged** particles.

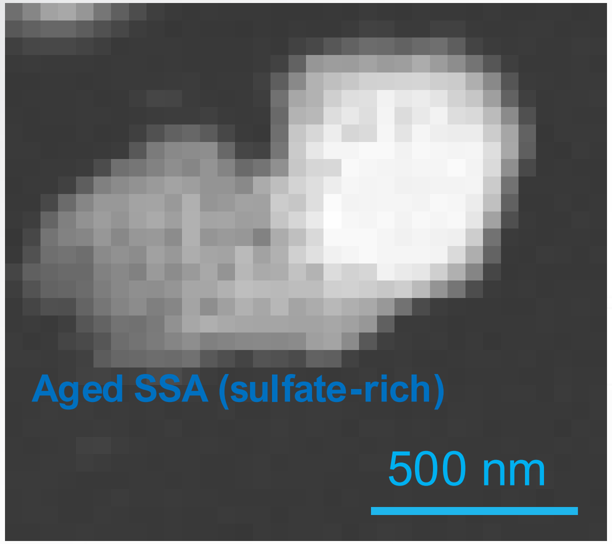
Elemental map of a sea salt particle

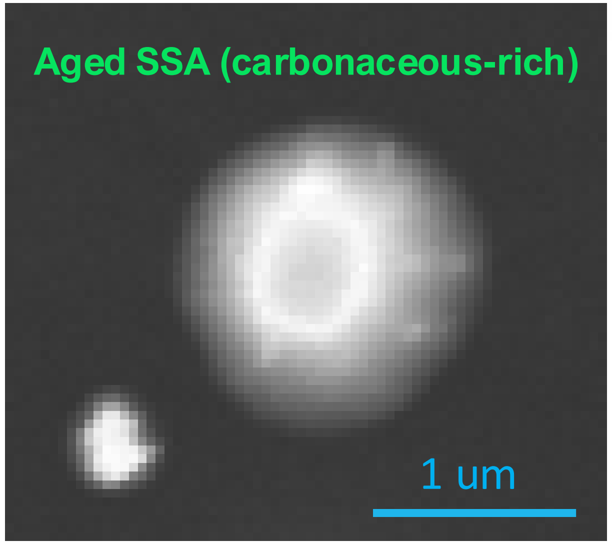


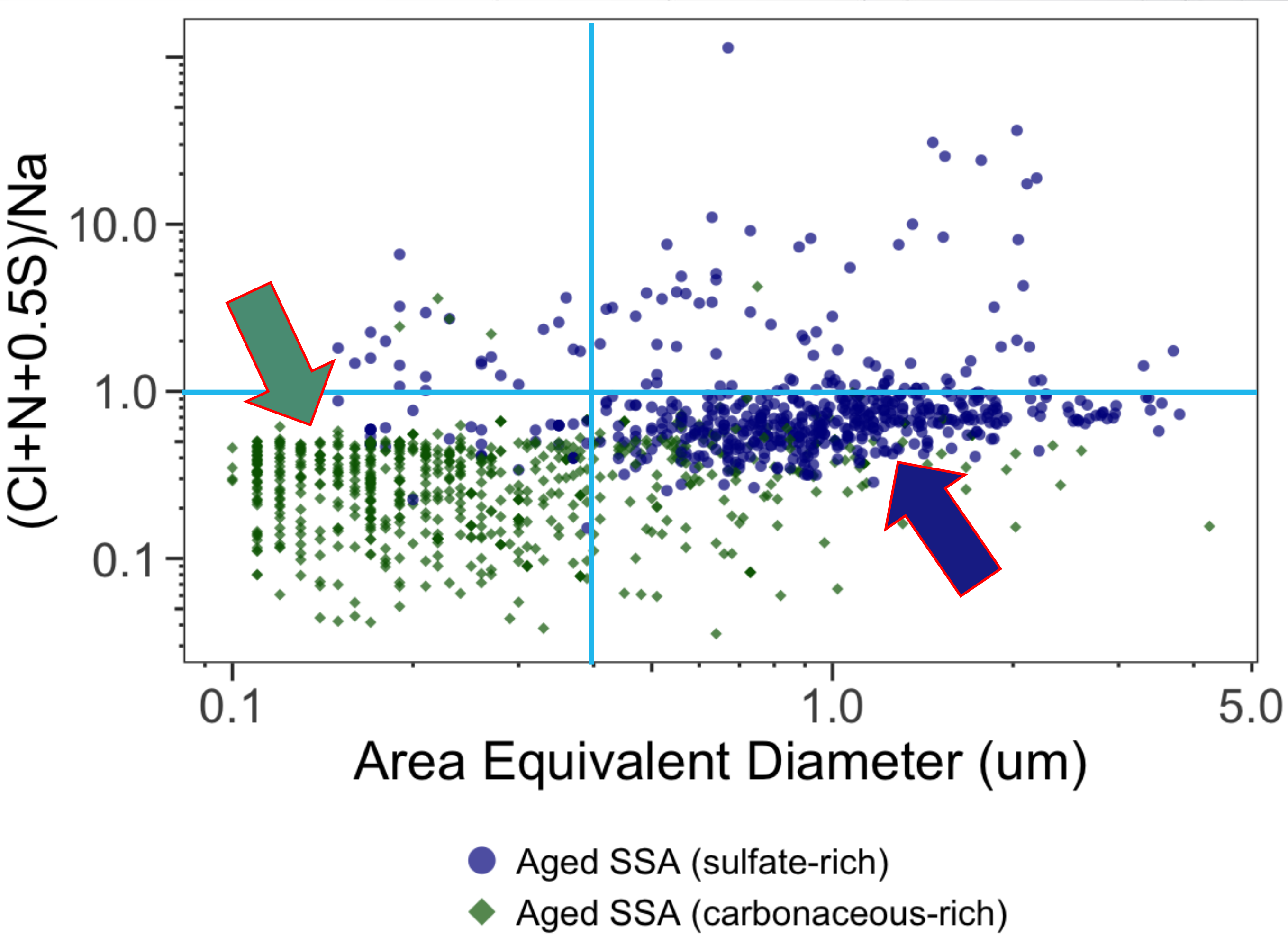
The organic shell is **enriched in Mg**, forming needle-like structure.

Aging mechanisms of SSA particles

Two morphologies of Aged SSA



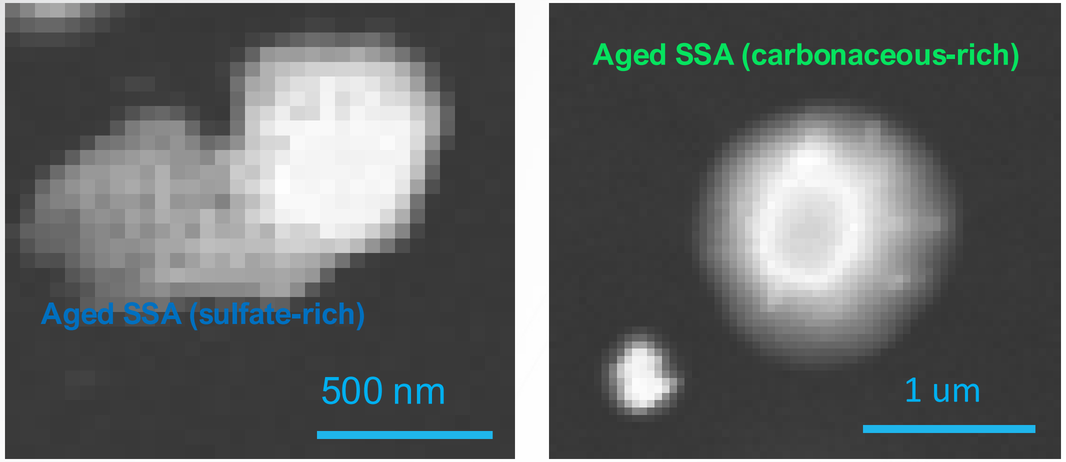




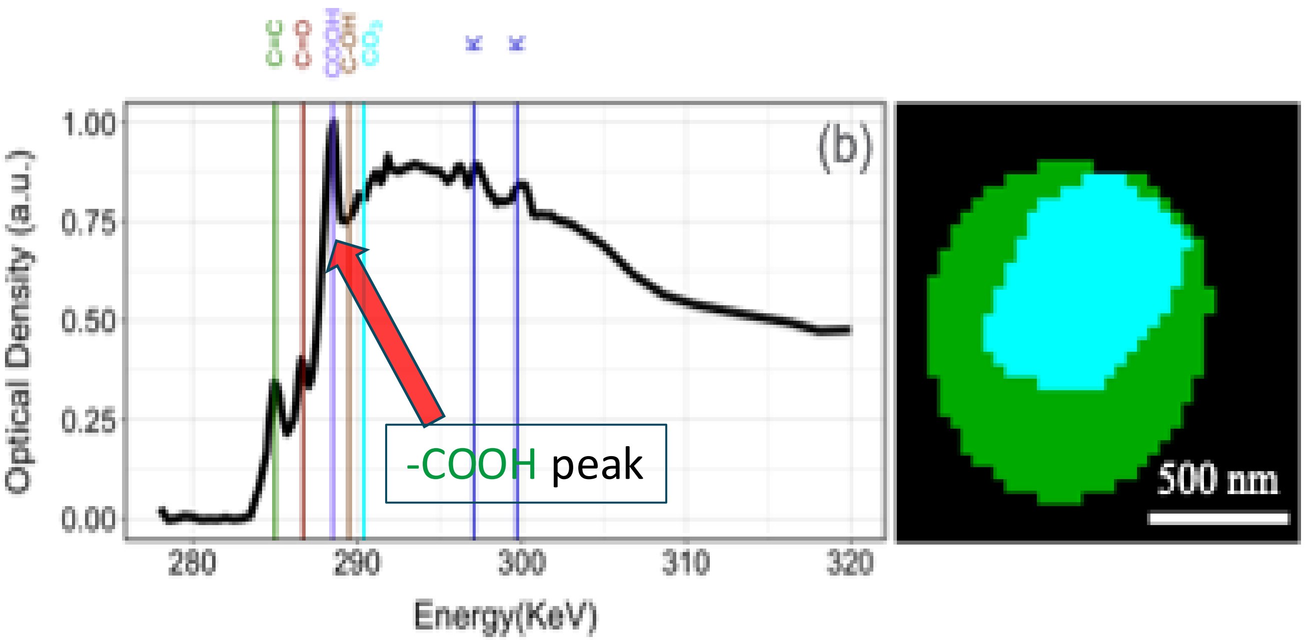
- **Aged SSA (carbonaceous-rich)** is generally **smaller** than **Aged SSA (sulfate-rich).**
- $\frac{Cl+N+0.5S}{Na}$ of **Aged SSA (sulfate-rich)** is close to **unity.**
- $\frac{Cl+N+0.5S}{Na}$ of **Aged SSA (carbonaceous-rich)** is smaller.

Aging mechanisms of SSA particles

Two morphologies of Aged SSA



Functional groups presented in aged SSA



- **Two aging mechanisms**:

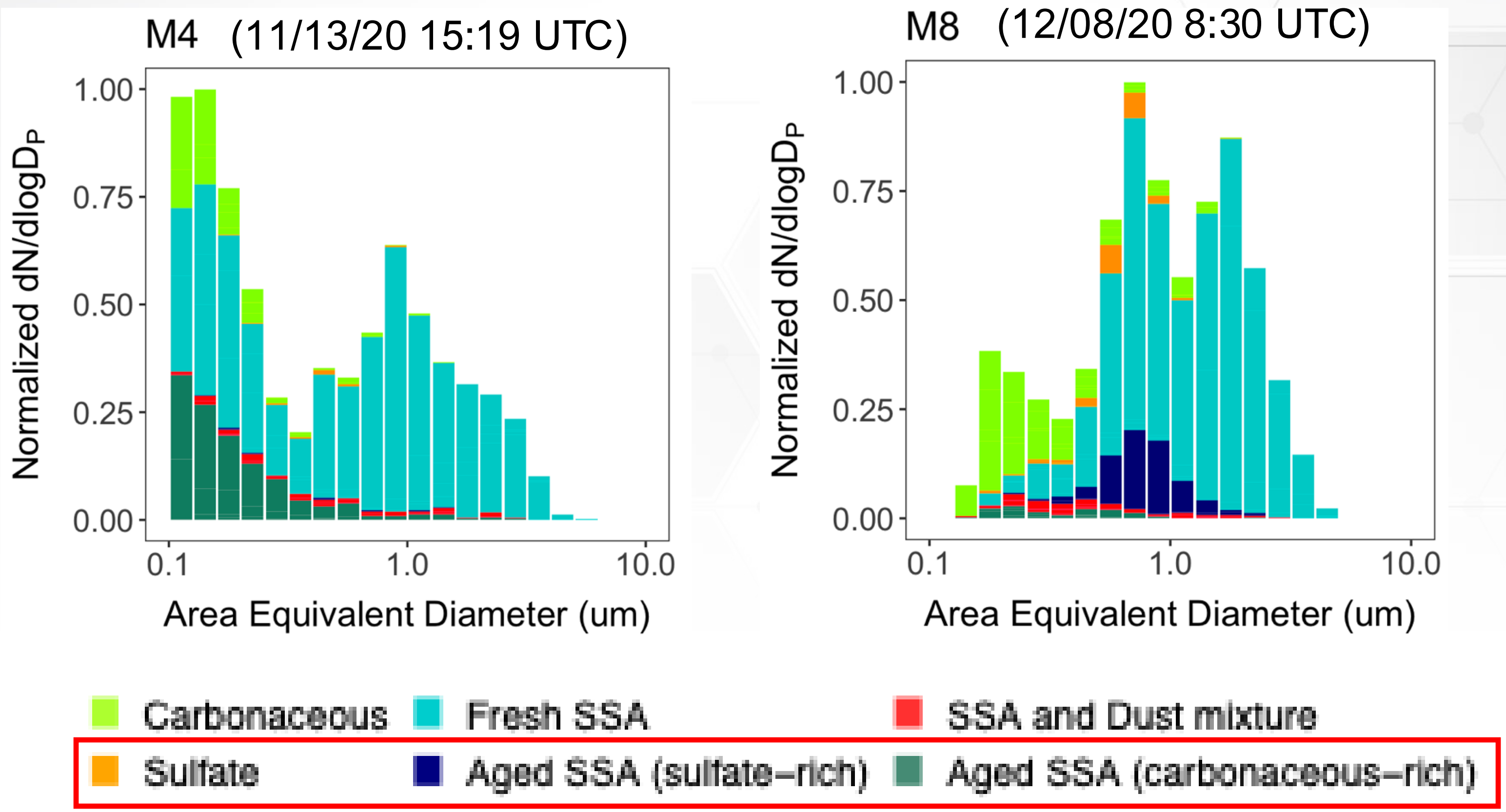
  (1) chlorine depletion by **organic acid**;

  (2) chlorine depletion by **sulfuric acid**.

- The presence of "**-COOH**" indicates the organic coating of SSA might contain **organic acid**.

Case studies

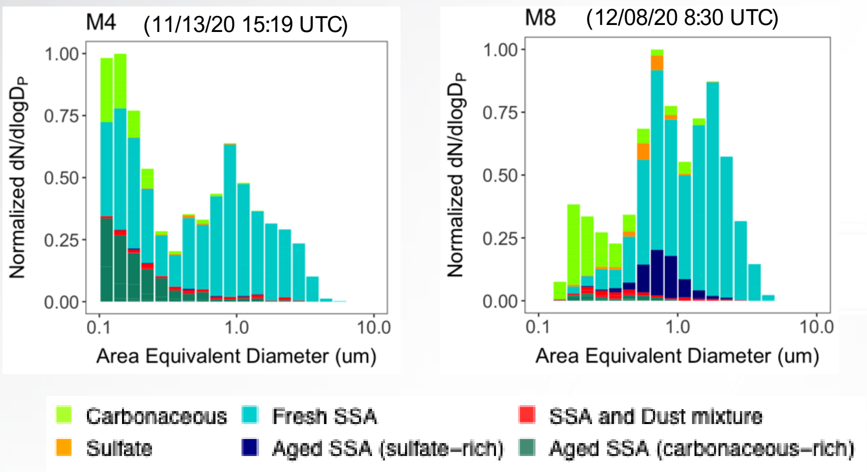
Size-resolved chemical composition



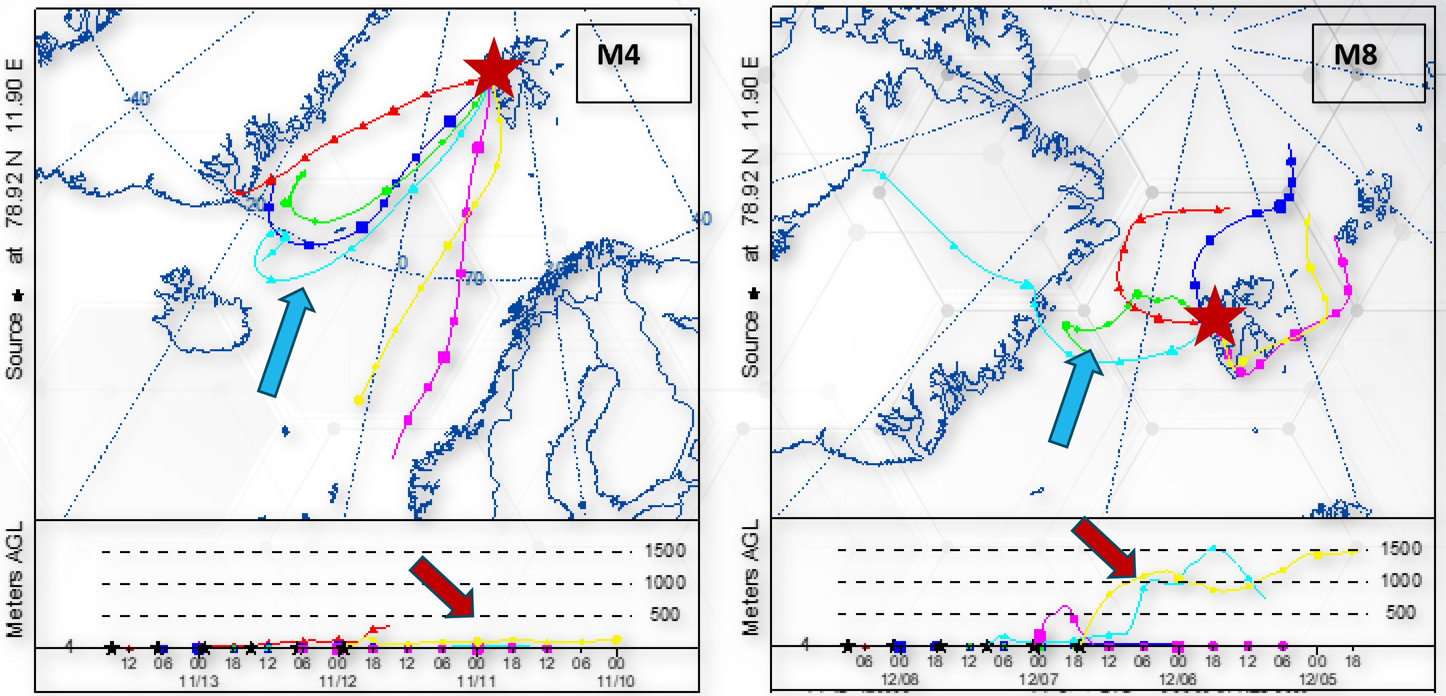
- M4: very low fraction of Sulfate and Aged SSA (sulfate-rich).
- M8: relatively low fraction of Aged SSA (carbonaceous-rich).

Case studies

Size-resolved chemical composition



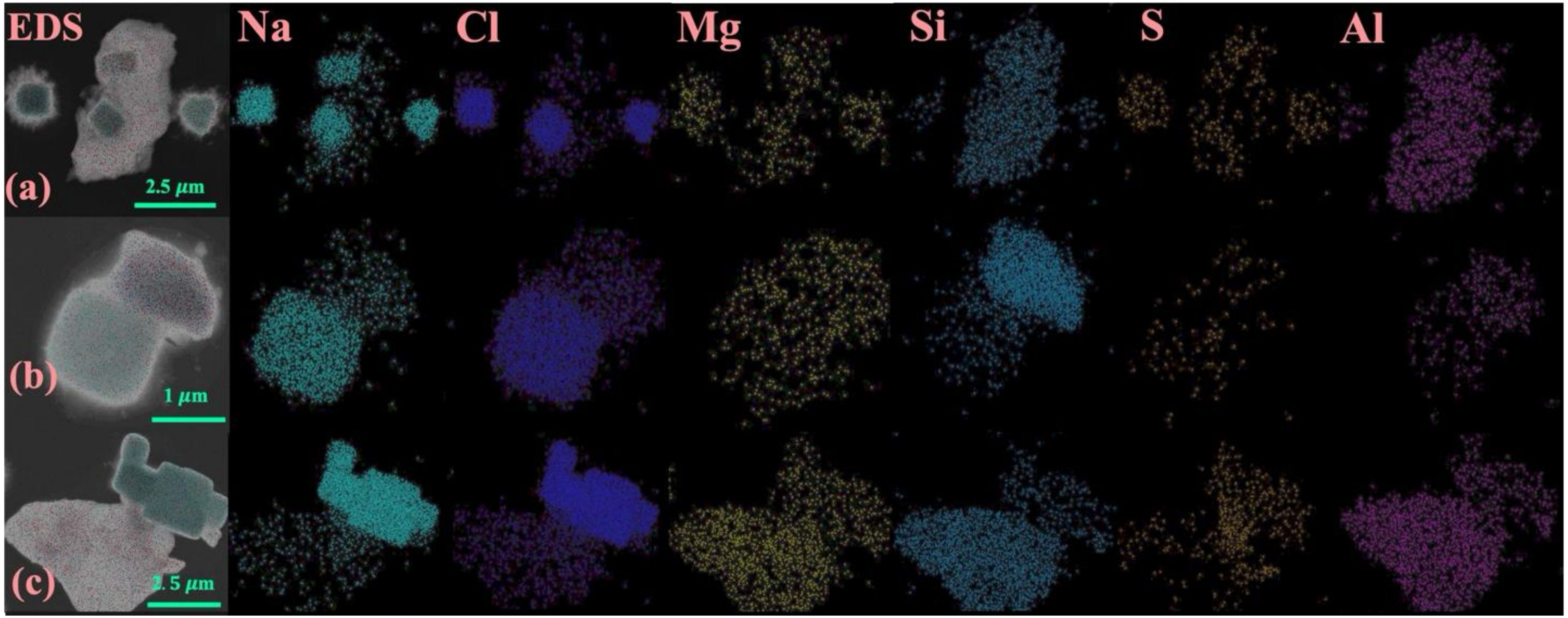
HYSPLIT back trajectory



- Air mass in case M4 traveled close to the ground, while in M8 it was lifted to above 1 km.
- M4 was mainly influenced by Artic Ocean, while M8 has the influence from the land.

SSA and dust mixtures

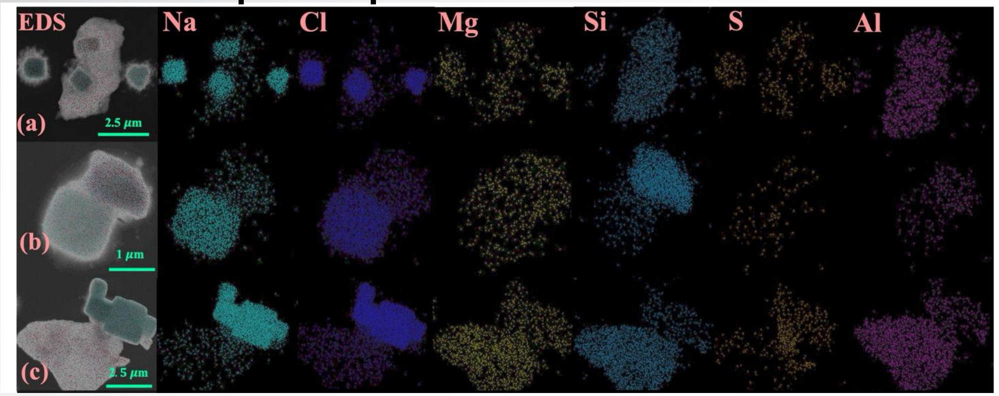
Elemental maps of representative SSA and dust mixtures



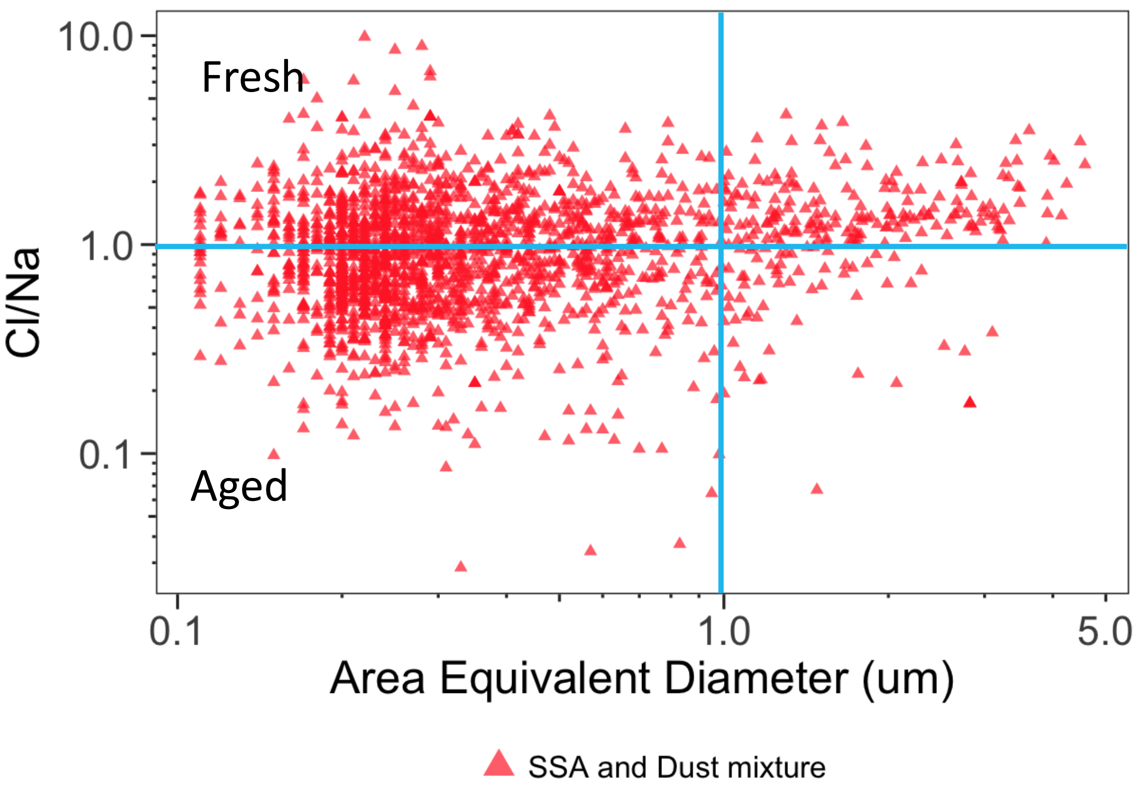
- **2063** out of 33448 (6%) analyzed particles are identified as "SSA and Dust mixtures".
- Mixture particles are not uniform in size and mixing states.

SSA and dust mixtures

Elemental maps of representative SSA and dust mixtures



Elemental ratio and size of individual particles



- Mostly occur in the submicron size.
- Independent of the aging state of SSA.

- Two **formation mechanisms**:

  (1) Formed during transport when SSA emissions intersect with dust emissions.

  (2) Results from the uplift of snow containing mineral dust deposited on the snow surface.

Conclusions

- **Fresh SSA** is the predominant species in Arctic aerosol (66%).
- **Organic coatings** are commonly found in both fresh and aged SSA particles.
- **HYSPLIT and spectroscopy** analysis reveals the potential aging mechanisms for each class: (1) chlorine depletion by organic acid; (2) chlorine depletion by sulfuric acid.
- **Dust is found to be commonly mixed with SSA** which might alter the ice nucleation ability of SSA.

Future Studies

- More single particle analysis is needed to reveal the mixing pattern of those mixture particles.
- The presence of SSA and dust mixtures calls for the need of future studies to quantify their ice nucleation abilities.

Acknowledgement

Environmental and Molecular Sciences Laboratory, Pacific Northwest National Lab

Advanced Light Source, Lawrence-Berkeley Lab

Institute of Polar Sciences of the National Research Council of Italy

Terrestrial-Atmosphere Process (TAP)
Earth & Biological Sciences

Left to right: Zhenli Lai (Joy), Nurun Nahar Lata, Mickey Rogers, Tania Gautam, Xena Mansoura, Valentina Sola, Zezhen Cheng (Jay), Swarup China, Gregory Vandergrift, Ashfiqur Rahman

Internship opportunities at EMSL, PNNL

Office of Science Graduate Student Research Program (SCGSR)

- U.S. Graduate students conduct part of PhD thesis research at a DOE National Laboratory
- Graduate researchers led projects in collaboration with a DOE National Laboratory scientist

Science Undergraduate Laboratory Internships (SULI) Must be currently enrolled full-time *or* 2 years after completion of an undergraduate degree prior to starting their internship-Graduate student can also Apply!

- Must be 18 years or older at the time of internship
- Must be a United States Citizen or Lawful Permanent Resident (LPR) at time of applying
- Two application cycles! Application is open now!

Contact information:

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Thank you