TNA User Report

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Project title	**Ice nucleation activity of marine aerosol particles**
Name of the accessed chamber	AIDA
Number of users in the project	4
Project objectives (max 100 words)	The aim of our project was to shed light on whether aerosols rich in organic matter, which may be produced by bubble bursting in Arctic open leads, are efficient ice nuclei at temperatures relevant for mixed-phase clouds with the ultimate aim of improving our ability to simulate such clouds and the surface energy balance of the high Arctic. Specifically we addressed the following questions: • What are the mechanisms that link marine biology, seawater chemistry, and aerosol physics? • How efficient might the particles generated in the open leads found in the high Arctic be as cloud condensation and ice nuclei?
Description of work (max 100 words):	During this project, we have determined the ice nucleating efficiency of seawater/phytoplankton culture samples (*Skeletonema marinoi* and *Melosira arctica*) grown under different conditions, as well as sea surface microlayer (SML) samples collected in the Arctic, using three independent methods. Aerosols were directed into the AIDA chamber/INKA continuous flow diffusion chamber (CFDC) following generation using a state-of-the-art sea spray simulation chamber, a nebulizer, an atomizer and through a spray nozzle. The ice nucleating particle abundance was also determined on the same culture/SML samples using the microliter nucleation by immersed particle instrument (µl-NIPI).

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[1] Physics; Chemistry; Earth Sciences & Environment; Engineering & Technology; Mathematics; Information & Communication Technologies; Material Sciences; Energy; Social sciences; Humanities.
[2] UNI= University and Other Higher Education Organisation; RES= Public Research Organisation (including international research organisations and private research organisations controlled by public authority); SME= Small and Medium Enterprise; PRV= Other Industrial and/or Profit Private Organisation; OTH= Other type of organization.
[3] UND= Undergraduate; PGR= Post graduate; PDOC= Post-doctoral researcher; RES= Researcher ENG= Engineer; ACA= Academic; TEC= Technician.
[4] Physics; Chemistry; Earth Sciences & Environment; Engineering & Technology; Mathematics; Information & Communication Technologies; Material Sciences; Energy; Social sciences; Humanities.
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*Reproduce the table for each user who accessed the infrastructure

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Trans-National Access (TNA) Scientific Report

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Instructions

Please limit the report to max 5 pages, you can include tables and figures. Please make sure to address any comments made by the reviewers at the moment of the project evaluation (if applicable, in this case you were informed beforehand). Please do not alter the layout of the document and keep in Word version.
The report will be made available on the eurochamp.org website. Should any information be confidential or not be made public, please inform us accordingly (in this case it will only be accessible by the European Commission, the EUROCHAMP-2020 project partners, and the reviewers). Please include:

- Introduction and motivation
- Scientific objectives
- Reason for choosing the simulation chamber/ calibration facility
- Method and experimental set-up
- Data description
- Preliminary results and conclusions
- Outcome and future studies
- References

Name of the PI: Matthew Salter
Chamber name and location: AIDA, Karlsruhe, Germany
Campaign name and period: Ice nucleation activity of marine aerosol particles (30-01-2017—24-02-2017)
Text:

Introduction and motivation

Clouds play an important role in the Earth's climate system due to their strong impact on the energy balance[1]. Clouds and their radiative effects are one of the major influences on the radiative fluxes in the atmosphere, but at the same time they remain the largest uncertainty in climate models[2].

In recent years the Arctic has warmed rapidly, a phenomenon called Arctic amplification, resulting in rapid sea-ice loss[2]. Unfortunately, our understanding of these rapid changes to Arctic climate is limited due to insufficient understanding of the physical processes controlling climate and sparse observational data in the region. However, what is clear is that clouds play a key role in the enhanced warming observed in the high Arctic through their effects on the energy budget and the subsequent melting and freezing of sea ice[3].

The high Arctic tends to alternate between cloudy and clear conditions, with each type persisting for several days before changes in moisture and heat cause a rapid transition between these states[4]. That cloudy conditions can persist for several days in the high Arctic is surprising. The clouds most often encountered in the high Arctic consist of a mixture of ice crystals and super-cooled water droplets, so-called mixed-phase clouds. This is a mixture which should be inherently unstable, to the extent that the clouds quickly glaciate and dissipate. These mixed-phase clouds are a major factor in the difficulty climate models have in simulating clouds in the high Arctic and the surface energy balance that depends upon it[5].

Ice crystals are frequently observed in low-level Arctic clouds at temperatures close to freezing which suggests that a source of very efficient ice-nucleating aerosol particles may be present in the region. Recently, it has been hypothesised that the open leads in the Arctic may be an important source of cloud and ice nucleating particles[6] via bubble bursting. However, quite how efficient the organic material associated with the phytoplankton blooms typical of the region is, remains unclear.

Scientific objectives

This project was designed to shed light on whether aerosols rich in organic matter, which may be produced by bubble bursting in the open leads, are efficient ice nuclei at temperatures relevant for mixed-phase clouds with the ultimate aim of improving our ability to simulate such clouds and the surface energy balance of the high Arctic.

As such, during this project we have addressed the following questions:

• What are the mechanisms that link marine biology, seawater chemistry, and aerosol physics?

• How efficient might the particles generated in the open leads found in the high Arctic be as cloud condensation and ice nuclei?

Reason for choosing the AIDA chamber for this project

Our project goal could only be achieved by combining our expertise in generating proxy sea spray aerosol with the unique opportunities to study aerosol-cloud interactions at the AIDA facility.

Method and experimental setup

During this project, we have determined the ice nucleating efficiency of seawater/phytoplankton culture samples (*Skeletonema marinoi* and *Melosira arctica*) grown under different conditions, as well as sea surface microlayer (SML) samples collected in the Arctic, using three independent methods. Firstly, aerosols generated using a state-of-the-art sea spray simulation chamber from seawater/phytoplankton cultures and sea surface microlayer samples were directed into the AIDA chamber. Here, the aerosol size distribution (SMPS/APS), composition (mass spectrometry), ice nucleating activity, and cloud condensation activity was determined. Most experiments were conducted in a temperature regime relevant for mixed-phase clouds. Secondly, the temperature-dependent abundance of ice nucleating particles was determined for the same aerosols using a continuous flow diffusion chamber (CFDC) called INKA (Ice Nucleation Instrument of the Karlsruhe Institute of Technology; Schiebel et al., in preparation), a new instrument recently installed at the AIDA facility. Thirdly, ice nucleating particle abundance was determined on the same culture/SML samples using the microliter nucleation by immersed particle instrument (μl-NIPI)[7].As an additional measurement, the CCN activity of the aerosol particles was determined using a dual column CCN counter (type CCN-200, DMT).

Data description

The AIDA chamber was operated for 20 days, typically with two expansions per day. As such, excluding reference expansion runs, we conducted 40 expansion experiments. A number of the experiments conducted were long-time experiments where the AIDA chamber was filled overnight using the sea spray simulation chamber.

The data available from the campaign consist of aerosol characteristics (number and size distribution) and estimated ice-active fractions from various instruments as a function of temperature and humidity.

Preliminary results and conclusions

Ice nucleation measurements using AIDA

The immersion freezing efficiency of the investigated samples (phytoplankton cultures and sea surface microlayer solutions) was probed in controlled expansion cooling experiments in the AIDA cloud chamber at temperatures between 258 and 237 K. The general procedure of such an experiment is illustrated in Fig. 1, showing an expansion cooling run with a *Skeletonema marinoi* phytoplankton culture that was sprayed into the AIDA chamber at 251 K with an ultrasonic nebulizer, yielding a median droplet diameter of about 0.9 μm (as measured with a scanning mobility particle sizer and an aerodynamic particle spectrometer). The controlled reduction of the chamber pressure (black line, panel A) led to a decrease in the gas temperature (red line, panel A) and an increase in the saturation ratios with respect to ice (S_{ice}, blue line, panel B) and supercooled water (S_{liq}, magenta line, panel B). The latter two quantities were measured with high accuracy in situ in the AIDA chamber by a tunable diode laser spectrometer. The formation of cloud droplets and ice crystals during the expansion was measured with optical particle counters, whose data are illustrated as a scatter plot in panel C (green dots). After the relative humidity has exceeded water saturation in the course of expansion cooling, the seed aerosol particles were activated to a cloud of supercooled water droplets (first vertical line). Ice crystal formation by immersion freezing initiated during continued pumping when the gas temperature further dropped below a threshold temperature of about 246 K (second vertical line). The nucleated ice particles were detected at much larger optical diameters than the cloud droplets and could therefore be separately counted by introducing an optical threshold size.

Ice nucleation measurements using INKA

Most samples that were probed in the AIDA chamber were also tested regarding their ice nucleation ability at constant temperatures and increasing relative humidities using the INKA cylindrical continuous flow diffusion chamber. Most investigations have been done above the homogeneous freezing limit (≥ -32°C) to enable direct comparison with AIDA cloud expansion data, while some samples have also been screened for deposition and homogeneous freezing behaviour at lower temperatures.

Figure 2 shows a typical INKA scan at -30°C (sample *Skeletonema marinoi*). During the scan, the relative humidity is slowly increased from about 80% to 115% relative humidity (upper panel) while the light scattering intensity of particles passing the detector is measured simultaneously (middle panel). A size threshold is chosen to discriminate bigger ice particles from interstitial aerosol or droplets (horizontal line in middle panel), which enabled to derive the number of ice crystals per volume of sample air (lowest panel).

For the samples under investigation during this campaign, ice nucleating particle concentrations have been below the detection limit at nucleation temperatures above -20°C. Even at lower temperatures, the observed ice nucleation was not very pronounced, which agrees with preliminary findings from the AIDA cloud chamber experiments and other recent observations[8].

Ice nucleation measurements using the µl-NIPI

The µl-NIPI[9] was used throughout the campaign to determine the ice nucleating particle (INP) activity of a range of collected microlayer samples and phytoplankton cultures. The µl-NIPI is a cold stage instrument, used with a substrate to probe the ice nucleation of µL volume droplets. Several sea surface microlayer samples used in the campaign were collected in the high Arctic during the ACCACIA campaign and their INP concentration was previously reported[7]. Wilson *et al.*[7] concluded that some component of marine organic material was found to nucleate ice under conditions relevant to mixed phase clouds. Phytoplankton and its exudates are likely candidates for this observed activity, and so this campaign provided an excellent opportunity to determine the ice nucleating ability of cultured phytoplankton alongside field samples.

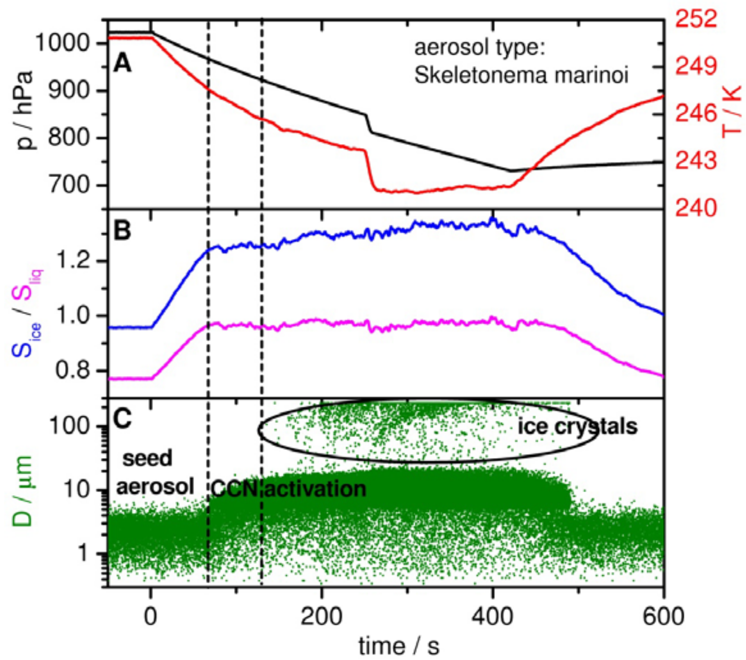

Figure 1. Ice nucleation characteristics of *Skeletonema marinoi* particles during an AIDA expansion cooling experiment started at 251 K. The contents of panels (A) to (C) are explained in the text.

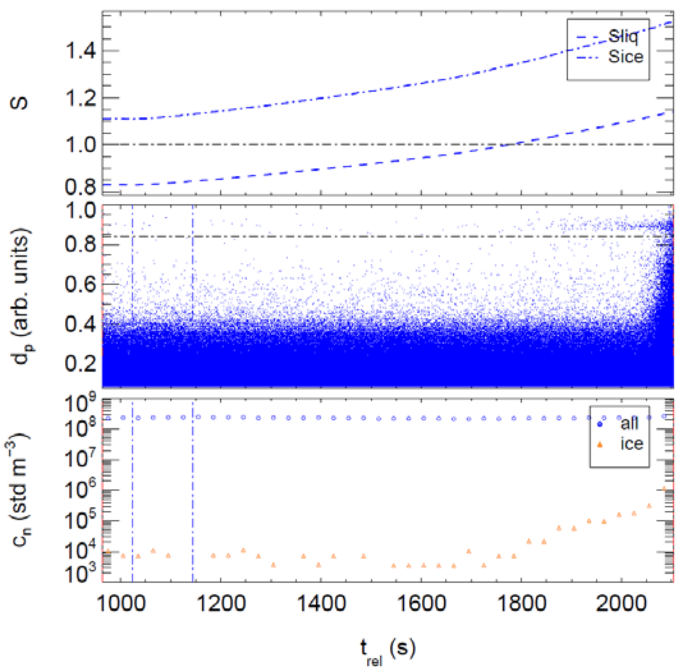

Figure 2. Typical INKA scan during the campaign (aerosol type: *Skeletonema marinoi* at -30°C). Upper panel: Water vapour saturation with respect to liquid and ice phase. Middle panel: Particle sizes as detected by the optical particle counter. Lower panel: Measured total particle and ice particle concentrations.

Figure 3 shows a fraction frozen curve, a measure of the fraction of droplets frozen at discrete temperatures, typical of those produced during the campaign. The curve at warm temperatures is an undiluted Arctic microlayer sample, SML5 described in Wilson et al[7]. The activity of this sample, compared to when it was collected in 2013, has not changed considerably after being stored at -80°C since summer 2013. This is important, because it is known that the freezing of microlayer samples affects their surface activity[10]. We then used µl-NIPI to quantify the INP concentration of water in the sea spray simulation chamber after microlayer and culture samples had been added to it. This allows us to relate the activity of the aerosolised samples in the AIDA expansion tests to the activity in the raw and sea spray simulation chamber samples. Bulk, surface and microlayer samples from the sea spray simulation chamber were collected, and the results of ice nucleation assays on the samples are shown in Fig. 3. It is clear that the ice nucleation activity has been reduced, consistent with the sample being diluted on introduction to the sea spray simulation chamber. But it can be seen that the bulk sample has lower activity than the microlayer sample, taken by dipping a glass slide vertically into the chamber and collecting the sample from the slide, and both have lower activity than the sample that was taken by scooping a falcon tube along the surface liquid. This indicates that organic INP material scavenged by bubbles resides at the surface and is surface active, and hence may become preferentially aerosolised during the bubble bursting process.

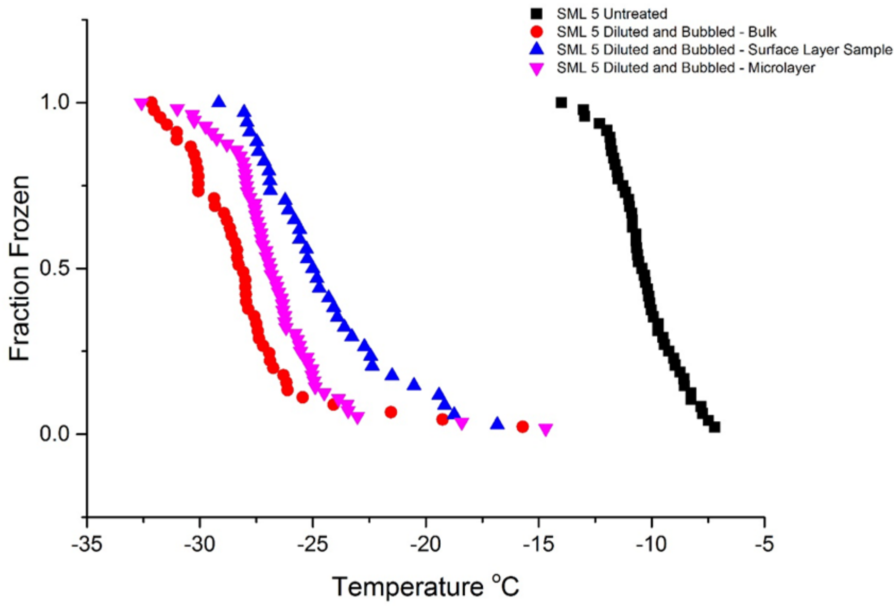

Figure 3. Fraction frozen curves for untreated and pure sea surface microlayer, SML5 from Wilson et al.[7] A fraction frozen curve shows the fraction of frozen droplets to unfrozen droplets. The coloured curves show the ice activity of SML5 after being diluted and run in the sea spray simulation chamber (samples were taken from the microlayer, surface layer and bulk of the chamber).

CCN measurements

Figure 4 shows a typical CCN measurement. Here, the sea spray simulation chamber was used to generate sea spray particles using a plunging jet. A known amount of the diatom *Melosira arctica* was added to 20 L of dissolved artificial sea salts (at a salinity of approx. 35 g/kg) to study the effect on the cloud forming potential. Plotting the activation ratio (# of CCN / # of total concentration) vs. supersaturation for a given diameter gives a critical supersaturation, Sc. From the Sc and the diameter one can calculate the hygroscopicity parameter, κ[11]. The processed data show that the diatoms and sea surface microlayer samples had very little effect on the hygroscopicity and CCN activity of pure inorganic salts.

Outcome and future studies

Data processing following the campaign is on-going. With regards the data from the AIDA chamber, the number concentration of nucleated ice crystals will be related to the seed aerosol number concentration to infer the ice-active fraction of the aerosol population. In a further analysis step, the ice nucleation efficiency will also be linked to the total amount of organic material in the aerosol particles. Such analyses will be done for all conducted experiments to systematically quantify the immersion freezing ability of the various samples and its dependency on the amount of organic material and the various techniques used to aerosolize the bulk solutions.

In the final step, the results from this project will be used to derive a parameterisation framework to describe the CCN and IN ability of local sources of aerosol particles in the high Arctic, which can be used in small scale and climate models to improve the representation of Arctic mixed-phase clouds.

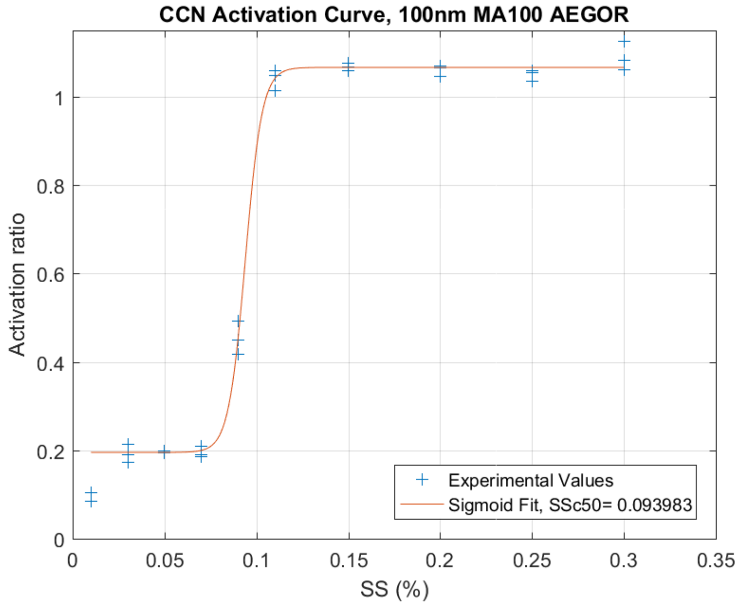

Figure 4. CCN activation curve for a 100 nm particle, where supersaturation (SS) was varied. Particles were generated from a sea spray simulation chamber (AEGOR) using a plunging jet. This sample contains *Melosira arctica* (MA100) and inorganic sea salts.

References

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