

Biogenic halocarbons in young Arctic sea ice and frost flowers



Anna Granfors ^a, Maria Andersson ^{a,1}, Melissa Chierici ^a, Agneta Fransson ^{b,2}, Katarina Gårdfeldt ^c, Anders Torstensson ^d, Angela Wulff ^d, Katarina Abrahamsson ^{a,*}

^a Department of Chemistry and Molecular Biology, University of Gothenburg, SE-412 96 Göteborg, Sweden

^b Department of Earth Sciences, University of Gothenburg, SE-405 30 Göteborg, Sweden

^c Centre for Environment and Sustainability, Chalmers University of Technology, University of Gothenburg, SE-412 96 Göteborg, Sweden

^d Department of Biological and Environmental Sciences, University of Gothenburg, P.O. Box 461, SE-405 30 Göteborg, Sweden

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ABSTRACT

The fate of halocarbons, naturally produced volatile halogenated organic compounds, in young Arctic sea ice was studied to better understand the role of sea ice in halocarbon cycling. In early spring, halocarbons were measured in sea ice frozen in core holes, during 12 days of formation and freezing. In order to understand which factors govern halocarbon concentration and distribution, salinity, temperature and biological parameters were monitored in the growing sea ice. It was found that sea ice participates in the cycling of halocarbons between sea and air. Sea ice concentrations and distributions of these compounds were influenced by production in the ice, where ice-inhabiting microorganisms caused local increases in halocarbon concentrations. Moreover, the halocarbon ice concentration decrease/change with time did not follow ice salinity, suggesting that additional removal processes caused sea ice to be a source of halogens to overlying air. The net production rate of bromoform in the surface of newly frozen ice was estimated to $14 \text{ pmol L}^{-1} \text{ d}^{-1}$ and the maximum removal rate was $18 \text{ pmol L}^{-1} \text{ d}^{-1}$. In addition frost flowers on newly formed sea ice were identified as contributors of halocarbons to the atmosphere with halocarbon concentrations in the same order of magnitude as in sea ice brine.

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1. Introduction

Halocarbons, naturally occurring volatile halogenated organic compounds, are a source of halogens to the atmosphere, and can be photolyzed to form reactive halogen species (Simpson et al., 2007). Reactive halogens are known to contribute significantly to the destruction of ozone in the polar stratosphere and the underlying troposphere. Halocarbons are biologically produced in marine environments by macro- and microalgae (Collén et al., 1994; Ekdahl et al., 1998; Carpenter and Liss, 2000). In ice-covered oceans, sea ice can act as a source of halocarbons when they are produced by microorganisms within sea ice brine (Sturges et al., 1992; Karlsson, 2012).

Although research has been undertaken into halocarbon production and distribution in marine environments, there are few studies of halocarbons in Arctic sea ice (Sturges et al., 1997). In addition, the processes that govern halocarbon concentration and distribution in sea ice during freezing are poorly understood. An understanding of these is necessary in order to estimate sources of halocarbons to

the atmosphere in this region. Sea ice has recently gained attention for its role in the interactions between ocean, sea ice, snow and atmosphere (Abbatt et al., 2012). Of special interest is the connection between ozone depletion and newly formed sea ice, where brine slush or frost flowers are possible sources of halogens (Jones et al., 2006; Simpson et al., 2007).

For some halocarbons, the activity of ice-living microorganisms, such as algae and heterotrophic bacteria, may affect their concentrations in sea ice. The interpretation of measurements of biogenic halocarbons in sea ice is complicated by variations originating from the influence of biological activity, and the fact that most samples from the field are spot samples from different locations. An attempt to overcome this problem was made in this study, which consists of a time series of samples from one experimental site in Ny-Ålesund, Svalbard, where the variability of halocarbons, and the biological parameters: microalgal and bacterial abundances, photosynthetic yield and bacterial carbon production were studied simultaneously during freezing of sea ice.

The aim of the study was to investigate the fate of halocarbons in Arctic sea ice, focusing on the potential flux to the atmosphere during freezing in a natural environment. Concentration and depth distribution of the measured parameters were studied in ice cores from old ice and from newly frozen ice (1 to 12 days old). Biological and

* Corresponding author.

E-mail address: k@chem.gu.se (K. Abrahamsson).

¹ Present address: ESSIQ AB, SE-421 50 Västra Frölunda, Sweden.

² Present address: Norwegian Polar Institute, Fram Centre, Tromsø, Norway.

physical processes affecting halocarbon concentrations and ice–air flux are discussed.

2. Materials & methods

2.1. Study site

The freezing experiment was carried out in Ny-Ålesund, Svalbard from 21 March to 2 April 2010. The experimental site was a small bay (Thiisbukta, Lat 78° 56' N, Long 11° 56' E), located 500 m from Ny-Ålesund village, and the ice was about two months old at the beginning of the experiment (personal communication with Bendik Halgunset, research advisor, Kings Bay AS). The total surface area of the bay was approximately 10,000 m², and approximately 1% was used as experimental site. At the beginning of the experiment the ice thickness was measured to be 27 cm. Thiisbukta was influenced by strong tides, which meant that the ice occasionally came in contact with the sediment. A few times during the experiment, sediment was found both in newly frozen and control ice cores. In the samples containing sediment, very high halocarbon concentrations were often detected and these samples were excluded from the displayed results.

2.2. Meteorological parameters

Air temperature and wind speed during the experiment were obtained from the Norwegian Meteorological Institute at their website; www.met.no (measuring site is 570 m from the study area), and photosynthetically active radiation (PAR) was obtained from our own radiation measurements (Fig. 1a–c). The average daily air temperatures

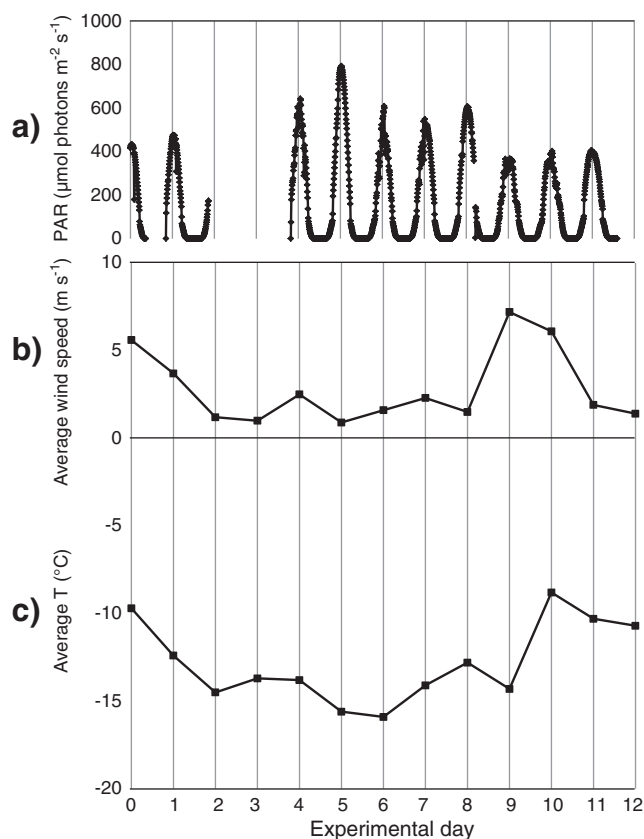


Fig. 1. Variability of a) photosynthetically active radiation (PAR), b) average wind speed and c) average air temperature (T) during the experiment.

were between $-10\text{ }^{\circ}\text{C}$ and $-15\text{ }^{\circ}\text{C}$, warmest on experimental day 10 and coldest on experimental day 6. Variation of average wind speed per day was between 1 and 7 m s⁻¹ with a maximum value on experimental day 9. The highest solar radiation was measured on experimental day 5. Radiation measurements in the ice showed that 8% of incident PAR remained at 35 cm depth (mid-day).

2.3. Sampling and experimental design

On experimental day 0, 50 holes were drilled through the ice using a gasoline powered ice-corer, with a diameter of 0.12 m. The holes were evenly distributed in an area of about 100 m². Care was taken so that no ice debris from the coring was left in the holes. The water in the holes was then left to re-freeze during the experiment, and re-frozen cores were sampled as single samples nearly every day (in total 10 occasions) during the 12-day experiment. Newly frozen ice-cores were randomly sampled from different boreholes for halocarbon measurements on experimental days 1, 2, 3, 5, 6, 8, 9, 10, 11 and 12. No hole was sampled more than once.

The old ice covering the bay was used as a reference (control) for halocarbon measurements, and was sampled on the same days as the newly frozen ice. This experiment treats the control ice as homogeneous with respect to location at the sampling site, since the cores drilled on day 0 all had the same thickness and structure (visually inspected), which implied that the area was covered with relatively homogenous ice. Moreover, a duplicate sample was analyzed from the control ice on day 0 and the variations between the inventory concentrations (mean bulk concentration) of the two cores were 17% (of average mean bulk concentration) for CH₂ClI, 2.7% for CH₂BrI and 7.4% for CHBr₃, which were found to be acceptable variations for our study.

The newly frozen ice cores, sampled from the various holes, were assumed to have the same concentration and depth distribution of halocarbons independent of their spatial distribution at the experimental site. This assumption was found to be reasonable for the experiment, since duplicate ice cores sampled at two occasions were similar in concentration and depth distribution. Variations (in percent of mean concentration) between duplicates at the same depth were on average 26% for CH₂ClI, 27% for CH₂BrI and 17% for CHBr₃. The duplicate samples of newly frozen ice cores were taken from the same sampling site, and were independent samples which were not included in the time series.

Ice cores for halocarbon analysis were divided into 5-cm sections and individually packed in gas-tight Tedlar® bags. After emptying the bags of surrounding air, the ice samples were thawed in darkness at room temperature for approximately 24 h. Sea ice temperature was measured immediately after the ice core was recovered at 5 cm intervals using a digital thermistor (Amadigit) with an accuracy of 0.1 °C. Separate ice cores were sampled for microbiological abundance and activity within 20 cm radius from the cores sampled for halocarbon analysis. These were immediately wrapped in black plastic to protect the algae from light stress.

Sea ice brine was collected at the study site in shallow (10 to 15 cm deep) “sack holes”. This means that the ice was partially cored to leave a core-hole in its surface. This was done to 10–15 cm depth in the ice to ensure that the seawater from below did not enter the hole. Brine that drained from the surrounding ice into the hole was sampled with a syringe. This method allowed sampling of large brine volumes but had the disadvantage that the ice volume feeding the collected sample was unknown. The sack holes were covered with a plastic lid, during brine seeping, to minimize gas exchange with the atmosphere. During our study the seeping time for brine was about 1 h to allow for sufficient sample volume, hence, some gas exchange may have taken place. Under-ice water (UIW) was collected with a glass bottle on a shaft (custom built) through the bore hole.

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