



## Short communication

## Clues that decaying leaves enrich Arctic air with ice nucleating particles

Franz Conen<sup>\*</sup>, Emiliano Stopelli, Lukas Zimmermann

Department of Environmental Sciences, University of Basel, Switzerland

## HIGHLIGHTS

- Ice nucleating particles (IN) were quantified at a coastal station in the Arctic.
- Longer backtrajectories over land coincide with greater IN number concentrations.
- Decaying leaf litter emitting biological particles is the most likely IN source.

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## ABSTRACT

Decaying leaves from Arctic regions have previously been reported to produce large numbers of ice nucleating particles (IN). Their atmospheric relevance is unclear. Our initial observations at a coastal mountain observatory in northern Norway reveal a tripling in concentrations of IN active at  $-15\text{ }^{\circ}\text{C}$  ( $\text{IN}_{-15}$ ) in oceanic air after about one day of passage over land (from 1.7 and 4.9  $\text{IN}_{-15}\text{ m}^{-3}$ , to 9.6 and 12.2  $\text{IN}_{-15}\text{ m}^{-3}$ ). Analysis of leaf litter collected near the observatory supports the earlier report of numerous IN associated with leaf litter on the ground ( $2 \cdot 10^2\text{ IN}_{-15}\text{ }\mu\text{g}^{-1}$  litter particles  $< 5\text{ }\mu\text{m}$ ). Together, both findings suggest that decaying leaves are a strong emission source of IN to the Arctic boundary layer.

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## 1. Introduction

Ice nucleating particles (IN) enable the formation of ice in clouds warmer than  $-36\text{ }^{\circ}\text{C}$ . Concentrations of IN vary by orders of magnitude in time and space. In the Arctic, IN are rarer than in most other places. Reported concentrations of IN active at  $-15\text{ }^{\circ}\text{C}$  ( $\text{IN}_{-15}$ ) are around  $1\text{ m}^{-3}$  winter and  $10\text{ m}^{-3}$  in summer (Bigg, 1996; Bigg and Leck, 2001). The Arctic summer values are two orders of magnitude smaller than what is typically found over a much wider geographic range (DeMott et al., 2010). As a consequence, removal of water from the Arctic atmosphere through ice growth and precipitation is IN-limited (Prenni et al., 2007). This limitation contributes to the persistence of optically thin low-level clouds that cause surface warming in summer (Tjernström et al., 2014).

Marine biological material aerosolized by breaking waves and bubble bursting is commonly considered a source of IN in the Arctic boundary layer (Schnell and Vali, 1975; Bigg, 1996; Bigg and Leck,

2001; Wilson et al., 2015). Biological particles emitted from Arctic land surfaces are rarely considered, although Schnell and Vali (1973) had found that a large number of IN can be obtained from well decayed leaves of Arctic origin when they are suspended in water. The same authors demonstrated in a later experiment, with an enclosure placed above plant litters, that such IN become airborne (Schnell and Vali, 1976). Leaf-derived IN are produced by bacteria and saprophytic fungi, are of subcellular size ( $<0.2\text{ }\mu\text{m}$ ), and lose their activity when heated above  $60\text{ }^{\circ}\text{C}$  (Schnell and Vali, 1972, 1973; Vali et al., 1976; Fröhlich et al., 2015; Pummer et al., 2015). Frequent freeze-thaw cycles positively select for microorganisms that produce ice nucleating proteins (Wilson et al., 2012). Hence, their relative abundance in the Arctic is larger than in milder climate zones (Schnell and Vali, 1973). Contributions of marine and land surface sources to the concentration of IN in air depend on the duration of previous contact with each kind of surface and on the extent of aerosolization of IN within the boundary layer. Here we report on initial observations of atmospheric IN at a coastal site in northern Norway during days with little and greater land contact of sampled air masses, in order to evaluate whether air is noticeably enriched in IN through contact with vegetated land.

<sup>\*</sup> Corresponding author.

E-mail address: [franz.conen@unibas.ch](mailto:franz.conen@unibas.ch) (F. Conen).

**Table 1**

Numbers of ice nucleating particles (IN  $\text{m}^{-3}$ ) observed in the atmosphere above Haldde observatory. Blank values are subtracted and numbers are adjusted to sea-level pressure. Blank values were on average 12% of gross values. Ice nucleating particles found in the litter sample are indicated per unit mass of litter particles  $< 5 \mu\text{m}$ .

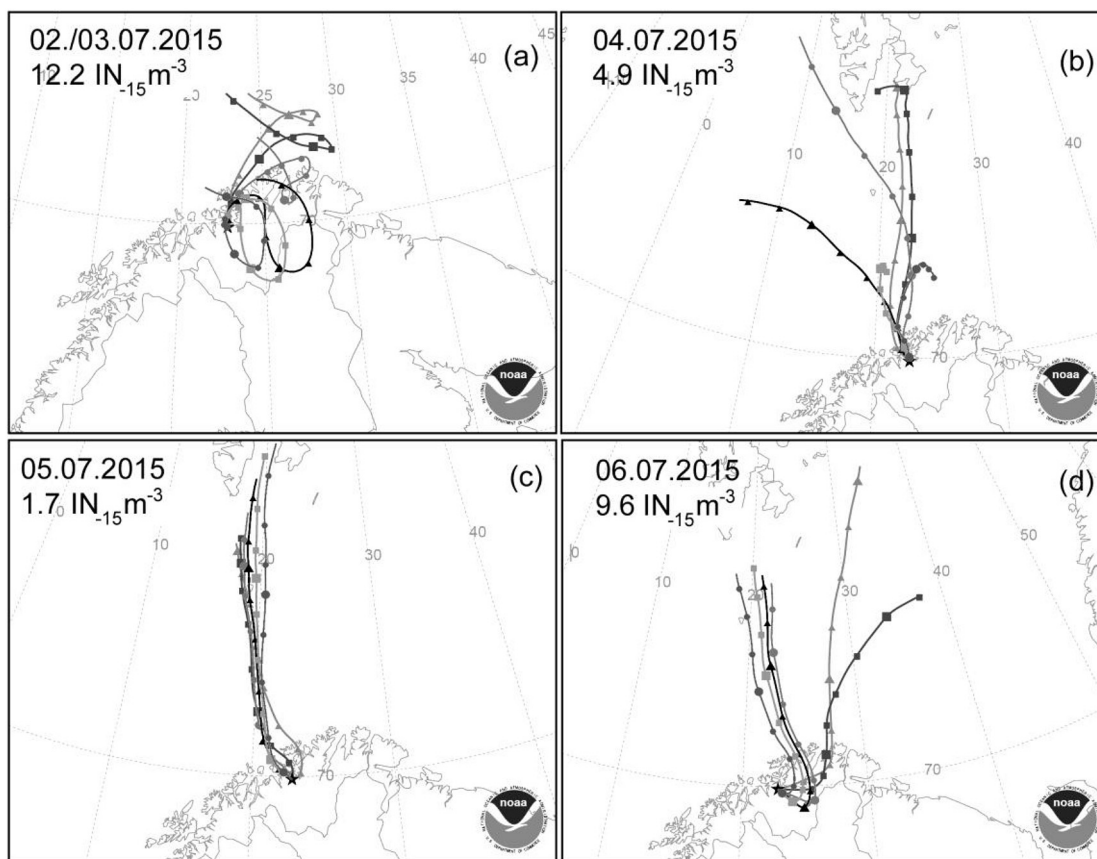
Temperature ( $^{\circ}\text{C}$ )	-7	-8	-9	-10	-11	-12	-13	-14	-15
Date	IN ( $\text{m}^{-3}$ )								
02./03.07.15	0.0	0.3	0.6	1.3	1.6	2.6	4.1	7.2	12.2
04.07.15	0.0	0.2	0.2	0.2	0.4	0.8	1.3	2.0	4.9
05.07.15	0.0	0.0	0.0	-0.1	0.1	0.0	0.2	0.8	1.7
06.07.15	0.0	0.2	0.2	0.9	1.2	1.8	2.2	5.5	9.6
	IN ( $\mu\text{g}^{-1}$ particles $< 5 \mu\text{m}$ )								
Litter	14	15	17	19	27	48	99	141	186

## 2. Material and methods

Atmospheric IN were collected between 02. and 06. July 2015 at Haldde observatory (Finnmark county,  $69^{\circ}55'45''$  N,  $22^{\circ}48'30''$  E, 905 m a.s.l.). The observatory was established by Kristian Birkeland in 1899 to study polar lights (Birkeland, 1908). From 1918 to 1926 Hilding Köhler stayed there to study cloud droplet formation (Köhler, 1937). It is situated on the top of a natural pyramid of rocks, 150 m above other rocks at a horizontal distance of 500 m. A portable  $\text{PM}_{10}$  sampler (PQ100, BGI, Mesa Labs Inc., Butler, NJ, USA) was installed with its inlet 2 m above the observatory tower. We sampled four times for a period of 24 h airborne particles on quartz-fiber filters (Pallflex® Tissuquartz, effective diameter 38 mm, Pall Corporation, Port Washington, NY, USA). The sampled air volume

was  $24 \text{ m}^3$  per filter at ambient conditions. Filters were analyzed at our laboratory in Basel following the method described in Conen et al. (2012).

Further, we collected a litter sample ( $5 \times 2 \text{ g}$ , bulked) from the surface of a melting snowfield about 300 m below, 2 km East-northeast of the observatory. Litter had been transported from the surroundings by wind and accumulated in the more sheltered location where snow was still present in July. The litter consisted of entire leaves and large fragments thereof, mainly from *Betula nana* and various grasses. Although they showed signs of decay to various degrees, their main structure was clearly discernible. We do not think this litter was any different from litter on snow-free surfaces. But, it was definitively easier to collect it there, than from other surfaces. At the laboratory we air dried it over night ( $23^{\circ}\text{C}$ ), then gently moved it by hand over a sieve ( $63 \mu\text{m}$ ) to separate small, lose particles. Total nitrogen (N) content was determined with an elemental analyser (model CN628, LECO Corporation, St. Joseph, MI, USA). Particles  $< 63 \mu\text{m}$  had a N content of 1.1%, similar to that of larger recognizable leaves in the sample (1.4% N) and very different from soil particles  $< 63 \mu\text{m}$  collected in the region (0.1% N). We suspended 0.05 g of litter particles  $< 63 \mu\text{m}$  in 15 ml 0.1% NaCl solution. The suspension was passed through a  $5 \mu\text{m}$  mesh filter (sterile cellulose acetate syringe filters, Sterlitech Corporation, Kent, WA, USA), diluted and analyzed on a droplet freezing apparatus (Stopelli et al., 2014). Additionally, a sub-sample was heated for 10 min to  $80^{\circ}\text{C}$  and another subsample passed through a  $0.20 \mu\text{m}$  filter (same type and supplier as  $5 \mu\text{m}$  filter) and analyzed for IN concentrations. The mass concentration of particles  $< 5 \mu\text{m}$  in the suspension was determined gravimetrically.



**Fig. 1.** Back trajectories starting at Haldde observatory every 4 h during a 24-h period of sampling aerosol on quartz-fiber filters (HYSPLIT Trajectory Model, NOAA, <http://ready.arl.noaa.gov/HYSPLIT.php>; 48-h trajectories; markers every 6 h; starting height 200 m above ground level). Sampling date and number of ice nucleating particles active at  $-15^{\circ}\text{C}$  (IN<sub>15</sub>  $\text{m}^{-3}$ ) associated with each of the four 24-h samples are shown in the panels.

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