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Size-resolved measurements of ice nucleating particles at six locations in North America and one in Europe

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ACPD
15, 20521–20559, 2015

Abstract

Detailed information on the size of ice nucleating particles (INPs) may be useful in source identification, modeling their transport in the atmosphere to improve climate predictions, and determining how effectively or ineffectively instrumentation used for quantifying INPs in the atmosphere captures the full INP population. In this study we report immersion-mode INP number concentrations as a function of size at six ground sites in North America and one in Europe. The lowest INP number concentrations were observed at Arctic and alpine locations and the highest at suburban and agricultural locations, consistent with previous studies of INP concentrations in similar environments. We found that 91, 79, and 63 % of INPs had an aerodynamic diameter > 1 µm at ice activation temperatures of −15, −20, and −25 °C, respectively, when averaging over all sampling locations. In addition, 62, 55, and 42 % of INPs were in the coarse mode (> 2.5 µm) at ice activation temperatures of −15, −20, and −25 °C, respectively, when averaging over all sampling locations. These results are consistent with six out of the seven studies in the literature that have focused on the size distribution of INPs in the atmosphere. Taken together, these findings strongly suggest that supermicron and coarse mode aerosol particles are a significant component of the ice nuclei population in many different ground-level environments. Further size-resolved studies of INPs as a function of altitude are required.

1 Introduction

Ice nucleating particles (INPs) are a unique class of aerosol particles that catalyze ice formation under atmospheric conditions. A variety of particle types have been identified as INPs, including mineral dust, black carbon, volcanic ash, glassy aerosols, and primary biological particles such as bacteria, fungal spores, and pollen (see reviews by Szyrmer and Zawadzki, 1997; Möhler et al., 2007; Ariya et al., 2009; Després et al., 2012; Hoose and Möhler, 2012; Murray et al., 2012; Yakobi-Hancock et al., 2013). Al-

ACPD
15, 20521–20559, 2015

though only a small fraction of aerosol particles nucleate ice (e.g. Rogers et al., 1998), INPs are important since they can lead to changes in the properties and lifetimes of mixed-phase and ice clouds, ultimately affecting climate and precipitation (Baker, 1997; Lohmann and Feichter, 2005; Baker and Peter, 2008; DeMott et al., 2010; Creamean et al., 2013).

Vali (1985) describes four modes of heterogeneous ice nucleation: deposition nucleation, where ice forms on the INP directly from the gas phase; condensation freezing, where ice nucleates during the condensing of water onto the INP; immersion freezing, where crystallization is initiated by an INP within a supercooled liquid droplet; and contact freezing, where the freezing of a supercooled liquid droplet is due to impaction by an INP. In this study we focus on freezing via the immersion mode in dilute solution droplets, which is relevant to mixed-phase cloud conditions.

Due to the importance of INPs for climate and precipitation, there has been a renewed interest in measuring the concentrations of INPs in the atmosphere (DeMott et al., 2011). While much of this work has focused on measurements of the total number concentration of INPs, there has been less emphasis on determining their size distributions in the atmosphere. Information on airborne INP size distributions may be particularly helpful in identifying the predominant INP sources. For example, information on the size distribution of INPs may help rule out or support the role of fungal spores in atmospheric ice nucleation since they are often in the supermicron range (Graham et al., 2003; Elbert et al., 2007; Sesartic and Dallafior, 2011; Després et al., 2012; Huffman et al., 2012). A similar approach can be used with black carbon particles, since they are mainly in the submicron (Clarke et al., 2004; Schwarz et al., 2008, 2013).

Previous modeling studies have shown that the transport and distribution of INPs, and aerosol particles in general, are sensitive to the size of the particles assumed in the models (Burrows et al., 2009; Wilkinson et al., 2011). Information on the size distributions of INPs are thus needed for accurate modeling of their transport and impact on climate and precipitation, as well as for the aerial dispersal of fungi and bacteria

ACPD
15, 20521–20559, 2015

(Morris et al., 2004; Hoose et al., 2010a, b; Sesartic et al., 2013; Haga et al., 2014; Spracklen and Heald, 2014).

Information on the size distribution of INPs is also needed to determine if techniques used to measure atmospheric INP concentrations capture the entire INP population. For example, the continuous flow diffusion chamber (Rogers et al., 2001b) is often used for measuring INPs (e.g. DeMott et al., 1998; Rogers et al., 2001a; Richardson et al., 2007; Pratt et al., 2009; Prenni et al., 2009; Eidhammer et al., 2010; Chou et al., 2011; Friedman et al., 2011; Hoyle et al., 2011; Corbin et al., 2012; Garcia et al., 2012; Tobo et al., 2013; McCluskey et al., 2014), but the aerodynamic diameter of particles measured with it is limited, from $\leq 2.4\,\mu m$ in some studies (e.g. Garcia et al., 2012) to $\leq 0.75\,\mu m$ in others (e.g. DeMott et al., 2003). Such techniques miss supermicron or coarse mode particles, the latter defined here as particles larger than $2.5\,\mu m$. The exact proportion missed may depend on temperature as well due to the potential dependence of ice-active size on temperature. Such online instruments have typically focused on measurements below approximately $-20\,^{\circ}C$ as sample volume considerations limit effective sampling of lower INP number concentrations at warmer temperatures.

Previous field studies of INPs as a function of size have been carried out using samples from Western Canada (Vali, 1966), the Pacific Ocean (Rosinski et al., 1986), the Gulf of Mexico (Rosinski et al., 1988), Eastern Europe (Berezinski et al., 1988), Italy (Santachiara et al., 2010), and the Central United States (Huffman et al., 2013). These studies are further discussed in Sect. 3.2. In the current study, we add to the existing body of size-resolved INP measurements by reporting ground-level INP size distributions from six locations in North America and one in Europe, investigating immersion freezing at -15, -20, and $-25\,^{\circ}C$. We found that both supermicron and coarse mode aerosol particles were a significant component of the INP population at all surface-based sampling locations. These results indicate that, when averaged over all locations and temperatures, the total INP number concentrations measured in similar ecosystems may be underestimated by a factor of 4.4 and 2.1 if supermicron or coarse mode particles, respectively, are not analyzed.

ACPD
15, 20521–20559, 2015

2 Methods

2.1 Sampling sites

The seven locations used in this study are detailed in Table 1 and shown in Fig. 1. Measurements using the sampling instrumentation described in the next section were made at five locations in Canada: Alert, Nunavut; the Labrador Sea near Newfoundland and Labrador; and Whistler Mountain, the University of British Columbia (UBC) campus, and Amphitrite Point in British Columbia. Measurements in Canada were conducted as part of the larger NETwork on Climate and Aerosols: addressing key uncertainties in Remote Canadian Environments project (NETCARE; http://www.netcare-project.ca/). Measurements were also made at Saclay, France and Colby, Kansas, USA.

2.1.1 Alert, NU, Canada

Arctic sampling was conducted at the Dr. Neil Trivett Global Atmosphere Watch Observatory in Alert, Nunavut, Canada (labeled 1 in Fig. 1; Cobbett et al., 2007) between 29 March and 23 July 2014. This Arctic research station is part of a global network for measuring chemical and physical perturbations of the atmosphere. Aerosol particles were collected through a total suspended particulate (TSP) inlet (Mesa Labs Inc., Butler, NJ, USA) and 0.9 m mast located on the upper level of an outdoor platform free of surrounding obstructions, and were stored in the dark at −15 or 4 °C for a period of 10–112 days prior to analysis.

2.1.2 The Labrador Sea

The Canadian Coast Guard Service vessel CCGS *Amundsen* serves as both an icebreaker for shipping lanes and an Arctic research vessel. One set of aerosol particle samples was collected from the top of the bridge of this vessel on 11 July 2014 while in the Labrador Sea off the coast of Newfoundland and Labrador, Canada (labeled 2 in Fig. 1). While sampling was within the marine boundary layer in the presence of

ACPD
15, 20521–20559, 2015

sea spray aerosols, back trajectories (not included) calculated using the Hybrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT4) model of the National Oceanographic and Atmospheric Administration (Draxler and Rolph, 2014) indicate that the sampled air mass spent the majority of the previous 72 h period over land. Air was passed through a louvered TSP inlet and 1.5 m mast during sampling, and collected aerosol particles were stored in the dark at 4 °C for a period of 45–46 days prior to analysis.

2.1.3 Whistler Mountain

The Whistler Peak High Elevation Site is located at the summit of Whistler Mountain in Whistler, British Columbia, Canada (labeled 3 in Fig. 1) and operated by Environment Canada (Gallagher et al., 2011; Macdonald et al., 2011). Aerosol particle collection at this alpine site occurred between 30 March and 23 April 2014. The louvered TSP inlet was located approximately 10 m from a chairlift operating station. Although there are no continuous combustion sources at the site, sampled air may have been influenced by engine exhaust for short periods of time due to nearby snowmobile operation. Samples were stored in the dark at 4 °C for a period of 1–4 days prior to analysis.

2.1.4 UBC campus

Four sets of aerosol particle samples were collected from a weather station on the roof of the five-story Earth Sciences Building on the UBC campus in British Columbia, Canada (labeled 4 in Fig. 1). The UBC campus is located on a peninsula and is surrounded by forest on three sides and ocean on the fourth. The site has been classified as suburban since it is less than 10 km from downtown Vancouver. Samples were collected through a TSP inlet and 0.5 m mast between 12 and 16 May 2014. The aerosol particles were stored in the dark at 4 °C for a period of 21–23 days prior to analysis.

ACPD
15, 20521–20559, 2015

2.1.5 Amphitrite Point

The coastal site at Amphitrite Point on Vancouver Island, British Columbia, Canada (labeled 5 in Fig. 1) is operated by Environment Canada, the BC Ministry of Environment, and Metro Vancouver for the continuous monitoring of aerosols and trace gases influenced by marine trajectories (McKendry et al., 2014; Yakobi-Hancock et al., 2014; Mason et al., 2015b). The mobile laboratory used during sampling was located approximately 100 m from the high tide line of the Pacific Ocean along a rocky shoreline, separated from the ocean by a narrow row of trees and shrubs approximately 2–10 m in height. Sampling took place from 6 to 27 August 2013 using a louvered TSP inlet and 3 m mast. Aerosol particles were stored at room temperature and analyzed within 1 day of collection.

2.1.6 Colby, KS

Aerosol particles were collected at the soybean and sorghum fields of the Kansas State University Northwest Research Center in Colby, KS, USA (labeled 6 in Fig. 1). One sample was collected at each location during combine harvesting from a distance approximately 3–10 m downwind of the field. A third sample was also collected at the sorghum field the night following harvest. Sampling took place on 14 and 15 October 2014 and samples were stored in the dark at 4 °C for a period of 41–46 days prior to analysis.

2.1.7 Saclay, France

Aerosol particle samples were collected at the Commissariat à l'Energie Atomique (CEA) Atmospheric Supersite (AS), CEA l'Orme des Merisiers. The CEA-AS Observatory is a suburban area located 30 km southeast of Paris in Saclay, France (labeled 7 in Fig. 1). The CEA-AS Observatory is surrounded by different sources of bioaerosols such as forest and agricultural fields, and is often influenced by marine or urban air

ACPD
15, 20521–20559, 2015

masses (Baisnée et al., 2014). Measurements were made as part of the BIODETECT 2014 intensive campaign, an intercomparison of bioaerosol detection methods (Sarda-Estève et al., 2014). During this study period, the site was heavily influenced by urban outflow. A large set of ancillary measurements was done to constrain all the particulate matter sources. Aerosol particles were sampled through a TSP inlet and 10 m mast between 15 July and 4 August 2014, and were stored in the dark at 4 °C for a period of 55–217 days prior to analysis.

2.2 Size-resolved INP number concentrations

INP number concentrations as a function of size and temperature were determined using the micro-orifice uniform deposit impactor-droplet freezing technique (MOUDI-DFT; Huffman et al., 2013; Mason et al., 2015a). This technique combines aerosol particle collection by a cascade inertial impactor with sharp size-cutoff characteristics (the MOUDI; Marple et al., 1991) with an established droplet freezing apparatus (the DFT) for determining immersion-mode freezing properties (Koop et al., 1998; Iannone et al., 2011; Haga et al., 2013). A similar approach has also been used to study deposition nucleation by particles collected from the atmosphere (Wang et al., 2012; Knopf et al., 2014).

2.2.1 Aerosol particle sampling

Size-fractionated aerosol particle samples were collected onto hydrophobic glass cover slips (HR3-215; Hampton Research, Aliso Viejo, CA, USA) using a model 110R or 120R MOUDI (MSP Corp., Shoreview, MN, USA). Previous work has shown that these hydrophobic glass surfaces do not cause significant heterogeneous ice nucleation (e.g. Haga et al., 2013, 2014; Wheeler et al., 2015). Substrate holders were used on the impaction plates of the MOUDI to reproducibly position the hydrophobic glass cover slips in regions where aerosol deposit particle concentrations did not vary significantly (for details see Mason et al., 2015a). At most locations MOUDI stages 2–9 were used, cor-

ACPD
15, 20521–20559, 2015

responding to particle size bins of 10–5.6, 5.6–3.2, 3.2–1.8, 1.8–1.0, 1.0–0.56, 0.56–0.32, 0.32–0.18, and 0.18–0.10 µm (50 % cutoff aerodynamic diameter; Marple et al., 1991), respectively. The range in particle size collected at each location is given in Table 1.

2.2.2 Calculating INP number concentrations

Samples were analyzed by the DFT to determine the number concentration of particles active in the immersion-freezing mode. Details of the experimental procedure can be found in Mason et al. (2015a). Briefly, samples were transferred to a temperature- and humidity-controlled flow cell coupled to an optical microscope equipped with a 5× magnification objective (Axiolab; Zeiss, Oberkochen, Germany). Water droplets with a diameter of 97 ± 42 µm (mean and standard deviation) were condensed onto the sample and monitored using a CCD camera recording a digital video. The freezing temperature of each droplet was then determined during cooling at a rate of −10 °C min^{-1} using the video timestamp and a resistance temperature detector located within the flow cell. Here we regard ice nucleation as a singular process (i.e. strictly temperature-dependent) but note that the stochastic (i.e. time-dependent) component to immersion freezing (Vali, 2014) may alter the median freezing temperature of a droplet by 0.5–2 °C per decade change in cooling rate (Murray et al., 2011; Welti et al., 2012; Wright and Petters, 2013; Wright et al., 2013; Wheeler et al., 2015).

Atmospheric INP number concentrations, [INPs(T)], were calculated using the following equation which accounts for the possibility of a droplet containing multiple INPs (Vali, 1971):

$$[\mathrm{INPs}(T)] = -\ln\left(\frac{N_\mathrm{u}(T)}{N_o}\right)\frac{N_o}{\left(\frac{A_\mathrm{deposit}}{A_\mathrm{DFT}V}\right)}f_\mathrm{nu}f_\mathrm{ne} \quad (1)$$

where $N_\mathrm{u}(T)$ is the number of unfrozen droplets at temperature T, N_o is the total number of droplets, A_deposit is the total area of the sample deposit on the hydrophobic glass

ACPD
15, 20521–20559, 2015

cover slips, A_{DFT} is the area of the sample analyzed by the DFT, V is the volume of air sampled by the MOUDI, f_{nu} is a correction factor to account for changes in particle concentration across each MOUDI sample (because the DFT analyzes only a fraction of the entire sample), and f_{ne} is a correction factor based on the analysis of Koop et al. (1997) to account for the uncertainty associated with the number of nucleation events in each experiment (see Mason et al., 2015a for details). Reported INP number concentrations at each location are averaged over all samples and have been adjusted to standard temperature and pressure.

3 Results and discussion

3.1 INP number concentrations

The total number concentration of INPs active at −15, −20, and −25 °C are shown for each site in Fig. 2. Freezing events were rare at temperatures warmer than −15 °C and are therefore not reported. Some of the DFT experiments proceeded such that all droplets froze at temperatures slightly below −25 °C. As this scenario prohibits calculation of INP number concentrations, −25 °C is the lowest temperature reported. As expected, INP number concentrations were found to increase with decreasing freezing temperature with the average concentration at −25 °C (3.77 L^{-1}) being more than an order of magnitude larger than at −15 °C (0.25 L^{-1}).

INP number concentrations were relatively low at the Alert, NU and Whistler Mountain sites with values of 0.05 and 0.10 L^{-1} at −15 °C, 0.22 and 0.16 L^{-1} at −20 °C, and 0.99 and 1.06 L^{-1} at −25 °C, respectively. These findings are consistent with previous measurements at similar locations. For example, Arctic measurements of Bigg (1996) and Fountain and Ohtake (1985) found mean INP concentrations of 0.01 L^{-1} at −15 °C and 0.13 L^{-1} at −20 °C, respectively, using a diffusion chamber, and Prenni et al. (2007) measured an average INP number concentration of approximately 0.33 L^{-1} between −8 and −28 °C. At high elevation sites, Bowers et al. (2009) at Mt. Werner in Colorado

ACPD
15, 20521–20559, 2015

and Conen et al. (2012) at the research station Jungfraujoch in Switzerland measured mean INP number concentrations of approximately 0.02 L^{-1} at −10 and −12 °C, respectively, using freezing assays. High elevation sites can receive large quantities of dust, which can be good ice nuclei at lower temperatures (Chou et al., 2011), but this was unlikely during our measurement period based on the low INP number concentrations.

INP number concentrations at Amphitrite Point were 0.23, 0.94, and 2.15 L^{-1} at droplet freezing temperatures of −15, −20, and −25 °C, respectively. Despite the predominance of marine air masses being sampled, the major source of INPs at Amphitrite Point during the study period was likely biological particles from local vegetation (Mason et al., 2015b). Similar values were measured in the Labrador Sea, where INP number concentrations at −15, −20, and −25 °C were 0.38, 1.32, and 2.79 L^{-1}, respectively. These concentrations are consistent with previous measurements within the marine boundary layer in regions influenced by air flow off of the nearby eastern coasts of continents, for instance those of Schnell (1977) off the coast of Nova Scotia, roughly 1100–1500 km southwest of our sampling site in the Labrador Sea, and Rosinski et al. (1995) over the East China Sea. However, INP number concentrations found during marine studies can vary by several orders of magnitude with location as summarized by Burrows et al. (2013).

The highest concentrations of INPs at a freezing temperature of −25 °C were found at the Colby, KS sites, where the average number concentration was 8.89 L^{-1}. Aerosol sampling was conducted adjacent to soya and sorghum fields during and following periods of combine operation. This high concentration of INPs is consistent with previous work of Garcia et al. (2012) that showed elevated concentrations of INPs downwind of corn fields during combine harvesting, and Bowers et al. (2011) who found greater INP concentrations in air above cropland than above suburban or forest sites.

The suburban sites of Saclay, France and the UBC campus also showed high INP concentrations, being 4.39 and 6.12 L^{-1}, respectively, at a freezing temperature of −25 °C. Both sites were likely influenced by multiple sources of INPs. For example,

ACPD
15, 20521–20559, 2015

both are in close proximity to major metropolitan centers and forest vegetation, which are potential sources of anthropogenic INPs (e.g. Hobbs and Locatelli, 1970; Al-Naimi and Saunders, 1985; Knopf et al., 2010, 2014; Ebert et al., 2011; Corbin et al., 2012; Cziczo et al., 2013; Brooks et al., 2014) and biological INPs (e.g. Vali et al., 1976; Kieft and Ruscetti, 1990; Richard et al., 1996; Hirano and Upper, 2000; Diehl et al., 2002; Prenni et al., 2009; Iannone et al., 2011; Pummer et al., 2012; Huffman et al., 2013; Tobo et al., 2013; Haga et al., 2014; Wright et al., 2014), respectively. The sampling site at Saclay, France was also within 1 km of agricultural fields, an additional source of biological aerosols that may act as INPs (e.g. Lindow et al., 1982; Hirano et al., 1985; Georgakopoulos and Sands, 1992; Möhler et al., 2008; Prenni et al., 2009; Bowers et al., 2011; Garcia et al., 2012; Haga et al., 2013; Morris et al., 2013; Hiranuma et al., 2015).

3.2 INP size distributions

Figure 3a shows the relative contribution of supermicron aerosol particles to the total measured INP population. Note that the same particle size range was not investigated at all locations with particles in the range of 0.10–0.18 µm not being measured at Whistler Mountain or Amphitrite Point. Averaging over all sampling locations, 91, 79, and 63 % of INPs had an aerodynamic diameter > 1 µm at ice activation temperatures of −15, −20, and −25 °C, respectively. At −15 °C, the percentage of supermicron INPs ranged from 78 % at Whistler Mountain up to 100 % at the Labrador Sea and Colby, KS sites. At lower temperatures, there was more variation between samples: at −20 °C the percentage of supermicron INPs ranged from 52 % at Whistler Mountain to 100 % over the Labrador Sea, and at −25 °C the percentage of supermicron INPs ranged from 39 % at Whistler Mountain to 95 % over the Labrador Sea.

Figure 3b shows the fraction of INPs that are in the coarse mode, calculated by assuming that half of the INPs found in the 1.8–3.2 µm MOUDI size cut were larger than 2.5 µm (i.e. INPs are uniformly distributed over this size range). Averaging over all sampling locations, we estimate that approximately 62, 55, and 42 % of INPs were in

ACPD
15, 20521–20559, 2015

the coarse mode at ice activation temperatures of −15, −20, and −25 °C, respectively. The percentage of INPs in the coarse mode was found to range from 38 % at Saclay, France to 91 % in Colby, KS at −15 °C, from 26 % at Whistler Mountain to 73 % in Colby, KS at −20 °C, and from 20 % at Alert, NU to 64 % at the Labrador Sea at −25 °C. Despite great diversity in the studied locations, each had a significant contribution from coarse mode particles to the measured INP population.

The median sizes of INPs at ice activation temperatures of −15, −20, and −25 °C are shown in Fig. 4 with the 25th and 75th percentile values. At −15 °C, the median INP size is relatively large at all locations, varying from 2.1 µm at Saclay, France to 4.7 µm at Colby, KS with an average of 3.2 µm. As droplet freezing temperature decreased, the median INP size also decreased, with the exception of samples from the Labrador Sea and the UBC campus. At −25 °C, the median size of INPs varied from 0.83 µm at Alert, NU to 3.1 µm at the Labrador Sea site with an average of 1.9 µm.

The median size of the INPs was > 1 µm in all cases with the exception of the Alert, NU and Whistler Mountain sites at a freezing temperature of −25 °C. Alert, NU, in the polar tundra at high latitude, and Whistler Mountain, at high elevation and periodically in the free troposphere, are unique sites in that they are remote with fewer local sources of aerosols. At times, long-range transport may strongly influence atmospheric composition at these two locations (Barrie, 1986; Worthy et al., 1994; Liang et al., 2004; McKendry et al., 2008; Shindell et al., 2008; Macdonald et al., 2011; Takahama et al., 2011; Fan, 2013). If INP sources are not local to these sites, removal of large particles via wet and dry deposition processes during long-range transport may account for the greater contribution from smaller INPs. Similar shifts in size distributions during long-range transport have been observed at other locations for total particles (Mori et al., 2003) and fungal spores (Fröhlich-Nowoisky et al., 2012). Transport losses may have also contributed to the small overall INP number concentrations at the Alert, NU and Whistler Mountain sites as shown in Fig. 2.

In Fig. 4, the difference between the 75th and 25th percentile sizes is relatively small at all locations at −15 °C. A narrow INP size distribution at −15 °C is consistent with

ACPD
15, 20521–20559, 2015

a single type or class of particles dominating freezing at this temperature. With decreasing temperature, the interquartile range significantly increased: the 75th and 25th percentile INP sizes decreased by an average factor of 1.2 and 3.7, respectively, between −15 and −25 °C, corresponding to a 63 % increase in the average interquartile range.

INP size distributions are further explored in Fig. 5, where the fraction of the measured INP number concentration found in each MOUDI size bin is shown. Colors on the red end of the scale illustrate that a large fraction of the INPs measured at a particular location belong to that particle size bin. Histograms of the INP size distributions are also available in Figs. S1–S7 in the Supplement.

Figure 5a shows that most INPs active at −15 °C were 1–10 µm in size. In particular, when averaged over all locations, 72 % of the INPs active at −15 °C were between 1.8 and 5.6 µm. Furthermore, the major mode (i.e. the global maximum in a size distribution) was always larger than 1.8 µm. At lower freezing temperatures the INP size distributions broadened with increased contributions from smaller aerosol particles, evident by the more uniform intensity of Fig. 5b and c. By −25 °C, six of the seven locations had a submicron INP mode, and at Alert, NU and Whistler Mountain this was the major mode. A general broadening of the INP size distribution with decreasing temperature would be expected if there were an increase in the number of particle types exhibiting ice activity with decreasing activation temperature.

Several previous studies have conducted size-resolved INP measurements. In most of these studies, the fraction of INPs larger than a given particle size was not reported, and in some cases the temperatures studied were different than in the current study. To better compare our data with these previous studies, we have used the literature data to calculate the fraction of INPs larger than either 1, 1.2, or 2.5 µm at temperatures as close as possible to the freezing temperatures we used. Details of the calculations are presented in the Supplement, and the results of the calculations are summarized in Table 2.

ACPD
15, 20521–20559, 2015

Table 2 shows that in six out of the seven previous studies, a large fraction (16–100 %) of the INPs was found to be supermicron in size, consistent with the current study. Note that the work of Rosinski et al. (1986) is considered as two separate studies given the change in the investigated mode of ice nucleation. The only measurements that did not observe a large fraction of INPs in the supermicron size range were the condensation freezing measurements by Rosinski et al. (1986), where approximately 1 % of the INPs were supermicron. However, for the same environment their data suggests that 100 % of INPs active in the immersion freezing mode were supermicron (when calculated for the size range > 0.5 µm). Taken together, measurements by Rosinski et al. (1986) suggest that the fraction of supermicron INPs may have a dependence on the mode of ice nucleation.

Kumai (1961) and Rucklidge (1965) also made direct measurements of INP size by measuring aerosol particle residuals found in collected ice crystals using electron microscopy. When multiple particles were present in their samples, the INP was assumed to be the largest particle. Kumai (1961) determined that all INPs were between 0.2 and 8 µm in crystals formed at temperatures of approximately −4 to −21 °C, with the size of maximum frequency ranging from 1–3.5 µm depending on the ice-crystal form. Rucklidge (1965), measuring INPs activated between −12 and −25 °C, found the mode of the INP size distribution to be 0.4–0.7 µm and approximately 86 % of INPs were smaller than 1 µm. The results of Kumai (1961) and Rucklidge (1965) are not included in Table 2 as INPs could not be conclusively identified in all samples given that multiple particles were occasionally present.

In addition to the previous studies that measured INPs as a function of size, several studies have investigated correlations between the concentrations of INPs and aerosol particles above a certain size (DeMott et al., 2010; Chou et al., 2011; Field et al., 2012; Huffman et al., 2013; Prenni et al., 2013; Tobo et al., 2013; Ardon-Dryer and Levin, 2014; Jiang et al., 2014, 2015). For example, using data from a variety of field measurements DeMott et al. (2010) observed a correlation between INP concentrations and the concentration of aerosol particles > 0.5 µm, and Ardon-Dryer and Levin (2014)

ACPD
15, 20521–20559, 2015

found that INP concentrations in Israel were better correlated to the concentration of aerosol particles 2.5–10 µm in size than those < 2.5 µm. These results are also consistent with INPs being relatively large in size.

4 Summary and conclusions

INP number concentrations in the immersion mode as a function of size and droplet freezing temperature were determined at six locations across North America and one in Europe. INP number concentrations varied by as much as an order of magnitude between locations, and were generally found to be lowest at the remote sites of Alert, NU and Whistler Mountain and highest at the agricultural sites of Colby, KS and the suburban sites of Saclay, France and the UBC campus, consistent with previous studies. Several key findings indicate the potential importance of large particles as a source of ground-level ice nuclei: (1) 91 and 62 % of INPs measured at −15 °C across all locations are supermicron or in the coarse mode, respectively, (2) at the lowest temperature analyzed, −25 °C, 63 and 42 % of INPs across all locations remained in the supermicron regime or coarse mode, respectively, (3) at −15 °C, the median INP size was relatively large at all locations, varying from 2.1 µm at Saclay, France to 4.7 µm at Colby, KS with an average of 3.2 µm; and (4) at −25 °C, the median size INP varied from 0.83 µm at Alert, NU to 3.1 µm above the Labrador Sea with an average of 1.9 µm.

Our measurements indicate that, when averaged over all studied locations and temperatures, 78 and 53 % of immersion-mode INPs may be missed if either supermicron particles or coarse mode particles are not sampled. As noted in Sect. 1, some instrumentation for measuring ambient INP number concentrations restricts the upper range of sampled aerosol particles. The data presented may be useful for estimating the fraction of INPs not measured with these instruments at ground sites and in different environments.

All measurements used in this study were conducted at ground-level and, apart from those at Whistler Mountain, were also close to sea level. As the large contribution of

ACPD
15, 20521–20559, 2015

supermicron and coarse mode INPs to the overall INP population noted here may not necessarily hold for higher altitudes, additional size-resolved INP measurements as a function of altitude are needed. In obtaining such data, careful consideration will be needed toward sampling issues with aerosol inlets and transfer through sample lines on aircraft platforms. It can be anticipated that without special inlets or care, supermicron particle transfer will be strongly restricted, and this may especially impact INP sampling.

One caveat to this study is that our measurements were confined to aerosol particle sizes greater than either 0.10 or 0.18 µm. If there were a significant contribution from INPs of smaller sizes (Vali, 1966; Schnell and Vali, 1973; Pummer et al., 2012; Augustin et al., 2013; Fröhlich-Nowoisky et al., 2015; O'Sullivan et al., 2015; Tong et al., 2015; Wilson et al., 2015), then the values presented would represent upper limits to the contribution of supermicron and coarse mode particles to the total INP population. Additional studies exploring the relative atmospheric abundance of INPs < 0.10 µm are therefore necessary (Hader et al., 2014).

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ACPD
15, 20521–20559, 2015

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Table 1. The seven locations used in this study and conditions during sampling.

Location	Environment	Coordinates	Elevation (m)	Sampling period	Number of samples	Average sampling time (h)	Average temperature (°C)	Average RH (%)	Particle size range (μm)*
Alert, NU, Canada	Arctic	82.45° N 62.51° W	12 m a.g.l.	29 Mar–23 Jul 2014	9	17.6	−17.4	77	0.10–10
The Labrador Sea, Canada	Marine	54.50° N 55.37° W	15 m a.s.l.	11 Jul 2014	1	6.2	10.9	75	0.10–10
Whistler Mountain, BC, Canada	Alpine	50.06° N 122.96° W	2182 m a.s.l.	30 Mar–23 Apr 2014	4	6.7	0.8	83	0.18–10
UBC campus, BC, Canada	Suburban	49.26° N 123.25° W	24 m a.g.l.	12–16 May 2014	4	6.3	15.2	70	0.10–10
Amphitrite Point, BC, Canada	Coastal	48.92° N 125.54° W	25 m a.s.l.	6–27 Aug 2013	34	7.8	13.8	97	0.18–10
Colby, KS, USA	Agricultural	39.39° N 101.06° W and 39.39° N 101.08° W	2 m a.g.l.	14–15 Oct 2014	3	4.5	17.0	48	0.10–10
CEA, Saclay, France	Suburban	48.70° N 2.14° E	10 m a.g.l.	15 Jul–4 Aug 2014	15	7.2	20.6	69	0.10–10

* Aerodynamic diameter based on the 50 % cutoff of the MOUDI (Marple et al., 1991).

ACPD
15, 20521–20559, 2015

Table 2. Previous size-resolved INP measurements.

Study	Location	Geographical description	Altitude	Particle sizes investigated (µm)	Mode of ice nucleation	Ice nucleation temperature (°C)[a]	Result
Vali (1966)	Alberta, Canada	Western Canada	Hail melt water	TSP[b]	Immersion	−12.8	16 % of INPs > 1.2 µm[c]
Rosinski et al. (1986)	Central and western South Pacific Ocean	Marine	Near sea level	0.5 to > 8	Immersion	−10.8	100 % of INPs > 1 µm
				< 0.5 to > 8	Condensation	−5 to −6	1 % of INPs > 1 µm
Rosinski et al. (1988)	Gulf of Mexico	Marine	Near sea level	0.1 to > 4.5	Condensation	−15 to −16	45 % of INPs > 1 µm
Berezinski et al. (1988)	European territory of the former Soviet Union	Eastern Europe	100–500 m a.g.l.	0.1 to > 100	Condensation	−15 to −20	37 % of INPs > 1 µm
Santachiara et al. (2010)	S. Pietro Capofiume, Italy	Rural Italy	3 m a.g.l.	TSP	Condensation	−17 to −19	47 and 30 % of INPs > 1 and 2.5 µm, respectively
Huffman et al. (2013)	Manitou Experimental Forest, CO, USA	Forest during/ after rainfall	4 m a.g.l.	0.32 to > 18	Immersion and deposition	−15 to −20	89 % of INPs > 1 µm
		Forest during dry periods	4 m a.g.l.	0.32 to > 18	Immersion and deposition	−15 to −20	46 % of INPs > 1 µm

[a] We used data at the temperatures of this study (−15, −20, and −25 °C) when available, otherwise the next closest temperature was used.
[b] TSP = total suspended particulate.
[c] Particle size reported for Vali (1966) is based on filter pore size. In all other studies, particle size is given as the aerodynamic diameter.

ACPD
15, 20521–20559, 2015

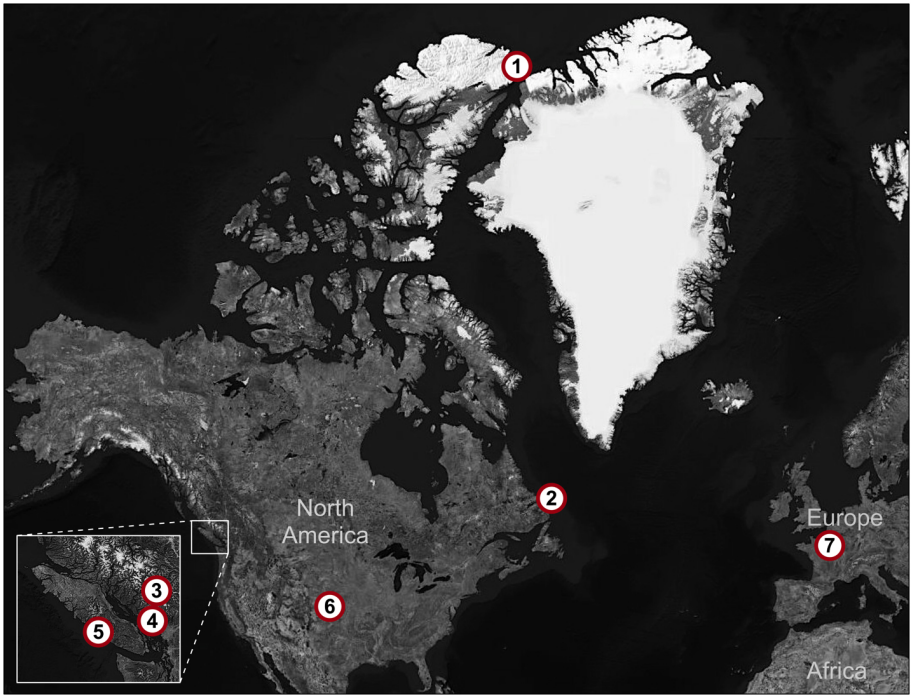

Figure 1. Sampling locations used in this study: (1) Alert, Nunavut, Canada; (2) the Labrador Sea, Canada; (3) Whistler Mountain, British Columbia, Canada; (4) the University of British Columbia campus, British Columbia, Canada; (5) Amphitrite Point, British Columbia, Canada; (6) Colby, Kansas, USA; and (7) Saclay, France. Site coordinates are given in Table 1 with details in Sect. 2.1. The image was modified from Bing Maps, 2014 (http://bing.com/maps).

ACPD
15, 20521–20559, 2015

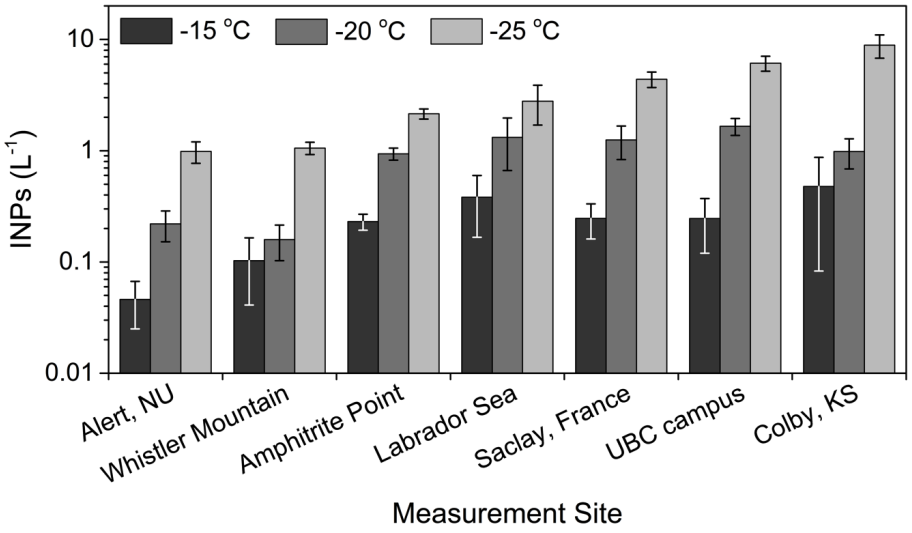

Figure 2. Mean INP number concentrations at droplet freezing temperatures of −15 °C (dark grey), −20 °C (intermediate grey), and −25 °C (light grey). Uncertainty is given as the standard error of the mean.

ACPD
15, 20521–20559, 2015

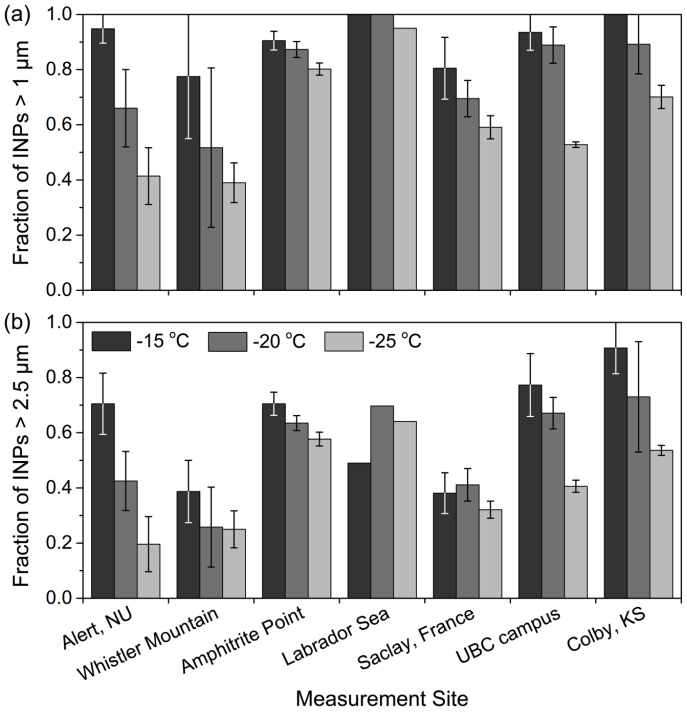

Figure 3. The mean fraction of INPs larger than **(a)** 1 µm and **(b)** 2.5 µm. Uncertainty is the standard error of the mean. Shading in the histogram corresponds to INP activation temperature: −15 °C is dark grey, −20 °C is an intermediate grey, and −25 °C is light grey. As 2.5 µm does not align with the size cut of a MOUDI stage, the fraction of INPs larger than 2.5 µm was found by assuming that number concentration of INPs 1.8–3.2 µm in size was uniformly distributed over that size range. As only one sample was available from the Labrador Sea, no uncertainty is reported.

ACPD
15, 20521–20559, 2015

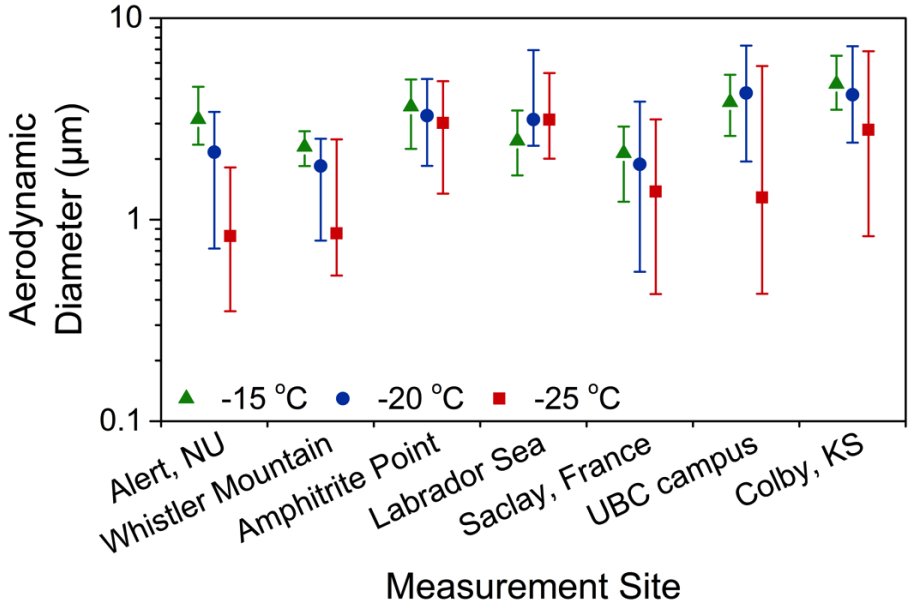

Figure 4. The median size of INPs at ice-activation temperatures of −15 °C (green), −20 °C (blue), and −25 °C (red) when averaged over all analyzed samples. Upper and lower uncertainties are the 75th and 25th percentiles, respectively.

ACPD
15, 20521–20559, 2015

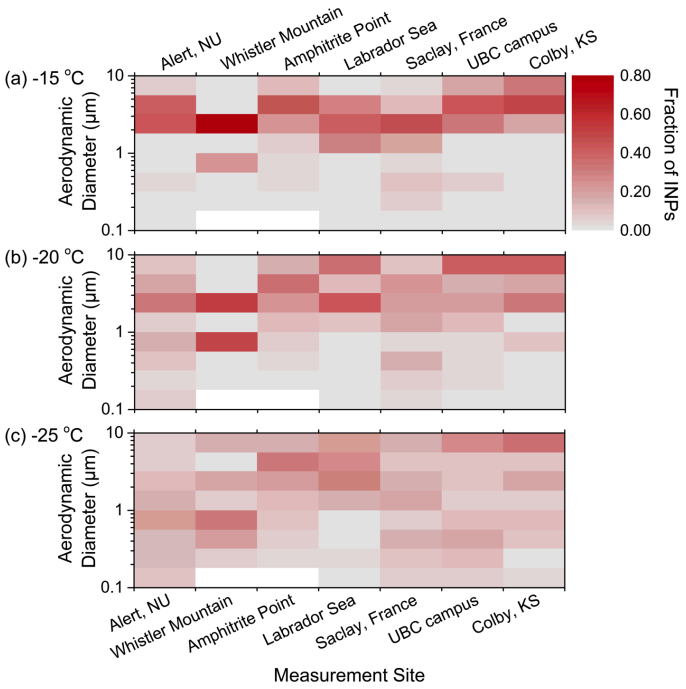

Figure 5. Fractional INP concentrations as a function of aerosol particle size, location, and activation temperature: **(a)** −15 °C; **(b)** −20 °C; and **(c)** −25 °C. The color bar indicates the fraction of INPs measured in each particle size bin. Aerosol particle sizes correspond to the 50 % cutoff aerodynamic diameters of the MOUDI stages (Marple et al., 1991). Missing sizes for the Whistler Mountain and Amphitrite Point sites are uncolored.

ACPD
15, 20521–20559, 2015