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Towards parametrising atmospheric concentrations of ice nucleating particles active at moderate supercooling

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Abstract. A small fraction of freezing cloud droplets probably initiates much of the precipitation above continents. Only a minute fraction of aerosol particles, so-called ice nucleating particles (INPs), can trigger initial ice formation at −15 °C, a cloud-top temperature frequently associated with snowfall. We found at a mountain top site in the Swiss Alps that concentrations of INPs active at −15 °C are different functions of coarse (> 2 μm) aerosol particle concentrations, depending on whether an air mass is precipitating, non-precipitating, or carrying Saharan dust and non-precipitating. Consequently, we suggest that a parameterisation at moderate supercooling should consider coarse particles in combination with air mass differentiation.

1 Introduction

Ice nucleating particles (INPs) affect clouds and their development by generating primary ice at temperatures between 0 and −38 °C. The difficulty of understanding and thus predicting atmospheric INP concentrations ([INP]) originates from the complex interplay of their rarity, their various sources, and nucleation temperatures (Paramonov et al., 2019). A current empirical parametrisation established by DeMott et al. (2015), hereafter D15, predicts [INP] based on nucleation temperature and number concentration of aerosol particles with diameters >0.5 μm ([$n_{0.5}$]) (DeMott et al., 2015). Although successful at colder temperatures, it remains "weakly constrained at temperatures >−20 °C, where much additional ambient and laboratory data are needed" (DeMott et al., 2015). Parametrising [INP] in the warmer temperature range is important because cloud-top temperatures associated with winter precipitation have a distinct temperature mode near −15 °C, as derived from close to 10^5 parallel observations of cloud-top temperatures and falling solid precipitation throughout the United States (Hanna et al., 2008). The same mode may also occur in other continental regions in the midlatitudes as it makes physically sense for various reasons (Wetzel and Martin, 2001). Recent findings from size-resolved measurements of atmospheric INPs show that those active at −15 °C or warmer ($INPs_{-15}$) are mostly >2.0 μm in diameter (Huffman et al., 2013; Mason et al., 2016; Creamean et al., 2015), a particle size that is poorly represented for instrumental reasons in the empirical data on which D15 and other parametrisations (Phillips et al., 2013) are based. A further finding is the increased atmospheric abundance of such INPs during precipitation

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(Bigg and Miles, 1964; Huffman et al., 2013; Hara et al., 2016; Conen et al., 2017). To test whether these findings can be reconciled with the general approach of D15 (i.e. parametrising INPs as a function of particles larger than a certain size), we collected and analysed aerosol samples from February to March 2019 at Weissfluhjoch, Switzerland, at average local air temperatures of −7.1 (s.d.±4.3) °C during sampling intervals. The site, surrounding mountains and nearby valleys were snow-covered, most of the lowland was not and it received precipitation as rain.

2 Material and Method

Between 11 February and 26 March 2019, we collected and analysed a total of 124 aerosol samples at Weissfluhjoch, Switzerland (46°49'58.670"N, 9°48'23.309"E, 2671.1 m a.s.l.) during the "Role of Aerosols and Clouds Enhanced by Topography on Snow (RACLETS)" campaign. Total aerosol was sampled through a heated inlet at a flow rate of 300 L min^{-1} for 20 min (i.e. 6 m^3 of air each) into 15 mL of ultra-pure water (Sigma-Aldrich, W4502-1L) using a high flow-rate impinger (Bertin Technologies, Coriolis®μ). Evaporative losses were compensated by replenishing the circulating water after 10 and 20 min. Samples were analysed immediately after collection in a drop freezing assay, with 52 droplets of 100 μL each, as previously described (Stopelli et al., 2014) and cumulative [INP] were calculated (Vali, 1971). The detection range of [INP] lies between $5x10^{-4}$ and 0.2 std L^{-1}. Samples outside this range (15 above, 1 below range) were not considered. Background measurements (n = 15) following identical procedure, but without turning on airflow, were below detection limit. Number concentrations of particles [n] were measured continuously with an Aerodynamic Particle Sizer (APS) spectrometer 3321 from TSI Corporation and were averaged over the time-period of the impinger samples. [$n_{0.5}$] and [$n_{2.0}$] are the sum of all counts of particles $\geq$ 0.542 μm and $\geq$ 1.982 μm, respectively. [n] and [INP] were adjusted to standard pressure conditions (P_{ref} = 1013.25 hPa). Five-day back trajectories were calculated using the Lagrangian analysis tool LAGRANTO (Sprenger and Wernli, 2015). For each sample, one trajectory was started at the full hour closest to the sampling time and from the exact sampling position. The driving wind fields are taken from the operational analysis of the Swiss National Weather Service (COSMO1; www.meteoswiss.ch) and the European Centre for Mid-Range Weather Forecasts (ECMWF; www.ecmwf.int). Started in the COSMO domain, the trajectories were extended based on ECMWF data at the time and location where they leave this domain. Their position was saved every 10 min. Along the trajectories, total precipitation was traced amongst others enabling us to determine the total precipitation amount along the last 6 hours prior to sampling.

3 Results and Discussion

From a total of 124 aerosol samples, about half (56) were collected from air masses that had precipitated at least 1.0 mm during the 6 hours prior to sampling (defined as "precipitating"). About half of these air masses were also precipitating when sampled at Weissfluhjoch, as observed by a precipitation gauge. A similar number of samples (57) were from air masses with less or without any prior precipitation ("non-precipitating") and 11 were from air masses carrying Saharan dust (SD) and no prior precipitation. Air masses were mainly reaching the sampling position from the West. Precipitating air masses were

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coming on a rather direct way from the Atlantic crossing the West of Europe with less detour than non-precipitating air masses whereas air masses carrying dust where coming from the direction of the Saharan desert passing the South of Europe (Fig. 1a-c). Precipitating air masses had the lowest [$n_{0.5}$] and the lowest concentration of aerosol particles with diameters >2.0 μm ([$n_{2.0}$]), but similar ratios as non-precipitating air masses of [$n_{2.0}$] to [$n_{0.5}$] (Fig. 1d-f). The largest ratio of [$n_{2.0}$] to [$n_{0.5}$] was in SD air masses (Fig. 1f). Atmospheric concentrations of $INPs_{-15}$ ([INP_{-15}]) in non-precipitating air masses were closest to the parametrisation of D15, based on [$n_{0.5}$]. Although the D15 parametrisation predicts the overall median [INP_{-15}] reasonably well (0.005 std L^{-1}, versus the measured 0.016 std L^{-1}), 72% of measured data was within a factor of 10, but only 29% within a factor of 2 of the prediction. Specifically, the D15 parametrisation tends to underestimate the median [INP_{-15}] in precipitating air masses by roughly one order of magnitude and overestimates those in SD air masses by about the same factor (Fig. 1g). Considering the fact that the observed size of $INPs_{-15}$ is mostly larger than 2 μm (Huffman et al., 2013; Mason et al., 2016; Creamean et al., 2015), we plot measured [$INPs_{-15}$] against [$n_{2.0}$], instead of [$n_{0.5}$], resulting in a more distinct separation of the data to the different air masses (Fig. 1h). The greater clarity of the separation confirms that [$n_{2.0}$] is a more powerful predictor of $INPs_{-15}$ when combined with air mass differentiation. The clearer separation goes hand in hand with the earlier observation that air masses influenced by SD carry less INPs active at moderate supercooling per unit mass of aerosol particles than European background air masses (Conen et al., 2015). It also reveals the known enrichment of the aerosol population with highly efficient INPs during precipitation (Bigg and Miles, 1964; Huffman et al., 2013). In each of the three categories of air mass [INP_{-15}] can be described as a function of [$n_{2.0}$] that is valid for a range of [INP_{-15}] from 0.0005 std L^{-1} to 0.2 std L^{-1} (Fig. 1h). The functions for data in SD and non-precipitating air masses appear to have similar slopes on a log-log plot. A possible explanation of additional INPs in precipitating air masses is the aerosolisation of INPs by rain itself, a mechanism that could be similar to the generation of bioaerosol by raindrop impingement (Joung et al., 2017). Since aerosolisation of efficient INPs during precipitation is likely independent of the background in [INP], we may describe [INP_{-15}] in precipitating air masses by adding a constant to the function fitted to the non-precipitating cases (Fig. 1h). The median difference between this function and measured [INP_{-15}] in precipitating air masses was 0.014 std L^{-1} (Fig. 1i). The relationship between these differences and [$n_{2.0}$] were weakly positive and not significant (Person correlation test, R = 0.0027 and p = 0.98), meaning that the absolute value of additional INPs during precipitation was independent of [$n_{2.0}$]. A consequence of this finding is that to air masses with low [$n_{2.0}$] and thus low [INP_{-15}], the addition of INPs aerosolised by precipitation makes a relatively large contribution to the overall [INP_{-15}]. The additional INPs during precipitation could be fungal spores emitted from the snow-free lowland of Switzerland and southern Germany, an explanation supported by observations of considerable presence of fungal spores in air during humid periods (Negron et al., 2020).

4 Conclusions

In summary, it is possible to reconcile two fundamental aspects of INPs active at moderate supercooling - increased abundance during precipitation and size - with a widely used approach to parametrise INPs active at colder temperatures. Our study is the first to demonstrate the potential of differentiating between air masses that are precipitating, non-precipitating, or carrying

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desert dust to predict INP concentrations. Future observations need to constrain the presented parametrisation functions in a wider geographical context. Particular attention has to be attributed to sampling at mixed-phase cloud height and including larger aerosol particles, as was done in this study. The more differentiated parameterisation including air mass history is a promising way towards predicting INPs active at around −15 °C, which probably trigger much of the precipitation above midlatitude continental regions during winter.

5 Data availability

https://www.envidat.ch/group/about/raclets-field-campaign

Author contributions. CM and FC conceived the study. CM and JW conduced the measurements. JW provided the aerosol data. MAS conduced the modeling. ZAK and JH hosted the entire measurement campaign. CM and FC interpreted the data and wrote the manuscript with contributions from JW, MAS, ZAK, JH and CA.

Competing interests. The authors declare no competing interest.

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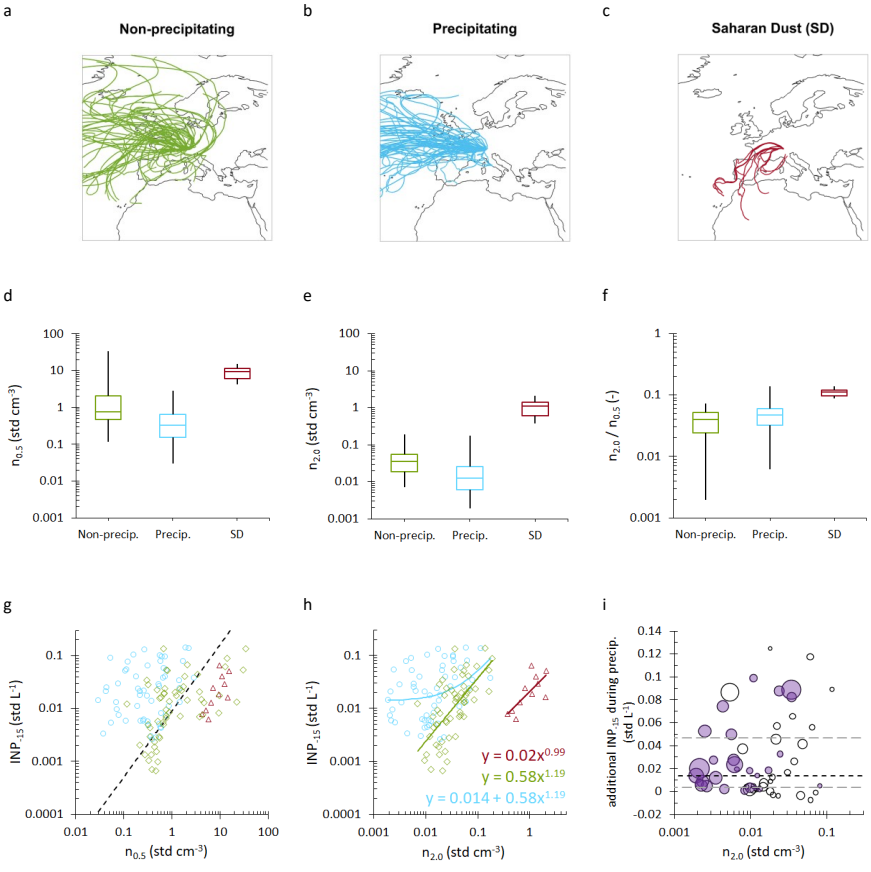

Figure 1. Separation of INP abundance in aerosol populations of air masses that were non-precipitating (green), precipitating (blue), and carrying Saharan Dust (SD, red). (**a**, **b**, **c**) Backward trajectories of air masses. Number concentrations of aerosol particles with diameters (**d**) >0.5 µm [$n_{0.5}$] and (**e**) >2.0 µm [$n_{2.0}$] and (**f**) their ratio. Concentrations of ice nucleating particles active at −15 °C or warmer [INP_{-15}] (**g**) versus [$n_{0.5}$] and (**h**) versus [$n_{2.0}$] for precipitating (circles), non-precipitating (diamonds) and SD air masses (triangles). The D15 parametrisation extrapolated to −15 °C is shown as a black, dashed line. Preliminary parametrisations based on [$n_{2.0}$] for non-precipitating and SD air masses are power functions. The function for precipitating air masses is the same as for non-precipitating air masses but with an added constant equivalent to 0.014 INPs L^{-1}. (**i**) Difference between [INP_{-15}] in precipitating air masses and the function fitted to the non-precipitating air masses. The median difference is 0.014 std L^{-1} (black line) and the lower and upper quartiles are 0.003 and 0.047 std L^{-1}, respectively (grey lines). Circle area is proportional to the amount of precipitation along the last 6 hours of the trajectory prior to sampling. Filled symbols are for samples precipitating at Weissfluhjoch.