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Influence of the ambient humidity on the concentration of natural deposition-mode ice-nucleating particles

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Abstract. This study reports measurements of deposition-mode ice-nucleating particle (INP) concentrations at ground level during the period July–December 2014 in Córdoba, Argentina. Ambient air was sampled into a cloud chamber where the INP concentration was measured at a temperature of −25 °C and a 15 % supersaturation over ice. Measurements were performed on days with different thermodynamic conditions, including rainy days. The effect of the relative humidity at ground level (RH_{amb}) on the INP concentration was analyzed. The number of INPs activated varied from 1 L^{-1} at RH_{amb} of 25 % to 30 L^{-1} at RH_{amb} of 90 %. In general, a linear trend between the INP concentration and the RH_{amb} was found, suggesting that this variability must be related to the effectiveness of the aerosols acting as INPs. From the backward trajectories analysis, it was found that the link between INP concentration and RH_{amb} is independent of the origin of the air masses. The role of biological INPs and nucleation occurring in pores and cavities was discussed as a possible mechanism to explain the increase of the INP concentration during high ambient relative humidity events. This work provides valuable measurements of deposition-mode INP concentrations from the Southern Hemisphere where INP data are sparse so far.

1 Introduction

Natural ice-nucleating particles (INP) are considered important in atmospheric processes since they induce freezing in clouds, thus initiating an efficient mechanism for cloud particles to reach a precipitating size. Consequently, many studies have been undertaken to determine their absolute concentrations, their origin and their chemical composition (DeMott et al., 2011; Hoose and Möhler, 2012).

Homogeneous freezing of supercooled droplets occurs at temperatures near −40 °C, but at temperatures warmer than approximately −36 °C, heterogeneous nucleation occurs (e.g., Cantrell and Heymsfield, 2005), which involves four different modes: condensation freezing, immersion freezing, contact freezing and deposition nucleation (Vali, 1985). Cloud condensation nuclei (CCN) can serve as INPs at temperatures below 0 °C during the condensation freezing mode. During the immersion-freezing mode, a particle suspended in the droplet initiates freezing. In the contact-freezing mode, ice is formed by the collision of cloud droplets with interstitial aerosols within the cloud. When the environment is supersaturated with respect to ice and subsaturated with respect to liquid water, ice deposits on the INPs directly from the vapor phase. This last mode is known as deposition nucleation. Measurements performed in the regime below water saturation are important in the understanding of deposition nucleation (DeMott et al., 2011).

Temperature and supersaturation ratio are the two main parameters determining deposition nucleation. Several reports have provided measurements of INP concentration for different temperature and supersaturation conditions. Studies linking INP concentration to temperature have shown an exponential trend in the dependence of INPs on temperature (e.g., Fletcher, 1962; Hussain and Saunders, 1984; Meyers et al., 1992; López and Ávila, 2013). This same correlation was found when studying INP concentration, regarding supersaturation (e.g., Rogers, 1982; Al-Naimi and Saunders, 1985; Hussain and Saunders, 1984; Cooper, 1980; López and Ávila, 2013).

Although temperature and supersaturation are recognized as the two main parameters in the activation of INPs, a significant divergence among the results of INP quantification is found in the literature when INP measurements are carried out at around the same temperature and supersaturation conditions (DeMott et al., 2011; Hoose and Möhler, 2012). In general, the results show that the ice-nucleating particles' content in the atmosphere varies considerably from day to day and from place to place. For instance, important anomalies of atmospheric ice-nucleating particles were observed under thunderclouds (Isono and Tanaka, 1966; Isaac and Douglas, 1973). Increments of the concentrations of ice-nucleating particles at ground level, by a factor of 10 or higher, have been measured during precipitation from convective storm precipitation. These anomalies were associated with downdrafts in and under the thunderclouds. Prenni et al. (2013) also studied the impact of precipitation at a forested site on the concentration and composition of INPs. They showed that at ground level, INP concentrations increase during rain events.

Thus, our knowledge of ice formation through nucleation modes is still scarce and limited. As a consequence, ice nucleation processes are currently very poorly represented in global climate models (e.g., Hoose et al., 2010). Furthermore, the study of ice cloud formation over a wide range of atmospherically relevant temperatures and humidity values is challenging (e.g., DeMott et al., 2011; Murray et al., 2012). Particularly, studies on anomalies in the concentration of atmospheric ice-nucleating particles are important, since they give us clues for solving problems on the origin and the nature of atmospheric ice-nucleating particles. The major goal of this work is the study of the factors that regulate the effectiveness of aerosols on deposition ice nucleation. Regarding the possible effect of precipitation on the INP concentration, this study reports measurements of INP concentration at ground level during the period July–December 2014 in Córdoba, Argentina. The experiments were performed on days with different thermodynamic conditions, including precipitation days, and the effect of the relative humidity at ground level on the INP concentration was analyzed. INP concentrations in deposition nucleation mode were determined at a temperature of approximately $-25\,^{\circ}\mathrm{C}$ and a supersaturation of approximately 15 % over ice. The ambient relative humidity at ground level was measured during all the experiments, in order to link this variable to the INP concentration.

2 Experiment

Measurements were carried out in Córdoba, a mediterranean city of Argentina, located in the center of the country, in a semiarid region (latitude $-31.4°$; longitude $-64.18°$; 470 m a.s.l.). Córdoba is the second largest city in the country, with approximately 1.3 million inhabitants. The climate is subhumid and the prevailing wind direction is NE. The annual average precipitation is close to 700 mm. However, the area is affected by severe and persistent dry periods that occur cyclically. Snowfall occurs infrequently; thus, precipitation mostly consists of rain. Rainfall is highly seasonal, concentrated mainly in summer (December–February) having accumulated values higher than 400 mm, while the dry winter season (June–August) has a cumulative precipitation depth close to 20 mm.

In a previous work we have quantified the INP concentration (López and Ávila, 2013) in deposition nucleation mode at ground level in Córdoba, for temperatures between -15 and $-30\,^{\circ}\mathrm{C}$, and ice supersaturation ranging between 2 and 20 %. In the present work the experiments were carried out following the same procedure as described by López and Ávila (2013) and a total of 63 measurements of INP concentration were carried out during 45 days, from 28 July until 10 December 2014 (winter and spring seasons). This procedure involves a cloud chamber, 46 L in volume, placed inside a cold room. Controlled volumes of humid air were injected from the exterior (at ambient temperature) in order to increase the relative humidity in the cloud chamber. Since the same volume of air is removed while this air is injected, the number of aerosols and the pressure inside the chamber are kept around constant. The increasing of the relative humidity over ice inside the cloud chamber ($\mathrm{RH_i}$) is explained according to the adiabatic isobaric mixing of two masses of air with different relative humidity and temperature (Curry and Webster, 1999). The supersaturation is achieved starting with a $\mathrm{RH_i}$ slightly lower than 100 % in the cloud chamber and by introducing controlled air injections from outside. Since the increase of $\mathrm{RH_i}$ is known for each air injection, it is possible to estimate the increase produced by a known number of injections, even once the vapor is supersaturated over ice. Once the supersaturation is reached, the INPs are activated and then grown at the expense of vapor until ice crystals fall down onto a dish placed on the floor of the cold chamber. This dish contains a supercooled aqueous solution of cane sugar. Once in the sugar solution, the ice crystals grow and reach a size large enough to be counted with the naked eye, as can be seen in Fig. 1. For a more detailed description of the procedure, see López and Ávila (2013).

In all the measurements carried out in the present work, the conditions of temperature (T) and supersaturation over ice (S_i) were settled at around $-25\,^{\circ}\mathrm{C}$ and 15 %, respectively. These values were chosen because, under these conditions, the number of activated INPs is significant and can easily be determined (López and Ávila, 2013). Furthermore, these T and S_i values are experimentally quick to reach, which facilitates the execution of the experiments. In order to determine $\mathrm{RH_i}$ and T in the cloud chamber, the Testo 435-4 multifunction measuring instrument was used. This instrument measures relative humidity in the temperature range $[-50, 150]\,^{\circ}\mathrm{C}$. When $\mathrm{RH_i}$ is measured over ice at temperatures below $0\,^{\circ}\mathrm{C}$, the resolution is 0.1 %. The experimental uncertainty associated with the determination of T and S_i was

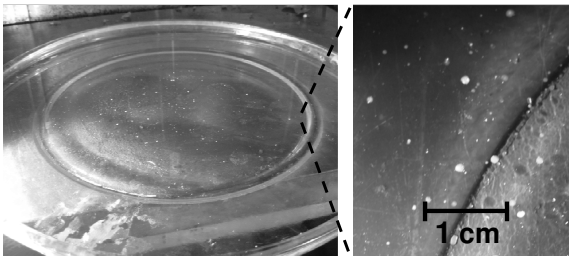

Figure 1. Images of crystals once they have grown on the sugar solution.

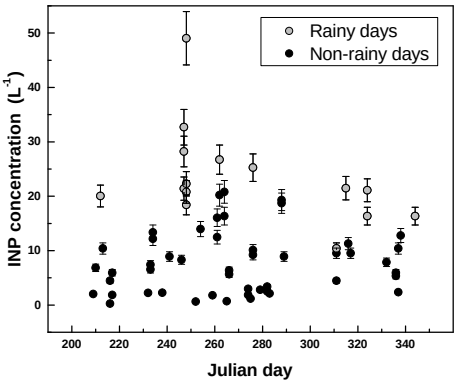

Figure 2. Data of INP concentrations measured throughout the period studied.

$\pm 2\,^{\circ}\mathrm{C}$ and $\pm 2\,\%$, respectively. These errors arise from variations in the experimental conditions involved in each measurement, such as the difference in the temperature between the beginning and the end of the air injections, the variability in the $\mathrm{RH_i}$ increase between the injections, the number of injections until reaching the particular S_i, and the accuracy of the humidity sensor.

Thus, considering that T and S_i were the same for all the experiments (apart from the inherent errors), the INP concentration was studied as a function of the ambient relative humidity at ground level ($\mathrm{RH_{amb}}$); this is the relative humidity of the air mass injected from the exterior. $\mathrm{RH_{amb}}$ was measured during all the experiments by using the instrument EE31 Series Model D (E+E Elektronik), which had a remote sensing probe for relative humidity measurements in the temperature range $[-40, 180]\,^{\circ}\mathrm{C}$. The uncertainty in the determination of $\mathrm{RH_{amb}}$ was $\pm 1\,\%$.

3 Results and discussions

The experiments were all performed for a supersaturated atmosphere with respect to ice and subsaturated with respect to liquid water; thus, only the deposition-mode INP concentrations were quantified. The INP concentration was determined at a temperature of approximately $-25\,^{\circ}\mathrm{C}$ and a supersaturation of approximately 15 % over ice. Figure 2 shows the temporal variations of the INP concentration values during the period under study. The gray points in the graph correspond to measurements performed during a rain event or immediately after rain had stopped. The error associated in the estimation of INP concentration did not exceed 10 %. There are some time periods without experimental data, mainly because the cold chamber was unable to achieve the expected temperature. The period of study is too short to perform a reliable statistical test showing seasonal behavior of INP concentration; nevertheless, the results do not seem to indicate that there is any particular trend during this period.

In order to study the effect of the $\mathrm{RH_{amb}}$ in the INP concentration, Fig. 3 displays the INP concentration as a function of the $\mathrm{RH_{amb}}$. Again, the gray points indicate measurements performed on rainy days. INP concentration values can be seen to increase with $\mathrm{RH_{amb}}$. The best linear fitting of the data yields values of 0.39 and -9.2 for the slope and ordinate, respectively, the square of the correlation coefficient (R^2) being equal to 0.71. The linear trend between the INP concentration and the $\mathrm{RH_{amb}}$ is remarkable and the dispersion of the experimental points is likely related to the natural variability in the INP concentration and to the inherent errors associated with the determination of T, S_i, $\mathrm{RH_{amb}}$ and INP number, which are present in each experiment.

The number of INP activated varies from $\sim 1\,\mathrm{L^{-1}}$ at $\mathrm{RH_{amb}} \sim 25\,\%$ to $\sim 30\,\mathrm{L^{-1}}$ at $\mathrm{RH_{amb}} \sim 90\,\%$. This variation is consistent with the results reported in López and Ávila (2013) on the same site, who found that INP concentration values varied between 2 and $35\,\mathrm{L^{-1}}$ at a temperature of $\sim -25\,^{\circ}\mathrm{C}$ and $\sim 15\,\%$ of supersaturation over ice. Thus, the variability in the INP concentration in the previous work could be related to the variability in $\mathrm{RH_{amb}}$ at the time the measurements were carried out (this variable was not recorded in such experiments). Current results are also in agreement with those reported by Prenni et al. (2013), who showed that, at ground level, the INP concentration rises by a factor of up to 40 during rain. Actually, here we observe that not only rainy days are required for increasing INP concentrations but also high relative humidity is needed, as shown in Fig. 3 during rainfall events with low humidity.

A variety of insoluble particles such as volcanic ash, mineral dust, soot, metallic particles or primary biological particles have been suggested as INP. However, the requirements needed for a surface to be an efficient INP are not completely understood (Pruppacher and Klett, 1997; DeMott, 2010; Hoose et al., 2010). Prenni et al. (2013), Hader et al. (2014) and Bigg et al. (2015) presented evidence that the rise of INP concentration during rain events can be related to the increase of biological particle concentration. It has been shown that biological particles can be released during precipitation through mechanical mechanisms. For instance, winds produced by storms can release biological particles from leaf surfaces and soil. The release of these biological particles includes biological INPs (bacteria, fungal spores and pollen),

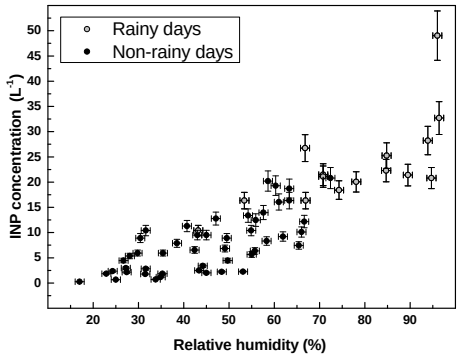

Figure 3. Data of INP concentrations at different ambient relative humidity levels.

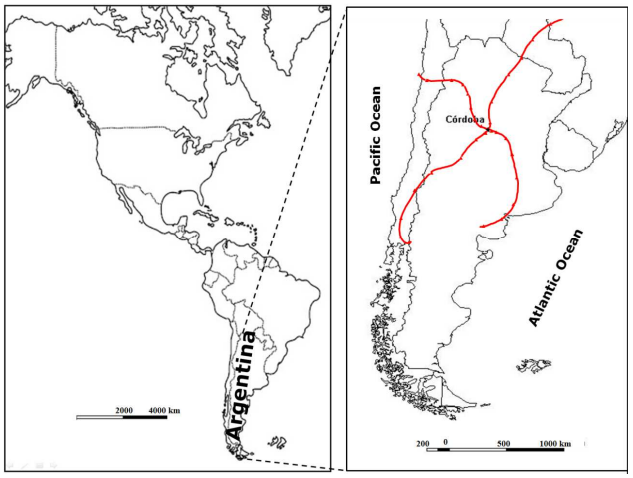

Figure 4. Location of Argentina and air mass pathways of different quadrants: northwest, northeast, southwest and southeast.

which can impact the INP population. This assumption was supported by different studies reporting both the release of biological particles during rain events and the role of these particles as sources of INPs (Huffman et al., 2012, 2013). Furthermore, Wright et al. (2014) provided observational evidence that the high relative humidity, coordinated with the arrival of a cold-frontal boundary on an observation site, triggered the active release of biological particles. Since mixing along the frontal boundary allows for the biological material to be entrained into the warm air that is being lifted over the cold front, they suggest that these biological particles can serve as immersion-mode INP while droplets cool during ascent.

It is important to remark that unlike the present study, where the deposition nucleation mode was examined, the previous studies on a relation between INP concentrations and relative humidity or rain events mainly focused on condensation and immersion-freezing modes. We have no direct evidence, so far, that the increase of the INP concentration with increasing humidity observed in the present work can be explained by biological particles.

Mineral dust aerosols seem to play a relevant role in deposition nucleation. They have active sites which are able to start ice nucleation. These active sites are linked to surface defects, like cracks, capillaries, pores or cavities, in which water vapor condenses under vapor pressures below saturation (Fukuta, 1966; Fletcher, 1969). Water vapor condenses in a capillary of a wettable particle under a pressure below water saturation (p), according to the Kelvin equation:

$$RTr \ln\left(\frac{p}{p_b}\right) = -2\gamma\nu_l. \quad (1)$$

R is the gas constant, T the absolute temperature, r the radius of the curved water surface, p_b the water vapor pressure over a flat water surface, γ the surface tension of water and ν_l the molar volume of liquid water. Recently, Marcolli (2014) hypothesized that ice nucleation below water saturation occurs predominantly in water confined between closely spaced surfaces, such as cavities and pores that may form in aerosol particles. In this sense, it is plausible to expect that as the RH_{amb} increases, more water molecules from the surrounding environment can be adsorbed in cavities and pores. As a result, the condensation and the subsequent freezing can occur in the cavity when the relative humidity is lower than 100 % outside. Considering this fact, a significant enhancement in ice nucleation efficiency should be expected at higher ambient relative humidity, which is consistent with the results reported here. Even though further work is needed to test and corroborate this hypothesis, the results presented in this work may support the idea that deposition nucleation can be pore condensation and freezing which occur in defects on the surface of some aerosol particles, as proposed by Marcolli (2014).

In order to relate the origin of the air masses to the behavior of INP measurements regarding RH, backward trajectories were calculated for all days when INP concentration measurements were carried out. Back trajectories were calculated by the NOAA HYSPLIT model (Hybrid Single-Particle Lagrangian Integrated Trajectory model) (Draxler and Rolph, 2003). Air mass back trajectories were calculated at 1000 m above mean sea level, 48 h before the arrival time to the sampling site. NOAA trajectories were calculated every day of INP concentration measurements. The air mass back trajectories were subdivided into four groups, regarding the quadrants from the compass rose. As an example, Fig. 4 shows air mass back trajectories representative of the four studied sectors, which will be referred to as the northwest quadrant, the northeast quadrant, the southwest quadrant and the southeast quadrant. The lower sample number was obtained coming from the NW quadrant (7.5 %), while the highest numbers of INP measurements were obtained coming from the NE quadrant (42.5 %). Trajectories coming from SE and SW quadrants were 22.5 % and 27.5 %, respectively. Figure 5 shows the INP concentration measurements

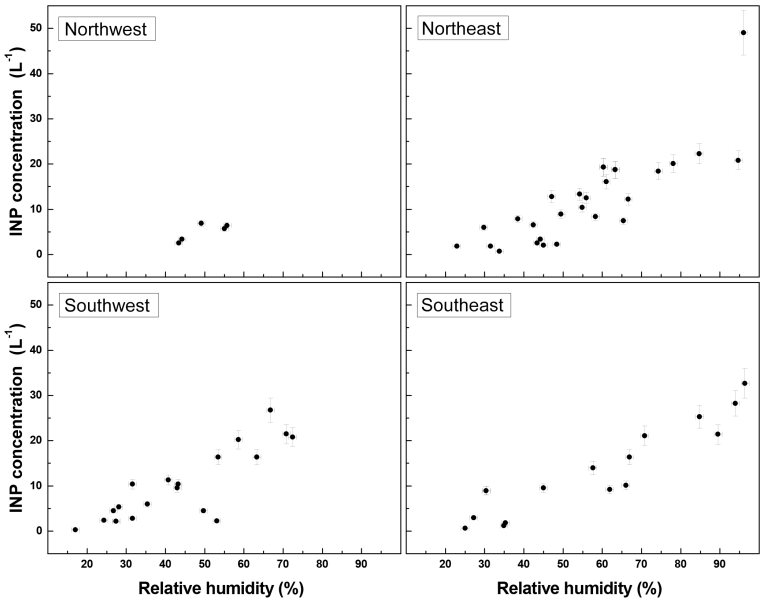

Figure 5. INP concentrations at different ambient relative humidity for the different origins of the air masses.

grouped following the previous classification of the origin of air masses. Observe that the number of INP measurements from the NW quadrant is too scarce to make an analysis. However, the analysis from the three remaining quadrants shows the increase of INP concentration with the increase in RH_{amb}, in the same way that it was observed in Fig. 3. Results show that, from the studied period, variations in RH_{amb} are not explained because of the origin of the air masses. This is to say that the relation between INP concentration and the ambient relative humidity is independent of the origin of the air masses. This fact discards the possibility that the increase in INP concentration is due to changes in the sources of the aerosols acting as ice-nucleating particles. Thus, HYSPLIT results seem to be consistent with the previous assumption regarding the importance of the ambient relative humidity on the ability of aerosols to act as INPs.

4 Summary and concluding remarks

Studies of heterogeneous ice nucleation with natural aerosols under a broad range of thermodynamic conditions are important, because they can provide clues for solving problems regarding the origin and the nature of atmospheric INP. This study reports measurements of deposition INP concentrations at ground level during the period July–December 2014 in Córdoba, Argentina. The experiments were carried out at a temperature of $(-25 \pm 2)\,^{\circ}C$ and a supersaturation of $(15 \pm 2)\,\%$ over ice. The effect of the relative humidity at ground level on the INP concentration was analyzed and a linear trend between the INP concentration and the RH_{amb} was found, suggesting that this variable must be related to the ability of the aerosols acting as INP. This assumption seems to be corroborated by the analysis of the backward trajectories of the air masses, which shows that the relation between INP concentration and the ambient relative humidity is independent of the origin of the air masses. Therefore, the variability observed on INP concentration cannot be explained regarding the changes in the sources of the aerosols acting as ice-nucleating particles.

The role of biological INPs and nucleation occurring in pores and cavities (as suggested by Marcolli, 2014) was discussed as a possible mechanism to explain the increase in the INP concentration during high ambient relative humidity events. More ice nucleation studies should be carried out to address the differentiation between these mechanisms.

This work contributes to the study of the factors affecting the IN concentration. Laboratory studies of heterogeneous ice nucleation with natural aerosols are important for the possibility of transferring the results into parameterizations for numerical models of clouds and climate. Furthermore, this work provides valuable measurements of deposition-mode INP concentrations from the Southern Hemisphere where INP data are sparse so far.

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