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The ice nucleating ability of pollen
Part III: New laboratory studies in immersion and contact freezing modes including more pollen types

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Abstract

Based on earlier experimental studies, the ice nucleating abilities of further pollen types were investigated in the immersion and contact freezing modes. The studies were carried out at the Mainz vertical wind tunnel with freely floating supercooled droplets down to − 28 °C. The pollen had diameters between 26 and 28 μm and correspondingly low sink velocities around 2.5 cm s^{-1}. The radii of the studied drops were calculated from the recorded wind velocity and for both freezing modes the radii of the observed droplets varied between 315 and 380 μm. Immersion freezing experiments were conducted with pollen particles added to the droplets while in contact freezing experiments supercooled droplets were subjected to a burst of pollen particles. The median freezing temperatures found in the immersion freezing mode were: − 13.5 °C (alder), − 21.5 °C (lombardy poplar), − 21.0 °C (redtop grass) and − 19.8 °C (kentucky blue). The median freezing temperatures in the contact freezing mode were found as: − 12.6 °C (alder), − 17.9 °C (lombardy poplar), − 18.7 °C (redtop grass) and − 16.1 °C (kentucky blue). The results show that the ice nucleating ability of pollen is not restricted to single pollen types but seems to be a general pollen property.

Keywords: Pollen; Ice nucleating ability; Immersion freezing; Contact freezing; Median freezing temperature

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1. Introduction

Field measurements have shown that biological aerosol particles are an essential part of the total atmospheric aerosol. Primary biological aerosol particles are for instance viruses, algae, plant fragments, spores, pollen and protozoa with diameters between 0.01 and 100 μm. The contribution of biological aerosol particles to the total atmospheric aerosol is about 25% both by concentration or by volume in urban or rural air masses (Matthias-Maser and Jaenicke, 1995; Matthias-Maser, 1998) and 10–20% in remote marine air masses (Gruber et al., 1998). According to Bogs (1996) and Gruber et al. (1998) the contribution of biological aerosol particles towards the total insoluble particles in rainwater and cloud water samples has been found to be up to 25% and 9–50%, respectively.

An important point is that biological particles are able to initiate freezing at warmer temperatures than other types of ice nuclei such as soot or mineral particles (e.g. Pitter and Pruppacher, 1973; Diehl and Mitra, 1998). A number of studies investigated the ice nucleating ability of different biological aerosol particles such as bacteria that show very high freezing temperatures up to -2 °C (Vali et al., 1976; Maki and Willoughby, 1978; Levin and Yankofsky, 1983) and marine plankton with freezing temperatures up to -4 °C (Schnell and Vali, 1976; Vali et al., 1976). Freezing studies with leaf litter resulted in similar freezing temperatures (Schnell and Vali, 1976; Schnell and Tan-Schnell, 1982). The highest activation freezing temperatures of free-living fungi were found to be at -2.5 °C and -1.9 °C for lichen species (Lee et al., 1995).

The initiation of freezing by biological particles sets in at temperatures not far below 0 °C, that would correspond to rather lower altitudes and at early stages of natural cloud development. Because of the release of latent heat after ice formation at warmer temperatures, biological particles could enhance updrafts in clouds allowing the clouds to rise to higher altitudes and thus attain lower temperatures where other ice nuclei such as minerals and soot particles might also become active. This can be calculated from numerical simulations for biomass burning situations (Diehl et al., 2004) where the ice nucleating abilities of various ice nuclei were considered. In Diehl et al. (2002) only four types of pollen were investigated. To demonstrate that efficient ice nucleation ability is not a property of only a few pollen species, further experiments were undertaken in the present study testing four more pollen species.

Pollen are large biological particles having diameters lying in the range from 10 up to 100 μm. On the surface of pollen grains one can find possible active sites like grooves and edges. Pollen with low sink velocities can be lifted to altitudes up to 3000 m and higher in the atmosphere and stay there for long time periods (Rempe, 1937; Gregory, 1978). According to Grosse-Bauckmann and Stix (1968) birch pollen are released near ground level in concentrations up to 1.5 l^{-1}. Following Rempe (1937) we can expect a continuous decrease in pollen concentration with height leading to a value of 1.2 l^{-1} at an altitude of 1000 m altitude and of 0.6 l^{-1} at 2000 m altitude. Balloon ascents in and above inversion layers show accumulations of pollen (Linkens and Jorge, 1986) which means that pollen may temporarily be present in the atmosphere in concentrations comparable to those of ice nuclei (Pruppacher and Klett, 1997). Therefore, for the new experiments pollen having rather low sink velocities of about 2.5 cm s^{-1} and thereby implying long atmospheric residence times were selected.

The investigations described in the present paper were carried out in the same way as mentioned in Diehl et al. (2002). The ice nucleating abilities of four more pollen types were studied in the immersion and contact freezing modes by using the facilities of the vertical wind tunnel, which allows to freely float supercooled droplets in a vertical air stream.

2. Experimental methodology

In the present studies two sets of laboratory experiments were carried out to determine the ice nucleating ability of two deciduous tree pollen, alder (*Alnus incana*) and lombardy poplar (*Populus nigra*) and two grass pollen species, redtop grass (*Agrostis alba*) and kentucky blue (*Poa pratensis*). These pollen types were chosen because of their small diameters between 26 and 28 μm which lead to their corresponding low sink velocities around 2.5 cm s^{-1} and longer atmospheric residence times (Straka, 1975; Gregory, 1978). The pollen grains were commercially available in a standardized dried form which is free from impurities and especially free from bacteria so that the results could not be influenced by the more efficient ice nucleating ability of bacteria. Fig. 1 shows photographs of the investigated pollen types.

The vertical wind tunnel at the University of Mainz allows one to float drops with radii between 20 μm and 4 mm in the vertical air stream of the tunnel. This wind tunnel represents the second generation of the type of tunnel built in 1968 at the UCLA (Pruppacher and Neiburger, 1968; Beard and Pruppacher, 1969). A detailed description of the Mainz vertical wind tunnel is given in Pruppacher (1988). Temperature, wind speed and relative humidity of the tunnel air are recorded continuously. For freezing experiments the tunnel can be cooled down to −30 °C in the observation section where the supercooled droplets are floated. The inlet air stream is first led through a set of particle filters to prevent possible ice nuclei present in the room air from influencing the experiments. Experiments with particle-free water droplets with radii between 200 μm and 400 μm in particle-free air showed no freezing of the drops at temperatures warmer than −28 °C.

For the present experiments the observation volume of the wind tunnel was thermally insulated to improve the floating conditions of small droplets at low temperatures and to prevent them from drifting toward the walls of the tunnel. The insulation also reduced the vertical temperature gradient in the observation section thereby reducing the systematic

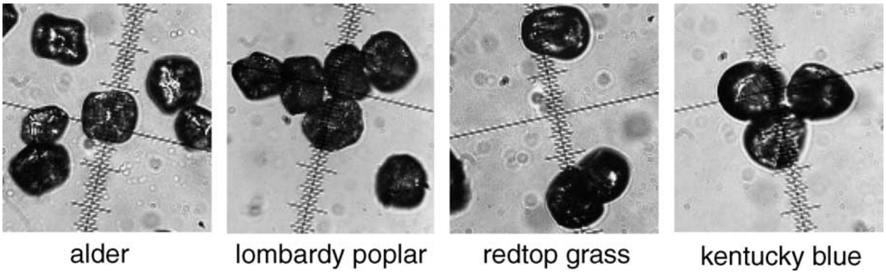

Fig. 1. Photographs of pollen grains used in the experiment: (a) alder, diameter ca. 26 μm; (b) lombardy poplar, diameter ca. 26 μm; (c) redtop grass, diameter ca. 28 μm; (d) kentucky blue, diameter ca. 28 μm.

difference between the temperature of the droplet and the temperature registered by the sensor to about ± 0.05 °C. In all experiments the air temperature in the observation section was varied and the temperature whether the drop froze or not was recorded and classified in 2 °C intervals. From the observed number of droplets the fraction of frozen drops per temperature interval were determined using a fitting curve applied to 2 °C classification intervals. For each pollen type the median freezing temperature (temperature at which 50% of the observed drops froze) was estimated from the fraction of frozen drops in both freezing modes.

The drop terminal velocity was obtained from the recorded wind velocity since in order to keep the drop floating in the observation section the terminal velocity must be equal to the wind tunnel velocity. From the measured terminal velocity and the environmental conditions such as temperature, pressure and relative humidity, the drop sizes were determined. For both freezing modes, the radii of the observed droplets varied between 315 and 380 μm. This variation in drop size has little influence on its freezing behavior.

In the first set of experiments the immersion freezing ability of all four kinds of pollen was studied. Drops, generated from purified distilled water to which a defined amount of pollen, 35 mg to 100 ml water, was added, were injected into the observation area of the wind tunnel by means of a syringe and needle. The droplets were freely suspended in the airstream at various temperatures down to − 28 °C and it was observed whether they froze. For each pollen type the number of drops observed was between 600 and 750. In this way a total number of about 3500 individual droplets were floated and investigated during the course of the study to provide a significant number for statistical analysis.

In the present study an attempt was made to document the freezing process of a droplet in the immersion mode by means of a video camera. Furthermore, to confirm earlier results of ice multiplication induced by freezing of supercooled drops (Pruppacher and Schlamp, 1975) observations were carried out during the immersion freezing experiments to determine whether the frozen drops split on freezing.

In the second set of experiments the contact freezing mode of the different pollen types was investigated. Pure water drops were freely suspended in the observation area of the vertical wind tunnel in temperature equilibrium with the ambient air at various temperatures down to − 28 °C. A small cloud of pollen was manually generated upstream of the droplet by blowing a short burst of air through a tube containing the pollen grains. While the particles passed the drop it was observed whether the drop froze after a collision with the pollen particles. The temperature intervals were also distributed in 2 °C intervals. The number of studied droplets was approximately 200 per investigated pollen type. This was less than that in the immersion freezing mode because of the more difficult experimental operation. Due to the burst of pollen grains the drops became very unstable.

3. Results and conclusions

The new results together with earlier results in Diehl et al. (2002) confirm the importance of biological aerosol particles as potential ice nuclei. The investigated pollen types were able to act as ice nuclei in the immersion and even more efficiently in the contact freezing mode.

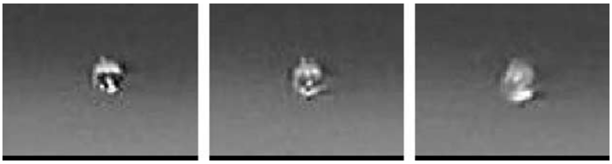

Fig. 2. A freezing droplet: not frozen drop (left), freezing drop (middle), frozen drop (right).

Fig. 2 shows a sequence of a freezing drop. Frozen drops were identifiable because they became opaque upon freezing. Because of the phase change of the droplets their regular shape became irregular with protrusions leading to an abrupt change of their terminal

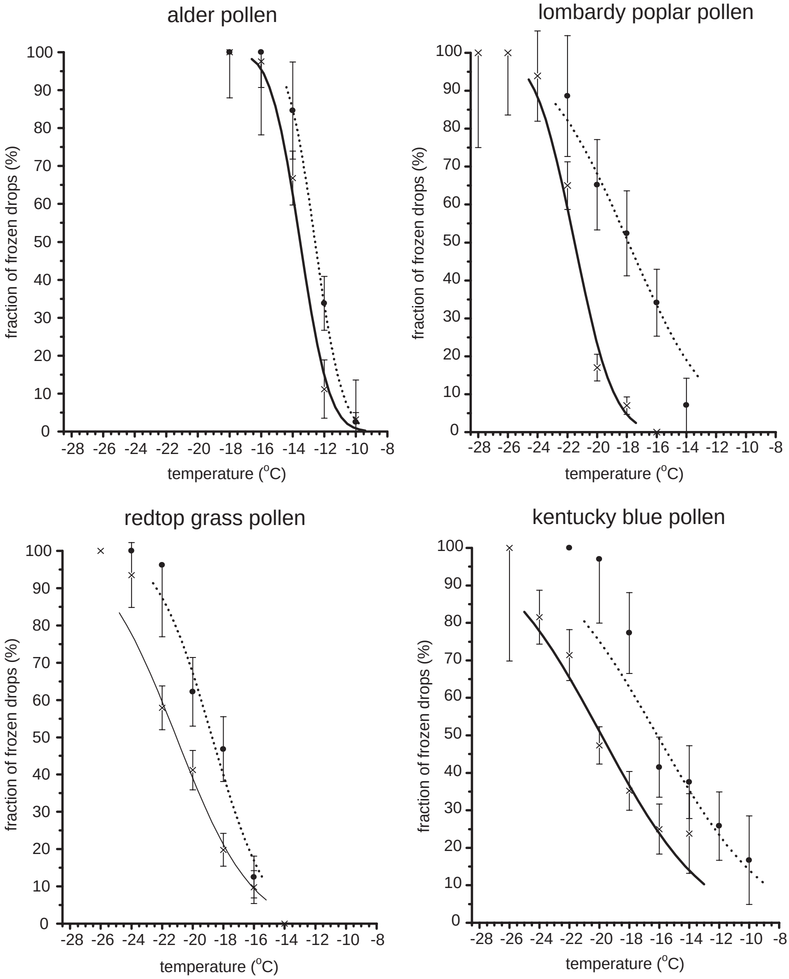

Fig. 3. Variation of the fraction of drops frozen due to immersion freezing (solid lines) and contact freezing (dashed lines) with temperature for alder, lombardy poplar, redtop grass and kentucky blue pollen.

velocity and falling mode: Some frozen droplets drifted horizontally and described a spiral track in a random fashion, some others became unstable and started spinning around.

It could be observed that for every investigated pollen type about 10% of the droplets split when they froze in the immersion mode. This leads to a multiplication factor of about 1.1 assuming that all split frozen drops break into two parts. This value agrees very well with the one observed by Pruppacher and Schlamp (1975) in their wind tunnel experiments with clay particles immersed in supercooled drops. The present results confirm their conclusion that immersion freezing does not contribute significantly to ice multiplication in the atmosphere compared to contact freezing and, in particular, ice multiplication during riming as shown by Hallett and Mossop (1974).

In Fig. 3 the results of immersion (solid lines) and contact (dashed lines) freezing experiments are summarized as a fraction of frozen drops in percent at different temperatures. The error bars are evaluated from the number of investigated drops in each temperature interval. From the distribution of the freezing temperatures into intervals the initiating freezing temperature was determined. Table 1 shows the median freezing temperatures and the initiating freezing temperatures for each pollen type and both freezing modes. For the immersion freezing mode alder pollen indicate the warmest freezing temperature of −13.5 °C, followed by kentucky blue pollen with a freezing temperature of −16.1 °C. Drops containing redtop pollen froze at a lower temperature of −21.0 °C while a still lower temperature of −21.5 °C was obtained for lombardy poplar pollen particles. The median freezing temperatures in the contact mode were always higher the corresponding ones in the immersion freezing mode, the difference was between 1 and 4 °C. Alder pollen possesses the highest contact freezing temperature of −12.6 °C and redtop grass demonstrated the lowest freezing temperature of −18.7 °C.

These results indicate that the ice nucleating ability of pollen is not restricted to single pollen types but seems to be a general pollen property. The median freezing temperatures of pollen species in immersion and contact modes which were investigated in Part II of the present paper series (Diehl et al., 2002) show median freezing temperatures between −14 °C and −16 °C for immersion freezing and −12 °C and −14 °C for contact freezing, i.e. the newly investigated pollen species show lower freezing temperatures (except alder) than others. Summarizing these results for pollen, it seems to be obvious that the ice nucleating efficiency of pollen is lower than that of other biological particles such as bacteria or fungi, but it is still higher than that of soot particles (Diehl and Mitra, 1998; Gorbunov et al., 2001) and higher or at least in the range of the ice nucleating ability of mineral particles as montmorillonite and kaolinite (Pitter and Pruppacher, 1973): Montmorillonite particles

Table 1
The initiating and median freezing temperatures of immersion freezing and contact freezing modes for each pollen type (2 °C classification).

Pollen type	Initial freezing temperature (°C)		Median freezing temperature (°C)	
	Immersion	Contact	Immersion	Contact
Alder	−9 to −11	−9 to −11	−13.5 ± 0.2	−12.6 ± 0.4
Lombardy poplar	−17 to −19	−13 to −15	−21.5 ± 0.4	−17.9 ± 1.2
Redtop grass	−15 to −17	−15 to −17	−21.0 ± 0.6	−18.7 ± 0.7
Kentucky blue	−13 to −15	−9 to −11	−19.8 ± 0.7	−16.1 ± 1.4

show median freezing temperatures of −19 °C in the immersion mode (drop radius 350 μm) and −8 °C in the contact mode, kaolinite −23 °C in immersion freezing and −12 °C in contact freezing (Pitter and Pruppacher, 1973). The ice nucleating ability of soot was determined by Diehl and Mitra (1998) to be −28 °C in the immersion mode. Thus, the role of biological particles on cloud and precipitation processes should not be neglected in comparison to mineral and soot particles.

As suggested in Diehl and Wurzler (2004) and Diehl et al. (2004) the consideration of different ice nucleating abilities of various ice nuclei in model simulations would allow for a more detailed description of the effects of freezing processes on, e.g., the vertical cloud dynamics.

However, still it is unclear why and how pollen act as ice nuclei. That means the dependence of the ice nucleating efficiency on morphology, cell surface structure, membrane properties, protein composition and intercellular spaces has to be investigated. On the other hand, together with the earlier pollen results of Diehl et al. (2002) the new results show a positive correlation between the ice nucleating efficiency and the time when pollen are released from the plants: except poplar pollen, the ice nucleating ability of pollen is higher the earlier in the year the pollen are released. Thus, the biological reason for the ice nucleating ability of pollen might be a kind of freezing tolerance, i.e. extracellular freezing starting not far below 0 °C protects the interior of the cells. This was discussed already in Diehl et al. (2002). Further investigation of more pollen types which complete a sort of ice nucleating ability could give more information about this hypothesis in conjunction with seasonal and geographic variations of pollen sources.

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