Response to Daniel O'Sullivan:

We thank Dr. B. Murray and Dr. D. O'Sullivan for their helpful comments. We may answer the comments point by point.

1) Similar to comment 4 suggested by Dr. C. Morris, we would like to draw the authors' attention to their discussion of the 'efficiency' with which the pollens nucleate ice. In particular, we highlight the authors' comparison of their measured rate coefficients to those of mineral dusts determined by Eastwood et al. (J. Geophys. Res. 2008, 113, D22203). On the basis of this comparison, which is graphically depicted in Fig. 5, the authors conclude that "many pollen species produce far more efficient ice nuclei than all studied mineral dusts" (p. 27288, lines 7-9), which is a striking claim. However, the rate coefficient measurements conducted by Eastwood et al. were for ice nucleation in the deposition mode and cannot be directly compared with measurements in the immersion mode. They cannot be compared in this way because they are determined for explicit RH values and J will vary strongly with RH at constant temperature. Eastwood's measurements also have different units. We suggest that the authors compare their rate coefficients to those of mineral dusts which have been measured in the immersion mode, such as performed by Murray et al. (Atmos. Chem. Phys. 2011, 11, 4191-4207). Murray et al. report J values in units of cm-2 s-1 but a comparison could be made if this were converted to cm-3 s-1 (units used by Pummer et al). This could be done for droplets of a particular size with a particular mineral concentration. It should be borne in mind that kaolinite is just one mineral type relevant to the atmosphere and other data suggest there are more 'efficient' minerals, although the results do not lend themselves to easy comparison with the format chosen by Pummer et al. units should be mentioned in caption of figure 5.

ANSWER: We removed the data of Eastwood et al. 2008 and inserted those proposed (Murray et al., 2011). According to our research kaolinite is an important kind of mineral, according to both abundance and IN efficiency (e.g. Zimmermann et al., 2008). To compare our data with those of Murray et al. 2011 we calculated:

$$J = J' \cdot \frac{\sigma}{V} \quad \text{Eq 1}$$

with J as the nucleation rate in Murray et al. 2010 (like in our paper), J' as the nucleation rate in Murray et al. 2011, V as droplet volume and σ as contact surface of IN per droplet.

Regarding 20 µm as average droplet diameter and using the highest listed value for σ ($3*10^{-5}$ cm^2), we calculate a conversion factor of 0.7162 cm^{-1}. By applying the linear fit by Murray et al. 2011 (see Eq. 2) we calculated J'-values and multiplied them with 0.7162 cm^{-1} in order to gain J-values.

$$\ln J' = -0.8802 \cdot T + 222.17 \qquad \text{Eq. 2}$$

As it can also be seen in the original Murray-paper, the curves for nucleation of pure water and kaolinite cross each other.

Eastwood, M. L., Cremel, S., Gehrke, C., Girard, E., and Bertram, A. K.: Ice nucleation on mineral dust particles: Onset conditions, nucleation rates and contact angles, J. Geophys. Res., 113, D22203, doi:10.1029/2008JD010639, 2008.

Murray, B. J., Broadley, S. L., Wilson, T. W., Bull, S. J., Wills, R. H., Christenson, H. K., and Murray, E. J.: Kinetics of the homogeneous freezing of water; Phys. Chem. Chem. Phys., 12, 10380-10387, 2010.

Murray, B. J., Broadley, S. L., Wilson, T. W., Atkinson, J. D., Wills, R. H.: Heterogeneous freezing of water droplets containing kaolinite particles, Atmos. Chem. Phys., 11, 4191-4207, doi:10.5194/acp-11-4191-2011, 2011.

Zimmermann, F., Weinbruch, S., Schütz, L., Hofmann, H., Ebert, M., Kandler, K., and Worringen, A.: Ice nucleation properties of the most abundant mineral dust phases, *J. Geophys. Res.*, 113, D23204, doi:10.1029/2008JD010655.

2) Fig. 5. Rather than using the natural logarithm as is stated on the vertical axis, the authors appear to have taken the base 10 logarithm of the rate coefficients from Eastwood et al. Is this also the case for the rate coefficients reported for the birch pollen? A table reporting the actual values might be useful.

ANSWER: We switched the axis format and now present J [$cm^{-3}*s^{-1}$]. We inserted a new table with numeric values for our measurement points.

3) P27221. ln 7-8. The Pratt result of 33% was for one out of a number of flights. Insert words 'up to'.

After E-Mail contact with Dr. Pratt we changed and expanded the paragraph:

"For an air mass sampled in the western United States and influenced by long-range transport and mineral dust, Pratt et al. (2009) observed biological particles to comprise a significant fraction of ice crystal residues. For ice nucleation measurements in the Amazon basin, Prenni et al. (2009) found biological particles to comprise a significant fraction of the ice nucleation-active particles, particularly at temperatures warmer than ~248 K. However, significant uncertainties remain in our understanding of the ice nucleation efficiencies of various bacteria, pollen, and other biological particles (Mohler et al 2007; DeMott and Prenni 2010). Further, it is likely that the relative importance of biological ice nuclei varies on regional and seasonal scales similar to that observed for biological aerosol (Burrows et al 2009; DeMott and Prenni 2010)."

Burrows, S. M.; Elbert, W.; Lawrence, M. G.; Poeschl, U.: Bacteria in the global atmosphere - Part 1: review and synthesis of literature data for different ecosystems, Atmos. Chem. Phys., 9, 9263-9280, doi:10.5194/acp-9-9263-2009, 2009.

DeMott, P. J.; Prenni, A. J.: New Directions: Need for defining the numbers and sources of biological aerosols acting as ice nuclei, Atm. Environ., 44, 1944-1945, 2010.

Möhler, O., DeMott, P. J., Vali, G., and Levin, Z.: Microbiology and atmospheric processes: the role of biological particles in cloud physics, Biogeosciences, 4, 1059-1071, 2007.

Pratt, K. A., DeMott, P. J., French, J. R., Wang, Z., Westphal, D. L., Heymsfield, A. J., Twohy, C. H., Prenni, A. J., and Praether, K. A.: In situ detection of biological particles in cloud ice-crystals, Nat. Geosci., 2, 398-401, 2009.

Prenni, A. J., Petters, M. D., Kreidenweis, S. M., Heald, C. L., Martin, S. T., Artaxo, P., Garland, R. M., Wollny, A. G., and Pöschl, U.: Relative roles of biogenic emissions and Saharan dust as ice nuclei in the Amazon basin, Nat. Geosci., 2, 402-405, 2009.

4) P 27223 ln 24. Why can droplet collisions 'ease nucleation events'?

ANSWER: The kinetic energy invested into the system by collision helps to surmount the activation barrier. A local increase in density of the supercooled liquid by impaction followed by sudden pressure release can enhance the number of ice-like clusters leading to crystallization (for nucleation by pressure relaxation see Cheftel et al., 2006).

Cheftel, J. C., Lévy, J., and Dumay, E.: Pressure-assisted freezing and thawing: principles and potential applications, Food Rev. Int., 16, 453-483, doi:10.1081/FRI-100102319, 2000.

5) Section 4. It would be helpful to include the isothermal data in the form a plot of fraction frozen (or unfrozen) verses time. Murray et al. (Atmos. Chem. Phys. 2011, 11, 4191-4207) showed plots such as this for kaolinite and Broadley et al. (Atmos. Chem. Phys. Discuss., 11, 22801–22856, 2011) showed results for NX – illite. The close to exponential decays in Murray et al. suggested the probability of all droplets freezing in a given time was approximately equal and that this was consistent with the single component stochastic model. Broadly et al found that for NX illite, a mixture of minerals thought to be representative of natural dust, the decay of liquid droplets is not exponential and indicated some droplets contained better ice nuclei than others. In which category does pollen fall? This is important because the expression used here (Eq 1) is a single component stochastic equation.

ANSWER: We added a plot with the raw data applied for our calculations to this response (see below). But we point out that our approach is an approximation of the equation presented by Murray et al. 2010, as we assumed a totally homogeneous distribution of IN among the droplets. We had to apply this approximation because of two reasons: First, we do not know the active IN surface per droplet, which is necessary to apply the formalism of Murray et al. 2011, and second, we directly want to compare heterogeneous nucleation results with homogeneous nucleation results (for which the heterogeneous formalism is inapplicable).

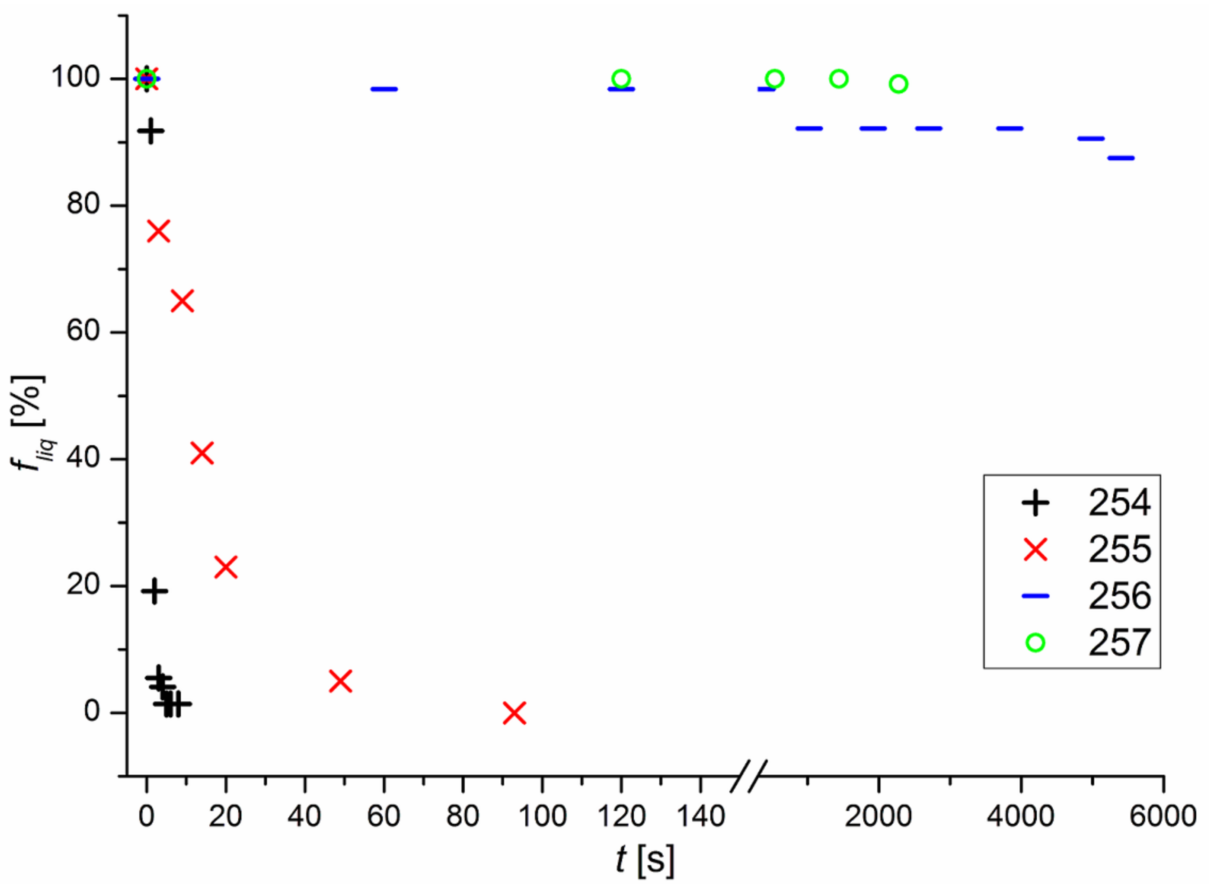

Figure 1. The dependence of f_{liq} (number fraction of liquid droplets) on time and temperature [K] for a sample of birch pollen grains in oil emulsion.

We thank Dr. K. Pratt for helpful discussion.