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Pollen grains are efficient cloud condensation nuclei

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Abstract

This letter presents a laboratory study investigating the ability of pollen grains to act as cloud condensation nuclei. The hygroscopicity of pollen is measured under subsaturated relative humidities using an electrodynamic balance. It is found, along with other results, that pollen exhibits bulk uptake of water under subsaturated conditions. Through the use of an environmental scanning electron microscope it was observed that the surface of pollen is wettable at high subsaturated humidities. The hygroscopic response of the pollen to subsaturated relative humidities is parametrized using κ-Köhler theory and values of the parameter κ for pollen are between 0.05 and 0.1. It is found that while pollen grains are only moderately hygroscopic, they can activate at critical supersaturations of 0.001% and lower, and thus pollen grains will readily act as cloud condensation nuclei. While the number density of pollen grains is too low for them to represent a significant global source of cloud condensation nuclei, the large sizes of pollen grains suggest that they will be an important source of giant cloud condensation nuclei. Low temperature work using the environmental scanning electron microscope indicated that pollen grains do not act as deposition ice nuclei at temperatures warmer than −15 °C.

Keywords: pollen, cloud condensation nuclei, CCN, hygroscopicity, electrodynamic balance, EDB

1. Introduction

Atmospheric particles influence the radiative forcing of the earth's atmosphere directly via scattering and absorbing solar and planetary radiation. In addition, aerosols can indirectly perturb the radiative balance by acting as cloud condensation nuclei (CCN) (Forster *et al* 2007). The concentration of aerosol dictates the concentration of droplets within clouds and hence the precipitation efficiency of the cloud (Rosenfeld *et al* 2008). Primary biological aerosols (PBAs) represent a significant fraction of the total aerosol burden; with the biosphere being a major source of primary aerosol (Jaenicke 2005, Huffman *et al* 2010, Wiedinmyer *et al* 2009). The number density of PBAs is insufficient to make a global impact on the direct aerosol effect. However, if pollen can act as CCN they would be sufficiently large to provide a disproportionate contribution to the development of precipitation within clouds by acting as coalescence embryos; also known as giant cloud condensation nuclei (GCCN) (Johnson 1982, Cotton and Yuter 2009).

PBAs found in the atmosphere include: pollen, bacteria, fungal, algae, moss and fern spores, viruses and fragments of animals and plants (Deguillaume *et al* 2008, Moller *et al* 2008, Després *et al* 2007, Möhler *et al* 2007). Pollen are produced by flowering flora and their purpose is to deliver the male gametes with which to pollinate the female plant. The largest pollen grain size is from the pumpkin family (∼200 μm) and the smallest pollen is from the forget-me-not (∼4 μm) (Wodehouse 1935). The density of pollen varies from species to species. Reported values for the density of the non-aqueous component of the aerosol vary between approximately 1000–2000 kg m^{-3} (Aylor 2002, Harrington and Metzger 1963, Laucks *et al* 2000, van Hout and Katz 2004). The size, shape and morphology of pollen are adapted, for the needs of each species, to allow for efficient pollination.

Anemophilous plants produce pollen which is wind delivered and entomophilous plants produce insect delivered pollen. Unsurprisingly, anemophilous pollen is more widely found in the atmosphere than entomophilous pollen. Pollen grains are designed to protect their genetic material cargo. To achieve this, they must be resistant to environmental stresses subjected to them during transport. The surface covering of pollen (exine) protects the pollen from injury due to excessive desiccation, photon damage and mechanical ageing. The exine is covered with waxes and proteins and therefore the pollen surface might be expected to be hydrophobic. The water composition of pollen is dependent upon the atmospheric relative humidity. At high humidity the pollen grain swells by taking up atmospheric water internally (Dingle 1966, Diehl *et al* 2001).

The presence of pollen in the atmosphere is dependent upon the pollination season of the investigated flora species. The dispersal and transport of pollen requires suitable meteorological conditions to produce uplift of air containing pollen. Atmospheric removal of pollen is determined by the wet and dry deposition rates. The dry deposition is controlled by the settling speed of the pollen grain which is a function of particle density and size (Aylor 2002). Both computer modelling and field studies have shown that pollen is capable of travelling large distances and can remain airborne on the order of days (Rousseau *et al* 2003, Sofiev *et al* 2006, Helbig *et al* 2004, Heise and Heise 1948).

The question of whether pollen can act as CCN was first asked by Dingle (1966), and the issue has recently been raised again in reviews concerned with PBAs and cloud activation (Moller *et al* 2008, Jaenicke 2005, Ariya *et al* 2009, Möhler *et al* 2007). Dingle indicates that pollen is hygroscopic at subsaturated relative humidities and that pollen grains are emitted wet during dehiscence (pollen emission) and are thus likely to be coated in a hygroscopic residue (Dingle 1966). Work by Diehl *et al* further demonstrated that pollen has a relative humidity dependent water uptake (Diehl *et al* 2001). The large size of pollen makes the possibility of them acting as CCN very likely (Dusek *et al* 2006), as long as the aerosol surface is wettable with water. If the pollen surface is hydrophobic even after internal water uptake it will not be able to act as CCN.

2. Methods

Four different types of anemophilous pollen were studied: daffodil (Narcissus spp.), water birch (Betula occidentalis), pussy willow (Salix caprea), and black walnut (Juglans nigra). The willow and daffodil pollen were freshly obtained from the Cambridge botanic gardens, and the walnut and birch were obtained from a commercial source (Sigma-Aldrich).

The hygroscopicity of the pollen samples were measured using the single particle electrodynamic balance (EDB) methodology. The critical supersaturation of aerosols can be estimated by the curvature of their subsaturated humidograms by κ-Köhler theory which parametrizes the humidogram using a one parameter (κ) fitting (Petters and Kreidenweis 2007). To investigate the wettability of the pollen surfaces an environmental scanning electron microscope (ESEM) was used. Hygroscopicity measurements, and κ-Köhler parametrizations, were performed on the birch, daffodil, and willow pollen samples. The ESEM technique was used to observe the birch and walnut specimens. The seasonal manual collection of the daffodil and willow samples precluded their study in the ESEM work which was performed after the pollination season.

2.1. Electrodynamic balance apparatus

The EDB is used to levitate single pollen particles, which contain a net charge, within a synthetic air environment at 295 K. The EDB design follows the double ring electrode architecture of Davis *et al* (Davis *et al* 1989), and a detailed description of the apparatus has been provided previously (Pope *et al* 2010a, 2010b). The changing mass of the particle can be followed by observing the electric force required to balance the weight of the particle. Neglecting forces other than the electric force and weight gives equation (1) where: m is the mass of the particle, g is the gravitational force at the earth's surface, V_{dc} is the balancing dc voltage, n is the number of elementary charges present on the particle, q is the elementary charge, z is the distance between the ring electrodes, and C is a geometric factor that depends on the shape of the EDB arrangement. Therefore the change in mass between time a and b can be followed by $V_a/V_b = m_a/m_b$. The data is typically represented as a humidogram where the change in mass is given as a mass growth factor, m/m_0, which represents the increase in mass when compared to the dry mass (m_0) acquired a 0% RH.

$$mg = (V_{dc}/z)nqC. \qquad (1)$$

The EDB measured the relative mass of the pollen particles under different subsaturated relative humidities (RH). The pollen particles were deposited into the EDB system by manually collecting pollen on the tip of a metal wire and subsequently tapping the wire above the EDB electrodes. Sufficient static charge was transferred from the wire to the particles to allow for electrodynamic trapping of the pollen. The EDB cell is thermally regulated by use of a chiller-circulator to pump coolant through veins in the EDB walls. The relative humidity of the cell was controlled by varying the ratio of dry and water-saturated air streams that were combined and passed into the EDB chamber. The accuracy of the relative humidity measurements are $\pm 1\%$. The water-saturated flow was achieved by passing air through a water bubbler held at constant temperature.

Labview software was used for experimental control and data acquisition. HPLC grade water (Rathbones) was used in the bubbler and synthetic air (BOC, zero grade) to generate the wet and dry flows. At least three individual experiments were conducted on each of the different pollen types.

2.2. Environmental scanning electron microscopy apparatus

The ESEM system (Phillips XL30 ESEM-FEG) was equipped with a Peltier cooled stage on which the sample discs holding the pollen could be placed. The pollen was placed on two different sample disc surfaces during the ESEM experiments: aluminium or aluminium with a thin hydrophobic polystyrene layer deposited upon it. The gaseous environment surrounding the particle is composed solely of water vapour, the pressure of which can be varied between 0.1 and 5 Torr. The environmental RH surrounding the particle was varied by changing the temperature of a fixed pressure gas surrounding the particle, which is assumed to be in thermal equilibrium with the Peltier device. Alternatively, the RH could be controlled by varying the gas phase water pressure at a set temperature. The electron beam accelerating voltage was either 5 or 10 kV.

2.3. κ-Köhler theory

The CCN ability of particles of known size and composition can be calculated using the Köhler equation which determines the vapour pressure over an aqueous droplet (Köhler 1936). The CCN ability is determined by two factors: the solute effect and the Kelvin effect. Use of the Köhler equation requires knowledge of the number of moles of solute within the droplet. Pollen grains are complicated biochemical systems and at present it is not possible to estimate the number of moles of solute. κ-Köhler theory has been developed that allows a relationship between a particles hygroscopicity and CCN ability to be established using a single hygroscopicity parameter κ (Petters and Kreidenweis 2007). The saturation ratio (S) over an aqueous droplet can be calculated from equation (2), where $p_w(D)$ is the water vapour pressure over a droplet of diameter D, and p° is the water vapour pressure over a flat surface at the same temperature T. a_w is the activity of water in a solution, $\sigma_{s/a}$ is the surface tension of the particle, M_w is the molecular weight of water, R is the gas constant, and ρ_w is the density of water. The exponential term is the Kelvin effect which is negligible for particles over 100 nm. Therefore for pollen the Kelvin effect is neglected and the saturation ratio is equal to the water activity. At equilibrium the activity of water in air is equal to the activity of the water associated with the pollen. The relative humidity describes the activity of atmospheric water as a percentage (RH $= a_w \times 100\%$).

$$S = \frac{p_w(D)}{p^\circ} = a_w \exp\left(\frac{4\sigma_{s/a} M_w}{RT\rho_w D}\right). \qquad (2)$$

The single parameter κ is defined in equation (3), where V_w and V_p are the volume of water in the particle and the volume of dry particulate matter respectively. The total volume of the particle, V_T, is then given by the sum of V_p and V_w.

$$a_w = \frac{V_w}{V_w + \kappa V_p}. \qquad (3)$$

Equation (3) can be rearranged into equation (4), with the knowledge that mass equals volume multiplied by density. Where m is the wet aerosol mass (RH $> 0\%$) and m_0 is the aerosol mass when dry (RH $= 0\%$), the mass of water associated with the wet aerosol is given by $m - m_0$. The density of the dry particle and water are given by ρ_w and ρ_p, respectively.

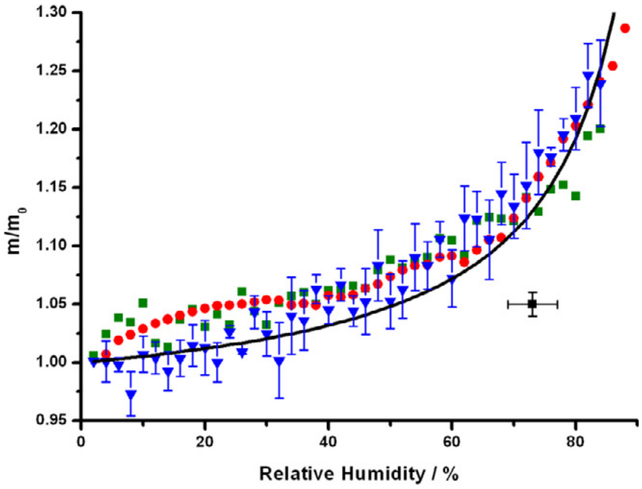

Figure 1. Humidograms of three pollen types measured by the EDB apparatus. Symbols: red circle—Salix caprea, blue triangle—Betula occidentalis, green square—Narcissus sp., black square—Betula alba (Diehl *et al* 2001). Line: modelled average humidogram of the three pollen types using κ-Kohler theory (Petters and Kreidenweis 2007). For clarity error bars (1σ) are only included for the Betula pollen species. The associated errors with the Narcissus and Salix pollen are similar to the Betula occidentalis.

$$a_w = \frac{\left(\frac{m-m_0}{\rho_w}\right)}{\left(\frac{m-m_0}{\rho_w}\right) + \left(\frac{\kappa m_0}{\rho_p}\right)} = \frac{\left(\frac{m/m_0-1}{\rho_w}\right)}{\left(\frac{m/m_0-1}{\rho_w}\right) + \left(\frac{\kappa}{\rho_p}\right)} = \frac{(m/m_0 - 1)}{(m/m_0 - 1) + \left(\frac{\rho_w}{\rho_p}\kappa\right)}. \qquad (4)$$

The value for κ can obtained by nonlinear curve fitting of the pollen humidograms (m/m_0 versus a_w) using a single parameter equal to $\kappa\rho_w/\rho_p$. Typical values of κ found within the atmosphere are in the range $0.1 < \kappa < 0.9$, with highly hygroscopic particles such as sea salt having values of $\kappa = 1.4$ (Petters and Kreidenweis 2007). As κ approaches zero the particle becomes non-hygroscopic. However, non-hygroscopic particles can still become activated at sufficiently large diameters as long as the surface is wettable. The critical droplet diameter, at which point the particle becomes activated, occurs when the saturation ratio goes through a maximum. This point is referred to as the critical supersaturation (s_c), where supersaturation is defined as $S - 1$.

3. Results and discussion

The average pollen humidograms for each of the three species (Betula, Narcissus, and Salix) investigated in the EDB are shown in figure 1. Each pollen species showed enhanced mass growth, of similar magnitude, at all RH greater than 0%. At 75% RH an average value for the mass increase, due to water uptake, was 16%. There are no significant differences between the mass growth curves measured under increasing RH when compared to the mass growth curves measured under shrinking RH. Hence there is no hysteresis observed within the humidogram. A preliminary hygroscopic measurement of a

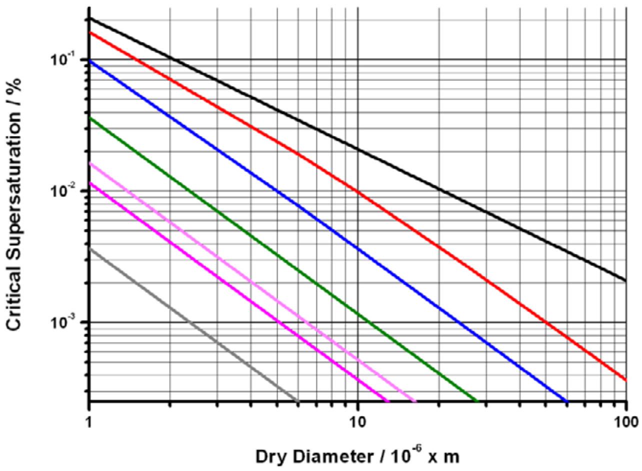

Figure 2. Diameter dependent critical supersaturations for CCN activation using different values of the kappa (κ) parameter. Density of dry particle is assumed to be 1000 kg m^{-3}. Lines: black—$\kappa = 1 \times 10^{-10}$ (wettable but non-hygroscopic), red—$\kappa = 0.0001$, blue—$\kappa = 0.001$, green—$\kappa = 0.01$, pink—$\kappa = 0.05$, magenta—$\kappa = 0.1$, grey—$\kappa = 1.0$.

different birch species (B. alba) was previously measured at 73 ± 4% RH showing a mass growth between 4 and 6% as shown in figure 1 (Diehl *et al* 2001).

The average hygroscopic data for all three pollen species, shown in figure 1, gives a best fit to the value for the single parametrization of $\kappa\rho_w/\rho_p = 0.048 \pm 0.002$. The use of the lower limit of pollen density (1000 kg m^{-3}) gives a kappa parameter value of $\kappa = 0.05$, and the upper limit of pollen density (2000 kg m^{-3}) gives a kappa parameter value of $\kappa = 0.10$. This range of κ indicates a low, but non-negligible, hygroscopicity of the bulk pollen sample. Figure 2 indicates that this κ value gives a lower limit for the critical supersaturation of pollen at ∼0.002%. This value assumes the lower density limit and the smallest sized pollen grain found in nature (4 μm). For larger pollen, such as used in this study, the required supersaturations are even lower. This upper limit for the critical supersaturation indicates that pollen grains could be very effective CCN. However, the activation to CCN will only occur if the surface of the pollen grain is hygroscopic and hence wettable.

The ESEM investigation on the Betula and Juglans pollen revealed that the Betula pollen is roughly spherical in shape with a diameter of approximately 25 μm. The Juglans pollen is more spheroidal in shape with a long diameter of 40 μm and short diameter of 30 μm. Both types of pollen possess prominent pores upon the grain surface. Experiments were conducted at temperatures above 0 °C to investigate the relative humidity dependent growth of pollen. In particular, observations were required to understand whether the pollen hygroscopicity was due to internal swelling and/or external uptake of water upon the pollen surface. Figure 3 shows the growth of a birch pollen grain under increasing humid conditions. It can be seen that between frame (A)–(D) (68%–93% RH) that the pollen grain swells internally, but there is no obvious sign of water uptake upon the pollen surface. At relative humidities greater than 95%, shown in frames E and F, liquid water can clearly be observed upon the particle surface. The choice of ESEM sample disc surface, aluminium or polystyrene coated aluminium, on which the pollen was deposited made no difference to the observations. The onset of surface water uptake was often observed to be initiated by water droplets forming over the pollen pores as shown in figure 4.

Three individual pollen particles from each of the Juglans and Betula species were investigated within the ESEM at temperatures down to −15°C. Within the temperature range investigated no depositional freezing of the pollen grains was observed. This is consistent with the observations of Diehl *et al* (2001). Hence the two pollen species investigated are not efficient ice nuclei in the depositional mode of freezing. However, further work by Diehl *et al* indicated that different modes of freezing are important for pollen.

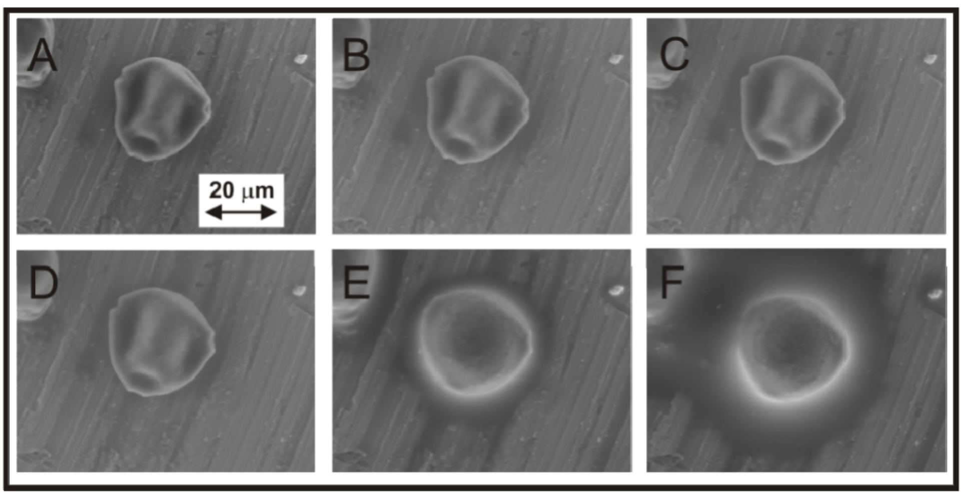

Figure 3. ESEM images of Betula occidentalis pollen at different relative humidities. Panels: (A)—68%, (B)—84%, (C)—91%, (D)—93%, (E)—95%, (F)—96%. The temperature of the sample was in the range 1.2–6 °C.

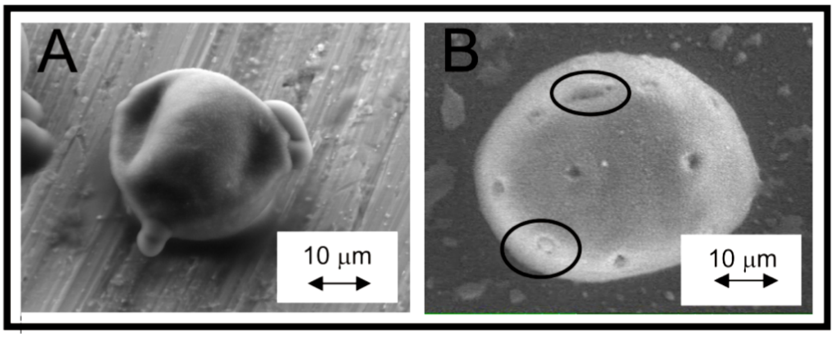

Figure 4. ESEM images of both Betula occidentalis (A) and Juglans nigra (B) showing initial uptake of water at pore sites. The image quality of the Juglans nigra sample is poorer because it was caught within the video mode of the ESEM, and hence the image capture time was shorter. The circles highlight the wetted pores on the Juglans sample.

4. Conclusions

The median supersaturation found within small or moderate cumulus clouds is approximately 0.1% (Pruppacher and Klett 1997). However, much higher supersaturations can be found under different cloud regimes. For pollen to act as GCCN and have a significant effect on cloud precipitation efficiency they need to have concentrations of $\sim 10^{-3}$ cm^{-3} (Cotton and Yuter 2009). This concentration is likely to be achieved by many pollen types during the spring pollination season. For example, Birch is a widespread Northern European tree species which produces pollen that can travel distances of $\sim 10^3$ km during dispersal. Furthermore, peak Birch pollen grain concentrations have been measured as high as 2–3000 pollen grains m^{-3} (Sofiev *et al* 2006). This work shows that whilst the volume averaged hygroscopicity of pollen is low; pollen will still act as effective CCN under very low supersaturations. During pollen seasons this extra source of GCCN is likely to be important for cloud processes. This result adds much impetus for further research into pollen and cloud interactions. In particular modelling studies need to describe how local meteorology affects pollen release mechanisms and the subsequent dispersion of the pollen within the atmosphere.

The observation of initial wetting of the pollen occurring on the pores of the pollen is intriguing. It is hypothesized that the small pore size (approximately 1 μm diameter at the opening) reduces the vapour pressure in this local region and hence capillary condensation occurs. Another hypothesis is that the initial uptake of water at the pores could be due to a difference in chemical composition within the pores compared to the rest of the particle surface. If the pore composition is more hydrophilic, there would be preferential water uptake upon these sites compared to the rest of the surface. Once the pores are filled with water then the rest of the surface is wetted. Further laboratory work is now being initiated within the laboratory to test these hypotheses.

No evidence was observed for deposition freezing to occur upon pollen (Juglans nigra and Betula occidentalis) at temperatures down to -15 °C and hence at these temperatures pollen does not form depositional ice nuclei. Previously, pollen grains have been shown to be efficient ice nuclei when the condensational, immersion and contact freezing modes have been available (von Blohn *et al* 2005, Diehl *et al* 2001, 2002).

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References

Ariya P A, Sun J, Eltouny N A, Hudson E D, Hayes T D and Kos G 2009 Physical and chemical characterisation of bioaerosols—implications for nucleation processes *Int. Rev. Phys. Chem.* **28** 1–32

Aylor D E 2002 Settling speed of corn (Zea mays) pollen *J. Aerosol Sci.* **33** 1601–7

Cotton W R and Yuter S 2009 *Aerosol Pollution Impact on Precipitation* ed Z Levin and W R Cotton (New York: Springer)

Davis E J, Buehler M F and Ward T L 1989 The double-ring electrodynamic balance for microparticle characterization *Rev. Sci. Instrum.* **61** 1281–8

Deguillaume L, Leriche M, Amato P, Ariya P A, Delort A M, Pöschl U, Chaumerliac N, Bauer H, Flossmann A I and Morris C E 2008 Microbiology and atmospheric processes: chemical interactions of primary biological aerosols *Biogeosciences* **5** 1073–84

Després V R, Nowoisky J F, Klose M, Conrad R, Andreae M O and Pöschl U 2007 Characterization of primary biogenic aerosol particles in urban, rural, and high-alpine air by DNA sequence and restriction fragment analysis of ribosomal RNA genes *Biogeosciences* **4** 1127–41

Diehl K, Matthias-Maser S, Jaenicke R and Mitra S K 2002 The ice nucleating ability of pollen: part II. Laboratory studies in immersion and contact freezing modes *Atmos. Res.* **61** 125–33

Diehl K, Quick C, Matthias-Maser S, Mitra S K and Jaenicke R 2001 The ice nucleating ability of pollen: part I. Laboratory studies in deposition and condensation freezing modes *Atmos. Res.* **58** 75–87

Dingle A N 1966 Pollens as condensation nuclei *J. Rech. Atmos.* **2** 231–7

Dusek U *et al* 2006 Size matters more than chemistry for cloud-nucleating ability of aerosol particles *Science* **312** 1375–8

Forster P *et al* 2007 *Climate Change 2007: The Physical Science Basis. Contribution of Working Group I to the Fourth Assessment Report of the Intergovernmental Panel on Climate Change* ed S Solomon *et al* (Cambridge: Cambridge University Press)

Harrington J B and Metzger K 1963 Ragweed pollen density *Am. J. Bot.* **50** 532–9

Heise H A and Heise E R 1948 The distribution of ragweed pollen and alternaria spores in the upper atmosphere *J. Allergy* **19** 403–7

Helbig N, Vogel B, Vogel H and Fiedler F 2004 Numerical modelling of pollen dispersion on the regional scale *Aerobiologia* **3** 3–19

Huffman J A, Treutlein B and Pöschl U 2010 Fluorescent biological aerosol particle concentrations and size distributions measured with an ultraviolet aerodynamic particle sizer (UV-APS) in central Europe *Atmos. Chem. Phys.* **10** 23215–33

Jaenicke R 2005 Abundance of cellular material and proteins in the atmosphere *Science* **308** 73

Johnson D B 1982 The role of giant and ultragiant aerosol particles in warm rain initiation *J. Atmos. Sci.* **39** 448–60

Köhler H 1936 The nucleus in and the growth of hygroscopic droplets *Trans. Faraday Soc.* **32** 1152–61

Laucks M L, Roll G, Schweiger G and Davis E J 2000 Physical and chemical (Raman) characterization of bioaerosols—pollen *J. Aerosol. Sci.* **31** 307–19

Möhler O, DeMott P J, Vali G and Levin Z 2007 Microbiology and atmospheric processes: the role of biological particles in cloud physics *Biogeosciences* **4** 1059–71

Moller B, Rarey J and Ramjugernath D 2008 Estimation of the vapour pressure of non-electrolyte organic compounds via group contributions and group interactions *J. Mol. Liq.* **143** 53–63

Petters M D and Kreidenweis S M 2007 A single parameter representation of hygroscopic growth and cloud condensation nucleus activity *Atmos. Chem. Phys.* **7** 1961–71

Pope F D, Dennis-Smither B J, Griffiths P T, Clegg S L and Cox R A 2010a Studies of single aerosol particles containing of malonic acid, glutaric acid, and their mixtures with sodium chloride. I. Hygroscopic growth *J. Phys. Chem.* A **114** 5335–41

Pope F D, Tong H-J, Dennis-Smither B J, Griffiths P T, Clegg S L, Reid J P and Cox R A 2010b Studies of single aerosol particles containing malonic acid, glutaric acid, and their mixtures with sodium chloride. II. Liquid-state vapor pressures of the acids *J. Phys. Chem.* A **114** 10156–65

Pruppacher H R and Klett J D 1997 *Microphysics of Clouds and Precipitation* (Dordrecht: Kluwer)

Rosenfeld D, Lohmann U, Raga G B, O'Dowd C D, Kulmala M, Fuzzi S, Reissell A and Andreae M O 2008 Flood or drought: how do aerosols affect precipitation? *Science* **321** 1309–13

Rousseau D-D, Duzer D, Cambon G, Jolly D, Poulsen U, Ferrier J, Schevin P and Gros R 2003 Long distance transport of pollen to Greenland *Geophys. Res. Lett.* **30** 1765

Sofiev M, Siljamo P, Ranta H and Rantio-Lehtimäki A 2006 Towards numerical forecasting of long-range air transport of birch pollen: theoretical considerations and a feasibility study *Int. J. Biometeorol.* **50** 392–402

van Hout R and Katz J 2004 A method for measuring the density of irregularly shaped biological aerosols such as pollen *J. Aerosol Sci.* **35** 1369–84

von Blohn N, Mitra S K, Diehl K and Borrmann S 2005 The ice nucleating ability of pollen: part III. New laboratory studies in immersion and contact freezing modes including more pollen types *Atmos. Res.* **78** 182–9

Wiedinmyer C, Bowers R M, Fierer N, Horanyi E, Hannigan M, Hallar A G, McCubbin I and Baustian K 2009 The contribution of biological particles to observed particulate organic carbon at a remote high altitude site *Atmos. Environ.* **43** 4278–82

Wodehouse R P 1935 *Pollen Grains: Their Structure, Identification and Significance in Science and Medicine* (New York: McGraw-Hill)