



RESEARCH LETTER

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Key Points:

- Pollen grains can rupture when wet to form submicron subpollen particles (SPP)
- Laboratory experiments show that SPP are hygroscopic and can act as CCN
- Pollen grains may contribute to CCN in northern midlatitudes

Supporting Information:

- Text S1 and Figures S1–S3

Correspondence to:

A. L. Steiner,
alsteiner@umich.edu

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Pollen as atmospheric cloud condensation nuclei

Allison L. Steiner¹, Sarah D. Brooks², Chunhua Deng², Daniel C. O. Thornton³, Michael W. Pendleton⁴, and Vaughn Bryant⁵

¹Department of Atmospheric, Oceanic and Space Sciences, University of Michigan, Ann Arbor, Michigan, USA, ²Department of Atmospheric Sciences, Texas A&M University, College Station, Texas, USA, ³Department of Oceanography, Texas A&M University, College Station, Texas, USA, ⁴Microscopy and Imaging Center, Texas A&M University, College Station, Texas, USA, ⁵Palynology Laboratory, Texas A&M University, College Station, Texas, USA

Abstract Anemophilous (wind-dispersed) pollen grains are emitted in large quantities by vegetation in the midlatitudes for reproduction. Pollen grains are coarse particles (5–150 μm) that can rupture when wet to form submicron subpollen particles (SPP) that may have a climatic role. Laboratory CCN experiments of six fresh pollen samples show that SPP activate as CCN at a range of sizes, requiring supersaturations from 0.81 (± 0.07)% for 50 nm particles, 0.26 (± 0.03)% for 100 nm particles, and 0.12 (± 0.00)% for 200 nm particles. Compositional analyses indicate that SPP contain carbohydrates and proteins. The SPP contribution to global CCN is uncertain but could be important depending on pollen concentrations outside the surface layer and the number of SPP generated from a single pollen grain. The production of hygroscopic SPP from pollen represents a novel, biologically driven cloud formation pathway that may influence cloud optical properties and lifetimes, thereby influencing climate.

1. Introduction

Pollen grains are released from terrestrial vegetation in large quantities to transmit the male genetic material for reproduction [Faegri and Iversen, 1989]. These coarse particles range in diameter from 5–150 μm with most airborne pollen ranging from 20 to 45 μm [Linder, 2000; Emberlin, 2008]. In the midlatitudes, vegetation disperses pollen through dominantly anemophilous mechanisms [Lewis et al., 1983], with different vegetation types delivering pulses of pollen grains into the atmosphere at varying times of the year. These pollen emissions are closely tied to meteorological (wind and relative humidity) and climatological (phenology and soil moisture) factors [Weber, 2003]. Aerobiology studies indicate that pollen can be transported up to 100–1000 km [Schueler and Schluenzen, 2006; Sofiev et al., 2006] and pollen grains have been observed both in the atmospheric boundary layer and free troposphere [Noh et al., 2013].

Prior studies have suggested that primary biological material such as pollen could alter cloud properties [Sun and Ariya, 2006; Moehler et al., 2007; Poeschl et al., 2010; Pope, 2010], and high ambient concentrations of biological particles are linked to rainfall events [Huffman et al., 2013]. Small, hygroscopic atmospheric particles (diameters < 200 nm) are known as cloud condensation nuclei (CCN) and provide a surface for the condensation of water vapor, enabling the growth of cloud droplets. Laboratory measurements indicate that whole pollen grains have the ability to act as CCN [Pope, 2010] and ice nuclei [Diehl et al., 2001; von Blohn et al., 2005] but to date, pollen has not been considered climatically relevant due to its large size and short residence time in the atmosphere. However, under high humidity or moist conditions, whole pollen grains can rupture and expel cytoplasmic material, forming smaller particles known as subpollen particles (SPP) [Taylor et al., 2002; Grote et al., 2003; Taylor et al., 2004]. SPP sizes range from several nanometers to several micrometers and are typically generated within a few minutes after contact with water [Suphioglu et al., 1992; Grote et al., 2001; Taylor et al., 2002]. These SPP are thought to be primarily composed of starch [Suphioglu et al., 1992; Laurence et al., 2011] and could be an additional source of organic carbon to the atmosphere. Recently, SPP have been shown to act as ice nuclei [O'Sullivan et al., 2015]. Here we provide laboratory evidence that SPP can act as CCN, providing a new pathway for a coupled vegetation-climate feedback through the production of small CCN-active material from pollen grain rupture.

2. Pollen Sampling and SPP Generation

Viable pollen samples were collected from six common anemophilous genera in the United States: (1) *Quercus polymorpha* Schltdl. and Cham. (oak; Austin, TX; intact grain in Figure 1a), (2) *Carya illinoensis*

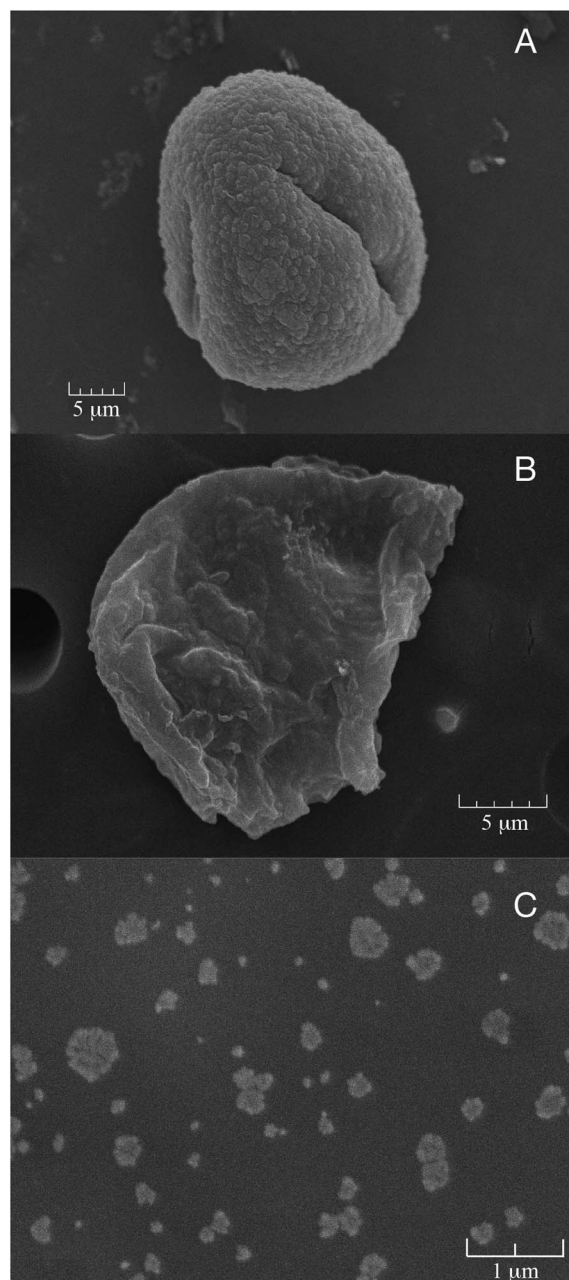


Figure 1. SEM images of (a) dry *Quercus polymorpha* coarse pollen grain, (b) ruptured *Quercus* grain present in solution, and (c) *Quercus* subpollen particles (SPP).

confirmed with SEM analysis (Figure 1b). Dry SPP samples were intercepted from the DMA exit stream by impaction on an aluminum foil impactor stage. Figure 1c shows the SPP size selected at 200 nm, where particles range from 50 to 300 nm likely due to impaction onto the SEM sampling stage and may be agglomerated or aggregated due to the sample collection technique. Together, this analysis shows that the laboratory technique can rupture whole pollen grains and produce SPP.

3. CCN Ability of SPP

CCN ability of SPP was analyzed by counting the total number of particles (or condensation nuclei; CN) and the number of particles that activate into cloud droplets (CCN) at specified saturation values. The CCN/CN

(Wangenh.) *K. Koch* (pecan; Austin, TX), (3) *Betula occidentalis* Hook (birch; Palo Alto, CA), (4) *Juniperus virginiana* L. (cedar; Norman, OK), (5) *Pinus taeda* L. (pine; Houston, TX), and (6) *Ambrosia trifida* L. (ragweed; College Station, TX). Full catkins (samples 1–3) or cones (samples 4 and 5) from flowering trees were collected. *Ambrosia trifida* (Giant ragweed; College Station, TX) was collected in 2012 by vacuum and stored as dry pollen. Organic pollen solutions were prepared by placing 2 g of pollen material (i.e., the entire catkin or cone) in 100 mL ultrahigh purity (UHP) water and immediately agitated [Pummer *et al.*, 2012]. Samples were allowed to soak in solution for 1 h, followed by filtering (Whatman 595 Paper filter; 4–7 μm pore size) to remove any large pollen particles. The resulting organic solution was atomized (TSI 3079 Atomizer Aerosol Generator) with a sample flow rate to generate approximately 2000–3000 particles cm⁻³. Atomized droplets were passed through three desiccators in series and sent to a Differential Mobility Analyzer (DMA; TSI Model 3080) to select particles (SPP) sized at 50, 100, and 200 nm diameters.

A Scanning Electron Microscope (SEM; JEOL JSM-6400 operating at 15 kV accelerating voltage and 15 mm working distance) was used to confirm the transformation from whole pollen grain to SPP. Intact coarse pollen grains (Figure 1a) were imaged from original live samples collected in the field. To observe the ruptured pollen grains, the organic solution was dried and passed through a series of 50% ethanol, 100% ethanol, 50% HMDS (hexamethyldisilazane), and 100% HMDS solutions on 13 mm polycarbonate membrane filters (5 μm pore size) mounted in a Swinney filter apparatus to ensure that rupture occurred in solution and not by surface tension forces during air drying. The rupture of intact grains is

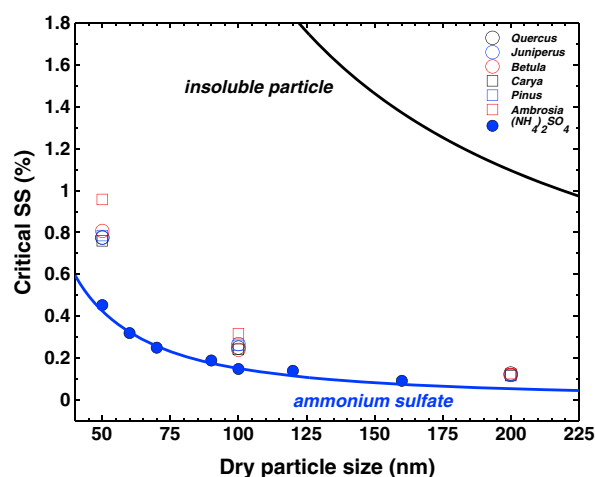


Figure 2. Critical supersaturation (S_c ; %) of six pollen types at three dry particle sizes (50, 100, and 200 nm) based on laboratory cloud condensation experiments. A theoretical range of S_c from very hygroscopic particle (blue line; ammonium sulfate ($(\text{NH}_4)_2\text{SO}_4$)) to an insoluble particle (black line) is shown, with $(\text{NH}_4)_2\text{SO}_4$ instrument calibration (blue circles).

CCN calibrations were performed using a range of size-selected aerosols generated from 0.1 wt% solution of $(\text{NH}_4)_2\text{SO}_4$ [Ma et al., 2013]. Within the CCN counter, SPP were exposed to a series of supersaturations with respect to water (Figure S1 in the supporting information). Each supersaturation stage was sampled for 6 min (200 nm) or 10 min (50 and 100 nm), and the concentration of particles that activated as CCN was determined using time averages of the 3–4 min and 5–8 min windows, respectively. In all cases, CCN/CN ratios were normalized to the maximum CCN/CN ratio observed for each pollen type and size. This relationship was then fit with a nonlinear function (MATLAB) to represent the transition to activation, and the S_c value for a SPP type and size was determined by the value of supersaturation when $\text{CCN}/\text{CN} = 0.5$.

For 200 nm particles, the average S_c was $0.12 (\pm 0.003)$ with little variation between pollen types (Figure 2). The average S_c was $0.26 (\pm 0.03)$ for 100 nm particles as compared to a value of 0.08 for ammonium sulfate, a standard reference material to describe highly efficient cloud droplet growth. The smallest particles (50 nm) exhibit the greatest variation in S_c , with most pollen types between 0.76 and $0.80 (\pm 0.02)$ and slightly higher S_c for *Ambrosia* (0.96). Errors on the nonlinear fit that determine S_c are relatively small, ranging from 0.00 to 0.03 (0.2–5.8%) of the S_c value. Due to the known size effect, larger particles have lower S_c than smaller particles. Among the six pollen types, similar size samples show little change in S_c with plant type (Figure 2). Recent studies suggest that biological nanoparticles may be important for the ice cloud nucleation [O'Sullivan et al., 2015]. Particles smaller than those sampled here (e.g., < 50 nm) are likely to be hygroscopic but based on the qualitative agreement of our observed size trend with Köhler theory, smaller particles would require supersaturations unlikely to occur in the atmosphere (>1%) and therefore are not atmospherically relevant.

We compare these results to two theoretical bounds on S_c : a lower limit for ammonium sulfate ($(\text{NH}_4)_2\text{SO}_4$), a very hygroscopic material and an upper limit for an insoluble, wettable particle such as dust. For ammonium sulfate (lower curve on Figure 2), S_c is calculated as in Seinfeld and Pandis [2006] and compared with the ammonium sulfate calibration. For an insoluble wettable particle such as dust (upper curve on Figure 2), droplets will form on the particles when the ambient supersaturation reaches the equilibrium vapor pressure and this upper limit is calculated with the Kelvin equation [Rogers and Yau, 1989]. Critically, this comparison shows the organic pollen solution generated small particles (50–100 nm) that are CCN active at supersaturations frequently encountered in the atmosphere.

4. Compositional Analysis of SPP

As cloud activation potential depends on particle composition and the solute effect, we investigated the chemical compounds present in SPP. While pollen walls (exines) are composed of a suite of proteins, lipids,

ratio determines the critical supersaturation (S_c), or the point at which the growing droplet is in equilibrium with the atmosphere and can spontaneously grow to form a cloud droplet. Atomized and desiccated droplets were sized at 50, 100, and 200 nm diameter with the DMA. While particles smaller than 50 nm have been shown to exhibit different thermodynamic properties than larger particles due to phase separation [Cheng et al., 2015], it is unlikely that changes in phase can compensate for reduced size, a significant disadvantage for CCN activation. Therefore we focus on the 50, 100, and 200 nm size particles as they are most relevant for CCN in the atmosphere [Dusek et al., 2006]. Outflow from the DMA was then split between a CCN counter (Droplet Measurement Technologies CCN-100) and a condensation particle counter (TSI-3010).

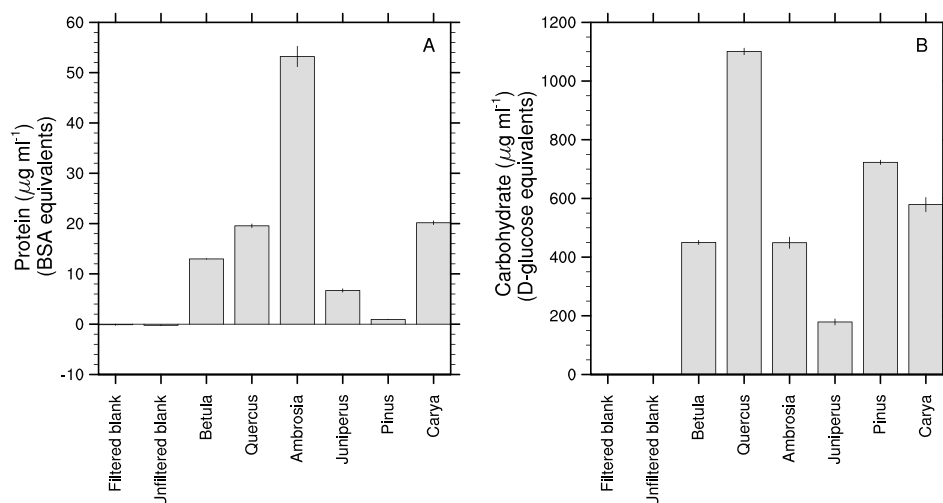


Figure 3. (a) Protein and (b) carbohydrate concentrations for the organic solutions based on the six pollen types (*Betula*, *Quercus*, *Ambrosia*, *Juniperus*, *Pinus*, and *Carya*). The pollen concentration in each sample was 10 g L^{-1} and error bars show mean \pm standard deviation ($n = 3$).

chromophores, and sporopollenin, the majority of the cytoplasmic material is starch [Laurence *et al.*, 2011]. To understand the potential chemical components responsible for CCN activity of the particles, SPP composition was evaluated with two methods. First, we determined the composition of SPP (sized at 200 nm; Figure 1c) with an electron microscope equipped with an Energy Dispersive X-Ray Spectrometer (EDS; IXRF Sedona SD Model SDD 3030-300C+). We collected 200 nm size SPP of three of the six pollen types (*Quercus*, *Pinus*, and *Carya*) and coated the SPP with an iodine vapor coating, as starch has the ability to exclusively absorb iodine vapor [Ellis and Pendleton, 2007]. The uptake of iodine by starch-containing *Quercus* samples is shown in Figure S2, with peaks in the 3.5–4.5 keV range. Other detected elements include carbon and oxygen (both present as organic material from the pollen grain), gold (used to coat the particles for improved visualization), and aluminum (the detection of the aluminum foil surface below the particle).

The second method analyzed the presence of protein and carbohydrates in the organic solution. Protein concentrations were measured using a spectrophotometer by the “Bradford assay” [Bradford, 1976] and the calibration curve was derived by dissolving bovine serum albumin (BSA) in UHP water, resulting in units of BSA equivalents. The calibration curve was linear ($r^2 = 0.995$) over the range $0\text{--}10 \mu\text{g protein mL}^{-1}$, with a limit of detection of $0.24 \mu\text{g protein mL}^{-1}$. Carbohydrate concentrations were measured using a spectrophotometer by the phenol sulfuric acid (PSA) method calibrated against D-glucose dissolved in UHP water [Dubois *et al.*, 1956; Thornton *et al.*, 2010]. The calibration curve was linear ($r^2 = 0.992$) over the range $0\text{--}80 \mu\text{g carbohydrate mL}^{-1}$, with a limit of detection of $0.21 \mu\text{g carbohydrate mL}^{-1}$. These analyses of the organic pollen solution found high concentrations of protein and carbohydrates (Figure 3), similar to Raman spectroscopy studies of organic pollen solutions [Pummer *et al.*, 2013]. Carbohydrate concentrations varied between the samples, with high relative magnitudes in *Quercus* followed by *Pinus*, *Carya*, *Betula*, and *Ambrosia*. Protein concentrations also varied between species, with the highest protein content in *Ambrosia*, *Carya*, *Betula*, and *Quercus*. Together, the EDS and chemical assays qualitatively indicate that SPP are composed of a large suite of biological macromolecules, including starch, other carbohydrates, and proteins. Field observations have observed similar organic components in-cloud droplets [Yttri *et al.*, 2007]. The observed size trend in CCN activation broadly follows Köhler theory, which implies that the composition of the particles is not likely changing with size.

5. Discussion and Conclusions

We propose the following mechanism for the ability of pollen grains to act as CCN: when pollen grains lofted into the atmosphere encounter sufficient moisture (e.g., either through processing within low clouds or if ambient atmospheric relative humidity is sufficiently high), the grains rupture and SPP composed primarily of starch are released into cloud droplets. We assume that the majority of pollen wetting and rupture will

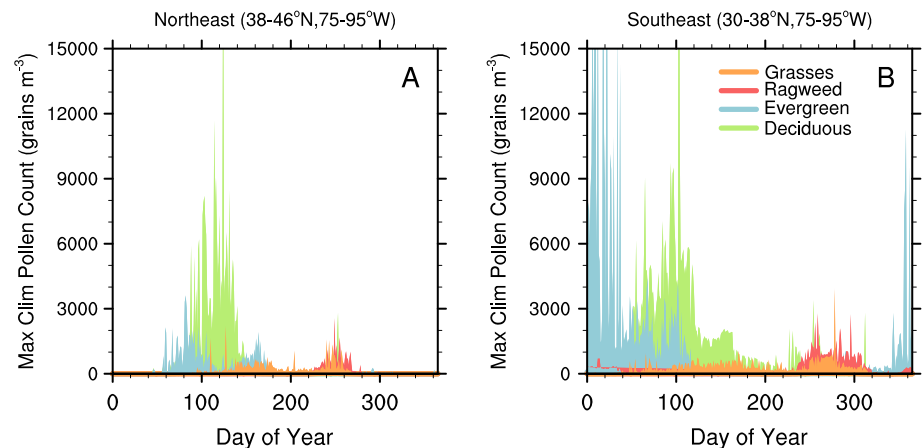


Figure 4. Climatological (2003–2010) surface maximum pollen counts (grains m^{-3}) from American Academy of Allergy, Asthma and Immunology (AAAAI) stations for grasses (orange), ragweed (red), evergreen (blue) forest, and deciduous broadleaf (green) forest over the (a) Northeastern United States (maximum concentrations based on 40 stations) and (b) the Southeastern United States (maximum concentrations based on 34 stations).

occur in cloud and that pollen grain rupture times are sufficiently fast to allow for in-cloud processing. Additional testing of *Quercus* pollen time in solution (ranging from 20 min to 7 h) had no effect on the S_c (Figure S3). After atmospheric drying, we assume that each droplet forms at least one SPP. Therefore, during pollen season, the cloud processing of pollen grains releases new small particles that can remain in and potentially alter the atmospheric environment.

The remaining question is whether pollen grains and resulting SPP can be a significant source of CCN in the atmosphere. While CCN closure studies can generally account for observed CCN number concentrations within a factor of 2 [Ervens *et al.*, 2010; Moore *et al.*, 2013], organic material of indeterminate origin can account for up to half of the total CCN number [Hitzenberger *et al.*, 1999]. To determine the potential quantity of pollen that can act as CCN, we use surface pollen count observations over the United States as provided by the American Association for Allergy Asthma and Immunology (AAAAI; stations utilized are listed in Text S1). Observed pollen counts are grouped into four plant functional types including (1) deciduous broadleaf trees and shrubs (including counts of *Acer*, *Alnus*, *Betula*, *Carpinus/Ostrya*, *Carya*, *Celtis*, *Fagus*, *Fraxinus*, *Juglans*, *Liquidambar*, *Morus*, *Platanus*, *Populus*, *Quercus*, *Salix*, and *Ulmus*), (2) evergreen trees (Cupressaceae, Pinaceae, *Pseudotsuga*, and *Tsuga*), (3) herbs, weeds, and grasses (*Artemisia*, Asteraceae, Amaranthaceae/Chenopodiaceae, Gramineae/Poaceae, *Plantago*, *Rumex*, and Urticaceae), and (4) ragweed (*Ambrosia*). Climatological averages of the four plant functional types were calculated for the Northeastern United States and Southeastern United States by averaging over multiple stations and multiple years, with the maximum climatological value for each category shown in Figure 4. Depending on land cover and meteorological conditions, atmospheric pollen counts reach up to several thousand grains per cubic meter (m^{-3}) of air during peak pollination season. The Northeastern US (38–46°N, 75–95°W) has the strongest contribution from the deciduous broadleaf trees such as *Quercus*, *Carya*, and *Betula* in April and May with peak spring pollen counts at some locations of 6000–12,000 grains m^{-3} (Figure 4a). In the Southeastern US (30–38°N, 75–95°W), maximum pollen counts reach up to 15,000 grains m^{-3} from evergreen trees *Juniperus* in the winter, followed by a deciduous peak of 5000–10,000 grains m^{-3} in early spring (Figure 4b). Lower quantities of grass and herb pollen are emitted ubiquitously throughout the summer (approximately 1000 grains m^{-3}), while *Ambrosia* peaks in the late summer and early fall with concentrations reaching up to 3000 grains m^{-3} . If we assume that a pollen count of 10,000 grains m^{-3} ruptures at the rate of approximately 1000 SPP to 1 pollen grain [Suphioglu *et al.*, 1992], this would result in approximately 10 CCN cm^{-3} . Using observed pollen counts from the atmospheric surface layer, the formation of 10^5 – 10^6 SPP per grain would be needed to provide a substantial contribution to CCN (100 to 1000 cm^{-3}) in the atmosphere, making the role of SPP in background CCN contributions relatively small. However, genus-level pollen studies indicate that 10^{10} – 10^{12} grains per tree are emitted annually [Hidalgo *et al.*, 1999; Gomez-Casero *et al.*, 2004], which is far greater than observed pollen counts typically based on

near-surface observations. Up to 1% of pollen generation has been estimated to be lofted into the atmosphere for deciduous broadleaf species [Sofiev *et al.*, 2006], indicating that pollen grains could provide CCN on the order of 10^2 – 10^3 cm⁻³ if pollen grains rupture in or above the atmospheric surface layer.

While the magnitude of SPP contribution is uncertain because the number of SPP produced per pollen is unknown, we predict that SPP can contribute to the total CCN count in the atmosphere in anemophilous pollen regions such as the northern midlatitudes during peak pollen season. Cloud processing likely plays an important role in the conversion of large pollen grains into smaller, CCN-active material, and the suspendable nature of pollen suggests that even more SPP could be formed during other atmospheric and surface wetting and drying cycles. The experimental results presented here provide novel evidence for the ability of SPP to act as CCN under conditions frequently encountered in the atmosphere and establish the ability of multiple species to produce particles of similar CCN activation potential. Because measurements typically do not observe composition of CCN or limit defining composition as “organic,” our estimates suggest that SPP can account for a fraction of the known organic CCN in pollen-influenced midlatitude regions. This population of biological aerosols may influence the optical properties and cloud lifetimes in anemophilous pollen regions and affect indirect estimates of cloud forcing.

Acknowledgments

Laboratory CCN, carbohydrate content, and protein content data are available from the corresponding author. We gratefully acknowledge the use of the American Association for Allergy Asthma and Immunology (AAAAI) pollen count data with individual acknowledgements listed in Text S1, and pollen count data are available upon request from the AAAAI. This material is based on work supported by the National Science Foundation under AGS 0952650 to A.L.S. Any opinions, findings, and conclusions or recommendations expressed in this material are those of the authors and do not necessarily reflect the views of the National Science Foundation. We thank Dale Kruse and Stephan Hatch of the Texas A&M Tracy Herbarium for their assistance in tree sample identification and Andrew Laurence of Texas A&M University for his input on the presence of starch in pollen grains.

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