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# Hygroscopic Properties of 11 Pollen Species in China

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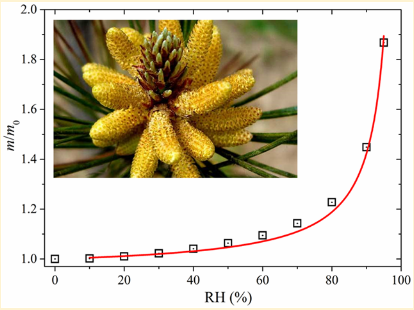
*Supporting Information*



**ABSTRACT:** Pollen, one of the most abundant types of primary biological aerosol particles, has significant impacts on human health, climate, and ecosystems. However, the hygroscopicity of pollen species in China remains to be unknown. In this work, we explored for the first time hygroscopic properties of pollen species widely found in China. Six anemophilous and five entomophilous pollen species (11 in total) were studied, and measurements were conducted as a function of relative humidity (RH, up to 95%) at two temperatures (25 and 37 °C). All 11 pollen species examined were found to show moderate hygroscopicity; the sample mass at 90% RH, normalized to that under dry conditions, was found to range from 1.33 to 1.43 at 25 °C, and the single hygroscopicity parameter ($\kappa$) was derived to be 0.036−0.048. No significant difference in hygroscopicity between anemophilous and entomophilous pollen species was observed, and the effect of the temperature (25 versus 37 °C) on pollen hygroscopicity was found to be small.

**KEYWORDS:** *primary biological aerosol particles, pollen, hygroscopicity, aerosol−water interaction, temperature effect*

## 1. INTRODUCTION

Primary biological aerosol particles (PBAPs) are of scientific and public concerns as a result of their effects on human health, climate, and ecosystems,[1−4] and their annual emission flux is estimated to be 10−1000 Tg.[5] Pollen, whose size typically varies between 10 and 100 $\mu$m, is one of the most abundant PBAPs, and its emission flux and atmospheric concentrations are around 47−84 Tg year$^{-1}$ and 10−1000 particles m$^{-3}$.[5,6] Figure 1 depicts the emission, transport, and impacts of pollen. Pollen grains can affect the formation and properties of clouds and precipitations because they are effective ice-nucleating particles (INPs) and cloud condensation nuclei (CCN).[7−9] Pollen can also lead to a series of allergic diseases, including allergic rhinitis, bronchial asthma, and dermatitis.[10,11] It was suggested that pollen could contribute to 5−20% of allergic reactions occurring in Europe.[12] In addition, pollen plays a key role in reproduction and dispersion of many plants[13] and, hence, significantly impacts ecosystem evolution.[6]

Hygroscopicity largely determines the mass and aerodynamic properties of aerosol particles (including pollen grains) under environmental conditions[14] and, therefore, affects their transport and deposition in the troposphere.[15] It also impacts the transport behavior of pollen grains and their fragments in the respiratory tract.[16] Moreover, the ability of aerosol particles serving as CCN and INPs is largely related to their hygroscopicity.[17,18] Therefore, hygroscopic properties of pollen species have been investigated in several previous studies.[19−22] For example, Diehl et al.[19] used an analytical balance to investigate water uptake of 11 pollen species at around room temperature, and the mass increase compared to that under dry conditions was measured to be 3−16% at 73% relative humidity (RH) and 100−300% at 95% RH. An electrodynamic balance was employed to study hygroscopic growth of eight pollen species at different RH.[20,21] They suggested that mass change of pollen species examined could be approximated by a modified $\kappa$−Köhler equation, and the single hygroscopicity parameter ($\kappa$)

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was found to be 0.05−0.1. Very recently, we investigated hygroscopic properties of six pollen species at different temperatures (5−37 °C), and the $\kappa$ values were measured to be 0.034−0.061 at 25 °C;[22] furthermore, in general, the temperature was found to have a small but negative effect on pollen hygroscopicity.

To our knowledge, all previous studies were focused on hygroscopicity of pollen species mainly found in North America and Europe, and hygroscopic properties of pollen species in China remain to be understood. Abundant pollen aerosols were found in ambient air in China, with higher concentrations observed in spring and autumn.[23,24] In the present work, we investigated hygroscopic properties of 11 pollen species from representative plants in China up to 90% RH (and even up to 95% RH for some pollen species). Experiments were also carried out at 37 °C (i.e., the physiological temperature) in addition to 25 °C, to help us better understand the transport and deposition of pollen in the respiratory tract. This work represents the first time that hygroscopicity of pollen species in China has been explored.

**Figure 1.** Emission, transport, and deposition of pollen and its impacts on human health, climate, and ecosystem.

## 2. MATERIALS AND METHODS

In this work, hygroscopic properties of 11 pollen species were investigated, all of which came from widely distributed plants in China (Flora of China, http://foc.iplant.cn/). Six plants are anemophilous, including *Pinus massoniana*, *Pinus tabuliformis*, *Pinus armandii*, *Pinus taiwanensis*, *Pinus bungeana*, and *Typha angustifolia*, and the other five plants are entomophilous, including *Pyrus* sp. (pear), *Amygdalus persica* (peach), *Malus pumila* (apple), *Prunus salicina* (plum), and *Brassica campestris* (canola). Pollen samples were collected from botanic gardens and then stored in pre-cleaned plastic vials, which were placed in a desiccator filled with silica gel for a few days to dry these samples. After that, these vials were sealed to store pollen samples for further experiments. The procedure used in our work to pretreat and store pollen samples (carried out at room temperature) was to minimize their interactions with moisture and reactive trace gases in the air, which may change their composition and properties.

Hygroscopic properties of pollen samples were investigated using a vapor sorption analyzer (Q5000 SA, TA Instruments), which has been applied to study hygroscopicity of atmospherically relevant particles in our previous work.[22,25,26] This instrument used a highly sensitive balance to measure the mass change of a sample as a function of RH at a given temperature to investigate its hygroscopicity. The sample mass could be measured in the range of 0−100 mg with a sensitivity of 0.01 $\mu$g. The temperature could be controlled in the range of 5−85 °C with an accuracy of 0.1 °C, and RH could be regulated in the range of 0−98% with an absolute accuracy of 1%. We routinely measured deliquescence relative humidities (DRHs) of NaCl, $(NH_4)_2SO_4$, and KCl to inspect the RH accuracy of this instrument, and the difference between measured and theoretical DRHs did not exceed 1%.

Hygroscopicity of all of the pollen samples was investigated at two temperatures (25 and 37 °C), and the experiments were carried out in triplicate. Figure 2 displays the change of RH and normalized sample mass with experimental time in a typical experiment. As illustrated in Figure 2, at a given temperature, the sample was dried at <1% RH and the sample mass under dry conditions was typically 1−5 mg (as a result, the average hygroscopicity was investigated for every pollen sample, which contained many pollen grains). After that, RH was increased to 90% step by step, and at each step, RH was increased by 10%; at each RH, the sample was considered to reach an equilibrium when its mass change was <0.05% within 30 min, and then RH was changed to the next value. For some experiments, RH was further increased to 95%. Because it could take up to 3 days to reach an equilibrium at 95% RH, we only conducted measurements at 95% RH for five pollen species (*P. massoniana*, *P. tabuliformis*, *T. angustifolia*, *Pyrus* sp., and *B. campestris*). At the end, RH was changed back to <1% to dry the sample again.

## 3. RESULTS AND DISCUSSION

Figure 3a displays the normalized sample mass (normalized to that at <1% RH, $m/m_0$) as a function of RH for *P. massoniana* pollen at 25 °C. In addition, the normalized sample mass at different RH is compiled in Tables S1−S4 of the Supporting Information for all 11 pollen species at 25 and 37 °C. Figure 3a suggests that a substantial increase in sample mass was observed for *P. massoniana* pollen at 25 °C, and the normalized sample mass was determined to be 1.045 ± 0.001 at 40% RH, 1.184 ± 0.009 at 80% RH, 1.343 ± 0.011 at 90% RH, and 1.718 ± 0.008 at 95% RH. As shown in Tables S1−S4 of the Supporting Information, a substantial increase in sample mass was observed for all of the pollen species considered in our work at both 25 and 37 °C. Diehl et al.[19] investigated hygroscopic properties of 11 pollen species at room temperature and found that the sample mass was increased by 3−16% (in comparison to that under dry conditions) at 73% RH; for comparison, sample mass was increased by 10−15% at 70% RH for the 11 pollen species examined in work. Therefore, the measured sample mass increases at around 70% RH were in good agreement between the work by Diehl et al.[19] and our study, although the variation in measured sample mass increase was significantly smaller for pollen species investigated in our work.

Table 1 and Figure 4a summarize normalized sample mass at 90% RH, $m(90\%)/m_0$, for the 11 pollen species that we studied. The values of $m(90\%)/m_0$ at room temperature were found to range from 1.325 ± 0.004 (for *P. taiwanensis* pollen) to 1.433 ± 0.015 (for *B. campestris* pollen). The small variation in the observed hygroscopicity could result from different compositions for different pollen species; for example, one previous study[22] suggested that pollen species with higher abundance of OH groups, relative to that of C−H groups, displayed higher hygroscopicity. In addition, the difference in microstructure of

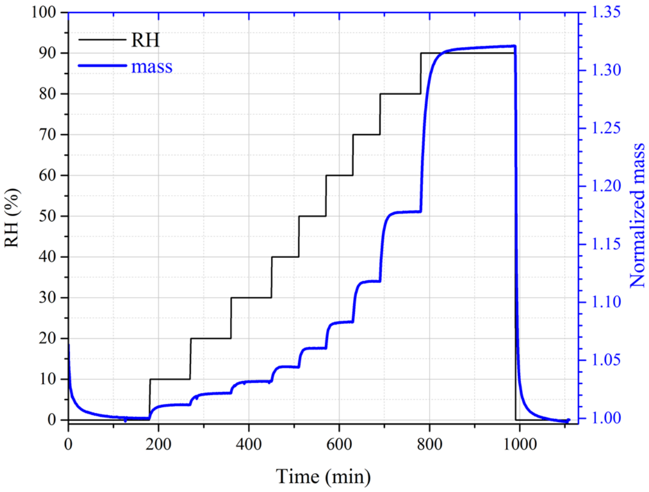

**Figure 2.** RH (black curve, left *y* axis) and normalized sample mass (blue curve, right *y* axis) as a function of experimental time during one experiment in which hygroscopic properties of *P. taiwanensis* pollen were examined at 25 °C.

pollen species may also contribute to the observed variation in their hygroscopicity. In our work, we examined pollen species from six types of anemophilous plants and five types of entomophilous plants, and the measured mass increases at 90% RH did not show significant difference between anemophilous and entomophilous pollen. Our previous study[22] investigated hygroscopic properties of six pollen species (*Populus tremuloides*, *Populus deltoides*, ragweed, corn, pecan, and paper mulberry), and $m(90\%)/m_0$ was found to vary between 1.293 ± 0.028 and 1.476 ± 0.094 at 25 °C. It appears that there is no significant difference in hygroscopicity for pollen species considered in these two studies.

Table 1 and Figure 4a compare $m(90\%)/m_0$ at 25 and 37 °C for the 11 pollen species investigated. A small decrease in $m(90\%)/m_0$ was generally observed for anemophilous pollen species when the temperature increased from 25 to 37 °C, except *P. massoniana* pollen. Our previous study[22] investigated six anemophilous pollen species at different temperatures (5−37 °C) and also found a negative dependence of hygroscopicity upon the temperature. In contrast, as shown in Table 1 and Figure 4a, increase in the temperature from 25 to 37 °C usually resulted in a small increase in $m(90\%)/m_0$ for entomophilous pollen, except *B. campestris* pollen. A negative dependence of pollen hygroscopicity upon the temperature indicates that water adsorption on this pollen species is exothermic and vice versa. Overall, our present and previous work[22] suggested that the effect of the temperature (25 versus 37 °C) on pollen hygroscopicity was very small.

Equation 1 is used to relate the particle mass increase at different RH to the single hygroscopicity parameter ($\kappa$)[17,22]

$$\frac{m}{m_0} = 1 + \kappa \frac{\rho_{\mathrm{w}}}{\rho_{\mathrm{p}}} \Big/ \left( \frac{1}{\mathrm{RH}} - 1 \right) \tag{1}$$

where $m$ and $m_0$ are the particle masses at a given RH and under dry conditions, respectively, $\rho_{\mathrm{w}}$ is the density of water (1 g cm$^{-3}$), and $\rho_{\mathrm{p}}$ is the density of the dry particle and is typically in the range of 0.5−2 g cm$^{-3}$ for pollen.[27] Equation 1 can be derived under the following two assumptions: (i) the particle is spherical, and (ii) the particle volume at a given RH is equal to the sum of the volume of water associated with the particle at this RH and the dry particle volume. Previous studies[20−22] suggested that eq 1 could approximate the mass change of pollen species at different RH. It was also found in our current work that the change in sample mass as a function of RH (at both 25 and 37 °C) could be fitted using eq 1 for all 11 pollen studied, and two examples (*P. massoniana* pollen at 25 °C and *Pyrus* sp. pollen at 37 °C) are displayed in Figure 3. Although previous studies[20−22] and our present work found that mass hygroscopic growth of a number of pollen species could be empirically described by eq 1, it should be pointed out that the underlying assumption in eq 1 is not fulfilled because pollen grains are not spherical. If we further assume that the density of dry pollen is equal to that of water (1 g cm$^{-3}$), the $\kappa$ value can then be derived from the fitting using eq 1. The values of $\kappa$ are displayed in Table 1 and Figure 4b, spanning from 0.036 ± 0.001 (for *P. taiwanensis* pollen) to 0.048 ± 0.002 (for *B. campestris* pollen) at 25 °C. Because the actual pollen density can differ from the density (1 g cm$^{-3}$) that we used to calculate $\kappa$ by a factor of ~2, the $\kappa$ values reported here are expected to have similar uncertainties. The $\kappa$ values were found to be between 0.034 and 0.061 for the six pollen

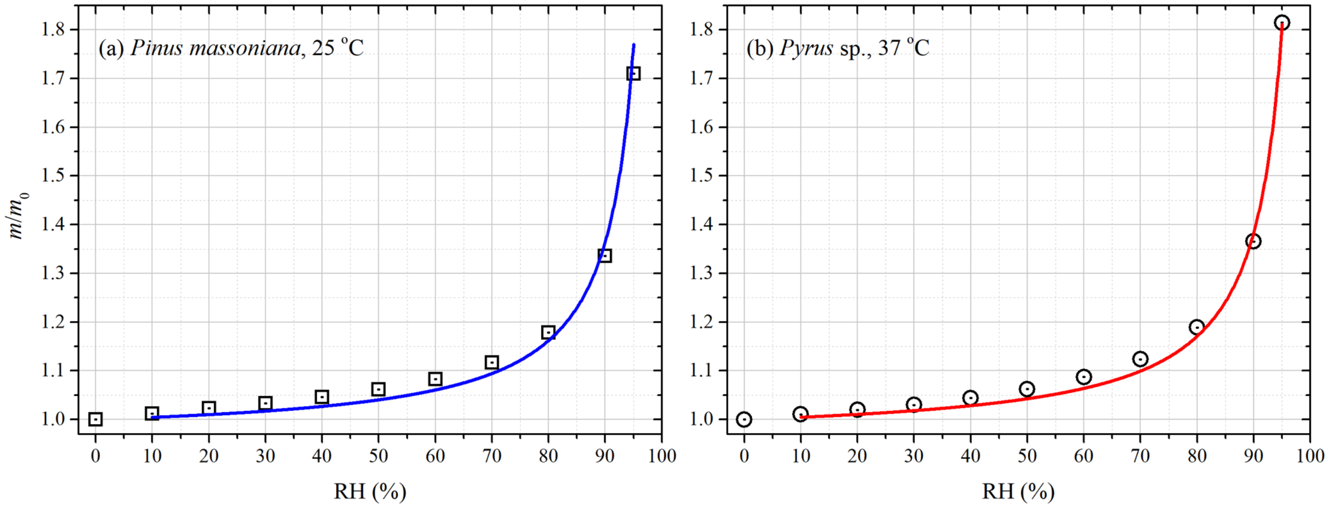

**Figure 3.** Measured change of sample mass (normalized to that at <1% RH, i.e., $m/m_0$) of pollen samples as a function of RH (up to 95%): (a) *P. massoniana* pollen at 25 °C and (b) *Pyrus* sp. pollen at 37 °C. The experimental data are fitted with the modified $\kappa$−Köhler equation (solid curve).

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Table 1. Normalized Sample Mass at 90% RH [Normalized to That at <1% RH, $m(90\%)/m_0$] and Derived Single Hygroscopicity Parameters ($\kappa$) in This Work for the 11 Pollen Species at 25 and 37 °C

| | $m(90\%)/m_0$ | | $\kappa$ | |
|---|---|---|---|---|
| pollen | 25 °C | 37 °C | 25 °C | 37 °C |
| *Pinus massoniana* | 1.343 ± 0.011 | 1.345 ± 0.001 | 0.038 ± 0.001 | 0.038 ± 0.001 |
| *Pinus tabuliformis* | 1.352 ± 0.003 | 1.307 ± 0.003 | 0.039 ± 0.001 | 0.034 ± 0.001 |
| *Pinus taiwanensis* | 1.325 ± 0.004 | 1.296 ± 0.002 | 0.036 ± 0.001 | 0.033 ± 0.001 |
| *Pinus armandii* | 1.375 ± 0.004 | 1.332 ± 0.015 | 0.042 ± 0.001 | 0.037 ± 0.002 |
| *Pinus bungeana* | 1.340 ± 0.002 | 1.301 ± 0.001 | 0.038 ± 0.001 | 0.033 ± 0.001 |
| *Typha angustifolia* | 1.328 ± 0.001 | 1.302 ± 0.001 | 0.036 ± 0.001 | 0.034 ± 0.001 |
| *Pyrus* sp. | 1.335 ± 0.008 | 1.353 ± 0.014 | 0.037 ± 0.001 | 0.039 ± 0.002 |
| *Amygdalus persica* | 1.336 ± 0.009 | 1.357 ± 0.010 | 0.037 ± 0.001 | 0.040 ± 0.001 |
| *Prunus salicina* | 1.345 ± 0.010 | 1.366 ± 0.009 | 0.038 ± 0.001 | 0.041 ± 0.001 |
| *Malus pumila* | 1.346 ± 0.007 | 1.374 ± 0.011 | 0.038 ± 0.001 | 0.042 ± 0.001 |
| *Brassica campestris* | 1.433 ± 0.015 | 1.389 ± 0.014 | 0.048 ± 0.002 | 0.043 ± 0.002 |

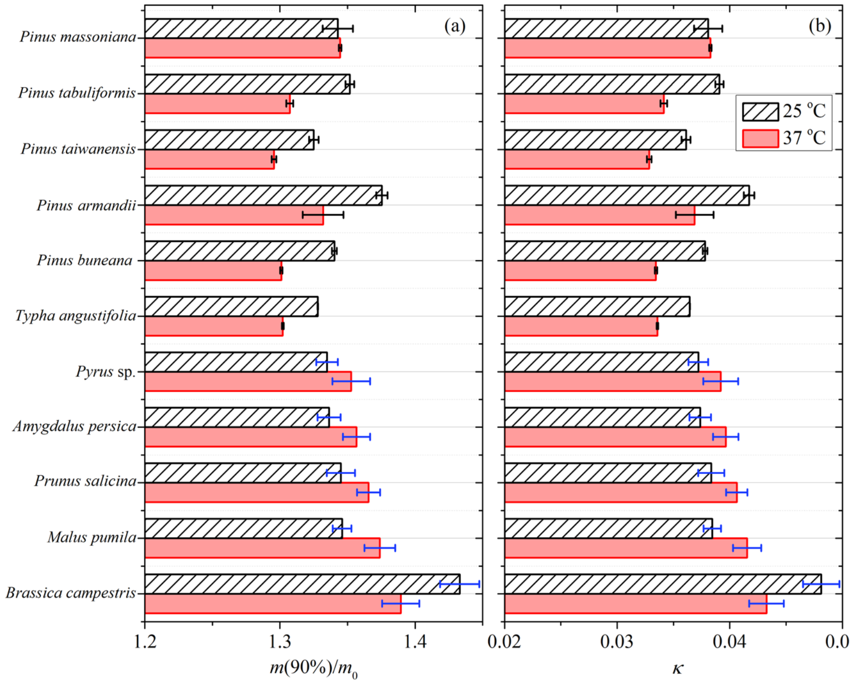

**Figure 4.** (a) Normalized sample mass at 90% RH [relative to that at <1% RH, $m(90\%)/m_0$] and (b) derived $\kappa$ values for 11 pollen species examined in this work.

species considered by Tang et al.[22] and between 0.05 and 0.11 for the eight pollen species considered by Pope[20] and Griffiths et al.,[21] reasonably agreeing with those determined in our present work. The $\kappa$ values of pollen species were significantly larger than those for fresh mineral dust (<0.01)[18] but slightly smaller than (or similar to) those for secondary organic aerosols (~0.1),[17] suggesting that pollen was moderately hygroscopic. Because the difference in normalized mass change at a given RH was small at 25 and 37 °C, the difference in $\kappa$ values at the two temperatures was also small, as expected.

## 4. CONCLUSION

Pollen is an important type of primary biological aerosol particles, significantly impacting human health, climate, and ecosystems. Hygroscopicity, which largely determines the transport and deposition of pollen particles in the air and is also closely related to their cloud condensation and ice nucleation activities, has been explored in several previous studies for pollen in Europe and North America. However, hygroscopic properties remain unknown for pollen species in China. In this study, we employed a vapor sorption analyzer to investigate hygroscopic growth of six anemophilous and five entomophilous pollen species (11 in total) widely found in China, and measurements were carried out up to 95% RH at two temperatures (25 and 37 °C). Significant increases in sample mass were observed for all of the pollen species at elevated RH for both temperatures. The sample mass at 90% RH, normalized to that at <1% RH, were found to fall between 1.325 ± 0.004 (*P. taiwanensis* pollen) and 1.433 ± 0.015 (*B. campestris* pollen) at 25 °C for the 11 pollen species examined, and correspondingly, the single hygroscopicity parameters were determined to be in the range of 0.036−0.048. No significant difference in hygroscopicity was observed for anemophilous and entomophilous pollen species, and the effect of the temperature on pollen hygroscopicity was found to be small. In addition, there was no obvious difference in hygroscopic properties between pollen species widely found in China and those typically found in Europe and North America. Because the number of pollen species investigated in the present work (as well as in previous studies) is rather limited, future work is encouraged to measure hygroscopic properties of more pollen species.

## ■ ASSOCIATED CONTENT

### Ⓢ Supporting Information

The Supporting Information is available free of charge on the ACS Publications website at DOI: 10.1021/acsearthspacechem.9b00268.

Normalized mass as a function of RH at 25 and 37 °C for the 11 pollen species examined in this work (Tables S1–S4) (PDF)

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**Notes**

The authors declare no competing financial interest.

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