

Comments by referees are in blue.

Our replies are in black.

Changes to the manuscript are highlighted in red both in here and in the revised manuscript.

Reply to Ref #1

Tang et al. investigated the hygroscopic growth of six pollen species and its temperature dependence. This study measured water uptake and growth factor by pollen grains using a vapor sorption analyzer and characterize pollen grains using FTIR. A hygroscopic parameter (k) was calculated from the measurements. The subject of this manuscript is within the scope of this journal. There are some minor issues that the authors may want to address before it can be accepted for publication.

Reply: We would like to thank Ref #1 for his/her insightful and detailed comments, which have largely helped us improve our manuscript. We have addressed all the comments adequately in the revised manuscript, as detailed below.

1. P5, L98-100, are these pollen species atmospheric relevant? Justification of using these pollen species needs further discussion.

Reply: These pollen species have been chosen in our work primarily because of their commercial availability; nevertheless, these plants are widely distributed in the globe and some of the pollen species, such as ragweed pollen, are well-known due to their impacts on human health. In the revised manuscript (page 5, line 101-105) we have added a few sentences to further justify why these pollen species were chosen in our work: “The six pollen species were chosen in our work primarily because they were commercially available. Furthermore, these plants are also widely distributed in the globe. For example, corn is the most produced grain in the world (International-Grains-Council, 2019), and up to 50% of pollen-related allergic rhinitis cases in North America are caused by ragweed pollen (Taramarcaz et al., 2005).”

2. P6, L114, what is the uncertainty of this moisture meter?

Reply: The sensor has an absolute uncertainty of $\pm 2\%$. In the revised manuscript (page 6, line 118-119) the sentence has been changed to “...and was monitored online using a moisture meter (CENTER 314) with an absolute uncertainty of $\pm 2\%$.”

3. P6, L130-132, which kind of temperature and humidity sensors that can achieve such high accuracy (± 0.1 K and 1% RH) at this temperature and RH range?

Reply: The temperature was monitored using a thermocouple, which could achieve a temperature accuracy of ± 0.1 K easily. The high accuracy of RH control was achieved by using mass flow controllers to precisely control the flow rates of the dry and humidified nitrogen flows used to regulate RH in the humidity chamber; the accuracy of RH control was routinely checked by measuring the DRH of standard compounds, and the difference in measured and theoretical DRH was always $<1\%$. In the revised manuscript (page 7, line 138-142) we have added a few sentences to explain how high accuracy in RH control was achieved: “RH in the humidity chamber was regulated by using two mass flow controllers to control the dry and humidified nitrogen flows very precisely. The accuracy in RH control was routinely checked by measuring the DRH values for a series of standard compounds, e.g., NaCl, $(\text{NH}_4)_2\text{SO}_4$, KCl, and etc., and the difference between the measured and theoretical DRH was always $<1\%$.”

4. P7, L137-138, Although it may be fine, I do not think this is an excuse that left the other temperature out. One can simply conduct a few more experiments for the missing points.

Reply: We agree with the referee, and would like to carry out measurements at 5°C for the other three pollen species. However, unfortunately due to a technical problem, the lowest temperature our instrument could reach was 15°C since the technical problem occurred. In the revised manuscript (page 7, line 146-149) the following change has been implemented for further clarification: “For each of the other three types of pollen species (corn, pecan and paper mulberry pollen), experiments were carried out at 15°C instead of 5°C , because the instrument could only be cooled down to 15°C due to a technical problem after we finished experiments for the first three pollen species.”

5. P8, L157-165, it is suggested to list these peak assignments in a table.

Reply: As suggested, we have included a table in the revised manuscript (Table 1, page 10) to summarize these peak assignments, and also made corresponding changes to text in Section 3.1.1 (page 8, line 168-169).

6. P9, L169-173, please justify the use of such ratio as a qualitative representation of OH groups. Have any other studies been using such proxy?

Reply: As stated in our original manuscript, one may expect that the amount of OH groups (relative to that of C-H groups) that pollen samples contain may affect their hygroscopicity. In addition, a number of previous studies found that heterogeneous aging of organic materials would lead to increase in hygroscopicity; in addition, they also found that the IR absorption intensity for

the O-H stretching mode would increase and the IR absorption intensity for the C-H stretching mode would decrease upon heterogeneous oxidation. Therefore, we used the intensity ratio of the O-H stretching vibration band to the C-H stretching mode to represent the amount of OH groups in a qualitative manner (not quantitatively, however) and explored if there was any correlation between this intensity ratio and measured hygroscopicity. In the revised manuscript (page 9-10, line 181-190) we have expanded our discussion to provide further justification: “OH groups and C-H groups in organic compounds are generally considered to be hydrophilic and hydrophobic, and one may expect that the amount of OH groups (relative to that of C-H groups) that organic samples contain may affect their hygroscopicity. For example, it was found in many previous studies (Eliason et al., 2003; Asad et al., 2004; Hung et al., 2005; Najera et al., 2009) that heterogeneous reactions of organic materials with O₃ and OH radicals would increase the IR absorption intensity for the O-H stretching mode and decrease the IR absorption intensity for the C-H stretching mode, meanwhile leading to the enhancement in their hygroscopicity. Therefore, in this work we use the intensity ratio of the O-H stretching vibration band (3000-3600 cm⁻¹) to the C-H stretching mode (2920 cm⁻¹) to qualitatively represent the amount of OH groups pollen samples contain.”

7. P11, L201-203, It only indicates that there is a correlation between water adsorption and OH groups in pollen samples. As discussed in L302-316, there may be other factors contribute to the water uptake. It is suggested to revise these related statements.

Reply: As suggested, in the revised manuscript (page 12, line 221-224) we have revised our discussion: “These results imply that water adsorption by pollen samples could be mainly contributed by OH groups of organic compounds they contained; in addition, other factors, such as porosity and internal structure, may also be important for hygroscopic properties of pollen grains.”

8. P12, L215, k parameter is just a fitting from the data. As for now there is no physical meaning for such equation. It is not really a theory.

Reply: We agree with the referee. In the revised manuscript (page 13, line 236) the title of Section 3.2.1 has been from “Theories” to “Hygroscopicity parameterizations”, and throughout the revised manuscript (e.g., page 1, line 34) “κ-Köhler equation” has been changed to “κ-Köhler equation”.

9, P16, L279-L284, As mentioned above, the k value is obtained from the fitting of these data points, of course, this should fit it well, otherwise k value is wrong. As for Freundlich approach, what are the A and B values? To compare these two different approaches, further discuss is needed.

Reply: We respect but do not quite agree with this comment, and would like to clarify it here. In our work we attempted to use both the κ -Köhler equation and the Freundlich adsorption isotherm to fit our experimental data. For the experimental data shown in Figure 4, if fitted with the κ -Köhler equation, the best fitting gave a $\kappa \cdot \rho_w / \rho_p$ value of 0.036 ± 0.001 , and as shown in Figure 4, the κ -Köhler equation fitted the experimental data very well; if fitted with the Freundlich adsorption isotherm, the best fitting gave an A value of 1.19 and a B value of 0.091, and as shown in Figure 4, the Freundlich adsorption isotherm failed to fit the experimental data. This is why we have stated in our original manuscript that the κ -Köhler equation described our experimental data very well but the Freundlich adsorption isotherm did not.

10, P17, L286-289, If use density of 1 g/cm³, that means k values may be 2 times higher or lower when considering range of 0.5 -2 g/cm³. This is a huge uncertainty. That mean you cannot really compare k values for different species unless they have very similar density.

Reply: As pointed out by the referee, our derived κ values have large uncertainties, mainly due to large uncertainties in pollen density. However, as shown in Eq. (5), at a given RH mass hygroscopic growth factors, m/m_0 , depend on $\kappa \cdot \rho_w / \rho_p$ rather than κ ; to compare the hygroscopicity of different pollen species, we should compare $\kappa \cdot \rho_w / \rho_p$ values rather than κ values; in our work to make the comparison mathematically simpler, we assume a pollen density of 1 g cm⁻³ and thus $\kappa \cdot \rho_w / \rho_p$ is equal to κ . As a result, when we compare hygroscopicity of the six pollen species examined in our work, we do not need to assume that they have very similar density.

11, P18, L298, It is not clear what does “All the errors are statistical only.” mean.

Reply: We actually mean that all the errors given here are standard deviations. In the revised manuscript (page 18, line 333) we have changed this sentence to “All the errors given in this work are standard deviations.”