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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Pre-deliquescent water uptake in deposited nanoparticles observed with *in situ* ambient pressure X-ray photoelectron spectroscopy" *by* Jack J. Lin et al.

Anonymous Referee #2

Received and published: 3 July 2020

In this work, Lin et al. study the uptake of water vapor onto the surfaces of impacted NaCl, sucrose and malonic acid aerosols relative humidities from 0 to 16% using X-ray photoelectron spectroscopy, a surface-sensitive technique capable of detecting subtle chemical occurring with adsorption of water molecules. They find that water is adsorbed onto NaCl and sucrose particles at low RH, well below the RH at which particles deliquesce, but sucrose does not. Generally, I think this work is very solid and deserves prompt publication. I congratulate the authors on very thorough statistical analysis and literature analysis regarding the results. However, the specific importance of this measurement on aerosols is not emphasized. Similar measurements have been performed for non-aerosol samples of the same substances. This is the major shortcoming of the

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work.

There are a few additional issues with the paper that frustrate the efforts of the reader to contextualize and cite the work. Furthermore, the fitting parameters, which are not included, should be more carefully documented and displayed somewhere in the paper or supplement. These improvements are necessary and could improve the impact and longevity of the work.

I recommend publication in ACP if these issues, and the following comments, can be addressed.

Major Comments

The literature review and discussion focus on aerosol water uptake, a phenomenon separate from water adsorption below deliquescence. More emphasis could be placed, in the introductory and concluding sections, on the chemistry occurring on surfaces of aerosol particles. Surface chemistry and reactions occurring during evaporation/condensation on aerosols is a separate and rapidly evolving branch of this science with many recent publications. This manuscript would have a larger impact if it included some references to recent developments in this topic. What reactions are promoted by adsorption of water molecules onto pre-deliquescent NaCl or carboxylic acids? This is an important question for this manuscript to discuss.

The conclusions are not prevalent enough. The assessment summarizing each qualitative/quantitative description are lost in each paragraph, although the results are presented with very good attention to detail and thoughtful analysis. I recommend improving the prevalence of these conclusion sentences, e.g. through their placement at the head of each paragraph, through an increased number of headings, or via another method. This would significantly improve the comfort of the reader and perhaps the breadth of the readership.

The peak fitting parameters are missing, although the fitting of peaks in this work is

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described with great care. Further, displaying all the peak fits in the main text may not be necessary. The statistical analysis of these peaks (peak area, peak width, peak shifts) are more interesting. More emphasis on the statistics would be helpful in interpreting the quality of the data and the conclusions presented.

Minor comments

1. The fitted peaks in each figure are visually blocking the data points. Please rectify the situation.

2. In the introduction or XPS measurement section, a brief description of the measurement mechanism and e.g. the meaning of the signal, for a slightly broader audience, would be appreciated. This topic is of great interest to various readers who do not use XPS.

3. "2.3 Data Analysis" – the peak fitting is described carefully but it is not clear to me how much the peak shape is derived from first principles and how much is empirical. If the shape of these peaks is not physically meaningful, less emphasis could be placed on justifying the process of fitting. Where there is a meaningful connection between the equation and the data, this could be emphasized.

4. Line 163 – "after calibrating . . . as described earlier" – this sentence is not needed, especially at the top of the paragraph.

5. Lines 164-165 – parameters like signal-to-noise and error bars on the fits – the omission of which I feel are a major detriment to the paper – should be included in a table somewhere, or in the supplement.

6. Lines 192-195 and lines 209-211– How does drying the aerosol influence the crystal form significantly? This is one important way in which the aerosol measurement may prove different from the non-aerosolized measurements. In keeping with what I believe is the major shortcoming of this manuscript, this connection between your work and the aerosol in the atmosphere is important to discuss in a location and/or under a heading

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where the reader can find it easily.

7. Lines 229-235 – the length of these statements could be reduced significantly.

8. Lines 242-245 – does curvature of the impacted particles resting on the substrate change the signal intensity corresponding to surface adsorption by virtue of the tilted angle of the sides of the particles? This is true of e.g. microscopy studies of impacted particles.

9. Lines 261-262 – specifically, how?

10. Figure 4 – the “COOH:COOH” looks very redundant here.

11. Lines 292-294, 301, and 303 – could these statements have come sooner in the section/paragraph?

12. Line 313 – please define “DP1” and “DP2”

13. Figure 8 – the peaks are very close together. It would be helpful to see a 95% confidence interval of the peak, or similar.

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