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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Investigation of the hygroscopic properties of $Ca(NO_3)_2$ and internally mixed $Ca(NO_3)_2/CaCO_3$ particles by micro-Raman spectrometry" *by* Y. J. Liu et al.

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General Comments: This paper describes hygroscopic behavior of Ca(NO3)2 and Ca(NO3)2/CaCO3 particles using micro-Raman spectroscopy. Although the paper highlights the usefulness of this technique in determining particle water content and structural changes upon phase transition, a few minor revisions/clarifications are suggested:

Comments: 1.The microscopic images in Figures 5 and 7 have very poor resolution. It would be helpful if the authors speciffied how these images were collected (i.e. type of instrument used).

Response: The images were collected by microscope (Olympus BX40) equipped to

micro-Raman spectrometry. A 50Œ NA0.5 long working distance objective was used (P10602, line 25). The microscopic images were recorded by an analog camera equipped to the micro-Raman system and connected to the computer. We have include the description in the revised version. The poor resolution of Figs. 5 and 7 was due to the poor resolution of the camera. We have replaced it with a high-resolution digital camera.

2. It would be helpful if the authors did not refer to the phase transition of Ca(NO3)2 between 11

Response: We agree that efflorescence refers to crystallization of a particle, it is an inappropriate term for the phase transition here. Thanks for the suggestion. "Dehydration process" will be used in the revised version.

3.The authors cite quite a few peak positions in the text, and refer to sharp shifts and changes in FWHM of the Raman spectra. It would be very helpful for the reader if a sample of these Raman spectra were shown.

Response: Thanks for the suggestion. The Raman spectra are included in the revised manuscripts.

4.The absence of a hysteresis effect between deliquescence and efflorescence processes is a fairly unusual result. Further discussion on the chemical reasoning behind this result would be appreciated.

Response: We are also very interested in the absence of a hysteresis effect between deliquescence and dehydration process of Ca(NO3)2 particles. Without knowing thermodynamic properties of amorphous Ca(NO3)2 particles, we could not provide the chemical reasoning behind this result. However, there is a scientific assumption. The hysteresis effect between deliquescence and efflorescence is usually explained by kinetic inhibition of critical germ nucleation in efflorescence process. However, there are also some other phase transitions that experience gentler rearrangement of atoms,

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called displacive transformation (Martin, 2000). These phase transitions don't require the formation of critical germ, and hence are not kinetically inhibited. Since the Raman spectrum of amorphous Ca(NO3)2 particles and that of Ca(NO3)2 supersaturated droplets have a relatively small difference, indicating similar chemical properties, transformation from Ca(NO3)2 solution droplets to amorphous particles is probably such a case. The above discussion and the reference will be included in the revised manuscript.

Reference: Martin, S. T.: Phase transitions of aqueous atmospheric particles, Chem. Rev., 100, 3403-3453, 2000.

Interactive comment on Atmos. Chem. Phys. Discuss., 8, 10597, 2008.

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