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Title: Atmospheric nanoparticles hygroscopic growth measurement by combined surface plasmon resonance microscope and hygroscopic-tandem differential mobility analyzer

We thank the anonymous referees for their valuable and constructive comments/suggestions on our manuscript. We have revised the manuscript accordingly and please find our point-to-point responses below.

Comments by Anonymous Referee #2:

General Comments:

Review to "Atmospheric nanoparticles hygroscopic growth measurement by combined surface plasmon resonance microscope and hygroscopic-tandem differential mobility analyzer". The authors present combined measurements of aerosol hygroscopic growth using an HTDMA and a new SPRM apparatus, targeting at the hygroscopic behavior of bulk aerosols and single particles of 100, 150, and 200 nm, respectively. Combined with the classification of chemical component from SEM-EDX investigations, the authors try to link the single-particle hygroscopicity of different chemical components and the non-uniform distribution of the bulk aerosol hygroscopic growth factor. This method is novel and fits into the scope of ACP. However, the significance of this combined hygroscopic growth study needs to be furtherly clarified, and more detailed information should be provided to make it a solid work. The reviewer recommends accepting this manuscript after addressing the following comments.

Response: We thank the reviewer for the constructive suggestions and comments. Point-to-point responses to comments and questions are detailed below. Following the reviewer's suggestions, we organized the manuscript in the clearer way and clarified the significance of combined SPRM and HTDMA measurements for particle hygroscopic growth studies. The new results and discussions are now included in the revised manuscript.

Major comments:

1) *What is the scientific question the authors want to address, based on the coupled SPRM and HTDMA measurement? To me, it looks like a closure study of aerosol hygroscopic properties based on the single-particle GF quantification and the bulk GF distribution for ambient aerosols. What type of additional knowledge it provides regarding the mixing state of aerosol chemical components?*

Response: We thank the reviewer's comment. Yes, it looks like a closure study but investigates aerosol hygroscopic properties from very different perspectives, i.e., single-particle and bulk aerosols. As the hygroscopic properties of ambient aerosols are not uniform but spreads among particles of the same size. To better understand the contribution of different aerosol components, we conduct combined hygroscopic growth measurements using a SPRM-ARI and an HTDMA and establish a link between the apparent hygroscopic properties of single particles and bulk aerosols, thereby providing more information about particle chemical composition and hygroscopic properties. We first identified individual particles with distinct hygroscopic growth behaviors from the SPRM single-particle probing and classified those particles into different categories including non-hygroscopic (NH), less-hygroscopic (LH), and more-hygroscopic (MH). The chemical compositions of individual particles were identified using SEM/ESD analysis, and the results likely agree with the apparent hygroscopic properties. Next, the mean growth factor (GF) of the three categories can be utilized to reproduce the GF distribution obtained from the HTDMA measurement, such that the number fractions of the three categories can be retrieved. We clarified this in the revised manuscript.

2) *The authors demonstrate the classification of the four groups (i.e., EC, fly ash, OC and AS+OC) in terms of ambient aerosol chemical components, based on the EDS mapping of SEM images. Please clarify the detailed approach of the classification and quantify how representative it is.*

Response: In this study, we take advantage of the SEM image and EDS spectra of individual particles, the relative abundance of key elements (e.g., C, O, and S) can be quantified for each particle. According to the particle morphology and elemental composition, the individual particles can be classified into different categories (Kirpes et al., 2018), i.e., organic carbon (OC), soot (mainly elemental carbon), fly ash and secondary aerosols (mainly OC and sulfate). The SEM and EDS analysis provides reference for potential particle chemical compositions.

3) *The low resolution of Fig. 3 makes the particle imaging at different RH levels blurred. Please provide a clear figure or equivalent statistics supporting the derivation of GF from GI intensity.*

<u>Response:</u> According to the reviewer's suggestion, we provided the SPRM-ARI figures (i.e, Fig. 3) with higher contrast, and now the variation of gray intensity under different RH conditions can be clearly observed.