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Comment on acp-2022-666

Anonymous Referee #1

Referee comment on "Atmospheric nanoparticles hygroscopic growth measurement by a combined surface plasmon resonance microscope and hygroscopic tandem differential mobility analyzer" by Zhibo Xie et al., Atmos. Chem. Phys. Discuss., <https://doi.org/10.5194/acp-2022-666-RC1>, 2022

The manuscript "Atmospheric nanoparticles hygroscopic growth measurement by combined surface plasmon resonance microscope and hygroscopic-tandem differential mobility analyze" by Xie et al. shows the new coupling of SEM-EDX/HTDMA and SPRM (also combined with chemical measurements of EC, OC and sulfate components) for the investigation of hygroscopic growth of real ambient 100nm, 150nm and 200 nm particles.

Such measurements are important and fits well into the scope of ACP, even when the methodological/instrumental part takes a large part in this manuscript and should actually be even longer in order to be able to follow all the details.

Partly the explanations are not fully comprehensible, also because some terms are not explained and some occurring abbreviations are explained only in the later part of the manuscript.

This is most critical for the main SPRM-ARI method. For the essential details, reference is made to other papers and the only visible result of the method is a very fuzzy Figure 3a. No information is given in the manuscript about the number of particles studied, so statistical significance is difficult to estimate.

Most of the fundamental data and conclusions are obtained from SEM and HDTMA measurements, so the paper as presented is more of a SEM/HDTMA as a SPRM-ARI/HDTMA coupling.

The presented main results of the manuscript are:

- Establish a link between hygroscopic properties of bulk aerosol and single particles respectively establishing a link between single particle composition and its hygroscopicity.
- The OC content of larger mixed AS/OC particles (100 nm vs. 200 nm diameter) increases.
- The used fitting reconstruction method has a good correlation with quantitative determined OC, EC and sulphate concentrations.

To 1) I am not sure if this link is reached in the manuscript by SPRM-ARI. Due to the missing statistics and the fuzzy Figure 3a, the mentioned advantage of SPRM-ARI over other methods mentioned in the manuscript (ESEM/ETEM) is not clear enough (see following discussion).

To 2) This observation seems to be correct for the 2 collections carried out at the specific sample location. To derive a general pattern from this is not permissible.

To c) I cannot share this statement from the data shown (also due to the points still to follow). I agree that there is no contradiction but there is not enough data given to make a correlation visible.

Further points:

Definition of subgroups: For the given particle diameters (100-200 nm) of an urban aerosol typically mixtures of secondary material (organic, nitrates and sulfates) and soot (which is a mixture of OC and EC) dominates. Often many of these components are internally mixed and the hygroscopic behavior of this mixture is given by the HTDMA curve in Figure 4.

Following the secondary electron images given in Figures 2 (figure2 legend is erroneous), 5 and 6 all shown particles (except the fly ashes) seems to be dominantly soot, respectively mixtures of soot and secondary material. As soot is a mixture of OC and EC components this does not necessarily contradict the given EC, OC subgroup definition. As a simplification, the approach of classifying all carbon-rich particles with low oxygen content as soot (dominant EC – will show no strong water uptake) and those with very high oxygen content as OC (low or no soot content) may be permissible. But the simplification of all secondary material as ammonium sulfate does not seem permissible to me. Maybe it should be called ambient secondary material. The shown EDX mappings in

Figure 2 are not helpful for the proof of ammonium sulfate as the shown count rates are too low and nitrate cannot be detected in EDX as a nitrogen peak may originate from ammonium or nitrate.

The division of the HDTMA curve into 4 sub curves based on the 4 self-defined subgroups seems uncertain to me because of the problems mentioned above. Also, it does not seem clear to me to what extent the SPRM-ARI data played a role here. The significance of these measurement must be shown and worked out more clearly or the statements must be adjusted accordingly.