

Interactive comment on “Hygroscopic growth and activation of HULIS particles: experimental data and a new iterative parameterization scheme for complex aerosol particles” by M. Ziese et al.

Anonymous Referee #1

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The manuscript presents laboratory results on the hygroscopic properties and CCN activity of HULIS and mixed particles. In previous studies, HULIS have been shown to be a significant fraction of atmospheric aerosols and attempts have been made to characterize their properties in terms of chemical composition, hygroscopic behavior and cloud nucleating ability. Whereas the properties of other inorganic and organic aerosol constituents can be well described by Koehler theory, the HULIS fraction cannot be easily parameterized as it is composed of many unknown compounds. Differences in growth factors between the present and previous studies highlight the ambiguity that exists in defining HULIS. The authors present an iterative procedure that has been applied in order to obtain a consistent parameter set to be used for the description of

the hygroscopic and activation behavior of their HULIS samples. They show that for more complex systems this procedure is not applicable and ascribe these deviations to the non-ideal mixture of inorganic/water-soluble and HULIS fractions in the sample. The topic is certainly appropriate for publication in ACP. However, I have numerous comments that address the organization of the manuscript and the presentation of the results in view of prior studies in order to make more useful statement about the difficulties in defining HULIS properties. In addition, there are several technical comments that should be addressed.

Comments

p. 13775, l. 9-26: This part needs better organization: If HULIS comprise 60% of total organics, the fraction in WSOC should be higher. A growth factor of 1.08 points to HULIS that is rather insoluble and, thus, is not a fraction of WSOC. Can the differences in surface tension be related to the age/source of HULIS? It would be useful here to introduce terms like 'aged HULIS'; and 'fresh HULIS' as this concept will be useful later in the manuscript in order to explain the differences in the presented results to prior studies.

p. 13776, l. 12: The promise that a consistent parameterization for HULIS will be presented is a bit bold. Make clear that it only refers to the HULIS samples analyzed in the present study (but also see my comments below).

p. 13777, l. 4/5: What is the time particles spend in the tube? How do you make sure that they reach equilibrium diameter?

p. 13777, l. 18: Is the refractive index for HULIS used here based on a similar HULIS sample? How has the refractive index of the mixed sample been derived?

p. 13778, l. 20 ff: Are there any differences expected between the first and the second HULIS sample? Are there any differences in the total composition between 'spring' and 'summer' samples? Fresh organic are usually less water-soluble than aged ones. How

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did you make sure that the Aerosol-Water-Extract indeed contained HULIS?

Section 3.2: This section can be shortened. E.g., the first two sentences can be combined; the same could be done with the sentences in l. 18-22. The last sentence of this section is a major finding in this study. I suggest to state the differences in HULIS depending on age/source etc in the abstract of the manuscript.

p. 13787, l. 15-18: The only variable parameter in $\rho(\text{ion})$ is the osmotic coefficient. One reason why it might change with increasing dilution is non-ideal behavior (as stated here). Another reason could be simply the fact that HULIS contains organic acids that dissociate. Is there any information available on functional groups in the HULIS samples? Repeat here what parameters are included in $\rho(\text{ion})$ in order to clarify that compounds with partial solubility exhibit a constant $\rho(\text{ion})$ as soluble mass is not included.

Section 5.1: It would be useful to apply the procedure in Section 4.2. to other HULIS samples e.g.. based on the studies by Dinar et al., 2007; and Gysel et al., 2005. Comparing these values to those obtained in the present experiments would reflect the range of HULIS parameters for aged vs. fresh HULIS.

p. 13787, l. 19 (Table 2, resp.). What are concentrations [g/l] of HULIS at the point of activation? Are these concentrations realistic for typical atmospheric samples, i.e. when HULIS only comprise a small fraction of the total mass?

Section 5.2: There are previous studies that addressed experimental studies on the hygroscopic properties of humic acid/inorganic mixtures (Badger et al., Atmos. Chem. Phys., 6, 755-768, 2006; Sjogren et al., J. Aerosol Sci. 38, 157-171, 2007). How do these compare to the Aerosol-Water-Extract analyzed here?

Technical comments

abstract: Define HULIS

p. 13776, l. 17/18: HH-TDMA and LACIS have been defined before. (Check the

remainder of the manuscript for redundant definition).

p. 13776, l. 22: Add unit to supersaturations.

p. 13777, l. 6: RH has been defined before.

p. 13778, l. 5: remove for

p. 13778, l. 9: Replace where by were

p. 13785, l. 5: Replace maybe by may be

p. 13778, l. 10: Replace higer by higher

p. 13779, l. 13: Replace them by the

p. 13792, l. 11: Replace Khler by Köhler

p. 13792, l. 22: Replace refactive by refractive

p. 13792, l. 28: Replace 99,1 by 99.1&

Figure 3: Replace lowes by lowest

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