

Interactive comment on “Influences on the fraction of hydrophobic and hydrophilic black carbon in the atmosphere” by G. R. McMeeking et al.

Anonymous Referee #2

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This paper introduces a novel experimental technique to study the hygroscopicity of black carbon (BC) soot aerosol as a function of aging. The technique combines the hygroscopicity tandem differential mobility analyzer with the soot photometer as the particle detector to obtain detailed information of the mixing state of BC-containing particles that few other methods can provide. This study is not routine and presents several interesting ideas that make the manuscript interesting to read. Among those ideas are the technical approach itself and a way to separate the contributions from the particle size, coating thickness, and hygroscopicity to the particle CCN ability.

The first half of the manuscript, including the Results section, reads nicely because it is well organized and clearly written. The Discussion section, however, raises a number of questions. First of all, the discussion on page 929 is very implicit. This reviewer had

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a hard time to understand what the authors tried to deliver. This section needs careful re-writing. A few simple formulae to support the written statements are required, such as the relationship between D_{bc} , D_{mob} , and the effective density. The coating thickness needs to be defined, i.e. whether it is based on volume equivalent, mass equivalent, or mobility diameters. Second, having understood the text on page 929, I realized that the cornerstone idea that supports the rest of this study is unjustified. Specifically, it is incorrect to derive the coating thickness from the difference between SP2-measured rBC diameters of non-hygroscopic and hygroscopic BC aerosol fractions because these two are largely unrelated. All they have in common is the mobility diameter according to which they were classified by the DMA. The hygroscopic BC fraction was not necessarily produced via coating the non-hygroscopic one. They are externally mixed BC aerosols of (possibly) different origin. To derive the coating thickness, one would have to perform the volatility analysis of the HTDMA-classified aerosol, using a third DMA in combination with the CPC and SP2, to measure the difference between mobility diameters of fresh and heated particles of the same hygroscopic growth factor. Considering the fractal morphology of fresh and partially processed soot, an instrument capable of non-destructive measurement of the particle mass (density), such as the aerosol particle mass analyzer or the couette centrifugal particle mass analyzer, is required instead of the third DMA to quantify the coating thickness (see for example, J. Pagels et al., *Aerosol Sci. Tech.* 43, 629, 2009; A. F. Khalizov et al., *J. Geophys. Res.* 114, D05208, doi:10.1029/2008JD010595, 2009).

I do not think that this manuscript in the present form is publishable and do not see an easy way of improving this manuscript to make it publishable without additional measurements. A number of other minor issues that need to be corrected are outlined below.

P. 923, L. 18: D_{BC} is introduced as “volume equivalent diameter” whereas in the rest of the manuscript it is referred to as “mass equivalent diameter”.

P. 924, L.7: What is the physical meaning of A in equation (1). Also, surface tension

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(sigma) is referred to, but does not appear anywhere in the equations.

P. 925, L.6 : Not always critical supersaturation can be obtained from hygroscopicity measurements at subsaturated relative humidity (RH). For instance, as many organic acids do not deliquesce below about 99% RH, their kappa factor determined at 90% RH would be essentially zero.

P.927, L.2-6: The way “non-hygroscopic mode” is used here makes this sentence somewhat confusing.

P. 928, L.7: Define abbreviation “a.g.l.” (above ground level?)

Figure 8: If number fractions shown in (a) and (b) summed together amount to 1, then either of them can be removed.

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