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Atmospheric Measurement Techniques Discussions

Interactive comment on "The white-light humidified optical particle spectrometer (WHOPS) – a novel airborne system to characterize aerosol hygroscopicity" *by* B. Rosati et al.

Anonymous Referee #2

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Retrieval of aerosol hygroscopic growth factors by comparing dry and wet size of aerosol particles is a common approach used to characterize the hygroscopicity of aerosol particles and therefore an airborne system capable of delivering such data would be highly welcome to atmospheric community. A simple and easy to use system that is described in this manuscript comprises mainly commercially available instruments (DMA and OPC) and is relatively fast compared to HTDMA. Another advantage over the HTDMA is the possibility to measure wet diameters of the size that is relevant for the optical properties of the hygroscopically grown aerosol particles. The manuscript provides all technical and theoretical details needed to follow the discus-

sion and the language is comprehensible. Some sections, however (the description of the retrieval algorithm, 2.2.2 and counting efficiency, section 3.1.1) are somewhat lengthy and would benefit from rewriting them in more precise and curt form. The operation principle of WHOPS is not new but well applicable, as demonstrated by the numerous publications that have emerged from the same group of authors and elsewhere recently. The novelty of the instrumental setup is ensured mostly by airborne nature of its application, and therefore I would recommend the manuscript to be accepted for publication, provided that the critical issues I address below are thoroughly discussed. Below I summarize my major concerns of a general nature, followed by a brief discussion of specific issues.

General comments

The method of growth factor determination described here has two steps. First, the index of refraction has to be determined from comparison of mobility and optical diameter of dry particles, and then the optical diameter of wet particles is measured using the response curve calculated for size dependent index of refraction. However, in this particular realization I see two major limitations of the method applicability.

1. As authors admit themselves, the size range of aerosol particle and the range of growth factors accessible with the described setup is pretty narrow. This size range is limited on the lower side by the smallest detectable dry particle size and by ambiguity of the WELAS response curve for growing aerosol particle (e.g., with size dependent index of refraction, as demonstrated for 300nm dry particles, figure 4 and discussion on the page 7334). On the other side, the largest particle selectable with the DMA sets the upper limit of the dry particle size to well below 1 μm (depending on the DMA model and flow conditions).

However, the chemical composition and therefore hygroscopicity of atmospheric aerosols is size dependent, as discussed, for example in (Zieger et al. Atm. Chem Phys. 2013). Both fine and coarse modes of atmospheric aerosol may contain low

hygroscopic (organics, mineral dust) or highly hygroscopic aerosols (inorganic salts, marine aerosols). With the setup presented here the fine mode is only partly accessible and the coarse mode is not accessible at all. I don't see how the measurements of a growth factor for a single dry size of 500 nm can be used to characterize the hygroscopic behavior of the total aerosol content within a certain air mass. Can the mean value of kappa obtained from such measurement be used for assessment of the optical properties of the total aerosol loading, like enhancement of scattering coefficient? This issue is actually addressed in the section 3.2 (First field measurements, pages 7345 - 7346 and Figure 11) of the manuscript, where authors make an attempt to infer the external mixture of the aerosol based on the WHOPS measurements of the growth factors. For example, the statement "On average, about 15% *of the particles were non- or slightly-hygroscopic with GF< 1.1, which could be explained with externally mixed dust (e.g. Herich et al., 2009), soot (e.g. Tritscher et al., 2011) or biological particles (e.g. Després et al., 2012)*" actually should read "15% of 500nm dry particles were non- or slightly hygroscopic. . ." and so on, which makes sense if considering that atmospheric soot, especially aged, has the size distribution centered around 100nm, and biological particles (bacteria, pollen and leaf residuals) have mostly supermicron size. Thus what left is the mineral dust which concentration in the measurement region depends strongly on the on the seasonal variability of the source fluxes.

Having said that, I agree that the data obtained with WHOPS as a part of a larger measurement system, including SMPS, AMS, APS, CCNC, wet nephelometer, etc. (you name it) would definitely help parameterizing the aerosol hygroscopicity needed for accurate characterization of aerosol optical properties. These additional measurements, though available on board of Zeppelin NT, are not used for the WHOPS data analysis, which I consider as a drawback and recomend discussing in the revised manuscript.

2. The described approach works for soluble aerosols (if the limitations mentioned above are to be lifted). The calculation of the response curve based on the assumption of effective index of refraction would not work, however, if the aerosol contains

non soluble particles, for example dust containing small fraction of soluble material, or aged soot. In this particular case amount of soluble material and therefore effective index of refraction cannot be inferred from mobility size measurements of a dry particle. Therefore, the problem becomes poorly constrained and a new optical model (coated sphere) has to be applied to interpret the observed growth rates. Introduction of a more complex optical model would in its turn require additional input parameters, that may be solved by using complementary measurements of aerosol size distribution, chemical composition and optical properties.

Specific comments arranged in order of appearance:

Section 2.1. Page 7327 lines 5 to 10: What is the purpose of a growth chamber? The time needed for hygroscopic aerosol particle to reach the equilibrium diameter is on the order of a second, which is probably ensured by the residence inside the humidifier anyway. At the same time, the growth chamber introduces a circulation cell, so that the actual growth time for individual particles can be anything from one second to minutes. It is not important for hygroscopic aerosols, but for bad guys like SOA the residence time can trigger kinetically limited effects that you probably don't want to deal with (and which will be difficult to characterize).

Section 2.2.1. I am somewhat irritated by the description of WELAS calibration procedure. It is stated that WELAS is not reporting the raw pulses heights (voltages), therefore they had to be calculated from the measured optical diameter values using the "time dependent voltage calibration factor Cv" (equation 7). At the same time, this factor itself is defined as a ratio of raw voltage values taken for calibration dust after certain time t (equation 8). But the raw voltage value is not accessible in WELAS (your statement), so to determine the raw voltage you need to know the value of Cv in the first place! Is there not a contradiction somewhere? It would be really helpful to rewrite this part more clearly.

Section 2.2.2. Page 7332: You cite (Kiselev et al. 2005) as a reference for retrieval of

index of refraction algorithm (inverse task), but this paper actually describes the white light OPC (which is incidentally called "WOPS") built for study of hygroscopic growth of aerosol particles in LACIS (Leipzig aerosol and cloud interaction simulator). This is an appropriate reference in different context, would be perfectly OK in the introduction section, but RI retrieval is not discussed there. Please correct.

Page 7334 and discussion of Figure 4: is there any particular reason why the refractive index of dry particle (the red curve in the figure 4) was chosen to be 1.45 and not 1.5 or even 1.55 which is more representative for atmospheric aerosol? Would you expect any ambiguities on the response curve in the range of the small growth factors in case of larger RI?

Section 3.1.2. Page 7340: The discussion of optical size measurement uncertainty is based on the propagating of 15% input error of scattering cross section and the response curve calculated with the FIXED index of refraction (RI). However, the optical size of the wet aerosol is calculated with the help of response curve calculated with account for gradual decrease of RI as aerosol particle takes up water. Hence these response curves and not the ones for fixed RI have to be used for error analysis. These response curves are less steep and for some combinations of dry size and refraction index are even ambiguous. For that reason I would expect larger errors than reported in the manuscript. For example, if we consider the green curve in figure 4b, the GF is almost constant between 300nm and 400nm, which is 33% error even if we forget about the initial 15% error of scattering cross section.

Caption of figure 8: In the text of manuscript (page 7340 line 20) the value of 15% uncertainty in the scattering cross section is used to evaluate the error of GF determination and obtain 9% on average. In the caption to figure 8 the "fixed uncertainty of $\pm$9% for the measured scattering cross sections" is mentioned. Which statement is correct?

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