Reply to Referee #2

We would like to thank the referee for his/her valuable and detailed comments. Our answers to the comments are given below in blue letters, while the referee comments are given in black italics. Additionally we added the changes made in the revised manuscript in blue bold letters.

- *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.*

  We revised the mentioned sections for a better comprehension. However, it was not possible to shorten them significantly since their content is essential for the manuscript, given the fact that this is a technical manuscript.

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.

The WHOPS provides measurements of aerosol hygroscopic properties in a size range that gives a strong contribution to the light scattering by atmospheric aerosols and that is not covered by other methods such as the HTDMA. The size range accessible by the WHOPS instrument is indeed restricted, more than anticipated when initiating the construction of this instrument. This is the very reason we highlight both the strengths as well as the limitations of the WHOPS in this technical paper. The revised manuscript has been adapted along the lines suggested by the referee:

Revised text: Page: 7345; line: 26-29:
On average, about 15% of 500 nm dry particles had 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). Mineral dust might be the most likely candidate because externally mixed soot particles are typically smaller and biological material is typically larger. However, we do not have supporting chemical analyses to support this speculation.

2. *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 recommend discussing in the revised manuscript.*

We simply repeat here our reply to the 5th comment by referee #1, who asked for using the additional measurements to support the interpretation and validation of the WHOPS measurements: The main topic of this paper is the technical description of the WHOPS instrument including technical design, data analysis procedures, uncertainty analyses and validation of accurate measurements – this was the very reason to choose AMT as a target journal for this manuscript as opposed to choosing a journal with a stronger focus on results and interpretation of atmospheric measurements such as e.g. ACP. Validation of new instruments is commonly done by the measurement of samples with known properties and behavior. In this case, PSL spheres and pure ammonium sulfate particles were used to validate the accuracy of optical sizing and hygroscopic growth factor measurements. Closure experiments, where the instrument response to a sample with a priori unknown behavior is compared against predicted behavior that is inferred from independent measurements of e.g. sample composition and with using a theoretical model, can only be used for instrument validation under certain conditions. The closure calculations proposed by the referee do not allow drawing any firm conclusions about the instrument performance; simply because the chemical characterization by the AMS is insufficient concerning a complete speciation (e.g. sea salt and dust components are not quantified). Therefore, we did not include such closure calculations. Nevertheless, the ambient measurements performed by the WHOPS were added to the manuscript to give an example on the kind of data that can be gathered during airborne measurements with the WHOPS. Comprehensive analyses of the aerosol and gas phase properties observed during PEGASOS flights in the context of air mass properties, vertical layering etc. are the subject of papers that are currently in preparation.

3. *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.

We agree and have tested a Mie code accounting for coated spheres. The results are added to the revised manuscript:

Page: 7333; line: 5:
The homogeneous sphere model used to generate the look-up-table, which accounts for the change in the refractive index caused by the absorption of water, is considered to be the most suitable choice for the analysis of the WHOPS measurements. A coated sphere model is expected to be more accurate for particles that contain insoluble material. However, in the size range between 300-500 nm dry diameters, the insoluble volume fraction is expected to be very small in the majority of the particles. Furthermore, the insoluble volume fraction is not available on a single particle basis, nor can it reliably be derived from the measured scattering cross section of the humidified particle. Sensitivity analyses comparing the homogeneous mixture to the core-shell model revealed that the discrepancies in the retrieved GF are generally less than 10% for particles with a soluble volume fraction of >70%. The largest discrepancies (~20% in GF) occur for particles containing a large fraction of insoluble material (70-90%) in the range of 1.1 < GF < 1.5.

4. ***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).*

Sjogren et al. (2007) stated that residence times >40s were required to reach equilibrium depending on the chemical composition. Therefore, we chose to include a growth chamber in order to give the particles residence times of ~10s in the humidifier itself and ~10s in the growth chamber. The timespan that the particles are exposed to high RH can indeed change the structure and properties of the particles but with regard to the GF measurement this should still allow for a clear separation of organic and inorganic material (see e.g. Duplissy et al., 2011 or Tritscher et al., 2011).

Revised Text Page: 7327; line: 8-10:
.. the dried and size-selected particles pass through a custom-built humidifier and finally enter a growth chamber with a residence time of about 10s. According to Sjogren et al. (2007) this time is needed in order to reach equilibrium.

5. ***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.

We made the following modifications in the revised manuscript:

Page: 7330, line 16:

In practice, the software calculates the voltage calibration factor internally by relating the raw voltage signal measured for quasi-monodisperse Caldust (calibration dust with a diameter of 850nm and an index of refraction similar to PSL; provided by the manufacturer) at a certain time t (user calibration), to the raw signal measured at a reference time t0 (initial factory calibration): ...

Page: 7332; line: 5-13:

Unfortunately, the WELAS instrument does not report the raw voltages and calibrated voltages corresponding to the reported optical diameter values. However, the calibrated voltages can be inferred from the optical diameters using the instrument's preset PSL-based calibration curve (D_{PSL} vs. V_{cal}; provided by the manufacturer). In a second step the raw voltages can be reconstructed from the calibrated voltages using Eq. (7) and the voltage calibration C_v from the last user calibration (stored in the instrument software) as noted during the user calibration.

6. ***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.*

Revised text: **Page: 7325; line: 4-7:**

The measurement principle is based on a combination of a DMA and OPSS (optical particle size spectrometer) which has been utilized also in previous techniques like for the LACIS (Leipzig Aerosol Cloud Interaction Simulator; Kiselev et al., 2005;Stratmann et al., 2004).

Revised text: **Page: 7332; line: 18-20:**

A similar approach was described in previous articles like in Flores et al., 2009 for WELAS measurements or in Hand and Kreidenweis (2002).

7. ***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?*

In the original manuscript we had chosen a scenario close to findings of the WHOPS (for the flight presented in Sect. 3.2 we found m=1.42±0.01). For the revised manuscript, we have added curves with m_{dry}=1.55 to Fig. 4 in order to bracket a range of possible indices of refraction.
This reveals that ambiguities occur for higher m_{dry}, as anticipated by the referee. We have improved the whole analysis and discussion of the sizing uncertainties and ambiguities by adding the new Fig. 9 with associated discussion to the revised manuscript.

The discussion of Fig. 4 in Sect. 2.2.2 has been adapted according to the additional curves in the revised Fig. 4. Besides, Sect. 2.2.2 has also been shortened according to the first comment put up by this referee (see above).

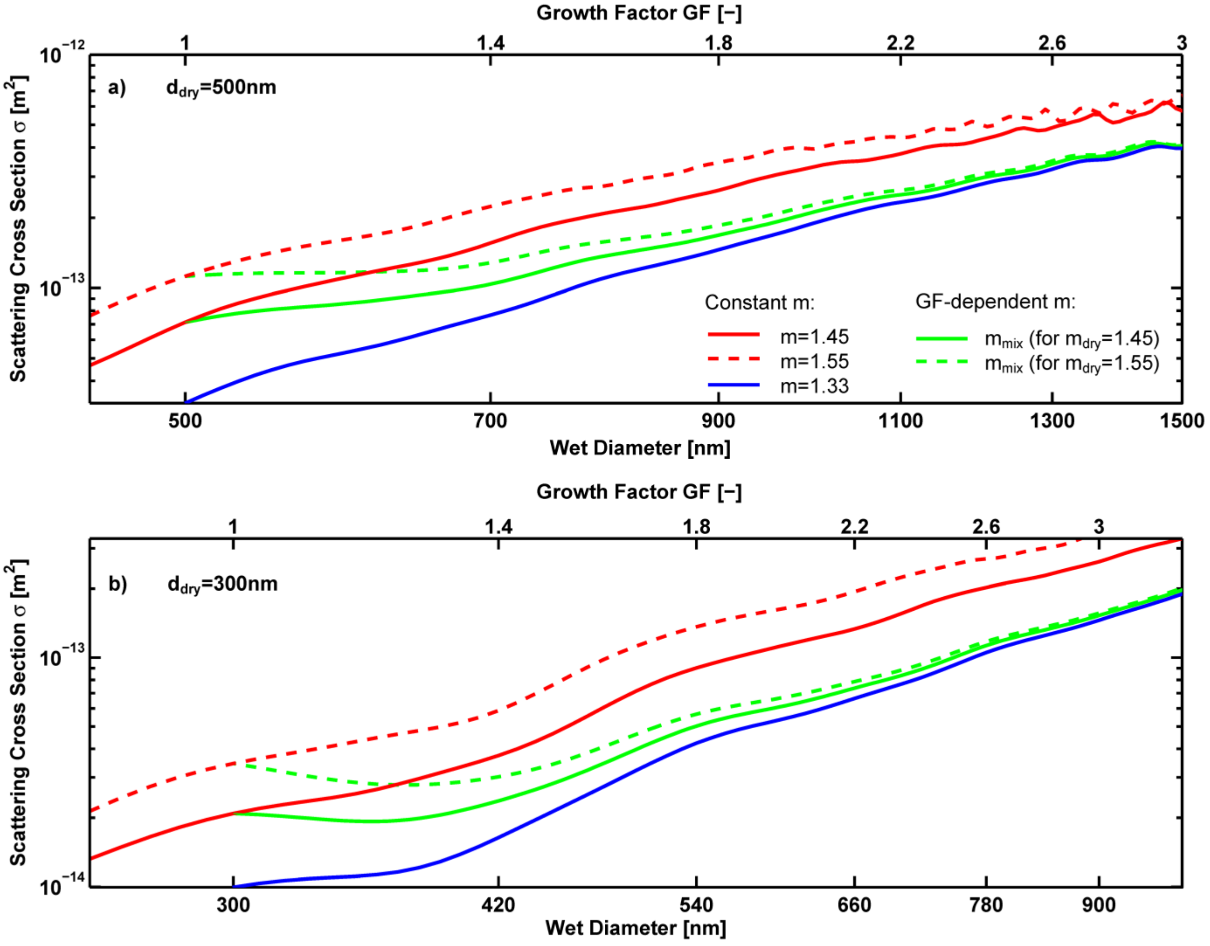

Figure AR1: New version of Fig. 4 with additional curves for m_{dry}=1.55.

Adapted Figure caption:

Calculated scattering cross section as a function of particle diameter (hygroscopic growth factor) for solution droplets (green lines) formed on particles with a dry diameter of (a) 500 nm (b) 300 nm. A volume weighting mixing rule is used to determine the index of refraction of the solution droplet (see Sect. 2.2.2 and Fig. 3). The Mie curves for pure water droplets (m_{H2O}= 1.333; blue lines) and particles with a constant index of refraction m_{dry} = 1.45 (solid red lines) and m_{dry}= 1.55 (dashed red lines) are also shown as a reference.

8. ***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.

Indeed, the approach to estimate the sizing uncertainty for grown solution droplet was not correct. This has been corrected and it is now addressed with the new Fig. 9 and associated discussion (for more details please see response 2 in the author reply for Referee #1).

9. **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 ±9% for the measured scattering cross sections” is mentioned. Which statement is correct?

There was an error in the caption of Fig. 8: the uncertainty in the scattering cross section is ±15% while the average uncertainty in the optical diameters for the stated indices of refractions amounts to ±9%.

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