Referee #2:

General comments:

My main concern is in the verification of the summation method (SM), which was used to extract the size-resolved growth factor. In this manuscript and in previous papers (Eichler et al., Atm.Environ, 2008; Achtert et al., J. Geoph. Res., 2009 and Birmili et al., Boreal Environ.Res., 2009) the authors state that DGF is representative of the metrics of atmospheric particle hygroscopic growth, but no experimental confirmations have been submitted. For the beginning I would suggest to compare HDMPS/TDMPD results with regular size-selected HTDMA measurements for different field conditions. It will then be possible to quantify the potential errors of the method used, and therefore more convincingly explain correlations between the aerosol particles hygroscopicity and their chemical composition.

We agree with your impression that the justification of the summation method has been incomplete. The main reason is that a technical paper on the H-DMPS/summation method has been conceived by us, but never submitted for publication. To disperse your concerns about the HDMPS/summation method and to illustrate its accuracy, we present to you unpublished results from that manuscript in the supplementary material added to the revised version of this manuscript.

Specific comments**:**

p.14299, line 23, Introduction Despite recent improvements in understanding the water uptake characteristics of organic-containing particles, in situ measurements are sparse.

I can't agree with this statement. See, for example, size-resolved hygroscopic measurements recently published by Kammerman at al.(ACP, 10, 10717, 2010); Ye et al.(Atmos. Environ., 99, 353, 2011), Fors et al. (ACP,11,8343,2011); Jurányi et al.(ACP, 13, 6431, 2013) and Laborde et al.(ACP, 13, 5831, 2013). These articles should be discussed in the context of this work especially the last two papers, which present the size-resolved HTDMA results obtained in Paris within the same time frame as the MEGAPOLI European project.

We changed "Despite recent improvements in understanding the water uptake characteristics of organic-containing particles, more in situ measurements are needed. Combining measurements of particle composition and their hygroscopic behavior, laboratory studies and hygroscopic growth models can be compared with field measurements." to "More in situ measurements with coupled measurements of particle composition and their hygroscopic behavior are needed. Conducting more of those measurements, laboratory studies and hygroscopic growth models can be associated with field measurements."

HDMPS and HTDMA measurements are not entirely comparable because of their different approach. Additionally, Jurányi et al. (2013) put a different emphasis and did not include data from chemical measurements. Therefore, we consider our paper a good complement to the paper of Jurányi et al. (2013).

A comparison can be drawn between the time series of the hygroscopic growth factor, Fig. 5 in Jurányi et al. (2013) and Fig. 6 in our paper. Even though we consider larger diameters (285 nm in comparison to 165 nm) the temporal evolution shows an increasing growth factor on 6th February 2010. From then on the growth factor is obviously higher.

What is also worth mentioning is the fact that the HTDMA data show a more internally mixed aerosol from 6th February 2010, because most particles are growing with a factor of approximately 1.6. This result indicates that the summation method can be successfully applied.

Laborde et al. (2013) also put a different emphasis on the measured data in Paris. Still, their Figure 7 comprises the same measurement interval as ours during the winter campaign and their examined diameter of 265 nm is close to ours with 285 nm as depicted in Figure 6. Some striking points in the time series of the growth factor are found in both time series. At the beginning of the campaign, a peak at the beginning of 17th January is followed by a sharp decrease with a minimum at the end of 17th. Subsequently, the time series show a general increasing trend of the growth factors until the 20th January. A sudden drop with immediate increase back to a value around 1.6 at the 21st is also remarkable. A minimum directly before the beginning of the 23rd around 1.2 is followed by a maximum at the end of the same day. Also, there is agreement in the second half of the campaign, e.g. a minimum at the end of 1st February and the following maxima in the beginning of 3rd and 4th and on 7th February. All three studies are good complements to each other, because even though they were measured at different sites in Paris, general trends of the humidified time series are found.

p.14299, line 26 The standard method for calculating hygroscopic growth from the aerosol chemical composition is based on volume-weighted water uptake by the individual chemical constituents, the so-called Zdanovskii–Stokes–Robinson mixing rule.

The ZSR rule is based on the water mass additivity (not volume). To convert this rule into a volume scale the volume additivity assumption is additionally used. This sentence should be reworded.

We reworded this sentence.

p.14302, line 10 2.3.2 The humidifying differential mobility particle sizer
p.14304, line 9 3.1 Determination of hygroscopic growth factors

These sections are actually complete copies of the corresponding sections in the already published papers (Eichler et al., Atm.Environ, 2008; Achtert et al., J. Geoph. Res.,2009 and Birmili et al., Boreal Environ.Res.,2009). There is no sense to repeat them again. Authors need only focus on the changes that have undergone measurement technique and data processing compare with earlier studies.

Of course, these sections repeat some of the content in the mentioned papers but they are not complete copies. If these sections were excluded or shortened it would be hard to follow the manuscript. The summation method is complex, so a detailed explanation is not redundant. In section 2.3.2 only the necessary fundamentals are included.

p.14305 line 26 “However, as long as no stepwise changes with particle size are observed in the average hygroscopic growth factors, the SM remains valid”.

As noted previously this statement must be quantitatively supported. It is necessary to indicate the uncertainty of the summation method using either independent size-resolved HTDMA data or (at worst) numerical experiments with aerosol models that are typical for urban and regional conditions.

Actually, our formulation about "stepwise changes" was somewhat misleading. Basically, we assume the summation method to be valid as long as changes in overall hygroscopicity (which includes the two sub-aspects external mixture as well as the absolute value of the growth factor) are modest compared to changes in particle diameter. An extreme case, which would bring the summation method to a certain breakdown, would be a monodisperse particle population on top of a wide size distribution with entirely different hygroscopic properties. Such a breakdown would, in any case, be seen in a sharp step in the DGF curve resulting from the allocation of number size distribution segments. There are several arguments against such a failure to be relevant in the case of our MEGAPOLI data set:

1) We did not observe sharp steps in any DGF curve resulting from the allocation of number size distribution segments.
2) The overall size-dependent changes in DGF were monotonous, steady, and rather modest during MEGAPOLI, typically increasing from a campaign average 1.1-1.2 at the lower end of the evaluated diameter range around 80 nm to 1.3-1.4 at the upper size distribution end (our Fig. 5).
3) The urban particle number size distributions during MEGAPOLI were flat and without major contours, suggesting the presence of an aerosol mixed from multiple sources, and from sources exhibiting wide particle modes.
4) In a continental aerosol, the particle modes typically exhibit a minimum spread (sigma of 1.4 and larger). In MEGAPOLI, concretely, black carbon was found to exhibit two rather broad modes in the sub-µm range (Healy et al., 2012).
These arguments will hopefully disperse the concerns that have risen during the review of the article.

We accordingly reformulated the text:

“The summation method is assumed to be valid as long as changes in overall hygroscopicity, which includes the two sub-aspects external mixture as well as the absolute value of the growth factor, are modest compared to changes in particle diameter.”

p.14310, line 28, “…. (Fig. 5a) with a volume fraction of 48 %”, while Figure 5a shows the value of 49 %.

The value of 49 % shown in Figure 5a is right. We changed the corresponding text to "with a volume fraction of 49 %".

p.14311, line 3 and Fig.5. RH? Please specify the RH used both in the text and in the figure caption.

We included this piece of information in the text "The size dependence of the corresponding DGF at an RH of 90 % is also represented in Fig.6a; as well as in b-d". We also added the information in the figure caption of Fig. 5 (Fig. 6 in the revised version).

p. 14312, line- p.14312, line 8.

The explanation of the discrepancy between the measured and calculated growth factors due to nitrates do not look convincing. In winter time from 16 to 20 and from 21 to 24 January the measured DGF are relatively well reproduced by the calculated GF, and the data presented in Fig.1b clearly show that nitrates do not fluctuate less than on other dates.

Additionally, expressions like "..strong diurnal variation.."(p.14310, line 15).., "..strongest variation in this time interval.."(p.14312, line17), "..the DGF shows a much higher variability in winter than in summer," (14313, line 2) etc., should be accompanied by respective statistical estimates. The graphic information presented in Fig.1 and Fig.6 does not permit estimating the validity of the proposed relationship between nitrate mass and aerosol particle hygroscopicity. In the context of this problem, I believe that a more profound study of the uncertainties caused by the experimental method used and model ZSR calculations needs to be included before the results presented in Section 4.5 can be compared.

To clarify the statement p.14310, line 15 we added another sentence: "In the course of the day the nitrate mass variation between daily minimum and maximum was about 4 $\mu g\ m^{-3}$ in the first, 13.3 $\mu g\ m^{-3}$ in the second and 6.6 $\mu g\ m^{-3}$ in the third day, whereas the nitrate mass varied by 1.8 $\mu g\ m^{-3}$ in maximum excluding these first days of the campaign."

We changed the expression in p.14312, line 17 to "…the DGF show a much higher variability in winter than in summer. The maximum deviation between the measured and the predicted GF is 37.1 % in winter and 27.1 % in summer. Also, the average value of the deviation of the wintertime data is 8.5 %, and thus higher than in summer with 6.5 %. For the wintertime data, the correlation coefficient is decreased to R=0.52 (Fig. 8c).

We changed "This is also evident in the DGF measurements of 285 nm particles which show the strongest variation in this time interval (Fig. 6a). Furthermore, the deviation of the measured and the calculated DGF is strongest during this period, whereas the rest of the time series fits well if one considers the use of a simplified growth model." to "This is also evident in the strongest deviation of the measured and the calculated DGF during this period (Fig. 8a), which has a mean value of 10.2 % for the beginning of the campaign and 5.4 % when excluding this period. Therefore, the rest of the time series agrees well if one considers the use of a simplified growth model."

Following the suggestion of reviewer #3, an estimate of the relationship between nitrate mass and aerosol hygroscopicity has been made (cf. Ref #3, 8./9.). We also tried to correlate the absolute NO_3 mass and the computed growth factor of the organic fraction. In both campaigns it is to say that for higher values of the NO_3 mass (> 3 $\mu g\ m^{-3}$ for summer and > 8 $\mu g\ m^{-3}$ for winter) the values of the organic growth factor calculated from this correlation are clearly smaller than unity, in some cases even negative. However, this correlation made clear that high nitrate is not the only factor for the discrepancy between measured and predicted growth factor. In some rare cases, the organic GF required to compensate for the difference between measured and computed growth factor is smaller than unity also when the nitrate mass is low.

The validation of the used H-DMPS/summation method can be found in supplementary material added to the revised version of the manuscript.
The H-DMPS/summation method and H-TDMA growth factors have been compared by using ammonium sulphate test aerosol under laboratory conditions. An inter-instrumental comparison for RH = 88% confirmed an agreement of both methods within +/- 0.05 in hygroscopic growth factor.

p.14326, Fig.2.

In Fig.2b the correlation coefficient has a negative value (- 0.52). The R, the fit line and fit equations should be disclosed in the figure caption.

You are right; it has to be a negative value. We included the linear fit equation and the corresponding correlation coefficient in the figure caption.

p.14330, Fig.6. : All symbols in Figs. 2b and 2c must be explained in the figure caption. What is meant by corr.low NO3=0.76? In Fig.2b, d the correlation coefficient was denoted as "R." Do red and blue points refer only to Fig.6b?

We changed the legend in the figure to make it clearer. Now, the high NOx periods are indicated by date in the legend and the figure caption labels that: "Red dots in the scatter plots denote periods under high NOx conditions."

Also, a small supplement has been added on page 14313: "Just like in summer, the correlation between measured and calculated growth factors improves from R = 0.52 to R = 0.63 if these periods (indicated in red in Fig. 8d) are not taken into account."

Technical comments

p.14311, line 18. …saltis reduced

We changed that, thanks.

References:

Healy, R. M., Sciare, J., Poulain, L., Kamili, K., Merkel, M., Müller, T., Wiedensohler, A., Eckhardt, S., Stohl, A., Sarda-Estève, R., McGillicuddy, E., O'Connor, I. P., Sodeau, J. R., and Wenger, J. C.: Sources and mixing state of size-resolved elemental carbon particles in a European megacity: Paris, Atmos. Chem. Phys., 12, 1681-1700, doi:10.5194/acp-12-1681-2012, 2012.

Jurányi, Z., Tritscher, T., Gysel, M., Laborde, M., Gomes, L., Roberts, G., Baltensperger, U., and Weingartner, E.: Hygroscopic mixing state of urban aerosol derived from size-resolved cloud condensation nuclei measurements during the MEGAPOLI campaign in Paris, Atmos. Chem. Phys. Discuss., 13, 2035-2075, doi:10.5194/acpd-13-2035-2013, 2013.

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