Answer to Referee #1:

The authors appreciate the time the reviewer have spent in assisting us to produce a high quality, understandable publication. All the requested corrections and suggestions are addressed and introduced to the revised version of the manuscript.

Comment: Page 15516, lines 4-5: when presenting the HR-ToF-AMS data, it would be good to add a brief statement on collection efficiency (CE) that I assume was applied for quantification. Did the author apply the standard CE = 0.5? Did they calculate their own CE? Please clarify on this subject.

Reply: A collection efficiency of 0.25 was applied compared to the volume concentration of DMPS. The CE is lower than the standard 0.5, which could be interpreted by the scavenged aerosol during long residence time in the sampling line (about 3m copper line, diameter 6mm) without extra suction flow. This was added in the text accordingly.

Page 15520, lines 17-20: the authors make the correct observation that the larger discrepancy between measured and calculated HGF for the smallest particles sizes is likely due to the fact that the AMS doesn't collect efficiently particles smaller than 50-60 nm due the aerodynamic lens design and cut-off. Perhaps add a brief statement somewhere here to explain this to the non-AMS expert reader; for example, you can re write as "...which is dominated by accumulation mode particles due to the cut-off of the standard aerodynamic lens for particles smaller than 80 nm (Williams et al., 2012)".

Reply: The text was corrected as: The HR-AMS measures the bulk chemical composition of submicron aerosol particles, which is dominated by accumulation mode particles due to the cut-off size of the standard aerodynamic lens for particles smaller than 80 nm (Williams et al., 2012).

Page 15520, lines 22-25: the authors should also note that the data on the y-axis of Fig 5 have a positive offset. Is this also related to the fact that the constant HGF and / or the mixing rule assumptions might be inadequate or there is something else going on? Later in the paper (page 15521, lines 10-14) when discussing the results of Figure 6, the authors show that the correlations improve when applying an O:C dependent HGF as presented in Massoli et al. (2010). Is the data offset in the y-axis also improving? Based on the slope values, it seems to me that that is the case, and that the offset is reduced even further when the authors apply their own parameterization. Perhaps a little phrase to point this out would be a nice addition given that the slopes are not shown in Figure 6.

Reply: We thank the reviewer for pointing this out. However, we don't know how to draw the conclusions from the offsets becoming closer to zero.

All the data points in Fig. 5 go through the 1:1 line, so we cannot conclude that there is a systematic discrepancy between HTDMA-measured and AMS-derived HGF. For the same reason, it is also difficult to give argument on the mixing rule assumptions based on the offset. From Fig. 6, the reduced offset was due to the improved slope when keeping the root-mean-square deviation (RMSD) minimum. Hence, we rely more on slope of the fitting line or R^2 value.

Figures 5,6,7: for extra clarity, the captions of these Figures should add that the dashed line is the 1:1 line, given that the slopes are not shown.

Reply: The caption of Fig. 5, 6, 7 was corrected in the revised manuscript accordingly.

Reference:

Willians et al., Characterization of an aerodynamic lens for transmitting particles greater than 1 micrometer in diameter into the Aerodyne aerosol mass spectrometer, Atmos. Meas. Tech., 6, 3271–3280, 2013, doi:10.5194/amt-6-3271-2013.

Answer to Referee #2:

The authors appreciate the time the reviewer have spent in assisting us to produce a high quality, understandable publication. All the requested corrections and suggestions are addressed and introduced to the revised version of the manuscript.

Major comment:

This study makes extensive use of O/C elemental ratio measurement with a HR-AMS. In a recent publication (Canagaratna et al., Atmospheric Chemistry and Physics 15, 253-272, 2015), such measurements have been evaluated by the developers of the AMS method. They conclude that an "Improved-Ambient" method provides a more accurate and precise measure of O/C ratio than the commonly used "Aiken-Ambient" method. The authors of the current manuscript should take this publication into account, since they base their study almost entirely on O/C measurements.

The authors should explicitly state the method used to determine O/C measurements (I'm guessing Aiken-Ambient) and put these measurements in the context of the Canagaratna publication. They should confirm that the Massoli paper they reference their results to performed the calculation in the same way.

Given the potential for confusion about O/C ratio going forward, I think it is highly advisable that the authors provide results using both calculations for O/C ratio (the current set could be included in the main manuscript and the alternative method could be put in supplementary information). I doubt that using Improved-Ambient vs. Aiken-Ambient will change the main conclusions of this manuscript, but given the importance of O/C ratio to this study, the authors must explore this possibility fully. Alternatively, the authors may want to recast their results in terms of carbon oxidation state (OSc), since the Canagaratna paper suggests that this is a more robust output of AMS measurements than O/C ratio.

Reply: We have reanalyzed the data using the new method by Canagaratna et al (2015) (Improved-Ambient) for O/C as suggested by the reviewer.

Figure 1 below repeats the comparison of derived HGF with measured ones, coloring with the O:C ratio, obtained from the 'Improved-Ambient' method. The new O:C ratio gave the same slope and the R^2 values as the ones in Fig. 5 in the manuscript. Hence, the main results from the paper do not change.

In Figure 2 and Figure 3 below (corresponding to Fig. 6 and Fig. 7 in the manuscript), new parameterization values are fitted using the new O:C ratio. The fit with the new method only marginally improved the results compared with the slope and R^2 values calculated with the old method.

In our paper, our results are compared with the results of Massoli et al. (2010), which used the 'Aiken-Ambient' method to estimate the O/C. For this reason we will keep the results from the old method and in addition we will briefly discuss the use of the new method in the main text.

Carbon oxidation state (OSc) is a better proxy for particle's oxidation level of AMS measurements than O:C ratio. Hence, we have reanalyzed the data using OSc instead of O:C. The results using OSc are shown in Fig. 4, Fig. 5 and Fig. 6, which are corresponding to Fig. 1, Fig. 2 (lower panels) and Fig. 3 using O:C, respectively. Comparing with the ones in Fig. 2 and Fig. 3 using O:C, the new fit does not improve the slopes and R^2 values using OSc. Hence, the main conclusions do not change.

Minor comments:

Do the nucleation mode particles have a higher sulfate volume fraction than Aitken/accumulation mode particles, and could this be related to the higher correlation between nucleation mode HGF and gas phase sulfuric acid concentration? Inorganic volume fraction is a less informative parameter for this correlation since it includes species other than sulfate.

Reply: it is possible that nucleation mode particles have a higher sulfate volume fraction than Aitken/accumulation mode particles. However, currently HR-AMS cannot measure the sulfate fraction in particles below 50 nm. Hence, we cannot directly conclude that the higher correlation between nucleation mode HGF and gas phase sulfuric acid concentration is due to the higher sulfate volume fraction in the nucleation mode particles.

Hansen et al., 2015 is mentioned a few times in the text, but no reference is included.

Reply: Hansen et al., 2015 was added in the reference.

Page 15522 line 10: should be “non-additive”.

Reply: text was corrected accordingly.

Reference:

Canagaratna, M. R., Jimenez, J. L., Kroll, J. H., Chen, Q., Kessler, S. H., Massoli, P., Hildebrandt Ruiz, L., Fortner, E., Williams, L. R., Wilson, K. R., Surratt, J. D., Donahue, N. M., Jayne, J. T and Worsnop, D. R.: Elemental ratio measurements of organic compounds using aerosol mass spectrometer: characterization, improved calibration, and implications, Atmos. Chem. Phys., 15, 253–272, 2015 doi:10.5194/acp-15-253-2015.

Kroll, J. H., Donahue, N. M., Jimenez, J. L., Kessler, S. H., Canagaratna, M. R., Wilson, K. R., Altieri, K. E., Mazzoleni, L. R., Wozniak, A. S., Bluhm, H., Mysak, E. R., Smith, J. D., Kolb, C. E and Worsnop, D. R.: Carbon oxidation state as a metric for describing the chemistry of atmospheric organic aerosol, Nature Chemistry 3, 133-139, 2011 doi:10.1038/nchem.948.

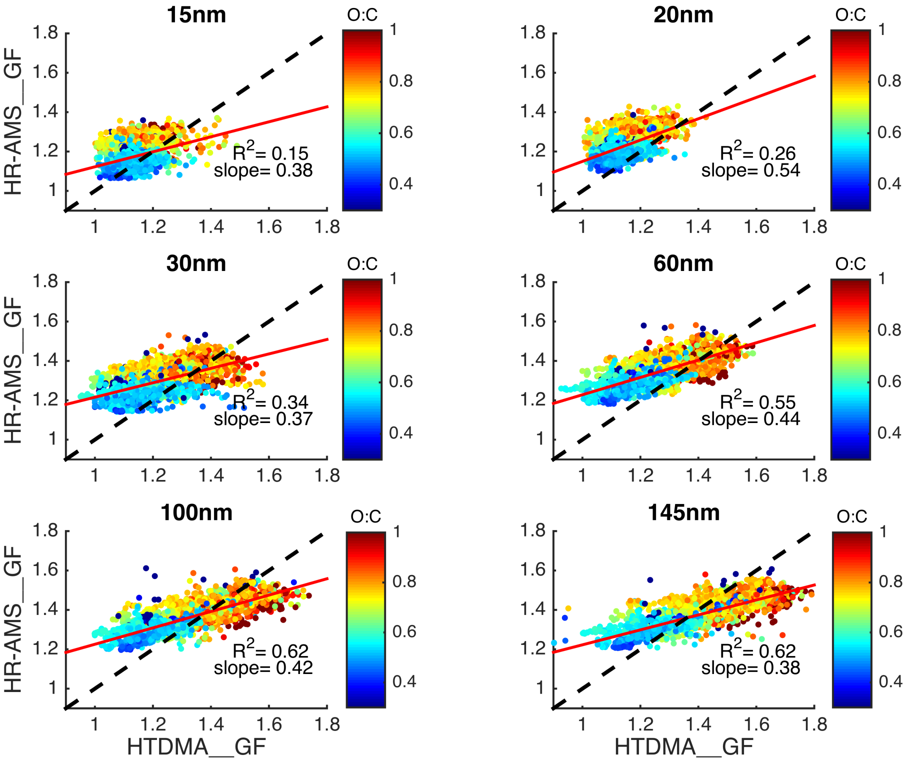


Figure 1. Comparison of HR-AMS-derived HGF with HTDMA-measured HGF of different-sized particles. O:C ratio was obtained from the 'Improved-Ambient' method.

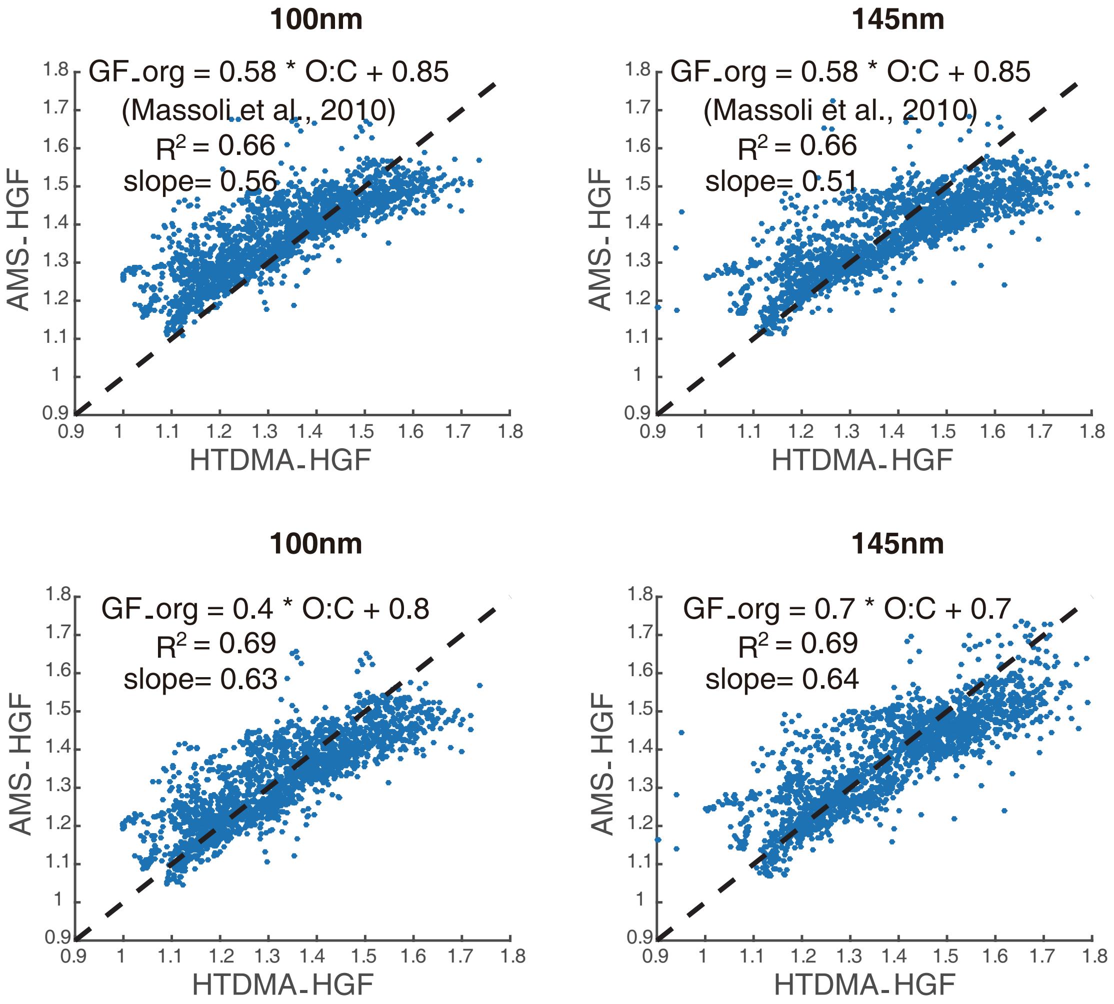


Figure 2. Comparison between AMS derived HGF with measured HGF when taking into account the influence of oxidation level of the organics on GF, with upper panels using the relation determined by Massoli et al. (2010), and lower panels by fitting the equation GF = $a \cdot \mathrm{O} : \mathrm{C} + b$ into our data. The O:C ratio from the lower panels were obtained from the ‘Improved-Ambient‘ method. The dash lines are 1:1 lines.

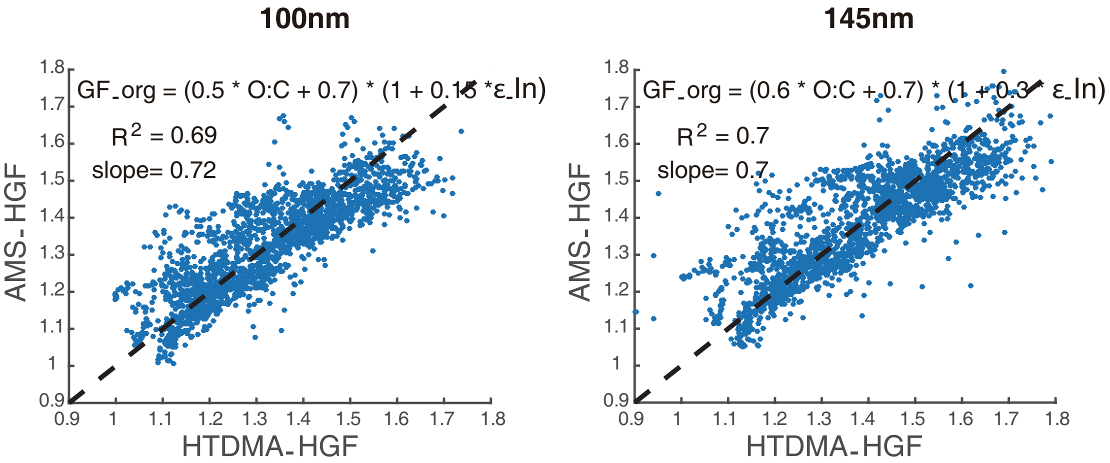


Figure 3. Comparison between AMS derived HGF with measured HGF when taking into account of both oxidation level and inorganic volume fraction on GF of the organics.

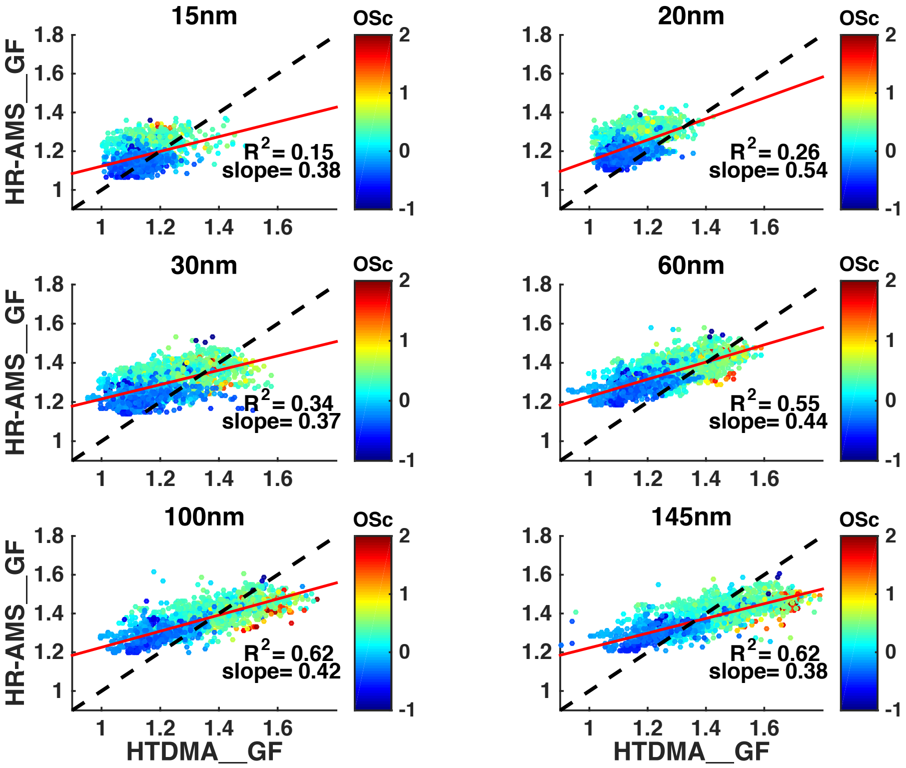


Figure 4: Comparison of HR-AMS-derived HGF with HTDMA-measured HGF of different-sized particles, coloring with oxidation state of particles from AMS measurement.

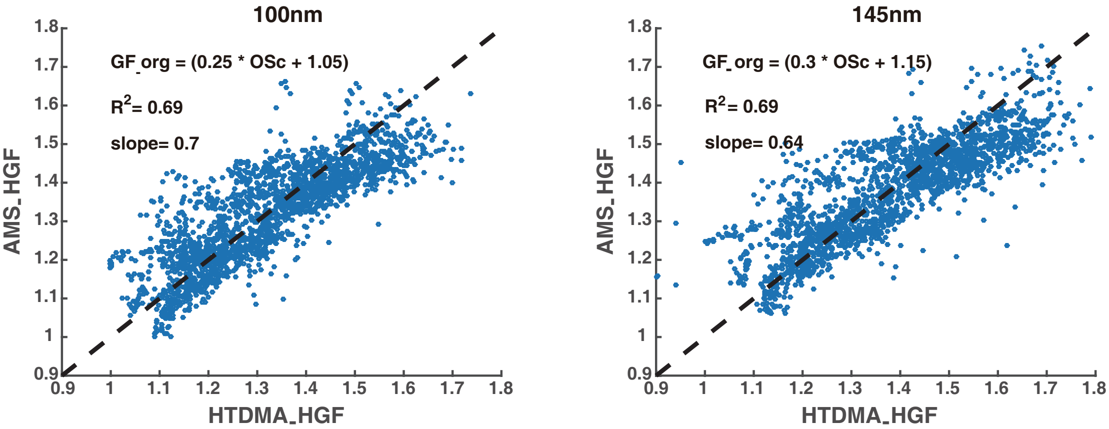


Figure 5. Comparison between AMS derived HGF with measured HGF when taking into account the influence of oxidation state of the organics on GF by fitting the equation GF = $a \cdot$ OSc + b into our data. The OSc were calculated as: OSc = 2 · O:C – H:C (Kroll et al., 2011).

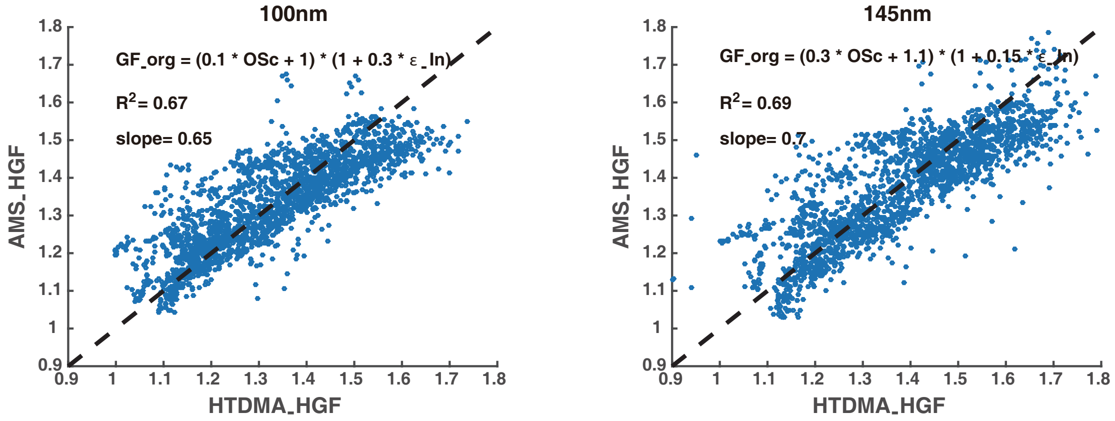


Figure 6. Comparison between AMS derived HGF with measured HGF when taking into account of both oxidation state (OSc) and inorganic volume fraction on GF of the organics.

List of changes:

For referee #1:

Page 2, line 2: A collection efficiency correction of 0.25 was applied to the HR-AMS. ‘ was added into the text.

Page 7, paragraph 2, line 4: Text was corrected as: The HR-AMS measures the bulk chemical composition of particles larger than 80 nm (Williams et al., 2012).’

The following article was added in the reference:
Williams, L. R.; Gonzalez, L. A.; Peck, J.; Trimborn, D.; McInnis, J.; Farrar, M. R.; Moore, K. D.; Jayne, J. T.; Robinson, W. A.; Lewis, D. K.; Onasch, T. B.; Canagaratna, M. R.; Trimborn, A.; Timko, M. T.; Magoon, G.; Deng, R.; Tang, D.; de la Rosa Blanco, E.; Prévôt, A. S. H.; Smith, K. A.; Worsnop, D. R.: Characterization of an aerodynamic lens for transmitting particles greater than 1 micrometer in diameter into the Aerodyne aerosol mass spectrometer, Atmos. Meas. Tech., 6, 3271–3280, 2013, doi:10.5194/amt-6-3271-2013.

Figure 5: ’The dashed lines indicate the 1:1 lines.’ was added into the figure caption.
Figure 6: ’The dashed lines indicate the 1:1 lines.’ was added into the figure caption.
Figure 7: ’The dashed lines indicate the 1:1 lines.’ was added into the figure caption.
Figure 8: ‘The dashed lines indicate the linear fit to the full data.’ was added into the figure caption.

For referee #2:

Page 4, from line 5: The sentence was corrected as: The O:C ratio was calculated from the relative mass concentrations of C and O in the whole ion fragments across the organic mass spectrum. This is named as ‘Aiken-Ambient’ method (Aiken et al., 2007). In addition, an ‘Improved-Ambient‘ method (Canagaratna et al., 2015), which provides a more accurate and precise measure of O:C ratio than the commonly used ‘Aiken-Ambient’ method, is briefly discussed in the result part.

Page 8, a new paragraph was added after paragraph 2:
Similar to Fig. 6 and Fig. 7, new parameterization values were also fitted using the O:C ratio obtained from the ‘Improved-Ambient‘ method. However, the new fit only marginally improved the results compared with the slope and R^2 values using the O:C ratio from ‘Aiken-Ambient‘ method (see S3 and S4). Massoli et al., (2010) also used the ‘Aiken-Ambient’ method to estimate the O:C ratio. For these two reasons, we present our data using the O:C ratio from ‘Aiken-Ambient’ method.

The articles below were added into the reference:

Canagaratna, M. R., Jimenez, J. L., Kroll, J. H., Chen, Q., Kessler, S. H., Massoli, P., Hildebrandt Ruiz, L., Fortner, E., Williams, L. R., Wilson, K. R., Surratt, J. D., Donahue, N. M., Jayne, J. T and Worsnop, D. R.: Elemental ratio measurements of organic compounds using aerosol mass spectrometer: characterization, improved calibration, and implications, Atmos. Chem. Phys., 15, 253–272, 2015 doi:10.5194/acp-15-253-2015.

Hansen, A. M. K., Hong, J., Raatikainen, T., Kristensen, K., Ylisirniö, A., Virtanen, A., Petäjä, T., Glasius, M and Prisle, N. L.: Hygroscopic properties and cloud condensation nuclei activation of limonene-derived organosulfates and their mixtures with ammonium sulfate, Atmos. Chem. Phys. Discuss., 15, 17317–17365, 2015, doi:10.5194/acpd-15-17317-2015.

Page 8, paragraph 2, line 8: text was corrected as: non-additive.

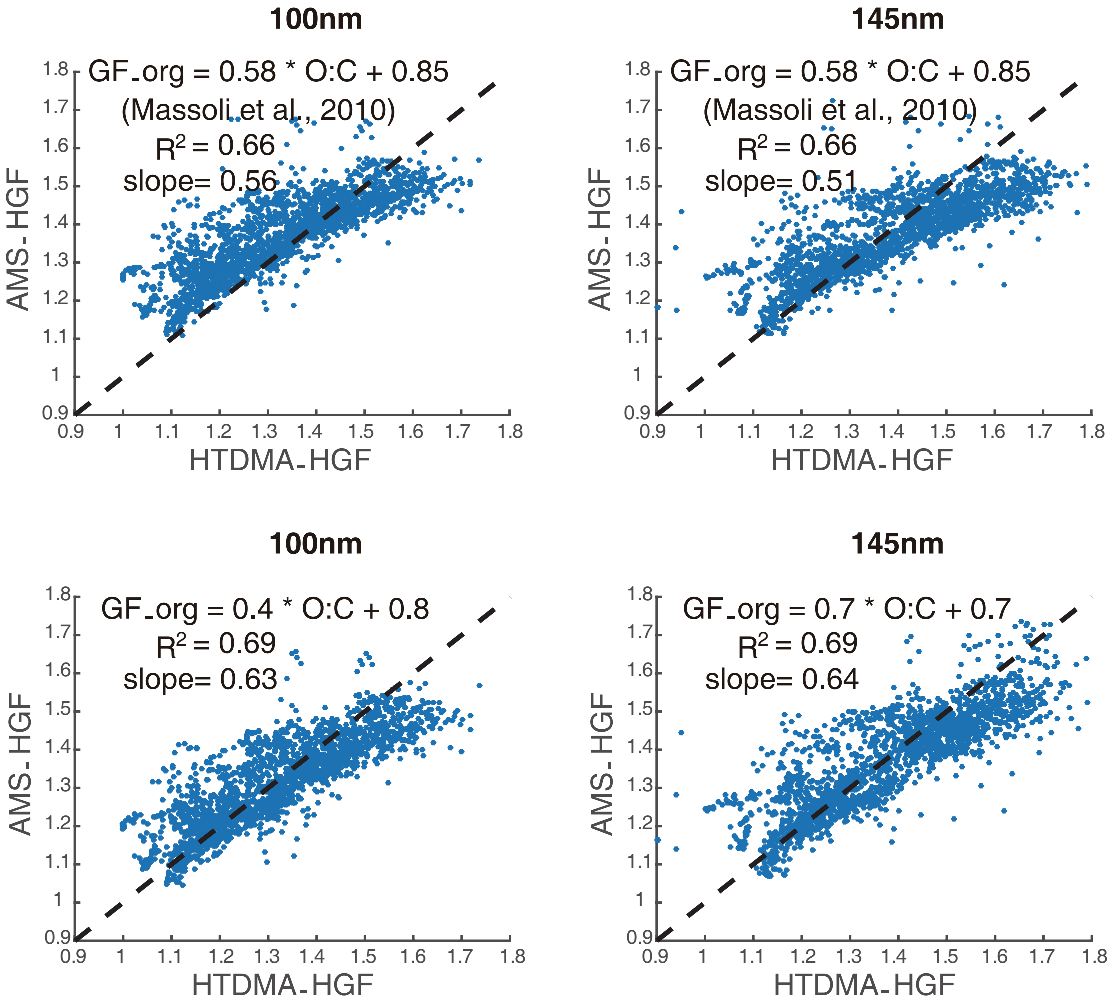
Page 9, paragraph 3, line 13: text was corrected as: Fig. S5

In the acknowledgements:
Text was changed as:

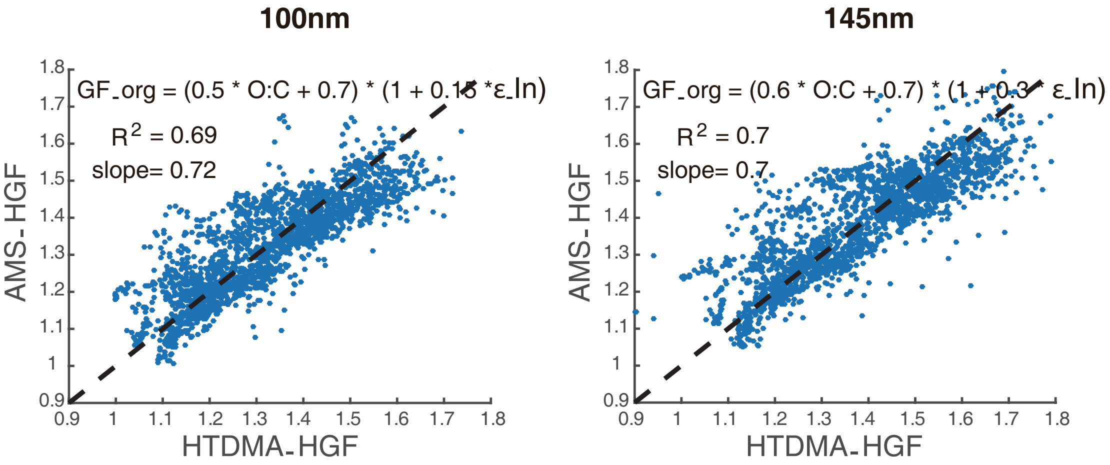
This work was supported by the Cryosphere-Atmosphere Interactions in a changing Arctic climate project (CRAICC) (N° 4720479), the Academy of Finland Center of Excellence (N° 1118615, N° 259005), European Research Council (ATM-NUCLE and Grant QAPPA 335478), University of Helsinki funds, and European Commission (ACTRIS, N° 262254) via the project 'Pan-European Gas-AeroSOls-climate interaction Study' under the Framework Programme 7 (FP7-ENV-2010-265148). Authors also wish to thank the staff at SMEAR II for their assistance throughout the whole intensive campaign.

For the supplementary material:

Two more figures were added as S3 and S4. The previous S3 was ordered as S5.



S3: Comparison between AMS derived HGF with measured HGF when taking into account the influence of oxidation level of the organics on GF, with upper panels using the relation determined by Massoli et al. (2010), and lower panels by fitting the equation GF = $a \cdot O : C + b$ into our data. The O:C ratio from the lower panels were obtained from the 'Improved-Ambient' method. The dashed lines are 1:1 lines.



S4: Comparison between AMS derived HGF with measured HGF when taking into account of both oxidation level and inorganic volume fraction on GF of the organics. O:C ratio were obtained from the 'Improved-Ambient' method. The dashed lines indicate the 1:1 lines.