

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. 3 and Fig. 4 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.

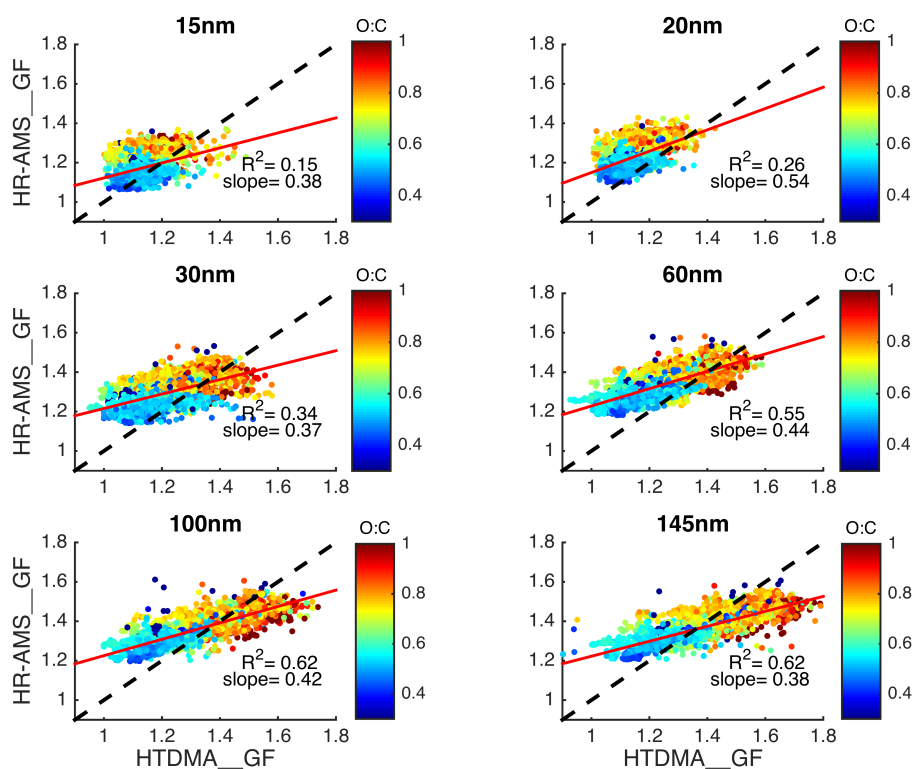


Figure 1. Comparison of HR-AMS-derived HGF with HTDMA-measured HGF of different-sized particles. O:C ratio was obtained from the ‘Improved-Ambient’ method.

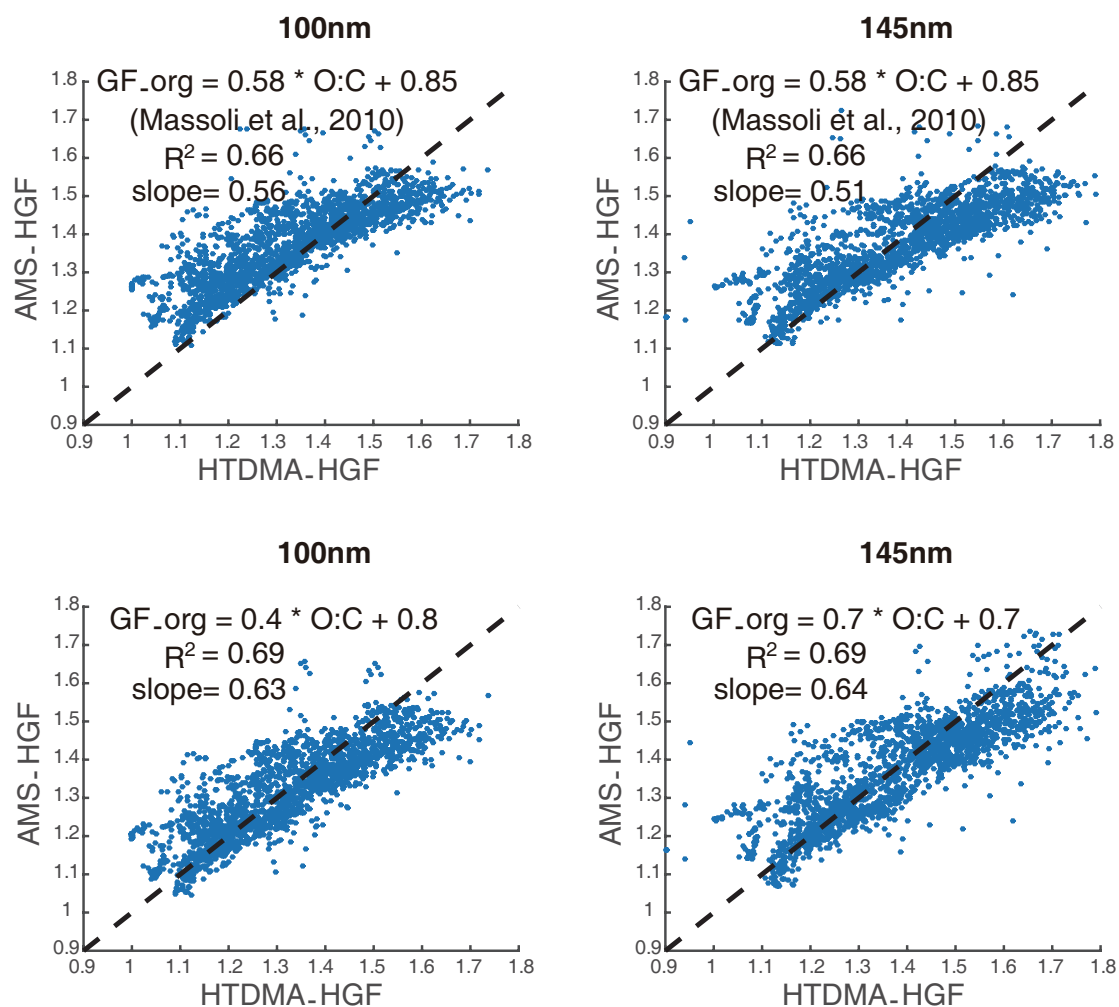


Figure 2. Comparison between AMS derived HGF with measured HGF when taking into account the influence of oxidation level of the organics on GF, with upper panels using the relation determined by Massoli et al. (2010), and lower panels by fitting the equation $GF = a \cdot O : C + b$ into our data. The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

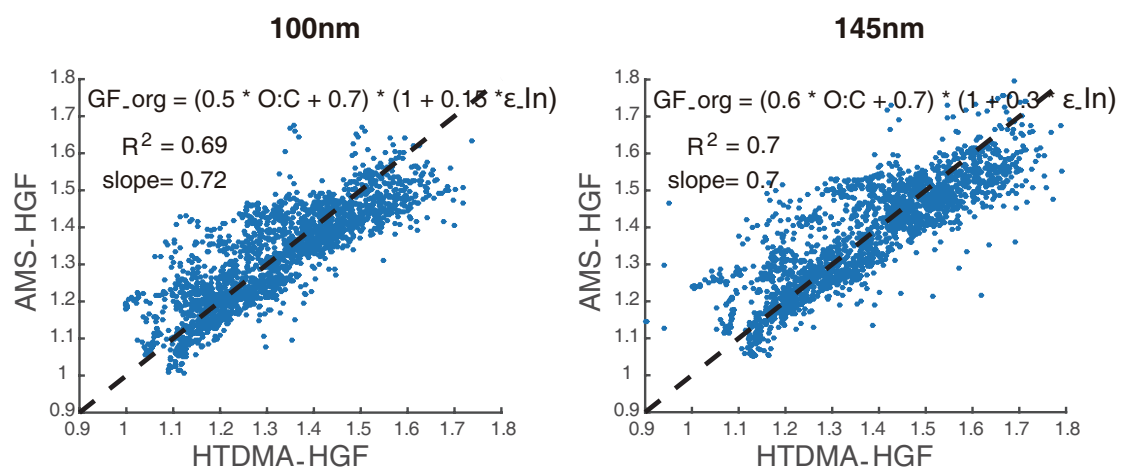


Figure 3. Comparison between AMS derived HGF with measured HGF when taking into account of both oxidation level and inorganic volume fraction on GF of the organics.

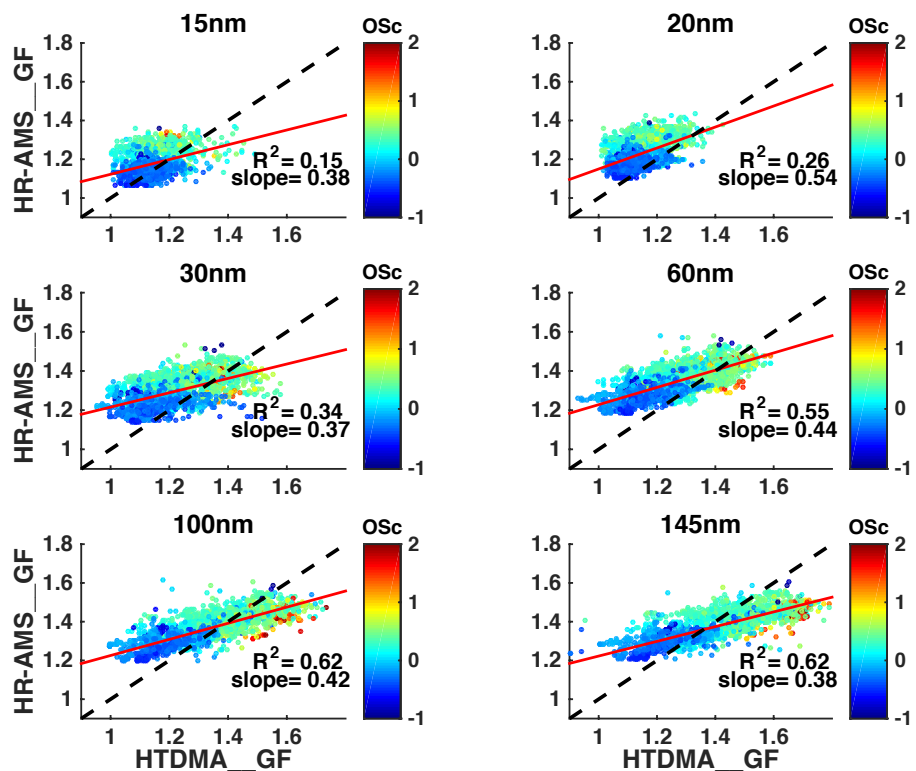


Figure 4: Comparison of HR-AMS-derived HGF with HTDMA-measured HGF of different-sized particles, coloring with oxidation state of particles from AMS measurement.

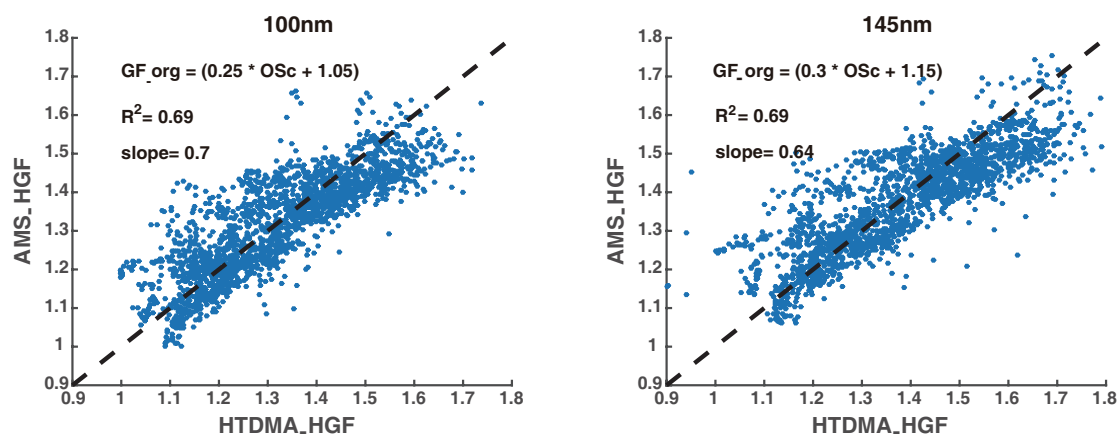


Figure 5. Comparison between AMS derived HGF with measured HGF when taking into account the influence of oxidation state of the organics on GF by fitting the equation $GF = a \cdot OSc + b$ into our data. The OSc were calculated as: $OSc = 2 \cdot O:C - H:C$ (Kroll et al., 2011).

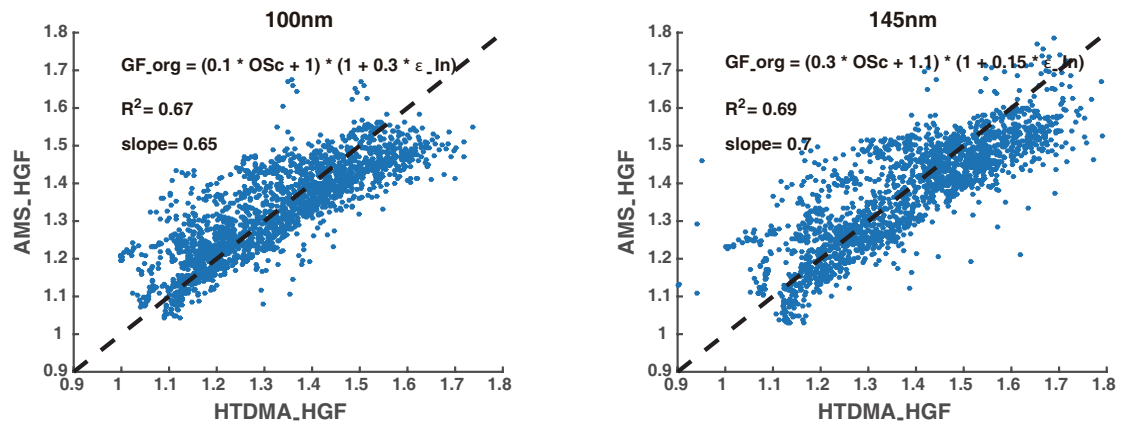


Figure 6. Comparison between AMS derived HGF with measured HGF when taking into account of both oxidation state (OSc) and inorganic volume fraction on GF of the organics.