

Referee #1

Alroe et al. describe a method for measuring the hygroscopicity of aerosols which separates the contributions of semi-volatile and low volatility components. Their approach combines measurements from an Aerosol Mass Spectrometer (AMS) and a Volatility and Hygroscopicity Differential Mobility Analyser (VH-TDMA), which together provide information about composition and hygroscopicity. The use of a thermodenuder (TD) allows the comparative impact of semi-volatile aerosol species to be assessed.

Overall the manuscript is interesting and within the scope of AMT. My major concern relates to data quality: while the extension compared to previous work is that the technique provides size-resolved (around 100 nm) and time-resolved data, the composition measurements do not appear sensitive enough to make this feasible in practice. Since all ambient and a large proportion of laboratory AMS measurements are reported to be below detection limit at this size range, it is not clear under what circumstances the full approach described is actually applicable to atmospheric measurements, or lab experiments. The authors therefore need to better justify their methodology and clarify the limitations of averaging over size bins and smoothing. I have some additional queries related to the analysis methods and overall clarity of the experiment descriptions which should be addressed before the manuscript is considered for publication.

Author

The authors thank the reviewer for the detailed comments and suggestions that have helped us to refine the manuscript. We agree that the detection limits reported in the original version of the manuscript significantly limited the value of the laboratory-based results. After further examination of our approach, we have identified several changes to our analysis which have significantly improved these results. The manuscript has been updated with a detailed discussion of this revised analysis, particularly in Sections 2.3, 2.6 and 3.1.

In short, the averaging time for the compositional measurements has been increased which has improved both the detection limits and the stability of the signal for all species except NH_4 . This has substantially improved the sensitivity of the measurement, raising a much greater proportion of the measurements above the detection limit. In addition, since the signal stability has improved, meaningful trends can be obtained without the need for any statistical smoothing. The detection limit for NH_4 remains high and the source of its variability has not been clearly identified, although it may be due to residual effects from high AS concentrations which had been sampled immediately prior to starting the filtered background measurements. A linear fit has been used to estimate the NH_4 concentrations, rather than discarding a significant source of AS mass. The resulting composition-based HGF models still obtain close agreement with the direct VH-TDMA measurements, so we are confident that this methodology offers meaningful results even when sampling rapidly changing, size-dependent aerosol.

The ambient marine analysis has not required substantial revision, since the concentrations of NH_4 , SO_4 and organics were well above their detection limits, NO_3 is not relevant to baseline marine aerosol, and Chl is primarily present in refractory compounds which cannot be efficiently detected by the AMS. The non-refractory species which exhibited mass fractions

and volatility consistent with characteristic marine aerosol, and gave good agreement with the hygroscopic measurements obtained by the VH-TDMA.

In summary, now that the chamber experiment results have been revised, the sampling system has demonstrated internally consistent findings and significant utility under two quite challenging scenarios. Given that Cape Grim receives some of the cleanest air in the world, we anticipate that much more detailed analysis will be possible when sampling atmospheric aerosols in other locations, or during less size-dependent or rapidly evolving laboratory studies.

Responses to the additional queries have been included below. Please note that text coloured in red refers to the added text in the manuscript. All page and line numbers refer to the revised manuscript (Revised_Manuscript_TrackedChanges.docx), or supplementary material (Supplement.docx), where all changes have been tracked. If the text has been significantly changed, only the section number is given in this document (e.g. “Section 2.6”).

General comments

Referee's comment

1. Experimental description:

Many more details are required as to how the chamber experiments were performed (section 2.5): For instance: How much n-butanol was added? How was RH introduced, and what purity of water was used? How was the HONO prepared and introduced? Was the chamber mixed? How was the chamber cleaned? What were the background concentrations (particles, AMS, CIMS...) prior to the experiment?

Author's answer

1. Section 2.6 has been extensively re-written to provide full detail about the chamber preparations and initial conditions of the experiment.

Referee's comment

2. Data quality:

The authors admit in section 3.1 (P7 L10-14) that using highly time- resolved and size-resolved AMS data result in “unstable” data “often falling below the detection limit”, even for lab experiments. If Fig 3 is a typical time series for the size range studied (130-180 nm), these concentrations seem more than sufficient for confidence in the AMS data. What is the source of this instability, then, and what is the size and origin of the background signal used to estimate the LOD? Can individual error bars be marked on in Fig 3? Without a more detailed discussion here it is difficult to be confident that the technique is viable and the results presented are meaningful in terms of quoted uncertainties.

Author's answer

2. The following discussion of detection limits and measurement uncertainties has been added to Section 2.1:

P2 L33: “Detection limits for each species are calculated as three times the standard deviation of their background concentration, observed when sampling particle-free air through a high-efficiency particle filter (DeCarlo et al., 2006). This accounts for the instrument’s background signal from stray ions and electronic noise. Uncertainties are given as the larger value of either the detection limit or the species-dependent measurement accuracy of the AMS. These accuracy estimates encompass uncertainties in the ionisation efficiencies, particle collection

efficiencies and the inlet flow rate and is commonly estimated as $\pm 37\%$ for organics, $\pm 35\%$ for SO_4 and Chl, and $\pm 33\%$ for NO_3 and NH_4 (Bahreini et al., 2008).”

The reviewer is correct that the bulk concentrations were more than adequate as seen in Fig 4a (previously Fig 3a). However, aerosol mass is strongly biased towards large diameter aerosol and aerosol in the desired size range ($130 < d_{va} < 180$ nm) represented only a small fraction of the total aerosol mass. In addition, during PTOF sampling, concentrations selected from this size range represent data from only a subset of the total sampling time. In short, the size-resolved measurements did not benefit from as much sampling time and signal averaging as the bulk measurements and, as a result, exhibited higher variability. In the original manuscript, both limitations were countered by smoothing the data with a non-parametric regression technique. After further examination, we have found that the compositional trends and detection limits are sufficiently improved by averaging to a time resolution of 12 minutes (6 minutes each of heated and unheated sampling).

The resulting 6-minute averaged PTOF measurements are shown in Fig S3 and uncertainties have been given as error bars. Detection limits were 0.123, 0.012, 0.023, 0.245 and 0.023 $\mu\text{g m}^{-3}$ for organics, NO_3 , SO_4 , NH_4 and Chl respectively. Section 3.1 has been substantially changed to include discussion of this revised analysis (P7 L9). In short, a large proportion of the measurements are now above these detection limits. The major exception was NH_4 , which was highly variable and had a correspondingly high detection limit. Its concentrations have been estimated with a linear fit to the size-resolved PTOF measurements.

Referee's comment

3. Smog chamber experiments – aerosol dynamics:

The authors note the rapidly changing conditions in the smog chamber experiments. However, substantially more information is required to understand the time series presented (Fig 3, 5-7). For instance, it is not clear how the aerosol mass loading ($\mu\text{g/m}^3$) and size distribution ($dN/d\log D_p$) actually evolve through the experiment – please present these as a function of time. It looks from Fig 5(a) that there are multiple maxima in the mass distribution – is this also the case in the number distribution, and if so why for a seeded experiment?

Related to this, one of the main driving forces for the rapidly decreasing HGF over time at 100nm is a decrease in sulfate, which is not even mentioned in the text. What are the causes of this? Can particle coagulation and wall loss rates be quantified, for instance?

I cannot reconcile the composition time series in Fig 3 and 5b. For instance, at 150 minutes in Fig 3, the sulfate and LV organic mass concentrations are comparable, while in Fig 5b, virtually no sulfate is present. I am wondering if Fig 3 presents the total AMS concentration rather than the size-resolved data as implied. If so its inclusion should be justified and thoroughly clarified. It would be necessary to see the raw size-resolved data also plotted and discussed in the context of “Data quality”, above.

Author's answer

3. Two time series have been added to the supplement demonstrating the total non-refractory aerosol mass (Fig S1), and the number size distributions (Fig S2) observed throughout the chamber experiment. Since the chamber did not have a mixing fan, it likely took at least 30 minutes to become uniformly mixed and this passive diffusional mixing may explain the initial rapid decrease in total particle mass and SO_4 concentration. Approximately 50 minutes after

the lights were switched on, SOA was condensing at a sufficient rate to drive up the total particle mass (Figs 3, S3 and S1) and increase the mode diameter of the particle number size distributions (Fig S2). An additional paragraph has been added to the start of [Section 3.1](#) commenting on these observations (P6 L38).

This chamber was constructed fairly recently and the chamber losses have not yet been fully characterised. Also, it is unclear when the chamber became uniformly mixed. So it is challenging to verify an appropriate initial reference concentration or loss rate for wall loss/coagulation corrections. However, when modelling composition-dependent HGFs, our calculations are based on the relative fractions of each species and so our results are not significantly dependent on loss-corrected particle concentrations.

As noted by the reviewer, there was a progressive decrease in AS volume fraction throughout the experiment (Fig 5b). Our analysis focused specifically on aerosol with $d_m = 100$ nm. Initially these were 100 nm AS seeds, but ongoing SOA formation allowed progressively smaller AS seeds to reach the 100 nm target size. As a result, the relative contribution of AS to 100 nm aerosol decreased over time, leading to a corresponding decrease in HGF.

The multiple maxima present in the mass distribution (Fig 5a), is likely due to two reasons. Firstly, compared to large diameter (high mass) AS seeds, only a relatively small quantity of condensed SOA is required to dominate the mass composition of small seeds. So the relative fraction of SOA is biased towards small sizes. In addition, the AMS mass distribution is presented in terms of d_{va} , which is density-dependent. So since the smallest particles have the greatest relative fraction of SOA, they likewise suffer the greatest reduction in density and d_{va} , causing separation between the AS- and SOA-dominant maxima in the mass distribution.

Referee's comment

4. Smog chamber experiments – derivation of HGFs: I have a number of queries about how the smog chamber hygroscopic growth factors were derived (P8 L1-22). Firstly, the ammonium sulfate HGF = 1.58 ± 0.03 from calibration experiments is low compared a range of previous measurements and the E-AIM and AIOMFAC models (1.7-1.8) e.g. (Denjean et al., 2014; Lei et al., 2014). Please discuss this discrepancy and how it might propagate given that AS is the dominant hygroscopic component. The contribution of LVOA is estimated based on a parameterisation of O:C vs HGF from Massoli et al. (2010). However, the authors cite recent work suggesting O:C may not be a good proxy for CCN activity and hygroscopicity. They also find the two OA components in their experiments have similar O:C. Why, then, was this parameterisation used? And more problematically, why only for the LVOA? The SVOA HGF was estimated via a residual approach, whereas the parameterisation would give the same HGF as LVOA. What value for HGFLVOA would be obtained using the same approach as SVOA, i.e. calculating a residual HGF in the heated sample after the sulfate contribution is accounted for? Would the model perform substantially less well with a single HGFOA, as is used for the ambient samples? Given the combined uncertainties in HGFLVOA and HGFSVOA (which overlap), and different estimation methods, the conclusion that the two fractions have different HGF needs to be better supported.

Author's answer

4. The authors would like to thank the reviewer for drawing attention to the low HGF used for AS in this experiment. After further investigation of the calibration measurements, we have found that the deliquescence point was reached at 82.0 ± 0.5 %RH, rather than 80 %RH (Tang

1991), suggesting that the H-TDMA humidity was overestimated by 2 %RH in both the calibration and subsequent chamber-based measurements. The HGF of AS is strongly RH-dependent above its deliquescence point, so this could account for the discrepancy between the observed and published HGF of AS. Since the humidity dependence of α -pinene SOA HGFs is not well defined, it is not feasible to correct for this discrepancy. Therefore all chamber-based HGFs must be considered to represent water uptake at 88 %RH. The HTDMA humidity has been corrected in the manuscript (P6 L14 and P8 L13)

The reviewer has questioned our use of an O:C parameterization for HGF. While we share their concern about its reliability as reported in other studies, our aim was to obtain an independent estimate for the LVOA contribution to HGF. With this, the AMS measurements could be used to estimate an independent composition-based HGF for the heated aerosol which could be compared against the direct VH-TDMA measurements. If a contribution had been derived via the residual (ZSR) method, it would not have been possible to verify agreement between the two instruments. A published value could have been used instead, but a very wide range have been published and the O:C parameterization gave some basis for the chosen value. We do agree that both OA components have very similar O:C values, which suggests that the same contribution may apply for both. However, near the end of the experiment, the heated aerosol has a negligible AS fraction and, as seen in Fig 6, the HGF is significantly higher the VH-TDMA-derived HGF of the semi-volatile component. Using this value for both OA components results in systematic underestimates of both heated and unheated HGFs.

Specific comments

Referee's comment

1. P1 L20-22: Please provide a general reference for this paragraph.

Author's answer

1. The following reference has now been provided:

Seinfeld, J. H., and Pandis, S. N.: Atmospheric Chemistry and Physics: From Air Pollution to Climate Change, 3 ed., John Wiley & Sons, Inc., New York, 2016.

Referee's comment

2. P1 L27: Clarify what is meant by “dynamic nature” – many volatile species are not particularly reactive or prone to condensation/evaporation. Perhaps indicate specific species of importance.

Author's answer

The authors agree that semi-volatile species are not necessarily “more reactive”. The term “dynamic”, was used to indicate that they more readily partition between particle and vapor phase due to changes in concentration and temperature. The associated sentences have been reworded as follows:

P1 L25: “Furthermore, many are volatile, Since many are semi-volatile, their relative partitioning between the particle and gas phase can be sensitive to concentration changes and local atmospheric conditions transitioning between the particle and gas phase in response to changes in their gas-phase concentration and local atmospheric conditions (Seinfeld and Pankow, 2003; Donahue et al., 2012). Furthermore, as their partitioning changes, they can become exposed to different phase-dependent chemical reactions. These volatile compounds

are often more reactive, subject to oxidative and oligomeric processes, and their dynamic changes dynamic nature complicates predictions of bulk aerosol properties.”

Referee’s comment

3. P3 L15: Replace “the two instruments” with “the two instruments (AMS and VH-TDMA)”.

Author’s answer

3. The line has been replaced, as per the recommendation.

Referee’s comment

4. P3 L17: When is additional drying required? Was it required in any of this work? If not, delete.

Author’s answer

4. Additional drying is required when the ambient temperature and/or sample humidity is too high for the nafion dryer to achieve the desired 30 %RH inlet humidity (such as when sampling in tropical environments or from a nebulised aerosol source). Additional drying was not used in either of the studies discussed in this paper, however the diffusion dryer was used instead of the nafion dryer for the Cape Grim coastal measurements. To reflect this, the manuscript has been updated as follows:

P3 L8: “... the aerosol is dried to a relative humidity (RH) of approximately 30 % using a membrane dryer (Nafion MD-700) or a silica gel diffusion dryer.”

P6 L12: “A nafion dryer was used to maintain the inlet RH at 32.6 ± 0.3 % throughout the experiment and the H-SMPS humidifier was set to 90 %RH.”

P6 L33: “The sampling inlet RH was consistently dried with a diffusion dryer to below 30 % ...”

Referee’s comment

5. P3 L26: The “Line A and B” terminology is confusing and is subsequently not used a great deal. According to Fig 1, A/B are not constant sampling lines but relate to the changing paths of the TD and unheated aerosol samples. Why not just use “TD” and “unheated” and remove “A/B” altogether? Similarly, for the wall-loss experiments (Fig 2), the exact path difference being monitored should be clarified.

Author’s answer

5. The authors agree with this recommendation and have updated the following lines in response:

P3 L14: Deleted the mention of Line A and B in parentheses

P5 L13: “... an additional solenoid valve on the TD line, ...”

P5 L24: “Figure 2 displays the relative transmission efficiency of the TD line compared to the unheated line.”

P9 L29: “...relative losses of up to 15 % are observed in the TD line at 120 °C (50 nm, AS).”

Figure 1: The labels “Line A” and “Line B” have been removed.

Figure 2: Caption has been updated, indicating that it depicts the “transmission rates of the TD sampling line compared to the unheated line”

Referee's comment

6. P3 L30-31: Was this “stepping” performed here? If so, for which parameters?

Author's answer

6. The RH and TD temperature remained constant throughout each of the studies discussed in this paper (i.e. no “stepping” performed). In the Cape Grim coastal study, the pre-selected diameter for the VH-TDMA was regularly cycled between 40, 100 and 150 nm. The diameter changes occurred after each pair of heated/unheated measurements. Results from the 40 and 150 nm samples have not been examined in this study as the focus was on aerosol large enough to be sampled by the AMS, but small enough to be close to the critical diameter for cloud droplet activation.

Referee's comment

7. P4 L1-2: What is the combined flowrate required, and hence the diluting flowrate? Given the interest in semivolatile partitioning, how was dilution of the gas phase accounted for?

Author's answer

7. Dilution does not significantly affect measurements for this system. There was no dilution in the AMS line. A vacuum supply ensured a continuous total sample flow of 1.0 L min^{-1} through the AMS sampling path, from which the AMS sampled at a rate of 0.1 L min^{-1} . For the VH-TDMA, dilution occurred after all sizing was completed (after passing through the electrostatic classifiers) and directly before the aerosol passed into each condensation particle counter (CPC). The exact dilution ratios depend on the model of CPC and the desired aerosol flow rate within each SMPS. In the case of these the two studies, the H-SMPS had a dilution ratio of 1:1 (aerosol vs filtered dilution air) and no dilution was used for the V-SMPS and. For the aerosol species examined in this manuscript, any volatilisation within the CPC itself would be unlikely to reduce the particle diameter below the instrument's detection threshold of 10 nm.

Referee's comment

8. P4 L8: “...all measurements were performed on aerosol with $d_m = 100 \text{ nm}$...”. This may be the case for all VH-TDMA measurements, but this is in the composition section and the AMS sample is not pre-classified according to Fig 1. Size-dependent number concentrations (Fig 4) and composition (Fig 5a) data are also shown later. Please clarify.

Author's answer

8. The authors agree that this wording was unclear. Size-resolved particle time of flight (PTOF) AMS measurements were used in the chamber-based experiment to restrict compositional analysis to a comparable range of vacuum aerodynamic diameters. However the Cape Grim aerosol did not exhibit strongly size-dependent composition and no size-selection was applied to the AMS measurements for that experiment. **Section 2.3 “Size-resolved composition”** has been significantly re-worded to avoid implying that all AMS analysis was size-selected.

Referee's comment

9. P4 L10: “If the aerosol is strongly size dependent”. Should this read “aerosol composition”?

Author's answer

9. This section (Section 2.3) has been significantly rewritten, and this comment has been incorporated into the new discussion.

Referee's comment

10. P4 L18-19: Was this linear correction factor applied to any data here? If so, which?

Author's answer

10. The linear correction factor was only applied to the chamber-based data. In that experiment, the composition was size-dependent and its analysis required size-resolved PTOF data including aerosol with diameters <100 nm. Since the coastal Cape Grim aerosol was not strongly size dependent, PTOF measurements were not used and therefore it was not necessary to apply any size dependent corrections to that dataset. To clarify this, the sentence regarding the linear correction factor has been reworded as follows:

P4 L19: “Since the chamber-based measurements, discussed in Section 3.1, required PTOF data from this diameter range, a linear correction factor was applied...”

Referee's comment

11. P4 L20-24: Which data were smoothed in this study? Mention here and in corresponding Fig captions. Please define “LOESS”.

Author's answer

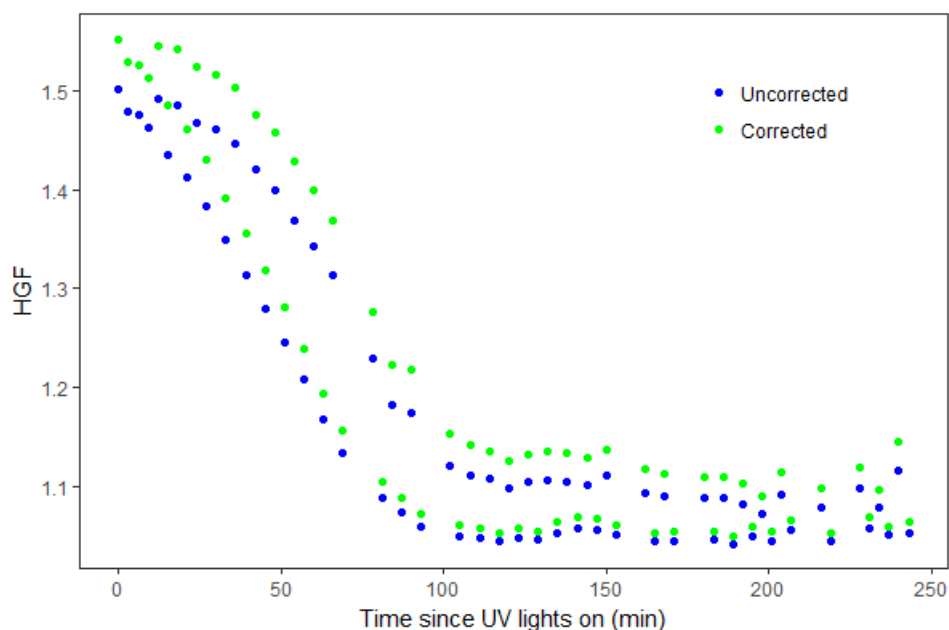
11. As discussed above, LOESS smoothing is not being applied to the chamber-based data now. Instead, the PTOF data from this experiment has been averaged to a 12 minute time resolution. When calculating AS concentrations for the HGF models, the NH₄ concentrations were approximated with a linear fit to the size-resolved NH₄ measurements. Section 3.1 has been significantly reworked to reflect these changes. No smoothing has been applied to the Cape Grim dataset.

Referee's comment

12. P4 L26-P5 L5: I am surprised that a Kelvin correction is required for a dry diameter of 100 nm. What was the magnitude of the correction?

Author's answer

12. The scale of the corrections are shown in the figure below. In short, the HGFs of the unheated chamber-generated aerosol increased by 0.6 – 3.5 % after the Kelvin correction. Diameters reduced by up to 30 % during heating, increasing the impact of the Kelvin correction and leading to HGF increases between 1.7 – 4.4 %. The largest corrections were observed during the first hour of the experiment because κ is partially dependent on the uncorrected HGF (Equation 2, P4 L33). Since the range of corrections for both heated and unheated HGFs exceed the 2% measurement uncertainty of the H-TDMA, Kelvin-corrected HGFs have been used in this analysis.



Referee's comment

13. P5 L30: Please change to "...transmission efficiency decreased linearly with increasing temperature...".

Author's answer

13. The sentence has been updated as recommended (P5 L26): "...transmission efficiency decreased linearly with **increasing** temperature..."

Referee's comment

14. P6 L12: Justify precision of [OH] – what is the uncertainty?

Author's answer

14. The authors agree that the original OH concentration was overly precise. Barmet et al. (2012) reported an uncertainty of 25% in the rate constant which relates [butanol-d9] to [OH]. In light of this, the manuscript has been updated to indicate that this is only an estimate of [OH] (P6 L15): "initial OH concentration of **approximately** 1.5×10^7 molecules cm^{-3} "

Referee's comment

15. P6 L16: The mention of Fig 3 here, without discussion, confuses section 3.1 of the results. I suggest this Figure is not mentioned until it is discussed in the results.

Author's answer

15. This sentence has been reworded and moved to **Section 3.1 (P6, L38)**. The figure numbering has also been updated, and the corresponding figure is now **Fig 4**.

Referee's comment

16. P6 L30: Again, reference to "all measurements" is confusing here. For instance, Fig 4 shows time series, not 6-hour averages.

Author's answer

16. Figure 4 uses a higher time resolution to demonstrate broader trends in composition and number size distribution on 2-3 March 2015. It is presented prior to any discussion of data analysis and its purpose is to support the choice of a restricted 6 hour time period for averaging and further detailed examination. To clarify this, the reference to “all measurements” has been reworded as follows:

P6 L30: “To account for the low aerosol concentrations, aerosol properties were averaged over this 6 hour period and the resulting mean values were used for all further analysis.”

Referee's comment

17. P7 L24: SVOA and LVOA are defined “for convenience” based on desorption at 120°C. How does this threshold relate, approximately, to well-known measures of volatility, such as saturation concentration/vapour pressure, for the mass loadings used here?

Author's answer

17. The authors acknowledge the value of reporting volatility in terms of equilibrium saturation concentrations, or the volatility basis set. However the thermodenuder used in this study has a short residence time of approximately 3 seconds, which is insufficient for the aerosol to reach equilibrium (Riipinen et al., 2010). In addition, the seeded α -pinene experiment was not repeated for different SOA loadings and only one thermodenuder temperature was used throughout the experiment. Under these circumstances, it is our understanding that it is non-trivial to determine meaningful equilibrium saturation concentrations and is outside the scope of this study. However, VBS analysis will certainly be a valuable addition for future studies using this methodology

Referee's comment

18. P7 L26: Please provide a plausible mechanism or literature precedent for dark SOA production in these conditions. The method section also implies the seeds and RH were added prior to alpha pinene, rather than in the 30 minutes afterwards.

Author's answer

18. The authors have not found literature supporting similar dark SOA formation. However in light of the improved analysis discussed above, it seems that there was relatively negligible organics present at the start of the experiment (Figs 5b and S3).

The smog chamber method section (Section 2.6) has been re-ordered to reflect the chronological sequence of events, shifting the sentence regarding injection of AS seeds (P6 L6) to directly before UV illumination of the chamber (P6 L9).

Referee's comment

19. P9 L1: Could organosulfates also contribute to this volatile sulfate (and organic) signal?

Author's answer

19. It is possible that organosulfates (OS) contributed to the sulfate mass fraction observed at Cape Grim, especially since OS formation is promoted by acidic aerosol (Surratt et al., 2007). Their fragmentation pattern within a unit mass resolution AMS is largely indistinguishable

from inorganic sulfates (Farmer et al., 2010), so it is not possible to conclusively identify an OS fraction in this Cape Grim dataset. However since OS compounds often have low volatility (Lukács et al., 2009; Liggio and Li, 2006), and other marine studies have reported relatively low OS contributions to the total organic mass (Hawkins and Russell, 2010; Claeys et al., 2010), it is unlikely that they contribute significantly to the volatile component in this study.

Referee's comment

20. P9 L9: *The heated particles lost 12% of their volume, but apparently around 50% of their mass (Fig 8). Please explain this discrepancy.*

Author's answer

20. The AMS relies on flash vaporization of aerosol at 600 °C and does not efficiently detect refractory compounds, such as sea salt. Therefore, while 50 % of the non-refractory mass was desorbed in the TD, it is likely that this comprised only 12 % of the total aerosol volume. The remaining 88 % of the aerosol volume was likely composed of refractory compounds (including sea salt), and non-refractory compounds which were not fully desorbed at 120 °C. To clarify this, P8 L37 has been reworded as follows:

49.5% of this non-refractory mass was desorbed at 120 °C, including the entire non-refractory organic fraction.

And the following sentence has been added to P9 L11:

The difference between the volume and mass fractions which remained after heating imply the presence of a substantial volume of refractory material (such as SSA) which could not be efficiently detected by the AMS.

Technical comments

Referee's comment

1. Fig 3: *Please label the panels (a), (b) and (c) respectively and refer to them as such in the manuscript.*

Author's answer

1. This figure has been renumbered as Fig 4, and the panels have been labelled as suggested.

Referee's comment

2. Fig 4 (b): *The dynamic range of the colour scale tops out at $\sim 10 \text{ cm}^{-3}$ to my eyes. Please adjust, or bin the data rather than using a continuous scale.*

Author's answer

2. This figure has been renumbered as Fig 3. Panel (b) has been replotted with an improved colour scale and expressing concentrations in terms of $dN/d\log(d_m)$. The y-axis has also been expanded to display the full diameter range measured by the SMPS (5 – 200 nm).

Referee's comment

3. Fig 5 (a): *Please add the units of $dM/d\log(dva)$.*

Author's answer

3. The y-axis label of Fig 5 (a) has been updated with appropriate units as follows: “dM/dlog(d_{va}) ($\mu\text{g m}^{-3}$)”

References

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