Anonymous Referee #1

We would like to thank both reviewers for their comments, which led to substantial improvements to the paper.

General/Major comments

1. It is of importance to assess to what extent the aerosol can be considered marine or influenced by anthropogenic or continental emissions during different time periods. Otherwise it is questionable what the presented results can be used for. It should be possible from the available data to come up with a better classification of the aerosol to replace the included speculation which is useless in this context.

This was a very helpful comment. In particular, we have now revised the trajectory analysis, arriving at a more detailed classification scheme for the air masses observed. This has allowed us to identify an initial period where the trajectories are likely to be influenced by both marine and continental sources (trajectories that arrive from the north having travelled along the west coast of Vancouver Island and the BC mainland), an intermediate period of descending, non-boundary layer air from the west, a period of air that is clearly from the marine boundary layer without continental influence, followed by a final period of air that was within the marine boundary layer, originated from the west (i.e. Pacific Ocean), and spent time over Vancouver Island. This new classification is helpful as it identifies the highest ambient kappa values as being clearly associated with the marine boundary layer air. Thanks for this suggestion.

We retain in the paper our initial discussion of the MSA/Sulfate ratio as an additional indication that the air mass has a marine influence. As well, from the NO_x and SO_2 data it is clear that there is a pollution source added to this air, likely from local marine traffic.

2. Size distributions were measured with an SMPS, but the size distributions are not presented nor is anything stated about particle number concentrations. Only the integrated particle volume is presented. The size distribution and number concentrations are very relevant for studies related to ambient CCN and the reported CCN activities – so they should be included in the paper.

The total number size concentration and particle size distribution as measured by the SMPS have been included as Figure 2. The typical number concentrations for both the 50nm and 100nm CCN measurements, i.e. as measured after the DMA, ranged from about 2 particles cm^{-3} to 20 particles cm^{-3}.

Please see the Specific Comments for details.

3. A lot of basic information about the experimental set up, the measurements and the methods is lacking. From the limited information presented it is not possible to reproduce a similar study in the same way. This issue is particularly pronounced with respect to the measurement conditions for the MOUDI since the results are likely to be very sensitive to changes in RH. Furthermore, nothing is stated about how the data analysis was carried out with respect to the ACSM. The specific comments related to these issues should be addressed when brought up below.

We have added additional experimental detail to the paper on both the MOUDI and ACSM measurements, as well as other aspects of the experiments.

4. The authors have decided to base several of their conclusions on results obtained on the basis of the assumption that there must be closure between the PM sampled with a MOUDI and size distributions measured with an SMPS. It is highly questionable whether it makes sense to expect such closure between the aerosol volume measured with an SMPS and the MOUDI under the given circumstances. Even under controlled lab conditions – with a well characterized aerosol - that is not an easy task – and it would require thorough calibrations that do not seem to have been carried out in the present study. In the present study the two instruments do not even share the same inlet. Furthermore, the results are most likely very sensitive to the diameter conversion – which in turn depends on a lot of assumptions. Some of the choices made in the calculations are not justified and errors and uncertainties are not estimated or even mentioned in this relation. The authors want to leave the impression that their results using this method are quite robust. In order to justify that a proper sensitivity analysis should be carried out.

The reviewer's perspective is correct. Without an external evaluation of the accuracy of this approach we deemed it better to remove the determination of the aerosol organic amounts from a comparison of SMPS to MOUDI data.

We still include the conversion of aerodynamic to mobility diameters in the Supplementary Material section, with a full error propagation to better address the critical aspects of the calculations.

5. The obtained results with respect to the organic fraction and kappa_org are presented without any critical sense – both in light of the comment above (4) related to the method – and the suspiciously high kappa_org presented. A kappa_org of 0.5 is highly unlikely – but that does not seem to be clear from the discussion/conclusion. It would be appreciated if the authors could justify those findings instead of referencing a paper where the highest kappa reported for a single organic compound is about 0.4. Furthermore, a significant fraction of the ambient kappa values are reported to be 0.3 or lower – how can that be explained if the kappa_org is in the range 0.3-0.5?

The estimation of κ_{org} using the combined SMPS and MOUDI datasets has been replaced with its inference using a comparison of $\kappa_{ambient}$ and the ACSM's organic to sulfate ratio on page 12536 and in Fig. 8. We feel that this relatively simple approach, which was qualitatively included in the original manuscript, is merited because it hinges upon only one major assumption (stated in the paper), i.e. that the ACSM PM1 composition is directly related to that of CCN-sized particles. While we arrive at similar conclusions as in the ACPD version of the paper, we agree with the reviewer that the potential errors in the SMPS-MOUDI comparison approach do not merit its inclusion in the final paper. As a result, we are now reporting a κ_{org} for the entire period of ACSM measurements

(κ_{org} = 0.3 ± 0.2, August 13 – 23). As well, we indicate that the κ_{org} value during the marine period (August 13, 18 – 20) may be just as large, but we clearly indicate that there are large uncertainties associated with its determination.

Specific comments

Structure: Order of figures: it would be easier for the reader if references where made to the figures so that the order of the figures correspond to when they are discussed in the text. Title: 'Hygroscopicity' traditionally refers to measurements of hygroscopic growth. Replacing 'Hygroscopicity' by 'CCN activity' would be more appropriate. 'Organic component' could be replaced by 'aerosol chemical composition'.

Thank you for mentioning this. The order of the figures has been double checked.
The title has been changed to "CCN activity of size-selected aerosol at a Pacific coastal location". Thank you for this suggestion.

Abstract: The results and statements related to the combination of MOUDI and SMPS data should undergo a more thorough sensitivity analysis before the findings are included in the abstract.

The comparison of MOUDI and SMPS data has been removed from the paper.

P 12526, L. 5-11: The main findings of the studies Albrecht (1989) and Twomey (1977b) are that an increase in CCN is likely to influence cloud properties and thus climate. That is not clear from the way these studies are cited.

Thank you for pointing this out. P 12526, L. 23-25 has been clarified as follows:

"Warm clouds can trap the Earth's outgoing infrared radiation, reflect incoming solar radiation, and influence the hydrological cycle and thus impact its climate (Albrecht, 1989; Twomey, 1977)."

P 12527, L. 9: "breaking waves" do generally not generate all the primary sea spray aerosol particles.

Thank you for pointing this out. P 12527, L. 8-10 has been rephrased as follows:

"Marine aerosol consists of two distinct sources: (1) primary sea-spray aerosol directly produced by breaking waves, consisting of inorganic salts and biogenic material such as surface-active microorganisms and exopolymer secretions…"

P 12528, L. 17-20: should be rephrased.

This sentence has been rephrased to:

"The CCN abilities of aerosol particles depend on both their sizes and compositional characteristics, such as water solubility and surface activity (Petters and Kreidenweis, 2007, 2013). By using this parameterization, these CCN abilities can be reported using a single standardized method."

P 12529: what was the altitude of the inlet above the sea surface?

The inlets were 25.5 m above sea level, as has been clarified on page 12529, L. 9:

"All instruments sampled air through a main inlet in a trailer's roof, with intakes about 25.5 m above sea level."

Are the sampled air masses influenced by the surf zone – depending on wind direction?

The wind and surf were sufficiently low that we do not believe we were influenced by the surf zone.

Is the tide pronounced?

The difference in low and high tide height ranged from 1.0 m to 3.1 m throughout August, as is now mentioned in the paper on P 12529, L 6-7.

Are there any local aerosol sources nearby which could affect the measurements?

There are few local anthropogenic sources on land. In particular, the measurement site is located at a quiet Coast Guard Station with only a handful of cars coming and going each day. It is 2 km from the small community of Ucluelet (population 1800), separated by a road with little traffic. This detail has been added to P 12529, L 5:

"The field campaign took place in August 2013 at a coastal field site which was situated roughly 100 m from shore and 2 km from the small town of Ucluelet…"

This new information is now added to the manuscript. We believe that the sources of local pollution come from the ocean. In particular, the following information was presented when interpreting the presence of NO_x, SO_2, and CO, beginning on P. 12531, L. 21:

"Fig. 4 shows a time series (PDT) of the daily sea-surface temperature, SO_2 and NO_x mixing ratios, and CO mixing ratios. Based on the presence of NO_x, SO_2, and CO, it is clear that the air masses were anthropogenically influenced at times. These three species tended to increase between 11am and 11pm on most days, with the maximum mixing ratios occurring between 5pm and 8pm (e.g. August 12 and 15). As a result, in the cases when the air masses were of type (c), i.e. western, within the boundary layer, and of marine origin (see Fig. S1), it is likely that these signals arise from boat traffic (e.g. whale watching, fishing, pleasure, and sight-seeing boats). The maximum mixing ratios, which occurred in the late afternoon, were possibly due to these vessels entering the harbor (3.5 km from the sampling site) and docking for the night."

P 12529, L 11: 'directly calculate the kappa' - is usually not possible from CCN spectra alone. Which assumptions were made e.g. with respect to temperature and thus surface tension?

Kappa was calculated with the assumption that the surface tension of water is 0.072 J/m^2, according to (Petters and Kreidenweis, 2007). The trailer's temperature was taken to be 25^oC, which was the mean temperature measured periodically inside of the trailer.

These assumptions have been added to P. 12529, L. 10 as follows:

"The focus of this work is on data that was collected using a cloud condensation nuclei counter (CCN, DMT 100), which was used to calculate the κ of the ambient aerosol ($\kappa_{ambient}$), with the assumption that the droplets' surface tensions were 0.072 J m^{-2} (Petters and Kreidenweis, 2007) and the trailer's temperature was constantly 25^{o}C."

P 12529, L. 12-14: How 'broad' were the selected particle distributions, and what were the typical number concentrations?

The typical number concentrations for both 50nm and 100nm particles ranged from about 2 particles cm^{-3} to 20 particles cm^{-3}. This has been added to P 12529, L. 14 as follows:

"Typical number concentrations for both sizes ranged from 2 to 20 particles cm^{-3}."

The breadths of the selected particle distributions were not measured.

P 12529, L. 14: 'monodisperse': How were multiply charged particles avoided?

Multiply charged particles were not avoided, i.e. we performed the CCNC scans using the output from the DMA. This point has now been made in the text on P 12529, L 19 as follows:

"As the CCN sampled directly from the output of the DMA, multiply charged particles were not avoided."

P 12529 L. 12+23: Which RH was the aerosol dried to?

The silica gel in the diffusion dryers was exchanged and dried in an oven every 24 hours in order to reduce the water content of the particles as much as possible. Although not measured on site in this campaign, we find that this always reduces the relative humidity to less than 20% and usually to less than 2% (Ladino et al., 2014). This has been added to P 12529, L. 14:

"The silica gel in the diffusion dryers was exchanged and dried in an oven every 24 hours in order to reduce the water content of the particles as much as possible. Although not measured on site in this campaign, we have found that this technique always reduces the relative humidity to less than 20%, and usually to less than 2% (Ladino et al., 2014)."

P 12529, L. 17: (0.07-1.2) -> (0.07-1.2 %)

Thank you for pointing this out. It has been corrected.

P 12529, L. 17: It is recommended that details about how the supersaturation of the CCNC was calibrated should be included (Rose et al., 2008)

Thank you for this suggestion. The following sentence has been added on P. 12529, L. 17:

"The CCN counter was calibrated twice during the campaign using ammonium sulfate according to the procedure of Rose et al. (2008)."

P 12529, L. 23-25: Was an impactor used in front of the SMPS?

A 0.071 cm impactor was used at the inlet of the SMPS. This has been included on P. 12529, L. 26:

“A 0.071 cm diameter impactor was attached to the inlet of the SMPS, which sampled from the same main inlet as the CCN counter and CPC.”

P 12530, L. 2: What does ‘wet’ refer to here?

Thank you for pointing out this lack of clarity. “Wet” refers to ambient particles that have not been dried has been clarified as follows:

“A micro-orifice uniform deposition impactor (MOUDI, MSP 110R) sampled directly from a separate main inlet, with a sample flow of 24 L min^{-1}, to collect ambient particles that had not been dried (i.e. wet aerosol particles) …”

P 12530, L. 11-12: if mobility diameters are reported here – then a comment about where to find the information of how they are derived should be given.

Thank you for pointing this out. The following sentence has been added on P. 12530, after L. 12:

“The derivation of the particles’ mobility diameters from their aerodynamic diameters can be found in Sect. 1 of the Supplementary Material.”

P 12530: It is misleading to report this collection efficiency – as pointed out by Ng et al.(2011) it depends on the aerosol being sampled. It should be stated which assumptions the ACSM data analysis is based on – and why.

This is a typical value of the AMS collection efficiency. While it is true that the value may depend on aerosol composition, the value is not relevant to the paper since we only rely upon the ratio of organic aerosol mass to sulfate aerosol mass. If the particles are internally mixed, as they commonly are to a large degree, then the collection efficiency does not affect this ratio.

P 12529-12530: Were the instruments kept in air conditioned surroundings at constant temperature or did the temperature fluctuate?

The temperature and relative humidity of the trailer containing the CCN, SMPS, and MOUDI setups did fluctuate. This has been added to P. 12530, L. 14 as follows:

“The temperature and relative humidity in the trailer containing the CCN, SMPS, and MOUDI setups were monitored for the duration of the campaign (Acurite 00891W3). The temperature was found to range from 21^{o}C to 30^{o}C (mean value of 25^{o}C), and the relative humidity varied from 28% to 58% (mean value of 45%) from August 7 to 23.”

The trailer’s temperature containing the ACSM and SO_2, CO, NO_x, and O_3 monitors was controlled to 22^{o}C. This has been included on P. 12530, L. 17.

P 12531, L. 3: ‘three types of air masses’ is this statement reasonable here? Can the type of an air mass alone be based on the backtrajectory? Showing just three backtrajectories is not very informative. It is recommended that more are included in the figure. The air masses seem to be classified according to the backtrajectories, but no information is provided about the classification scheme.

Thank you for pointing this out. We have now revised the trajectory analysis, arriving at a different classification scheme for the air masses observed, rather than a simple classification based on trajectory direction alone. This has been clarified on P. 12531, in place of the old air mass classification scheme’s description:

“During the entire campaign there were four types of air masses that arrived in Ucluelet: (a) air that was influenced by both marine and continental sources (trajectories that arrive from the north having travelled along the west coast of Vancouver Island and the BC mainland), (b) descending, non-boundary layer air from the west, (c) air predominantly from the west that was clearly from the marine boundary layer without continental influence, and (d) air that was within the marine boundary layer, originated from the west (i.e. Pacific Ocean), and spent time over Vancouver Island.”

P 12531, L. 14-17: why this discussion about ‘marine’ when the ratio clearly depends highly on many parameters that cannot be quantified here – as pointed out at the end of the paragraph?

This discussion is presented as an attempt to interpret the ratio of MSA to nss-SO_4^{2-} that was found in this study. We did not intend to give the impression that this ratio uniquely defines the marine content on an air mass, which is why we went on to point out that this ratio depends on a number of parameters. Reporting the MSA to nss-SO_4^{2-} ratio is analogous to the widely used practice of reporting acetonitrile or CO values during a biomass burning episode. There are widely varying values of those quantities prevalent during biomass burning periods, but elevated levels generally indicate a biomass burning influence.

P 12531, L. 21-12532, L. 7: The question here is: can these measurements in combination with wind direction and other time series be used to classify which air masses that are highly influenced, moderately influenced or not influenced by anthropogenic emissions? This would be useful for the interpretation of the aerosol characterization.

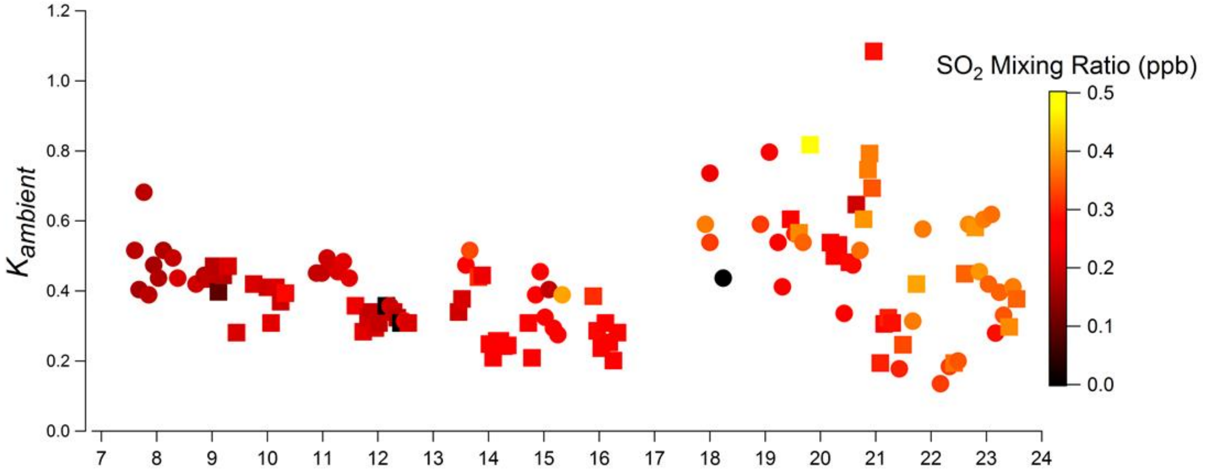
We attempted to use the SO_2, NO_x, and CO measurements to classify the degrees to which the air masses were influenced by anthropogenic emissions, with the end goal being to determine a relationship between each of these gases and $\kappa_{\mathrm{ambient}}$. The figure below (now Fig. S3 in the Supplementary Material) shows the variation in the average SO_2, NO_x, and CO mixing ratios as $\kappa_{\mathrm{ambient}}$ varied with time. However, a relationship was not found between these variables. This is now included on P. 12535, in the last paragraph:

“Finally, the relationships between $\kappa_{ambient}$ and the mixing ratios of SO_2, NO_x, and CO were investigated using Fig. S3 of the Supplementary Material; however, no correlations were found.”

As described above, the back-trajectories have been used to re-classify the corresponding air masses according to whether they were (a) influenced by both marine and continental sources (trajectories that arrive from the north having travelled along the west coast of Vancouver Island and the BC mainland), (b) descending, non-boundary layer air from the west, (c) clearly from the marine boundary layer without continental influence, or (d) within the marine boundary layer, originated from the west (ie. Pacific Ocean), and spent time over Vancouver Island. This classification scheme has been used to re-interpret κ_{ambient} in Fig. S1 in the Supplementary Material and on P. 12533, as follows:

"Through the use of the air mass classification scheme described in Sect. 3.1, four periods of air masses and $\kappa_{ambient}$ values are identified. First, an initial period where the trajectories were likely to be influenced by both marine and continental sources (i.e. air mass type (a), August 7 – 12, representing trajectories that arrive from the north having travelled along the west coast of Vancouver Island and the BC mainland); second, an intermediate period of descending, non-boundary layer air from the west (i.e. air mass type (b), August 14 – 16 and 23); third, a period of air that was clearly from the marine boundary layer to the west without continental influence (i.e. air mass type (c), August 13 and 18 – 20); and finally, a period of air that was within the marine boundary layer, originated from the west (ie. Pacific Ocean), and spent time over Vancouver Island (i.e. air mass type (d), August 21 – 22).

Using this air mass classification, the highest $\kappa_{ambient}$ values, and most hygroscopic particles, (August 18 – 21) are identified as being associated with the marine boundary layer air without continental influence (air mass type (c)). In contrast, air masses that were not necessarily only of marine origin (e.g. influenced by the continent or descended from above the marine boundary layer) were associated with lower $\kappa_{ambient}$ values. When the $\kappa_{ambient}$ values during each of the four air mass time periods were averaged, it was found that the average $\kappa_{ambient}$ values for both 50 nm and 100 nm particles were 0.41 ± 0.08, 0.33 ± 0.10, 0.57 ± 0.16, and 0.35 ± 0.16, during air mass types a, b, c, and d, respectively, where the uncertainties represent standard deviations."


SO₂ Mixing Ratio (ppb)
0.0
0.1
0.2
0.3
0.4
0.5
K_ambient
0.0
0.2
0.4
0.6
0.8
1.0
1.2
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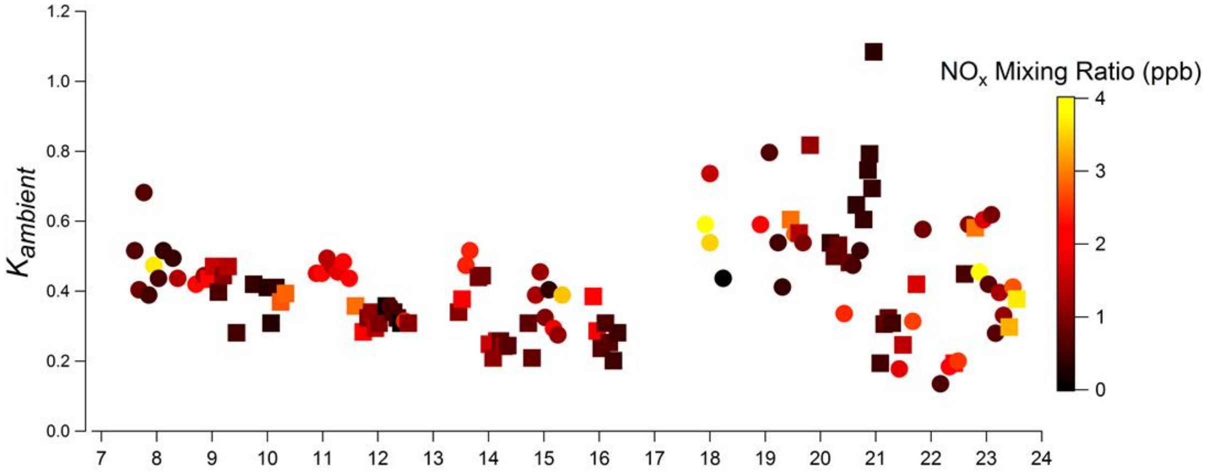
NOₓ Mixing Ratio (ppb)
0
1
2
3
4
K_ambient
0.0
0.2
0.4
0.6
0.8
1.0
1.2
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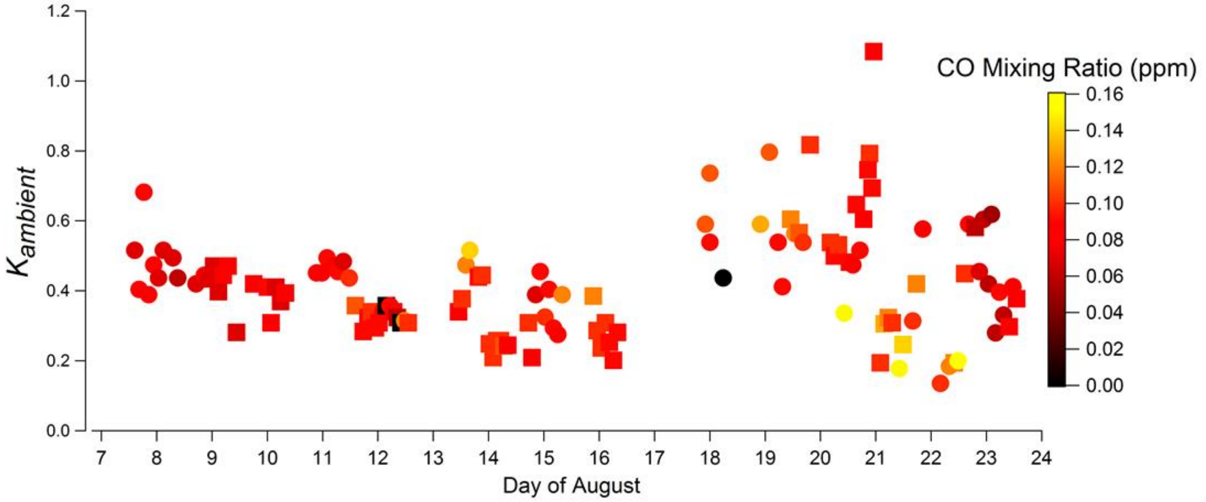
CO Mixing Ratio (ppm)
0.00
0.02
0.04
0.06
0.08
0.10
0.12
0.14
0.16
K_ambient
0.0
0.2
0.4
0.6
0.8
1.0
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Day of August

P 12532, L. 2-7: where are the mentioned harbor, local industry and forests located relative to the measurement station? Is there any signature in the data series from these sources?

The small harbor was 3.5 km from the measurement site. The contribution of boat traffic to the results was likely due to the vessels passing the measurement site on their way to the harbour rather than being in the harbour itself. The area is entirely forested, so any air mass that passes near the coast has the potential to be impacted by forest emissions. These sources have been clarified on P. 12532, L. 2:

"The maximum mixing ratios, which occurred in the late afternoon, were possibly due to these vessels entering the harbor (3.5 km from the sampling site) and docking for the night."

The nearest industrial centre (i.e. pulp and paper mills) was 80 km inland from the sampling site. As a result, it probably did not have a substantial influence on the results, and so that potential source has been removed from the text.

P 12532, L. 8-11: Is sea salt or sea salt sulfate detected by the ACSM? According to Ng et al.(2011) only non-refractory species are detected (at least in a quantifiable way). Please clarify how nss-sulfate is obtained or estimated.

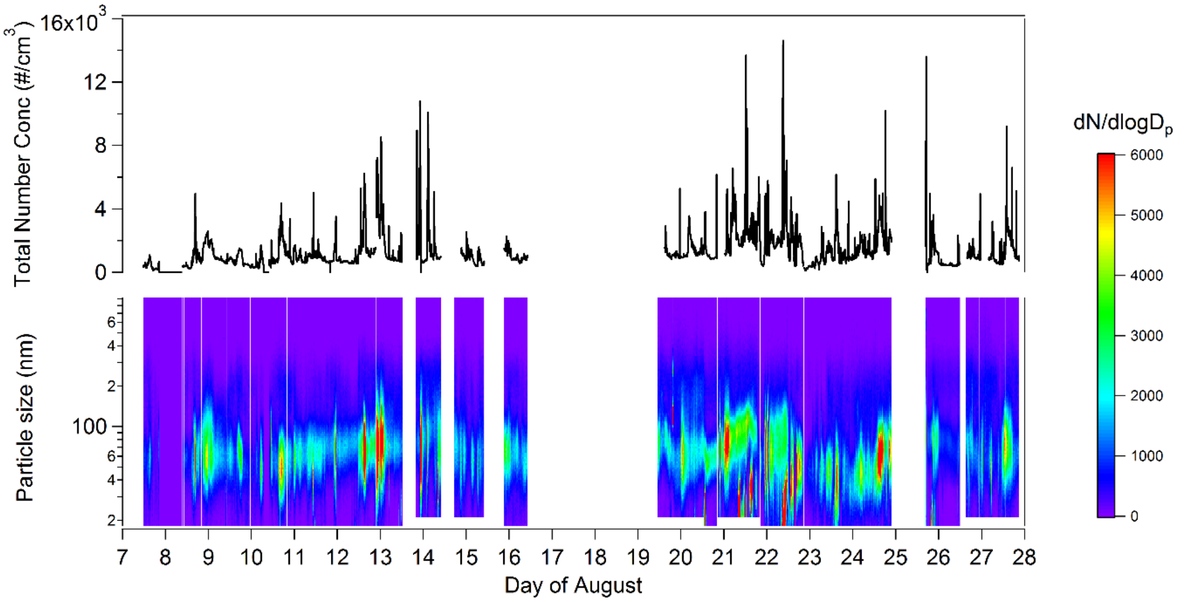
The ACSM detected total sulfate but did not detect sea salt. Na^+ (measured by ion chromatography) from the MOUDI samples was used as a tracer to detect nss-SO_4^{2-} according to Warneck (1988). Using this method, the mass concentration of sea salt SO_4^{2-} can be approximated by the mass concentration of Na^+ as follows:

$$[\text{nss-}SO_4^{2-}] = \text{total } [SO_4^{2-}] - [\text{sea salt } SO_4^{2-}] = \text{total } [SO_4^{2-}] - 0.27[Na^+],$$

where 0.27 is the ratio of the fraction of sulfate to sodium in seawater.

P 12532, L. 13: Why is only total volume from the SMPS data shown? What were the total number concentrations? Which particle size modes dominated? It would be useful for the interpretation of the results to show number size distributions.

Below are the total number concentrations as a function of time and the particle size distribution for the study's duration. The dominant particle size range was typically 40 – 100nm. These have been included as Fig. 2 (shown below).



P 12532, L. 16-19+Fig.3: Is the time in Fig. 3 (and all other figures with time series) given in local time?

All of the time series are given in Pacific Daylight Savings Time (PDT). This has been added to the captions of all of the figures.

P 12533, L. 6: "where the solution is assumed to be water" – at which temperature?

The temperature was assumed to be 25°C, which as an appropriate assumption based on the temperature measurements in the trailer. This detail has been added to P. 12533, L. 6 as follows:

"… where D_d is the dry particle's mobility diameter, $\sigma_{s/a}$ is the surface tension of the solution/air interface (where the solution is assumed to be water at 25°C)…"

P 12533, L. 8: 'particle size (10%)' -> does the 10% represent the standard deviation of the size selected aerosol? Or is it related to possible systematic errors?

This refers to the error that is associated with size selection using DMAs. This has been clarified in the sentence as follows:

"Based on the uncertainties in the particle size (±10%), temperature (±10°C), and instrumental and calibrated critical supersaturations (±10)…"

P 12533, L. 9: It is not reasonable to calculate the error in Kappa based on the error in diameter and supersaturation alone. The error in kappa depends on the error in the critical super saturation, which also depends on the quality and resolution of the activated fraction vs SS curves. Such a figure could be shown in Supplementary material. Nothing is stated about how multiply charged particles are accounted for.

Doubly and multiply charged particles cause a plateau to occur at smaller diameters in CCN activation curves (Rose et al., 2008). In order to determine if these particles had a substantial effect on the corresponding kappa, the fraction of activated doubly charged particles was determined from the height of this plateau. Because this fraction was assumed to be constant over the entire particle size range, the activated fraction of singly charged particles was calculated according to Rose et al. (2008):

$$\left(\frac{N_{CCN}}{N_{CN}}\right)_1 = \frac{N_{CCN} - N_{CN} \cdot \left(\frac{N_{CCN}}{N_{CN}}\right)_2}{N_{CN} - N_{CN} \cdot \left(\frac{N_{CCN}}{N_{CN}}\right)_2}$$

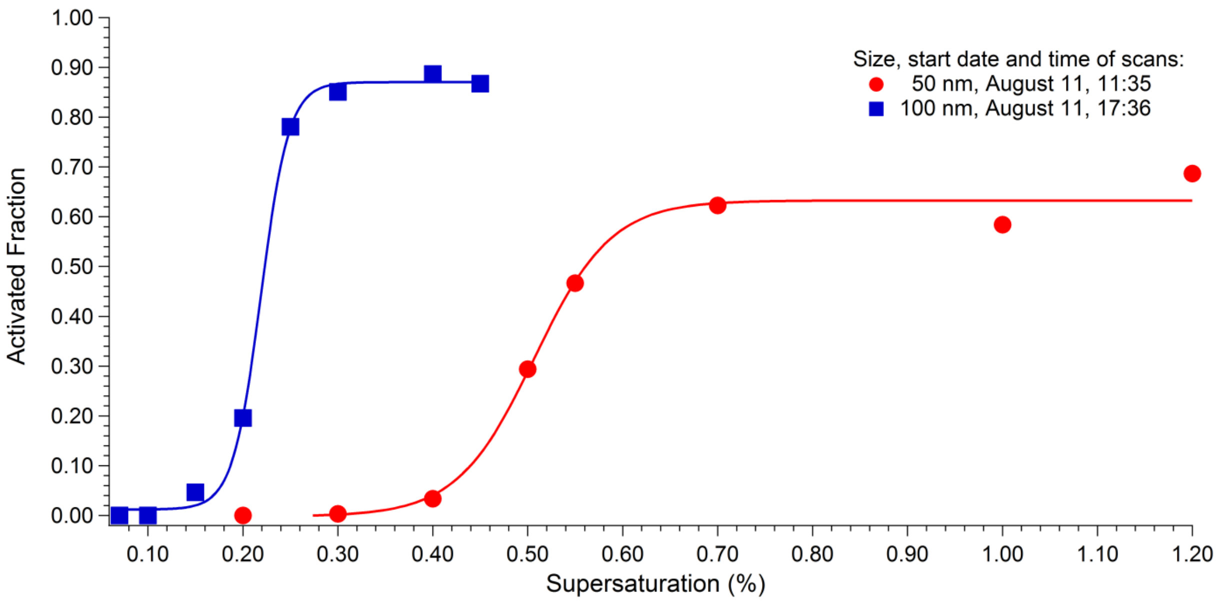
Where N_{CCN} represents the number concentration of CCN, N_{CN} represents the total number concentration of particles, $(N_{CCN}/N_{CN})_1$ represents the corrected activated fraction, and $(N_{CCN}/N_{CN})_2$ represents the uncorrected activated fraction (Rose et al., 2008).

Using this method, several corrected kappa values were calculated and compared to the corresponding uncorrected kappa values in order to determine whether the presence of doubly or multiply charged particles had a substantial effect on the kappas. It was found that the effect was insignificant since the kappa values changed by a maximum of 0.05. As a result, uncorrected kappa values have been reported in the study.

This has now been included on P. 12529, L. 19:

"As the CCN sampled directly from the output of the DMA, multiply charged particles were not avoided. However, the effect of multiply charged particles on the results (i.e. $\kappa_{ambient}$) was investigated according to a method summarized in Rose et al. (2008). It was found that the effect was insignificant, and so the uncorrected $\kappa_{ambient}$ values have been reported."

Below are examples of the activated fraction versus supersaturation curves for 50nm and 100nm particles. This figure has been added as Fig. S2 to the Supplementary Material.



Because the quality of the curves was very good, the error in the instrumental critical and calibrated supersaturations was estimated to only be ±10%. This has been clarified on P. 12533, L. 9 as follows:

"Based on the uncertainties in quantities such as the particle size (±10%), temperature (±10°C), and instrumental and calibrated critical supersaturations (±10%)…"

Please note that if the activated fraction's plateau was lower than 0.65 the scan was not used as the air was likely significantly polluted. This has been stated on P. 12529, L. 17:

"Note that if the activated fraction's plateau was lower than 0.65 the scan was not used as the air was likely significantly polluted, and the focus of this work was on the marine component of the aerosol. It is possible that low plateaus arise from an externally mixed population of low hygroscopicity carbonaceous particles from nearby marine pollution sources, but we have no direct evidence this is the case."

P 12533, L. 19: 'two time periods' -> it is not clear which two time periods are being referred to… is it 18-21 Aug and then the time periods before and after combined? Please clarify.

Thank you for pointing this out. The phrase "two time periods" has been removed and replaced with the actual dates throughout the paper.

P 12533, L. 23-24: Again it is not clear what 'first period' and 'second period' refers to.

Thank you for pointing this out. The phrases "first time period" and "second time period" have been removed and replaced with the actual dates throughout the paper.

P 12533, L. 24-12534, L. 6: It is not clear how kappa values higher than 0.7 can be explained by biological activity/photochemistry.

Thank you for pointing out this lack of clarity. Enhanced biological activity can lead to the enhanced production of DMS by e.g. phytoplankton, which is then converted into sulfate through photochemistry, as was described on P. 12534, L. 1-2. This sulfate can then be present in various forms, including $(NH_4)_2SO_4$ and H_2SO_4, which have high kappa values of 0.61 and 0.90, respectively. Therefore, it is possible that the relatively sunny time period that occurred from August 18 to 20 resulted in an enhancement of photochemistry, and therefore an enhancement of H_2SO_4 production, which would have driven up the aerosol's $\kappa_{\mathrm{ambient}}$ value.

This has been clarified and moved to P. 12536, before L. 10:

"The presence of sunlight would then have enhanced the photo-oxidation of DMS to SO_4^{2-}, the levels of which were particularly high during this time period as shown in Fig. 5, whereas the organic mass loadings were not so affected. Because acidic sulfate has high kappa values (e.g. 0.90 for sulfuric acid), it is likely that the relatively sunny time period (August 18 to 20) led to the aerosol's $\kappa_{ambient}$ values to be driven up to values exceeding 0.7."

P 12534, L. 3-6: The discussion relating cloud coverage to CCN activity is speculative – support the discussion on findings in the data/literature or leave it out.

This discussion has been removed.

P 12534, L. 7-13+Fig.7: Be careful here –PM1 composition is being directly related to measurements carried out around 50 nm and 100 nm.
P 12534, L. 9: 'Increasing sulfate' – in Fig. 7 only a ratio is included – which does reflect the change in sulfate alone.

Thank you for both of these comments. This discussion has been moved to P. 12536, two paragraphs before L. 10, and the point that the comparison of kappa to the organic to sulfate ratio is between PM1 and 50 and 100 nm particles has been clarified, and that sulfate is only increasing relative to the organic material has also been mentioned:

"By inspection of Fig. 8 (bottom), where $\kappa_{ambient}$ is considered as a function of the ACSM's PM1 organic to SO_4^{2-} ratio from August 13 to 23, it is observed that some of the variability in the $\kappa_{ambient}$ data may be explained through the relationship between $\kappa_{ambient}$ and the PM1 organic to SO_4^{2-} ratio of the aerosol, where increasing SO_4^{2-} in relation to organics resulted in increasing $\kappa_{ambient}$ values."

P 12534, L. 9-13: 'dependency of the CCN activity' – of course the CCN activity depends on the chemical composition – please clarify what is meant here.

Previous studies have demonstrated that aerosol sulfate content, in particular, has a dominant influence on the CCN properties of marine aerosol. This discussion has been moved to P. 12536, two paragraphs before L. 10 and clarified:

"The particular dependence of the CCN activity of coastal and marine aerosol on sulfate content is supported by previous studies that partially attributed the CCN properties of marine aerosol to the presence of this species (Andreae and Rosenfeld, 2008b; Matsumoto et al., 1997; Quinn and Bates, 2011, and references therein)."

P 12535, L. 8-13+Fig.8: if the authors wish to include such a figure in the paper – they such provide more information in this section about the method and assumptions/errors involved. It cannot be justified to present fig. 8 as it is without a more thorough analysis.

Thank you for pointing this out. Fig. 8 and its discussion have been replaced with a plot of the molar concentrations of ionic species present in the two size bins, as measured by the MOUDI/IC (see Fig. 7 and discussion on P. 12535, L. 8). In addition, the estimation of the organic content using a comparison of the SMPS and MOUDI has been removed, and Sect. 1 of the Supplementary Material has been considerably enhanced in order to clarify all of the assumptions and errors in the conversion of the MOUDI's aerodynamic diameters to mobility diameters. Finally, Table S2 has been modified to only include the assumed densities of the ionic bearing constituents (ρ) used in the calculations described in Sect. 1 of the Supplementary Material.

P 12535, L. 21-24: This very general statement is not true – and it is not what is shown in the cited studies. O'Dowd et al.(2004) clearly show that the marine aerosol composition is highly dominated by sea salt – only during periods with high biological activity the particulate matter below roughly 500 nm is dominated by sulfate and organic species.

The authors did not intend to imply that all aerosol in the previous studies was dominated by sulfate and organic material. However, it has been reported that particle size ranges that contain the size ranges considered in this study (42 – 75 nm and 78 – 141 nm) are dominated by sulfate and organic material. Because the estimation of organic content using the MOUDI and SMPS has been removed from the manuscript, this portion of the discussion now focusses on the content of sulfate, and not organic, species, and the sentence has been clarified accordingly:

"The observation that sulfate species dominate the composition of particles of the size ranges considered in this study agrees well with previous studies of the chemical composition of marine aerosol (Good et al., 2010; Hawkins et al., 2008; Moore et al., 2012; O'Dowd et al., 2004; Phinney et al., 2006; Prather et al., 2013)."

P 12536, L.1-9: If the aerosol is dominated by anthropogenic or continental sources – then a correlation between wind speed and kappa_ambient cannot be expected. Also if the organics are primary marine organics and supposedly high in concentration mixed with sea salt – then it is not clear that a correlation should be observed between wind speed and kappa_ambient. It seems more reasonable to investigate a relationship between particle number concentrations and wind speed to the power of ~3 (with winds approaching from the ocean and no expected influence from anthropogenic emissions) – if a signature of primary marine aerosol is to be investigated.

In addition to wind speed, the relationship between wind speed cubed and the total number concentrations was investigated, and no correlation was found. If found, this correlation could have arisen from either the formation of primary marine aerosol or the enhanced ventilation of DMS from the ocean. This has been clarified on P. 12532:

"For the majority of August there was a clear diurnal trend in wind speed and power (i.e. wind speed cubed, not shown), with mid-day winds weaker than those occurring later in the day and into the night. Although this could be expected to cause a corresponding trend in the particle mass loadings if there is a large primary source, there was not a clear correlation between wind speed, wind speed cubed, and either the total number concentrations or total particulate volume concentrations shown in Fig. 2 (top) and Fig. 3 (top), respectively."

P 12536, L.12-13: PM1 usually does not cover the 'total aerosol loading'.

Thank you for mentioning this error. It has been corrected on P. 12536, L. 10, as follows:

"By inspection of Fig. 8 (bottom), where $\kappa_{ambient}$ is considered as a function of the ACSM's PM1 organic to SO_4^{2-} ratio…"

P 12536, L. 16-18: This is NOT a proper way to estimate the error on kappa_org! Either meaningful error propagation calculations should be carried out – or in this case a reasonable sensitivity study is probably more adequate.

Thank you for point this out. Because κ_{org} is now estimated using the ACSM's PM1 organic to sulfate ratio, the estimation of its error from this calculation has been removed from the manuscript.

P 12537, L. 20-23: It is claimed that it is likely to find a mixture of ambient organic compounds with a kappa around 0.5 with a reference to Petters and Kreidenweis (2007). The highest reported kappa for organic species reported in that study is about 0.4 – so it is not fair to base that statement on that reference. The authors should do a better job in justifying a kappa_org of 0.5 – if they really want to claim that is a result of their study.

The Petters and Kreidenweis (2007) reference was not referring to the kappa_org of 0.5. It was referring to the statement that molecules having higher kappas are "as an upper limit, smaller, more water soluble, or more surface active than" other organic molecules. However, this sentence has been removed from the manuscript.

A substantial fraction of the kappa_ambient datapoints is below 0.3 – how is that explained if kappa_org is above 0.3?

κ_{org} is now estimated using through the comparison of $\kappa_{ambient}$ and the ACSM's organic to sulfate ratio (Fig. 8 and discussion on page 12536). According to this method, when data from August 13 to 23 is used, κ_{org} is estimated to be 0.3 ± 0.2, indicating that the organic kappa may be as small as 0.1.

P 12538, L. 4-8: rephrase

This sentence has been rephrased:

"A cloud condensation nucleus counter was used to investigate the CCN activity of ambient aerosol particles on the west coast of Canada (Ucluelet, BC) in August 2013 as one aspect of NETCARE."

P 12538, L. 16-19: It is not clear what 'in the past' refers to here. It was e.g. suggested by Charlson et al.(1987) that secondary aerosol could play an important role as CCN in marine environments.

Thank you for pointing out the lack of clarity in this sentence. It has been rephrased as follows:

"The dominant presence of secondary ionic components such as nss-SO_4^{2-} (13 – 39%), and the low concentrations of Na^+ (a tracer for sea salt), highlights the importance of these secondary species in general, in addition to the primary components that have been reported to be drivers of the CCN properties of marine aerosol by previous studies (e.g. Andreae and Rosenfeld, 2008)."

Fig.1: It would be useful to show the variance in backtrajectories and indicate the criteria for classification into North, West and South as shown e.g. in Fig S1.

Fig. 1 has been removed, and the air masses have been re-classified into four types, as are now described in Sect. 3.1: (a) air that was influenced by both marine and continental sources (trajectories that arrive from the north having travelled along the west coast of Vancouver Island and the BC mainland), (b) descending, non-boundary layer air from the west, (c) air that was clearly from the marine boundary layer without continental influence, and (d) air that was within the marine boundary layer, originated from the west (i.e. Pacific Ocean), and spent time over Vancouver Island.

In addition, Fig. S1 has been included in the Supplementary Material, where trajectories are shown for local noon on each day of the study.

Fig.3: It is hard to tell what the volume concentration is from the upper panel. Do the spikes reflect noise that has not been removed – or is it real? It would be very useful to show the number concentration and number size distributions also/instead. It may be useful to indicate in the lower panel when the wind direction was classified as 'west'.

Please see pages 10 – 11 of this response for the figure of the total number concentrations and size distributions as a function of time.

Thank you for pointing out the spikes in the plot of volume concentrations. We do not know the source of these spikes but they could well arise from either a small boat close to shore or one of the few cars on site – the parking lot is about 100 m away.

In addition, in the bottom plot, the wind direction has been classified according to whether the air was (a) influenced by both marine and continental sources (trajectories that arrive from the north having travelled along the west coast of Vancouver Island and the BC mainland), (b) descending, non-boundary layer air from the west, (c) clearly from the marine boundary layer without continental influence, or (d) within the marine boundary layer, originated from the west (i.e. Pacific Ocean), and spent time over Vancouver Island.

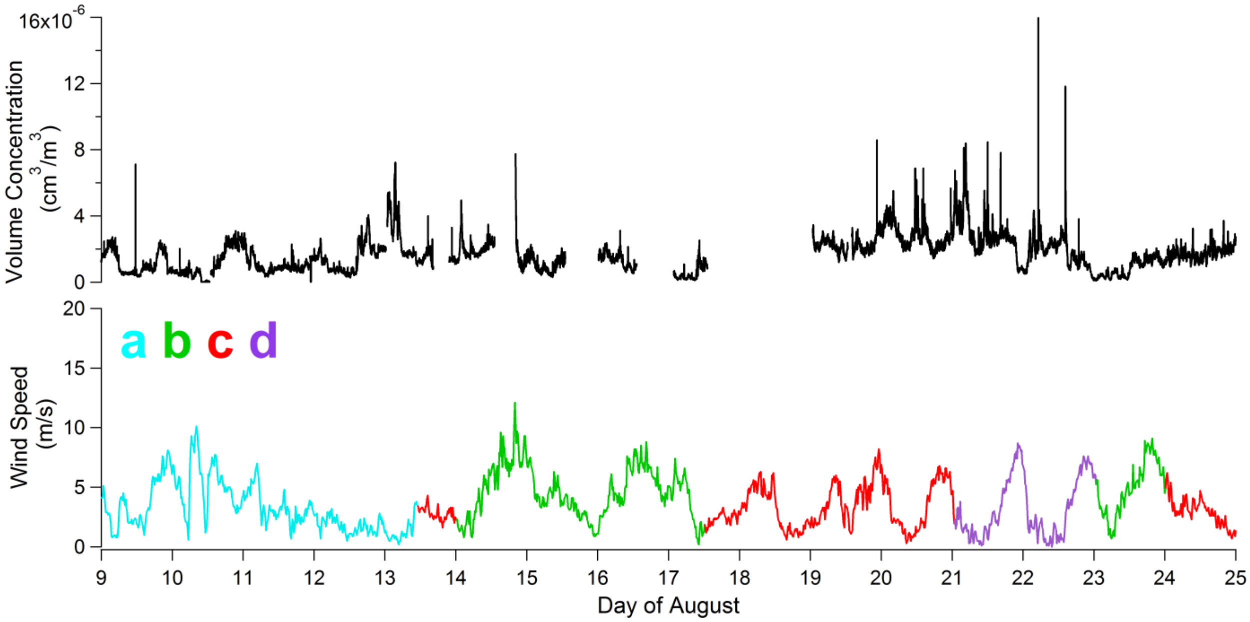


Fig.4: It seems like there is a trend in the SO2 concentration – is that an instrument artifact?

It is true; there is a trend in the data that is likely an instrument artifact. Indeed, the minimum values in this plot may reflect a baseline issue in the measurement. The value of the plot is to indicate that there are only small SO2 pollution spikes sitting on top of this baseline.

Fig.5: It is hard to read this figure. Please use a different scale for the different species – or use a logscale.

Thank you for pointing this out. The mass concentrations of the organics, NO_3^-, and SO_4^{2-} have been plotted on three separate axes in Fig. 5 for clarity, where the Organics axis has been cut off at 5 μg/m^3:

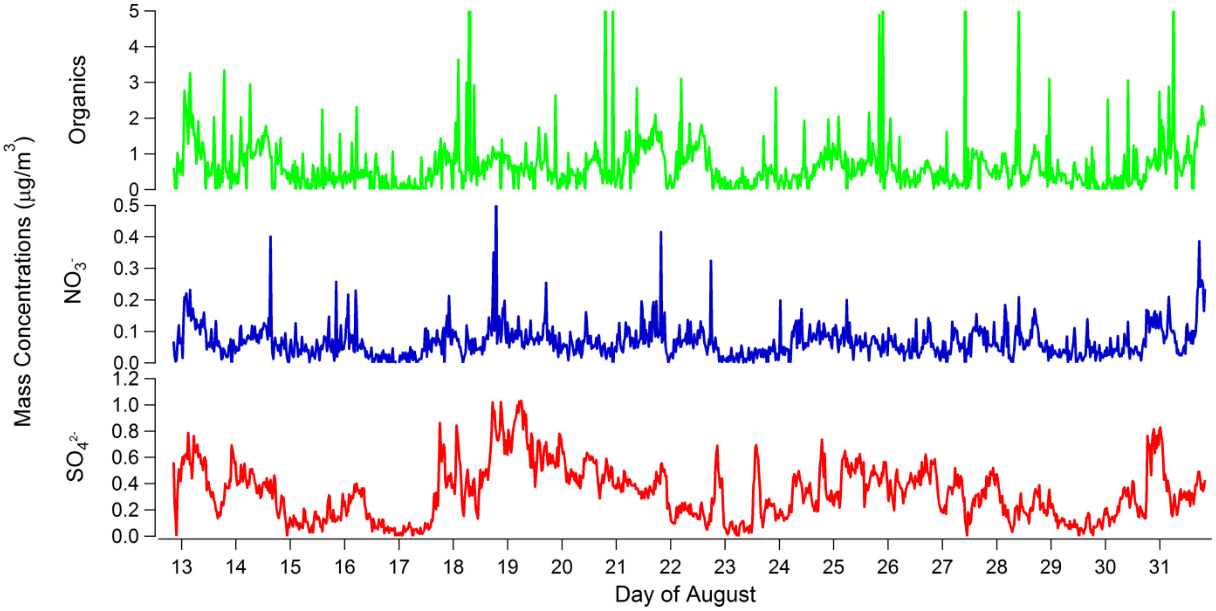


Fig. 6: Caption: "dry mobility diameters of 50 nm (red circles) and 100 nm (blue squares)". In the figure also red squares and blue circles are presented! With 1.5 to 2 h per scan there should ideally be 12 to 16 datapoints per 24 h. Are datapoints missing due to problems interpreting the CCN spectra or only due to instruments failing?

Thank you very much for pointing out the red squares and blue circles. The authors did not notice this mistake and have corrected it (shown below). The CCN did fail periodically, which resulted in missing data points.

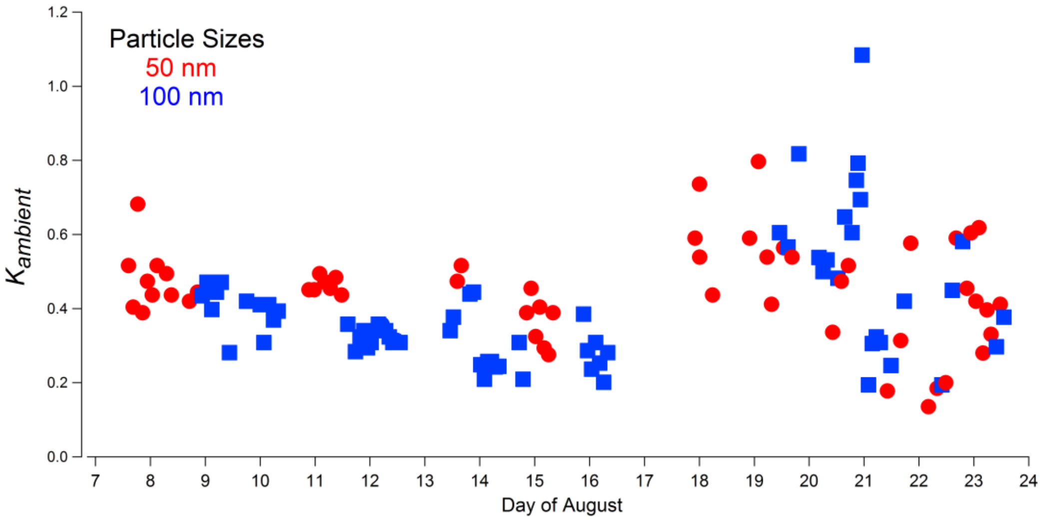


Fig.7: It is questionable whether this figure contains useful information – see comments above.

We feel there is value to this figure (now Fig. 8) as a rough indication of the dependence of the ambient kappa on the chemical nature of the aerosol, showing that more sulfate rich PM1 particles have a higher ambient kappa. While we recognize that the ACSM is sensitive to larger particles than those under study, it is likely that the gas-to-particle processes that govern the accumulation mode particles are similar to those prevalent for smaller particles, especially when the aerosol is largely secondary as in this case. That the relationship between the hygroscopicity of 50 nm and 100 nm particles is only very rough has been clarified on P. 12536, at the end of the first paragraph:

"We stress that, because the ACSM is sensitive to PM1, with an upper size range close to 1 μm, i.e. much larger than the size of CCN, this can only be viewed as a rough estimation of κ_{org}, with the assumption that the chemical compositions of PM1, and 50 nm and 100 nm particles are similar to one another."

Fig.8: It is highly questionable whether this figure contains meaningful results – with respect to the organic fraction.

This figure has been replaced by Fig. 7, which shows the two-day average ionic molar concentrations as measured by the MOUDI. In addition, the estimation of the organic concentration, using the comparison of the MOUDI and SMPS has been removed from the manuscript.

Supplement:

Page 1:

1) More information about the measured RH please.

   The RH monitor was an Acurite 00891W3, as is now indicated on P. 12530, L. 14.

2) The last sentence of this paragraph is very problematic:
   I: it is simply not true that this sensitivity test validates the 'aforementioned assumption' – particularly not when it is being claimed elsewhere that about 80% of the mass is highly hygroscopic organics.
   II: a doubling of water present - which change in RH does that correspond to?
   III: If the size range of the MOUDI corresponds to a dm, which is on average just 10 nm smaller than included in the calculations – then this will most likely have a huge influence on the calculated organic fraction presented in Fig. 8 – and thus the inferred kappa_org.

These are good points. While it is true that the organics may be hygroscopic, they are not as hygroscopic as the inorganic salts. However, without prior knowledge of their hygroscopicity we cannot confidently account for the amount of condensed phase water that they contribute to the particles. For that reason we performed the sensitivity analysis where we doubled the amount of condensed phase water. Note that these calculations are no longer used to estimate κ_{org}.

Page 2:
A lot of highly uncertain assumptions are made – without much/any justification and the authors want to give the impression that the results are very robust. Please provide proper references for the choices made and report/estimate errors. Please perform a proper sensitivity test based on reasonable error estimates: e.g. how does a 10-20 nm lower dm (or whatever is reasonable) affect the results?

Thank you for this suggestion. A much more detailed description of the calculations, their corresponding sources of error, and error propagation is now included in Sect. 1 of the Supplementary Material in place of a sensitivity analysis. Also, as noted already, these calculations are now not related to the derived value of the organic kappa.

Anonymous Referee #2

We would like to thank both reviewers for their comments, which led to substantial improvements to the paper.

1) Please include more description (or at least some references) of the size-resolved CCN measurements, and the derivation of $\kappa_{\mathrm{ambient}}$.

Thank you for these suggestions. More details have been included in the description of the CCN measurements on P. 12529, according to the following questions.

How was the activation fraction derived?

Details of the derivation of the activated fraction have been added to P. 12529, L. 17:

"The number concentrations measured by the CCN and CPC (N_{CCN} and N_{CPC}, respectively) were used to calculated the activated fraction, AF = N_{CCN} / N_{CPC}, examples of which are shown in Fig. S2 of the Supplementary Material. Note that if the activated fraction's plateau was lower than 0.65 the scan was not used as the air was likely significantly polluted, and the focus of this work was on the marine component of the aerosol."

Did SMPS step or scan through different sizes?

As has been elaborated upon on P. 12529, CCN experiments were conducted on either 50 nm or 100 nm particles (i.e. the DMA was fixed at one size for a period of time), and while the size was fixed, the supersaturation within the chamber was increased in a step-wise manner from 0.07% to 1.2%.

How was the multiple charged particles taken into consideration?

Doubly and multiply charged particles cause a plateau to occur at smaller diameters in CCN activation curves (Rose et al., 2008). In order to determine if these particles had a substantial effect on the corresponding kappa, the fraction of activated doubly charged particles was determined from the height of this plateau. Because this fraction was assumed to be constant over the entire particle size range, the activated fraction of singly charged particles was calculated according to Rose et al. (2008):

$$\left(\frac{N_{CCN}}{N_{CN}}\right)_1 = \frac{N_{CCN} - N_{CN} \cdot \left(\frac{N_{CCN}}{N_{CN}}\right)_2}{N_{CN} - N_{CN} \cdot \left(\frac{N_{CCN}}{N_{CN}}\right)_2}$$

Where N_{CCN} represents the number concentration of CCN, N_{CN} represents the total number concentration of particles, $(N_{CCN}/N_{CN})_1$ represents the corrected activated fraction, and $(N_{CCN}/N_{CN})_2$ represents the uncorrected activated fraction (Rose et al., 2008).

Using this method, several corrected kappa values were calculated and compared to the corresponding uncorrected kappa values in order to determine whether the presence of doubly or multiply charged particles had a substantial effect on the kappas. It was found that the effect was insignificant since the kappa values changed by a maximum of 0.05. As a result, uncorrected kappa values have been reported in the study.

This has now been included on P. 12529, L. 19:

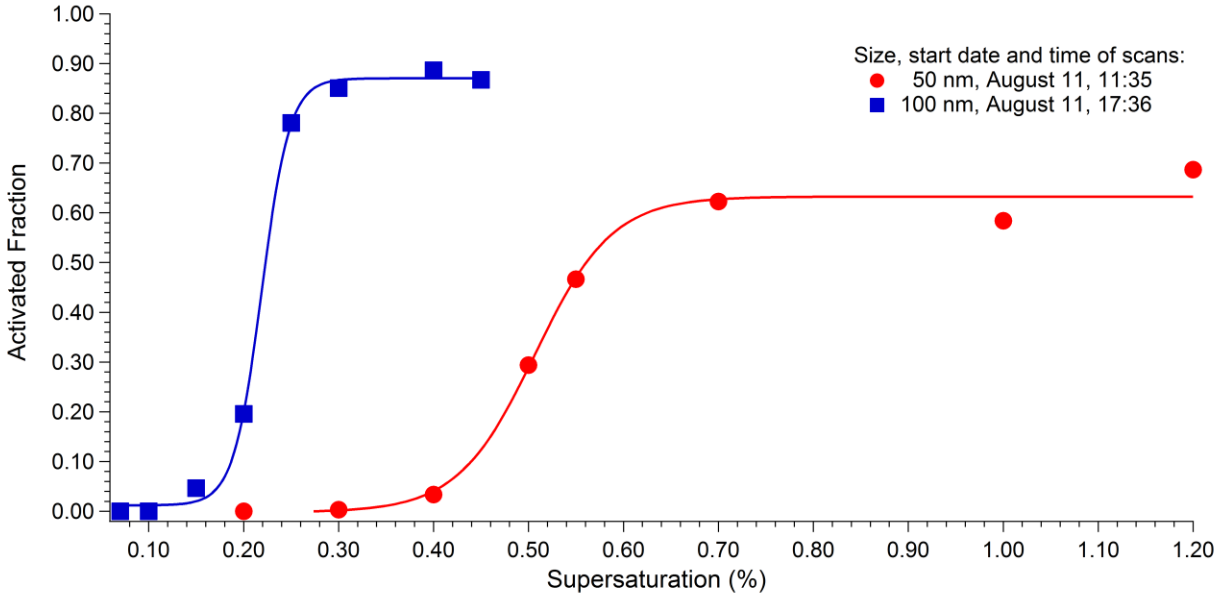
"As the CCN sampled directly from the output of the DMA, multiply charged particles were not avoided. However, the effect of multiply charged particles on the results (i.e. $\kappa_{ambient}$) was investigated according to a method summarized in Rose et al. (2008). It was found that the effect was insignificant, and so the uncorrected $\kappa_{ambient}$ values have been reported."

2) It took about 1.5 to 2 hours for a full scan. Did aerosol properties vary substantially during 1.5 - 2 hour periods, given the impact of frequent boat traffic (Fig 4a)? If so, how would this affect derived $\kappa_{\mathrm{ambient}}$?

Because the scans lasted 1.5 to 2 hours it was assumed that the aerosol was chemically consistent over this time period. This was a reasonable assumption based on the activation curves (examples of which are shown below and in the Supplementary Information as Fig. S2), i.e. the curves are generally characterized by stable plateaus and clear sigmoidal shapes. Had the overall particle composition been changing during a supersaturation step-wise scan, the plateaus would not have been flat and the sigmoidal shapes would not have been produced.

This is now included on P. 12533, L. 15:

"In addition, it should be noted that because the activation curves (examples of which are shown in the Supplementary Material as Fig. S2) were generally characterized by stable plateaus and clear sigmoidal shapes, it was assumed that the CCN-active aerosol was chemically consistent and internally mixed during each experiment."



3) Page 12532, Lines 24-25: Again, please provide more details about how Sc was derived. The current approach appears to be based on the assumption that particles are internally mixed. Does the size resolved CCN measurement suggest internal mixture at DMA selected sizes?

Evidence that the aerosol was externally mixed was provided in that the activated fractions' plateaus rarely reached 1. However, because the curves were characterized by stable plateaus and clear sigmoidal shapes, it was assumed that the CCN-active aerosol was chemically consistent and internally mixed during each experiment. This is now included on P. 12533, L. 15:

"In addition, it should be noted that because the activation curves (examples of which are shown in the Supplementary Material as Fig. S2) were generally characterized by stable plateaus and clear sigmoidal shapes, it was assumed that the CCN-active aerosol was chemically consistent and internally mixed during each experiment. However, there was evidence that the aerosol as a whole was externally mixed, containing both soluble and insoluble components, because the activated fractions' plateaus did not often reach 1."

4) Page 12533, Lines 8-9: Uncertainties in the particle size (10 %) and instrumental supersaturation (5 %) should lead to much greater uncertainty in $\kappa_{\mathrm{ambient}}$ according to Eq. (1). Please include more details on how the uncertainty in $\kappa_{\mathrm{ambient}}$ was derived.

Thank you for this comment. The uncertainty in $\kappa_{\mathrm{ambient}}$ was re-calculated through the error propagation of the equation shown below:

$$\kappa_{ambient} = \frac{4A^3}{27D_d^3 \ln^2 S_c}, \text{ where} = \frac{4\sigma_{s/a}M_w}{RT\rho_w},$$

D_d is the dry particle's mobility diameter, $\sigma_{s/a}$ is the surface tension of the solution/air interface (where the solution is assumed to be water at 25^{o}C), M_w is the molecular weight of water, R is the gas constant, T is the temperature, and ρ_w is the density of water.

By assuming typical errors of 10%, 10%, and 10^{o}C in the particle size, critical and calibrated supersaturation, and temperature, respectively, it was found that the systematic error in $\kappa_{\mathrm{ambient}}$ is 37%. The details of how this uncertainty was derived have been included on P. 12533, L. 8 – 10:

"Based on the uncertainties in quantities such as the particle size (±10%), temperature (±10^{o}C), and instrumental and calibrated critical supersaturations (±10%), the relative error in $\kappa_{ambient}$ is calculated to be ±37% through error propagation."

The error $\kappa_{\mathrm{ambient}}$ has been included in the caption of each applicable figure rather than as error bars in the figures themselves.

5a) Page 12533, Line 19 - Page 12534, Line 2: Does the composition measurement in these two size ranges indicate enhanced sulfate concentration/fraction as suggested?

Yes, the ACSM data (Fig. 5) clearly show high levels of sulfate during this period. This point is now made on P. 12536, before L. 10 of the paper:

"The presence of sunlight would then have enhanced the photo-oxidation of DMS to SO_4^{2-}, the levels of which were particularly high during this time period as shown in Fig. 5, whereas the organic mass loadings were not so affected."

5b) Many $\kappa_{\mathrm{ambient}}$ values during August 18-23 are greater than 0.6. Could the larger values of Kappa be due to higher NaCl contribution during this period?

Thank you for this comment. Significant NaCl concentrations were not observed during this time; however, $\kappa_{\mathrm{ambient}}$ values greater than 0.6 could be attributed to the presence of sulfate. This has been clarified on P. 12536, before L. 10 as follows:

"The presence of sunlight would then have enhanced the photo-oxidation of DMS to SO_4^{2-}, the levels of which were particularly high during this time period as shown in Fig. 5, whereas the organic mass loadings were not so affected. Because acidic sulfate has high kappa values (e.g. 0.90 for sulfuric acid), it is likely that the relatively sunny time period (August 18 to 20) led to the aerosol's $\kappa_{ambient}$ values to be driven up to values exceeding 0.7."

6) Page 12536, Line 22 - Page 12537, Line 3: There have been a number of studies that derive κ_{org} from CCN/ size resolved CCN measurements. For example, Lathem et al., 2013 and Mei et al., 2013 a,b. Please include previous studies in the discussion.

Thank you for this comment. It is true that a number of studies have derived κ_{org} in primarily continental (Lathem et al., 2013) and urban (Mei et al., 2013a, b) settings; however, the purpose of this discussion was to examine the work that has been done on deriving κ_{org} in a marine setting that was not substantially influenced by continental or anthropogenic sources (e.g. large biomass burning events or industrial centres).

The suggested references have been included as follows:

P. 12537, L. 10-13:
"As was the case for Bougiatioti et al. (2009), this range in κ_{org} was comparable to the hygroscopicity parameters of oxidized organic material of continental origin (e.g. Dusek et al., 2010; Lathem et al., 2013; Petters and Kreidenweis, 2007; Shantz et al., 2008)."

P. 12527, L. 20-26:
"While an enormous amount of effort has been applied to understanding and characterizing the aerosol hygroscopicity and CCN properties of continental biogenic, anthropogenic, and primary sea-spray aerosol (e.g. Andreae and Rosenfeld, 2008; Bigg, 2007; Chang et al., 2010; Coggon et al., 2012; Fuentes et al., 2011; Hegg et al., 2009; Kanakidou et al., 2005; Langley et al., 2010; Leck and Bigg, 2005a, 2005b; Mei et al., 2013a, 2013b; Moore et al., 2011; Orellana et al., 2011; Ovadnevaite et al., 2011; Prather et al., 2013; Roberts et al., 2006; Shantz et al., 2010; Sun and Ariya, 2006)"

7) Given the large variation in $\kappa_{ambient}$ and very different backtrajectories, I would suggest derive κ_{org} for different air mass types, instead a single average value. Including comparison and discussion of aerosol hygroscopicity, size distribution, and composition among different air masses would also strengthen the paper.

Thank you for this suggestion. In the new version of the manuscript, κ_{org} is estimated using a comparison of $\kappa_{ambient}$ and the ACSM`s PM1 organic to sulfate ratio (see Fig. 8 and the discussion on page 12536). Using this method, the κ_{org} for August 13 to 23, during which time both marine and continental air was measured, has been estimated to be 0.3 ± 0.2. The κ_{org} for clean marine air (August 13 and 18 – 20) appears to be at least 0.3, but with very high uncertainties, which is notable since according to the MOUDI/IC data, the aerosol was largely not composed of NaCl, or sea salt.

8) Page 12537, Line 20: I think the range of κ_{org} (0.3-0.5) is too high. I am struggling to find an example organic compound that has an effective Kappa in this range.

Thank you for this comment. With the new analysis (from the ACSM data only), the κ_{org} values for both continental and marine air (0.3 ± 0.2), while large, is in reasonable agreement with the literature.

9) Supplementary Information, section 1: The mass concentration of organics is derived as the difference between SMPS and MOUDI measurements. As the two measurements are based on very different principles, this might lead to substantial uncertainties in derived organics concentration. I am wondering if this approach can be validated by deriving the total organics mass concentration, which can then be compared to direct measurements from ACSM.

Thank you for this comment. Because of the difficulties in validating the comparison of SMPS and MOUDI measurements, this section of the manuscript has been removed and substituted by a discussion of the MOUDI ionic concentrations (Fig. 7) and the relationship between $\kappa_{ambient}$ and the ACSM's organic to sulfate ratio (Fig. 8), as was previously mentioned.

10) Supplementary Information, section 1: The derivation of particle density (Eq. S1) is based on the assumption that organics don't take up water. This appears to be inconsistent with very high κ_{org} derived. What is the impact of water uptake by organics on derived particle density, and particle composition?

These are good points. While it is true that the organics may be hygroscopic, they are not as hygroscopic as the inorganic salts. However, without prior knowledge of their hygroscopicity we cannot confidently account for the amount of condensed phase water that they contribute to the particles. For that reason we performed the sensitivity analysis where we doubled the amount of condensed phase water. We note that these calculations are no longer used to estimate κ_{org}.

11) Figure 3: Please consider changing the unit from cm^3/m^3 to $\mu m^3/cm^3$, which is equivalent to $\mu g/m^3$ when density is $1 g/cm^3$.

Thanks for the suggestion, which has been implemented.

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