

Response to Reviewer #1

This paper describes the detection and quantification of sea salt aerosol with a TOF-ACSM equipped with a capture vaporizer. The authors performed calibration experiments with size-selected NaCl particles and used the results to analyze ambient data from two field campaigns as well as chamber data with NaCl seeds. This paper is a nice contribution to the literature and should be accepted after consideration of the following comments:

Response:

We thank the reviewer for the positive assessment of our work and for recognizing its contribution to the literature. We also appreciate the constructive comments provided. We have carefully considered all points and revised the manuscript accordingly. All author responses are marked in blue, and changes in the revised manuscript are indicated in red.

Major comments

1. The calibration experiments suggest that the response at 23, 58 and 60 is more proportional to aerosol surface area than to aerosol mass. Aerosol surface area is not commonly reported. How useful is it? Do modelers use it? I think you need some explanation for why this is an important quantity to report.

Response:

We agree that aerosol surface area is not as commonly reported as aerosol mass. However, our calibration experiments demonstrate that the ACSM response at m/z 23, 58, and 60 scales with particle surface area rather than particle mass.

We attribute this behavior to incomplete evaporation of NaCl in the capture vaporizer, which operates as a surface-limited process, since only the fraction of the particle in direct contact with the vaporizer is efficiently vaporized and detected. Consequently, surface area represents the most direct and assumption-free quantity derived from the ACSM signal under these conditions. Since NaCl does not completely evaporate in ACSMs/AMS measurements, this likely also applied to standard vaporizers and any calibration with respect to mass would only be valid for a similar size distribution as the calibration aerosol. Therefore we report NaCl surface area, even though it is not as useful as mass concentration.

NaCl aerosol mass can be calculated from the measured surface area, if prior knowledge or estimates of the NaCl size distribution is available, which can e.g. be gained from impactor measurements for certain regions.

Abstract (line 23-27):

The calibration experiments show that the ACSM response (ions/s) at characteristic NaCl fragments scales with the available particle surface area, yielding a surface-normalized signal that is only weakly dependent on particle size for monodisperse NaCl aerosol. This indicates that the CV-ToF-ACSM provides a direct and robust measure of sea salt aerosol

surface area. If a prior estimate of the NaCl size distribution is available this signal can be converted to aerosol mass concentrations.

Introduction (line 60 - 70):

ACSM instruments typically use a standard vaporizer (SV). To diminish the effects of particle bounce, the capture vaporizer (CV) was developed (Xu et al., 2018). The CV contains an inner cavity to reduce the effects of particle bounce, therefore improving the collection efficiency (CE) of the instrument to ~ 1 . Although the CV improves CE compared to the SV, its structure increases aerosol particle residence time, which increases thermal decomposition and results in distinct fragmentation patterns (Zheng et al., 2020). Sea salt is not routinely measured in ACSMs, because it does not fully vaporize due to its refractory nature and vaporization temperature requirements ($> \sim 600^\circ\text{C}$) (Bahreini et al., 2003; Zhang et al., 2011). Nevertheless, partial evaporation of ambient sea salt at temperatures of $\sim 600^\circ\text{C}$ had been demonstrated earlier (O'Dowd and Smith, 1993). It should be noted that the vaporization efficiency in the CV may exhibit a size dependence, as the process is governed by the surface-to-surface contact between particles and the vaporizer. This implies that the fraction of particle material that is effectively vaporized can depend on particle size.

2. If you apply Eq 1 to the ambient measurements for the marine air, how well does the calculated surface area agree with the surface area measured by the SMPS for that site, assuming all of the aerosol is sea salt aerosol? This would be an important confirmation of your approach.

Response:

We thank the reviewer for this valuable suggestion. We agree that, in principle, a comparison between ACSM-derived surface area and SMPS-derived surface area would provide an important consistency check. However, such a comparison is not directly feasible in our case.

First, NaCl represents only a small fraction of the total aerosol population at our coastal site, even under marine air mass conditions NaCl contributes less than 10% to the total aerosol population. Since the SMPS measures the size distribution of the entire aerosol population, the resulting surface area is not directly related to that of NaCl.

Second, sea salt particles typically contribute significantly at the upper end of, or beyond, the SMPS size range. The SMPS does not capture the particle fraction between 800 nm and 2.5 μm , where a substantial portion of sea salt mass (and associated surface area) resides. This leads to an incomplete representation of sea salt aerosol in the SMPS-derived surface area.

Due to these two factors, assuming that the SMPS-derived surface area represents NaCl alone would introduce substantial bias and is not appropriate for validating the ACSM-derived NaCl surface area.

3. Do you have any marine air masses at CESAR and what slopes do you get for the 23, 58 and 60 ions?

Response:

We investigated this by analyzing the data from the CESAR site in Cabauw as a function of wind direction. While we selected sectors corresponding to southwestern to northwestern winds to represent marine influence, it should be noted that Cabauw is located approximately 45 km inland from the North Sea coast and the air masses pass densely populated areas from the North Sea to the measurement site. Therefore CESAR does not experience purely marine air masses.

For these marine-influenced wind sectors, the signals at m/z 23, 58, and 60 remain very low, and no clear correlations between these marker ions are observed. We interpret this as an indication that sea salt concentrations at Cabauw are generally too low and close to or below the detection limit of the instrument under these conditions.

As a result, it is not possible to derive meaningful slopes between the NaCl marker ions for CESAR data.

4. The ions at 35, 36, 37 and 38 are also used to quantify Cl compounds. Did you look at those ions in the calibration experiments and/or the ambient measurements? It might be interesting if there were different ratios for marine (NaCl) and continental (NH_4Cl) air masses. In addition, not all CVs have signal at 58 and 60, so a fingerprint at the Cl ions would be useful.

Response:

We thank the reviewer for this valuable suggestion. We investigated the Cl-related ions (m/z 35, 36, 37, 38, as well as m/z 81 and 83) during the chamber experiments.

We found that the signals at m/z 37, 38, 81, and 83 were generally close to the detection limit of the instrument and therefore not suitable for quantitative analysis. The signals at m/z 35 and 36 were sufficiently above the detection limit; however, they were strongly influenced by the injection of α -pinene and subsequent SOA formation.

This indicates that these ions are not stable tracers under the investigated conditions, as they are subject to interference from organic aerosol formation and oxidative processing. As a result, they are less suitable as robust markers for NaCl compared to m/z 23, 58, and 60. We agree that Cl-related ions could be of interest for distinguishing between different chlorine-containing aerosol types, but our results suggest that their applicability is limited under realistic ambient conditions with significant organic aerosol formation.

5. With the standard vaporizer, calibrations for NaCl are done with a mixture of NaCl and NH_4NO_3 so that the NO_3 is an internal standard. Did you try that calibration technique? How well did the results agree with assuming $\text{CE}=1$? It seems clear from your reported size dependence that the CE is not actually 1 for NaCl particles even in the capture vaporizer.

Response:

We agree that the effective CE for NaCl in the capture vaporizer is well below 1 and is not fully constant across particle sizes. We did not perform mixed NaCl calibrations with NH_4NO_3 as an internal standard in this study. Our calibration approach focussed on directly relating the ACSM response to the known NaCl particle surface area. Within this framework, the measured response reflects the combined effects of collection efficiency and ionization efficiency, which cannot be independently separated using our experiments alone. A comparison between the NaCl response and typical NH_4NO_3 calibration responses suggests that the effective sensitivity for NaCl is substantially lower, consistent with incomplete

vaporization and $CE < 1$ for NaCl, even in the capture vaporizer configuration. We agree that this point deserves further emphasis and have revised the manuscript to clarify that the capture vaporizer improves, but does not fully eliminate, incomplete collection and vaporization effects for NaCl particles.

Results (line 201-205):

The derived slopes (-0.22 , -0.066 , -0.021 , and -0.31 for m/z 23, m/z 58, m/z 60, and the summed NaCl fragments, respectively) indicate a consistent size-dependent decrease in signal response. This trend is consistently observed across all NaCl marker fragments, suggesting a uniform reduction in ACSM response with increasing particle size. This indicates that the CE for NaCl in the CV is well below 1, which is expected due to the refractory nature of NaCl.

6. I am not completely clear on the point of including the AIDA data. You show that the α-pinene SOA does not have organic fragments at 23, 58 and 60 but, on the other hand, you clearly do have organic interference at 58 and 60 in the continental air masses in some seasons. In addition, for chamber experiments, you should adjust the frag table to account for the NaCl seed instead of lumping the 58 in with organics. If you want to keep the AIDA data, I would put it after the calibration and ambient data which are more important.

Response:

We agree that the AIDA chamber data fits better after the calibration and ambient data, and have made changes to the order of the manuscript accordingly.

In the chamber experiments, besides the default organics signal, the separate signals for M 58, 60 and 23 are displayed. Figure 5 shows that these three signals together make up the largest part of the organics signal when measuring pure NaCl aerosol. This is something we want to demonstrate in this manuscript, therefore we would prefer to show the uncorrected signal. When using these fragment ions to identify or quantify NaCl in ambient data or in chamber experiments, it should be taken into account that when measuring marine air masses, the signal at M 58 will not be organic and the fragmentation table can be updated accordingly.

Minor comments

Lines 56-58: You need another sentence or two explaining what the vaporizer is.

Response:

We have added additional sentences to clarify the function and role of the vaporizer.

Introduction (line 60 - 70):

ACSM instruments typically use a standard vaporizer (SV). To diminish the effects of particle bounce, the capture vaporizer (CV) was developed (Xu et al., 2018). The CV contains an inner cavity to reduce the effects of particle bounce, therefore improving the collection efficiency (CE) of the instrument to ~ 1 . Although the CV improves CE compared to the SV, its structure increases aerosol particle residence time, which increases thermal decomposition and results in distinct fragmentation patterns (Zheng et al., 2020). In addition, the vaporization efficiency in the CV may exhibit a size dependence, as the process

is governed by the surface-to-surface contact between particles and the vaporizer. This implies that the fraction of particle material that is effectively vaporized can depend on particle size.

Line 58: Be more specific about the impact of the CV on thermal decomposition. Instead of “can alter” say “increases.”

Response:

This change has been made to the text

Introduction (line 63 - 64)

Although the CV improves CE compared to the SV, its structure increases aerosol particle residence time, which increases thermal decomposition and result in distinct fragmentation patterns (Zheng et al., 2020).

Line 60: The difference in temperature between the SV at 600 C and the CV and 550 C has very little impact on fragmentation. It is really about the structure and residence time. I would delete the end of this sentence.

Response:

The suggested change has been made to the text.

Introduction (line 63 - 64):

Although the CV improves CE compared to the SV, its structure increases aerosol particle residence time, which increases thermal decomposition and results in distinct fragmentation patterns (Zheng et al., 2020).

Figure 1 caption: I don't see a purple shaded area. The org signal does not go to zero at the end. Is that 58 or something else?

Response:

The figure caption has been changed to correctly mention only existing lines and shaded areas. The organics signal not going to zero is because the instrument was still turned on after disconnection from the chamber, and sampling lab air. The figure caption has been updated to include this.

Results (lines 333 - 340):

Figure5: Time series of the fragment signals at m/z 23, m/z 58, and m/z 60 (light blue, yellow, and red, respectively), and the organics signal as assigned by default in the fragmentation table of the TOF-ACSM, during the AIDA chamber experiment on 18 January 2024. The pink shaded area represents the injection of NaCl particles from a 0.1 mass percent (wt%) aqueous solution. The polydisperse aerosol had an average number mode diameter of ± 222 nm as derived from the measurements with the two SMPS instruments. The purple lines represent the injection of 872 ppb ozone, the injection of 10 ppb α -pinene, and the injection of 34 ppb ammonia, respectively. α -pinene and ammonia were injected

with 10 standard liters per minute (slm). The black dashed line represents the time at which the ACSM was disconnected from the chamber, after which laboratory air was sampled.

Lines 197-198. I don't see how 58 can be the most dominant ion in Org after the α -pinene injection.

Response:

We thank the reviewer for pointing this out. We would like to clarify that the manuscript states that the fragment at m/z 58 is the most dominant contributor to the total OA signal after injection of NaCl and *before* injection of α -pinene, not after the α -pinene injection. We have rewritten the specific sentence to avoid confusion.

Results (line 346 - 348):

The fragment at m/z 58 is the most dominant contributor to the total OA signal after injection of NaCl. After the injection of α -pinene, the total OA signal increases substantially due to secondary organic aerosol formation.

Figure 2. You need some kind of legend for the different concentrations. The y-axes should be labeled with mass and surface area because it is easy to miss the change in units. Scale both y-axes from 0. I don't understand what the /sc and /mc in the legend are referring to.

Response:

We have changed the x-axis title to represent more clearly which one is about mass and surface area. Also, the legend was changed to include the different concentrations and marker sizes.

Line 221 and line 234. I don't think these slopes are useful to report since you are not using them to calculate anything and you would need the intercepts, too. You say there's a factor of two change for mass concentration. State what the relative change is for surface area.

Response:

We agree that the slopes do not necessarily need to be reported and have removed them from the text. Also, we have stated the relative change factor for the surface area of the particles.

Results (line 199-204):

For NaCl aerosol particles with a diameter of 150 nm, the normalized signal is approximately a factor of two higher than that for particles with a diameter of 250 nm, while the relative change in surface area is a factor of 1.67. The derived slopes indicate a consistent size-dependent decrease in signal response. This trend is consistently observed across all NaCl marker fragments, suggesting a uniform reduction in ACSM response with increasing particle size.

Line 228. "it is impossible" seems a bit strong. The size dependence does introduce some uncertainty but one could still report concentration.

Response:

We agree that the original wording was too strong. The text was changed accordingly.

Results (line 209 - 211):

Due to the observed size dependence, deriving ambient sea salt mass concentrations from the ACSM signal requires prior information on particle size and is otherwise subject to substantial uncertainty.

Line 235 “confirming the absence” is also a bit strong. There is a size dependence when normalized by surface area. It is just less pronounced.

Response:

Also here, we agree that the original wording was too strong. We have revised the text and made improvements regarding the wording.

Results (line 214 - 217):

Linear regression analysis yields slopes close to zero for all NaCl marker fragments (-3.9×10^{-3} , -1.2×10^{-3} , and -3.8×10^{-4} for m/z 23, 58, and 60, respectively) as well as for the summed NaCl signal (-5.4×10^{-3}), indicating that the size dependence is substantially reduced when normalized by surface area.

Line 261 and Figure 3 legends. Do you really think you have three significant digits in R²?

Response:

We thank the reviewer for this comment. We agree that reporting three significant digits for R² is not warranted. We have revised the values in the text and in Figure 3 caption to 2 significant digits (lines 236 - 243).

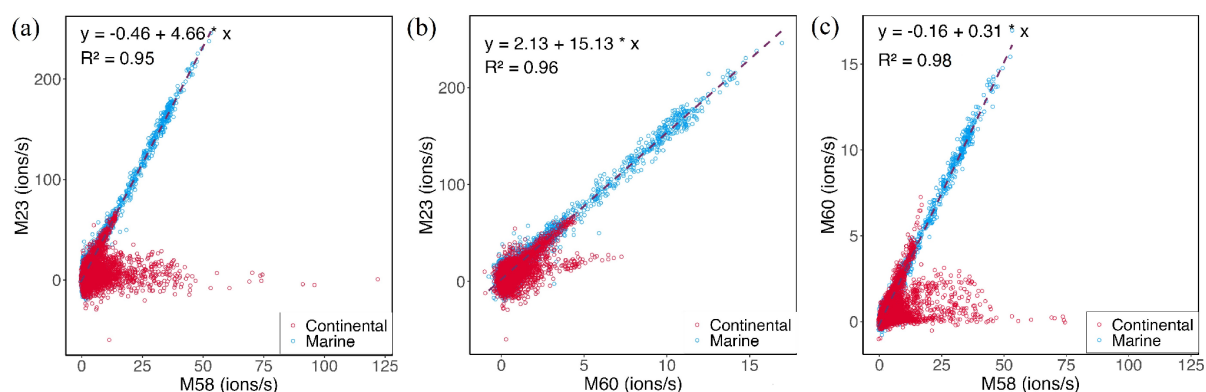


Figure 2: Correlations between the signals of m/z 58 and m/z 23 (a), m/z 60 and m/z 23 (b), and m/z 58 and m/z 60 (c) in ions/s as measured at the Lutjewad measurement station. The dashed lines indicate the regression lines for the marine air data points.

Figure 2 presents correlations between the ACSM signals of m/z 23 and m/z 58, m/z 23 and m/z 60, and m/z 60 and m/z 58, respectively. A clear dependence on air mass origin is observed. Under marine-influenced conditions (onshore wind), absolute signal intensities

are significantly higher than under continental influence (offshore wind). Furthermore, for marine air masses, the signal ratios remain highly consistent, with R^2 ranging from 0.95 to 0.98.

Lines 296-299. This is an interesting observation. Do you see enough SO₄ or NO₃ to confirm? Do you see different slopes at Cabauw?

Response:

Indeed, at our measurement site we almost always see high enough concentrations of SO₄ and NO₃ to facilitate NaCl ageing. At Cabauw we have an absence of the NaCl signal due to the site not being near enough to the coast, and therefore we cannot check for the slopes.

Line 343. The background signal also changes with time, even for a given instrument.

Response:

We thank the reviewer for this comment and agree that the background signal can vary over time, even for a given instrument. We have revised the text to reflect this temporal variability.

Results (line 325-326):

As the background signal at m/z 23 changes over time and is instrument specific, it must be determined individually for each CV-ToF-ACSM prior to application of this method.

Line 347 and line 364. I don't understand this comment about NaCl before aging. Do you think aging changes the surface area? How is what you report related to the actual aerosol?

Response:

The equation assumes that each of the sodium ions is coupled with a chloride ion, so no depletion of chloride is considered in the equation. The text has been changed to state this more clearly.

Results (lines 327-330):

Equation 1. is also strictly only applicable to above-background m/z 23 signals. Using this relationship for marine air masses at the coastal site, we show significant concentrations of NaCl up to $121 \mu\text{m}^2 \text{cm}^{-3}$. These concentrations correspond to NaCl before any chloride depletion, assuming that each detected Na ion is paired with a Cl ion.

Line 355. This claim of minimal interference is contradicted later in the paragraph. Do you mean for marine air masses?

Response:

We agree that the phrasing was ambiguous. The statement regarding "minimal interference" refers specifically to the controlled chamber experiments, where α -pinene SOA formation and oxidative ageing did not produce significant contributions at the NaCl marker ions. In contrast, interferences at m/z 58 and 60 can occur in ambient continental air masses, as discussed later in the paragraph.

We have revised the text to clarify this distinction.

Conclusions (line 369 - 377):

These fragments exhibited high signal strengths and minimal interference from organic fragments during controlled chamber experiments, including α -Pinene SOA formation and oxidative ageing. Laboratory experiments revealed that the ACSM response to NaCl aerosol is more closely related to available particle surface area than to total aerosol mass. A slight concentration dependence of the ACSM signal normalized to aerosol surface concentration was observed, with lower aerosol concentrations resulting in proportionally weaker signals. Field measurements at a coastal site confirmed strong positive correlations between the fragment signals at m/z 23, m/z 58 and m/z 60 when sampling marine air. In continental air masses, interferences from organic fragments at m/z 58 and m/z 60 are sometimes present, but can be distinguished by a lack of correlation between m/z 58 and m/z 60.

SI line 3. “inclusion in the analysis” rather than “data acquisition”

Response:

The suggested change has been made to the text.

SI:

After each change in NaCl aerosol concentration, the signal was allowed to stabilize before inclusion in the analysis.

Figure S1. Can you use different symbols for the different sizes? Those colors are very hard to tell apart. Also, “analyzed” rather than “collected” in the caption.

Response:

Figure S1 has been adjusted with different symbols for the three particle mobility diameters. Also, the figure caption has been adjusted.

SI

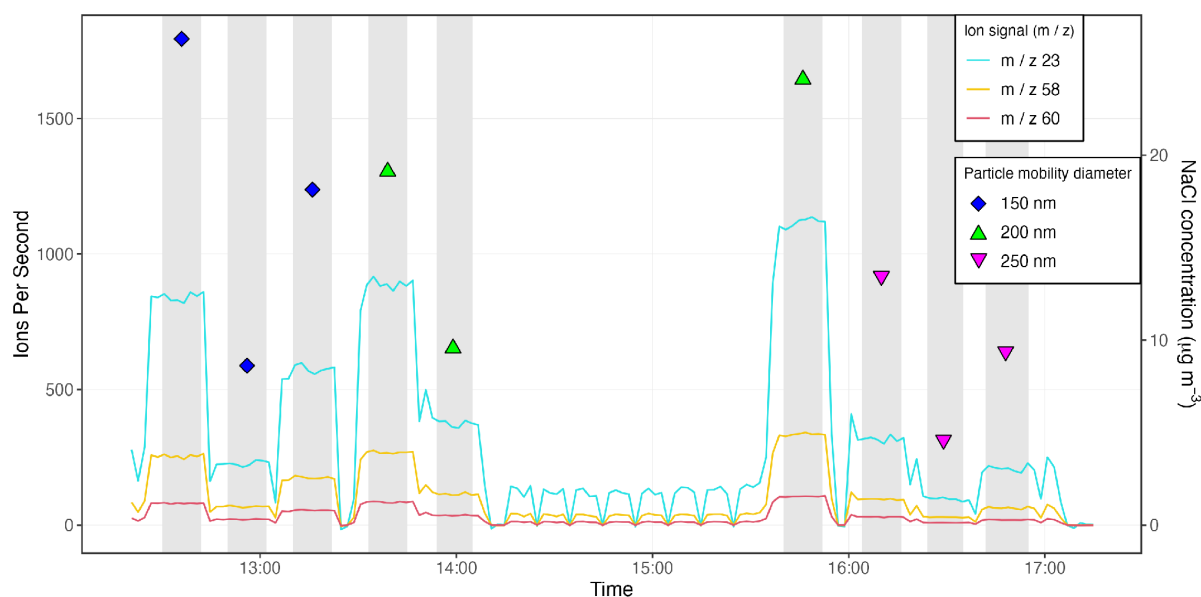


Figure S1: Time series of the fragment signals at m/z 23, 58 and 60 recorded during the laboratory experiment. The grey rectangles indicate the time period during which data was analyzed. The diamonds indicate the NaCl aerosol mass concentration and particle mobility diameter for each measurement.

Figure S2. I don't see a purple shaded area. Do you know why the organic is rising before you inject α -pinene?

Response:

We thank the reviewer for noting this. The figure caption has been corrected to accurately describe the shading.

The increase in organic signal after NaCl injection and prior to the α -pinene injection is due to the presence of some organic contaminants in the chamber background, which can undergo oxidation in the presence of ozone and cause the observed increase in organic aerosol.

SI

Figure S2: Time series of the fragment signals at m/z 23, 58 and 60 (light blue, yellow, and red, respectively), and the organics signal as assigned by default in the fragmentation table of the TOF-ACSM, during the AIDA chamber experiment on 19 January 2024. The pink shaded area represents the injection of NaCl particles from a 0.1 wt% aqueous solution. The polydisperse aerosol had an average number mode diameter of ± 223 nm as derived from the measurements with the two SMPS instruments. The purple lines represent the injection of 783 ppb ozone, the injection of 10 ppb α -Pinene, the injection of 34 ppb ammonia, a second injection of 34 ppb ammonia, and the injection of 77 ppb NO₂, respectively.

Typographical/grammatical errors:

Response:

All proposed changes have been made to the text.

Response to Reviewer #2

Van den Born et al. present the first systematic calibration of a CV-ToF-ACSM for the detection and quantification of sea salt aerosol, using the typical m/z 23, 58 and 60 sea salt marker ions and demonstrating that the instrument calibration response scales with particle surface area. The study provides a useful addition to the existing literature, most substantive points have already been addressed by reviewer 1, leaving limited additional issues to raise.

Response:

We appreciate the reviewer's favorable evaluation of our manuscript and their recognition of its relevance to the literature. We are also grateful for the constructive feedback provided. Revisions made in response to the comments are highlighted in red in the

Comments

In Ovadnevaite et al 2012, I understand that a scaling factor of 51 was retained for $^{23}\text{Na}^{35}\text{Cl}$ at m/z 58 as the simplest way to estimate sea salt mass concentration in $\mu\text{g m}^{-3}$ which was confirmed through offline filter comparison. The authors here have eight months of near-continuous data at the Lutjewad coastal station, if offline filter measurements or ion chromatography data are available for this period, even at coarser time resolution, a similar validation would considerably strengthen the paper and is encouraged. If no such collocated filter data exist, the authors could attempt to reproduce Figure 5 from Ovadnevaite et al. (2012). Examining whether the typical power law relationship shows up between CV-ToF-ACSM sea salt m/z tracers and wind speed during stormy marine episodes would be a nice addition in the supplementary.

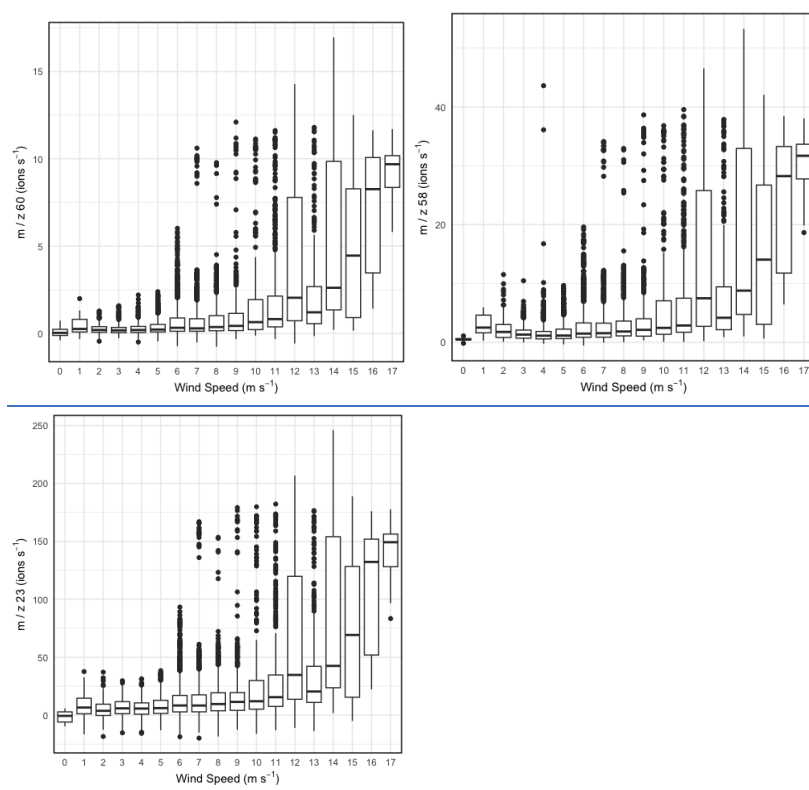
Response:

We agree that offline filter measurements or ion chromatography data would have been a valuable asset to this study. Unfortunately, no offline filter measurements or ion chromatography data are available for the Lutjewad measurement period, and therefore a direct validation of the ACSM-derived sea salt signal against independent sea salt mass measurements is not possible.

We would also like to thank the reviewer for the valuable suggestion to investigate the relationship between the NaCl marker ion signals and wind speed. Following a similar approach to Ovadnevaite et al. (2012), we can show that the sea salt marker signals generally increase with wind speed and show a tendency toward the expected power-law behavior as found in Ovadnevaite et al. (2012). However, this behavior is less pronounced in our dataset because the measurement period did not include high wind speed and storm conditions as reported in Ovadnevaite et al. (2012), where wind speeds reached up to 25 m s^{-1} .

In addition, Lutjewad is located in the Wadden Sea region, which is characterized by shallow and sheltered coastal waters with fewer breaking waves compared to open-ocean environments. Consequently, sea spray production through bubble bursting is expected to be lower than the exposed marine sites.

We therefore decided not to include these figures in the manuscript, as they do not provide substantial additional quantitative insight beyond the existing analysis. Nevertheless, we appreciate the reviewers suggestion and agree that such an analysis would be highly valuable for this work.



Given that the CV here operates at a different temperature ($\sim 550^\circ\text{C}$) and has a longer particle residence time than the standard vaporizer ($\sim 650^\circ\text{C}$ used in Ovadnevaite et al. 2012), it is conceivable that the CV produces a different fragmentation pattern, potentially generating CV-specific sea salt-related ions? The sea salt family in the HR-ToF-AMS included a rich set of ions: ^{23}Na , Mg , ^{25}Mg , ^{26}Mg , ^{35}Cl , H^{35}Cl , ^{37}Cl , H^{37}Cl , K , ^{41}K , Mn , Fe , Na^{35}Cl , $^{24}\text{Mg}^{35}\text{Cl}$, Na^{37}Cl , $^{26}\text{Mg}^{35}\text{Cl}$, $^{25}\text{Mg}^{37}\text{Cl}$, $^{26}\text{Mg}^{37}\text{Cl}$, $^{40}\text{Ca}^{35}\text{Cl}$, $^{40}\text{Ca}^{37}\text{Cl}$, Br , $\text{Na}_2^{35}\text{Cl}$, $\text{Na}_2^{37}\text{Cl}$, and several MgCl_2 isotopologues. Could the authors include one or more representative mass spectra in the supplementary material, from the laboratory calibration or ambient sampling and briefly comment on this?

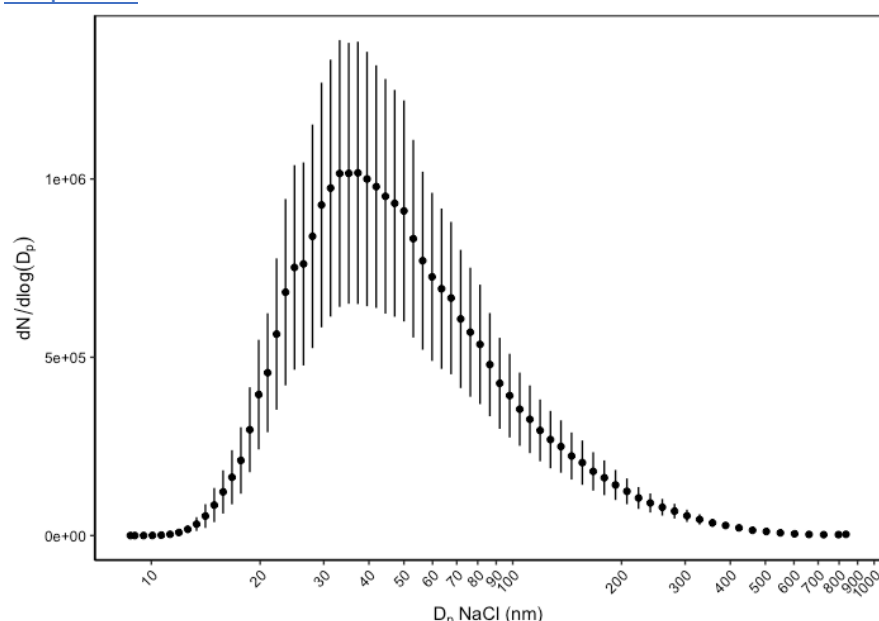
Response:

A representative mass spectrum from an ambient high sea salt episode has been added to the supplementary material. The sea salt family we found in the ACSM has a substantially different composition than the aforementioned one of the HR-ToF-AMS, likely because of the more pronounced fragmentation. However, we do see peaks at m/z 35-38, possibly corresponding to ^{35}Cl , H^{35}Cl , ^{37}Cl , H^{37}Cl , and a very small peak at m/z 59 that could be $^{24}\text{Mg}^{35}\text{Cl}$, although with the unit mass resolution of the ACSM a definite assignment of these peaks is difficult.

Finally, the authors selected three mobility diameters of 150, 200 and 250 nm for their calibration experiment. While the use of discrete DMA-selected sizes is standard practice, the authors could briefly justify why this particular size range was chosen, and whether the derived calibration relationship is expected to hold outside the 150-250 nm range. Sea salt particles are highly hygroscopic and carry multiple charges when nebulized, inversion could potentially misassign counts from larger multiply-charged particles into the size range used

for surface area calculation, biasing the normalization. Could the authors confirm that multiple charging corrections artefacts were not an issue for the sea salt calibration?

Response:



We thank the reviewer for this important comment. The selected mobility diameters (150, 200, and 250 nm) were chosen to be representative of submicron NaCl particles while remaining within the effective transmission range of the PM_{2.5} aerodynamic lens of the ACSM (> ~110 nm). At the same time, these sizes were selected to minimize, as far as possible, contributions from multiply charged particles. Based on the limited size dependence of the surface concentration normalized signal, we expect that this would extend to larger particle sizes.

Originally, we had not accounted for the presence of multiply charged particles and we are grateful for the reviewer to raise this issue. The contribution of doubly and triply charged particles to the mass and surface concentration was not entirely negligible in our case. After accounting for this, the absolute number in Fig. 1 and equation 1 have slightly changed, without affecting the general trend.

Results (line 191)

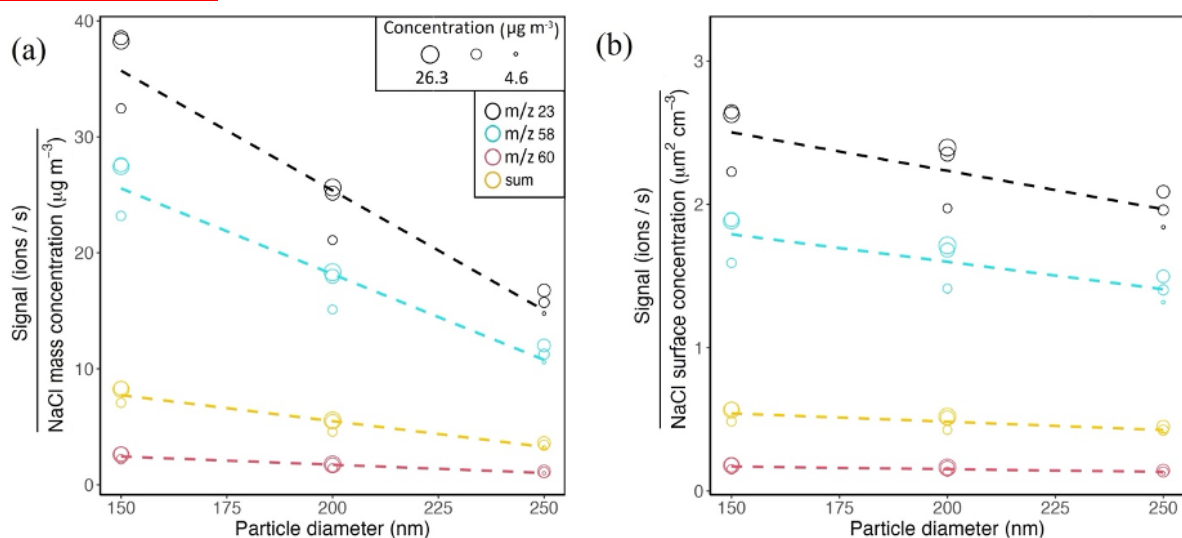


Figure 1: Fragment signals normalized to NaCl mass (a) and surface concentration (b) at m/z 23, m/z 58, and m/z 60 as a function of different aerosol concentrations and particle sizes. The size of the markers is proportional to the NaCl aerosol concentration ranging from 4.6 $\mu\text{g m}^{-3}$ to 26.3 $\mu\text{g m}^{-3}$. The black, light blue, red and orange markers respectively indicate the sum of the normalized signals at m/z 23, 58, 60 and the sum of these three signals (ions/s).

(Line 325)

The corresponding sensitivity is $0.625 \mu\text{m}^2 \text{cm}^{-3}$ per ions/s, with an R^2 of 0.995.