

## **Response to Referees' Comments:**

### **Anonymous Referee #1**

Received and published: 24 April 2018

(My ratings for both scientific and presentation quality are between "Good" and "Fair", so I give one "Good" and one "Fair".)

#### General Comments:

The authors performed 4 month-long field measurements across 53\_N-53\_S over the Atlantic Ocean from 2011-2012 and reported convincing source apportionment based on adequate and well-processed datasets obtained from HR-ToF-AMS and other techniques. Due to few number of similar studies that covered long time series and/or oceanic regions, the findings in this paper certainly provide valuable insights into the sources and origins of marine submicron atmospheric aerosols. Overall, the paper discussed relevant scientific questions within the scope of ACP journal with novel methods and datasets, and the results are generally (but not all) supportive to the interpretations and conclusions, in spite of some technical concerns and vague presentation or expression that need to be further supported, examined, or re-phrased. Therefore, I would recommend this paper be accepted for publication once the following specific comments are completely addressed.

#### Reply:

Thank you very much for your encouraging comments. We tried our best to improve the weak parts on both technical and language aspects. The detailed comments are responded point-to-point in the following text.

#### Suggestions for major improvements and revision:

1. The author(s) should have made best use of their valuable datasets as well as the

previously published studies, and emphasized the significance of their findings, if they also agree that they haven't done this enough in the abstract and the in the introduction. Besides, the author(s) should also, on one hand, carefully refer to previous studies that used similar techniques for marine aerosol, and on the other hand, include necessary comparisons (if available) in their own discussions. For example: The authors should add proper references to the sentences ended in: Page 10 Line 5; Page 11, Line 25; Page 12, Line 27; Page 12, Line 34; Page 13, Line 6; Page 16, Line 24 (Might be useful: Charlson et al., 1987, Nature; Bonsang et al. 1992, GRL; Yassaa et al., 2008, Env. Chem.; Shaw, Gantt, and Meskhidze, 2010, Advances in Meteorology).

Reply:

Thank you very much for your kind comments. According to referees' comments and co-authors' suggestion, we changed the title of this paper to "Organic aerosols over the Atlantic Ocean from 53°N to 53°S: a similar contribution of ocean and long-range transport". The abstract and introduction have been re-written to better stress 1) the key findings on Atlantic organic aerosol sources; 2) the motivation of this study; and 3) previous studies on the same topic or field.

We added the references to the sentences mentioned in examples, as well as in the not mentioned but necessary places. However, the number of references in the original version reached 144, which is too many as pointed out by the other referee. So we removed unnecessary references during re-organizing the whole manuscript. The detailed revisions to each mentioned sentence are showed below:

Page 10 Line 5: (Now in Page 8 Line 26) "Similar seasonal variation of the marine biogenic tracer MSA was also observed (Huang et al., 2017), suggesting the biogenic sources (i.e. phytoplankton) contributed significantly to sulfate (Charlson et al., 1987; Hoffmann et al., 2016)"

Page 11 Line 25: (Now in Page 10 Line 5) "This factor is well correlated with the marine tracer MSA ( $R^2 = 0.83$ , Figure 3), consequently linked to oxidation of DMS emitted by phytoplankton (Charlson et al., 1987; Gondwe et al., 2003)"

Page 12, Line 27: The original sentence “The diurnal cycle of MOA might have been weakened by averaging because the biological activities in autumn are usually lower than in spring.” is subjective and not well supported by the chlorophyll a satellite map offered by the referee. We deleted the sentence and replaced it by the new one in Page 10 Line 32: “In order to focus on the atmospheric behavior of MOOA and exclude the influence from other chemical composition, a “MOOA dominating period” is selected for a case study (about 57 h from 19:40, 18.11.2012 to 04:20, 21.11.2012).”

Page 12, Line 34: (Now in Page11 Line 7) “Similar diurnal cycles are observed for MSA and sulfate, suggesting that MOOA, MSA and sulfate are formed via the same secondary pathway (Charlson et al., 1987; Gondwe et al., 2003; von Glasow and Crutzen, 2004).”

Page 13, Line 6: This sentence was deleted when re-organizing the whole paragraph.

Page 16, Line 24: The original sentences were: “In addition, the particles measured in the range from  $\sim 15^{\circ}\text{N}$  to  $15^{\circ}\text{S}$  (close to the west and middle Africa) showed external mixing state by the HTDMA measurements (details in an accompany paper by Wu et al., in preparation). It is the typical property of BB emissions.” Considering the mentioned paper will provide more detailed explanation and it is also arbitrary to define the “typical property of BB emissions”, we deleted these two sentences in the end of paragraph.

2. Conclusions discussing causality or reasoning must be carefully examined. Just give a few examples: Page 9, Line 5: The authors attributed “insufficient offline samples” to the weak correlation between AMS and offline sea salt. Actually this might not be a reasonable explanation especially if they used AMS data collected from the exactly same periods of time during the offline filter sampling. The data size itself should not affect the  $R^2$ , and the authors should also examine p-value of correlation for “meaningfulness”. Furthermore, in this case, the authors should also clarify how they measured sea salts using the individual techniques and why they applied the method

from Ovadnevaite et al. (2012). For example, what ions were included as sea salts? Did they count Na<sup>+</sup>, Cl<sup>-</sup>, SO<sub>4</sub><sup>2-</sup>, K<sup>+</sup>, Mg<sup>2+</sup>, etc. in both? If NaCl accounted for different fraction from that in Ovadnevaite et al. (2012), was the scaling factor of 51 still suitable? Otherwise, the “therefore” in Line 6, did not explain why the same scaling factor was applied, considering the correlation and the coverage of time (“full year measurements in the reference”) discussed above was not supportive, or not relevant.

Reply:

Thank you for your comments and suggestions. We reorganized the paragraph about sea salt estimation and add more technical details including:

1) We agree that the “insufficient offline samples” was not a reasonable explanation for the weak correlation. According to referee’s suggestion, we performed the significance test (Spearman’s correlation test because of non-normal distribution of the data) and the resulting p-value is 0.009, indicating the sea salt concentrations from two techniques (AMS and offline) are significantly correlated, that is, the correlation is meaningful. Now relative sentence is in Page 7 Line 32: “the p-value of the regression of AMS-derived sea salt with offline results is 0.009 (Spearman's correlation test), indicating that sea salt concentrations from AMS and offline methods are correlated significantly.”

2) For the sea salt estimation based on PM<sub>1</sub> filter measurements, we use Na<sup>+</sup> and Cl<sup>-</sup> ions to derive the sea salt concentration as applied in previous studies (Bates et al., 2001; Quinn et al., 2001): sea salt [ $\mu\text{g m}^{-3}$ ] = Cl<sup>-</sup> [ $\mu\text{g m}^{-3}$ ] + Na<sup>+</sup> [ $\mu\text{g m}^{-3}$ ] x 1.47 ; where the factor of 1.47 is the seawater ratio of (Na<sup>+</sup> +K<sup>+</sup> +Mg<sup>2+</sup> + Ca<sup>2+</sup> +SO<sub>4</sub><sup>2-</sup> + HCO<sub>3</sub><sup>-</sup>)/Na<sup>+</sup>. So this estimation method can prevent the inclusion of non-sea-salt K<sup>+</sup>, Mg<sup>2+</sup>, Ca<sup>2+</sup>, SO<sub>4</sub><sup>2-</sup> and HCO<sub>3</sub><sup>-</sup> in the sea salt mass, and also allow for the loss of chloride mass through chloride depletion processes (Bates et a., 2001). The estimation method was added to the caption of Figure S6.

3) For the sea salt estimation using HR-ToF-AMS, we use the method from Ovadnevaite et al. (2012). It is the first reported studies of sea salt estimation using AMS containing both laboratory calibrations using artificial sea salt and ambient

measurements, and the later studies (e.g. Schmale et al., 2013) followed the method (applied the similar scaling factor). The sea salts certainly include many ions once fragmented by AMS, e.g. Na<sup>+</sup>, Cl<sup>-</sup>, SO<sub>4</sub><sup>2-</sup>, K<sup>+</sup>, Mg<sup>2+</sup>, NaCl<sup>+</sup>, Na<sub>2</sub>Cl<sup>+</sup> and so on (see details in Table 1 from Schmale et. al, 2013, shown below). However, the particulate sea salt mass concentration cannot be the simple sum of the ions on the list, because most of them could be contributed by non-sea-salt sources (e.g. Cl<sup>-</sup>, SO<sub>4</sub><sup>2-</sup> and K<sup>+</sup> can be from continental transport) and some of the ions have too low intensity to be detected in ambient situation (e.g. metals and isotopes). Also, one of the main ions, Na<sup>+</sup>, can vary significantly with the AMS vaporizer temperature (Ovadnevaite et al., 2012; Schmale et al., 2013). Therefore, Ovadnevaite et al. (2012) only used the NaCl<sup>+</sup> (m/z 57.95) as a surrogate of sea salt rather than the sea salt family ions, and we followed it for the same reason. In our study, the correlation slope between offline sea salt and AMS NaCl<sup>+</sup> ion was 62 (± 6), not very far from the reference value, but with a mild correlation coefficient ( $R^2 = 0.38$ ). We still prefer to use the scaling factor of 51 from the literature for several reasons: first, this factor could lead to better coherence (slope = 1.01) of estimated sea salt concentration between filter and AMS; second, most scattering dots (with factor of 51) are distributed within the uncertainty range derived from literature (Figure S6); and third, the factor of 51 is not too far from the slope derived in this study (62 ± 6) but can make our study consistent with the previous studies. Of course, the estimated sea salt mass concentration should be used and discussed with caution and we need to always be aware of its uncertainty.

According to the comments, we re-organized the paragraphs and removed the improper conjunction phrase “therefore”. The revised sentence is now in Page 7 Line 28: “To be consistent with the literatures (Ovadnevaite et al., 2012; Ovadnevaite et al., 2014; Schmale et al., 2013), the scaling factor of 51 from the reference is applied to the sea salt surrogate (NaCl<sup>+</sup>) to estimate the sea salt mass concentration in this study.”

Table 1. Ion fragments considered within the sea salt family (Schmale et. al, 2013)

Ion fragment	Exact mass	Ion fragment	Exact mass
Na	22.99	Na <sub>2</sub> <sup>35</sup> Cl	80.95
Mg	23.98	Na <sub>2</sub> <sup>37</sup> Cl	82.94
<sup>25</sup> Mg	24.98	<sup>54</sup> Fe <sup>35</sup> Cl	88.91
<sup>26</sup> Mg	25.98	Zr	89.90
NaMg	46.97	Fe <sup>35</sup> Cl	90.90
Mn	54.94	Fe <sup>37</sup> Cl	92.90
OK	54.96	<sup>94</sup> Zr	93.91
Fe	55.93	Mg <sup>35</sup> Cl <sub>2</sub>	93.92
Ni	57.94	Mg <sup>37</sup> Cl <sup>35</sup> Cl	95.92
Na <sup>35</sup> Cl	57.96	<sup>25</sup> Mg <sup>37</sup> Cl <sup>35</sup> Cl	96.92
Mg <sup>35</sup> Cl	58.95	Mg <sup>37</sup> Cl <sub>2</sub>	97.92
Na <sup>37</sup> Cl	59.96	<sup>25</sup> Mg <sup>37</sup> Cl <sub>2</sub>	98.92
Mg <sup>37</sup> Cl	60.95	Fe <sup>35</sup> Cl <sub>2</sub>	125.87
Cl <sub>2</sub>	69.94	Fe <sup>37</sup> Cl <sup>35</sup> Cl	127.87
<sup>37</sup> Cl <sup>35</sup> Cl	71.93		

Page 10, Line 18: The author stated “The ammonium concentrations didn’t follow a clear seasonal trend, although its precursor ammonia could be emitted from ocean (Ikeda, 2014; Johnson et al., 2008). The absence in seasonality suggests particulate ammonium during Polarstern cruises was contributed by both anthropogenic and biogenic sources.” The deduction did not support their conclusion.

Reply:

Yes the causality of the sentences was not clear. We replaced them with the new ones: “The ammonium concentrations did not exhibit a seasonal difference between spring and autumn. The highest median value was found over the tropic Atlantic followed by the North Atlantic, while the lowest ones were in the South Hemisphere. Both continental pollutants via long-range transport and marine organism could be the origin of the ammonium or its precursor ammonia in MBL (Adams et al., 1999).” (Now in Page 9 Line 11).

Page 13, Line 29: “The diurnal variation of NOA shows clear peak in the afternoon, reaching the maximum while the global radiation starts decreasing (Figure 4), indicating that the NOA factor is certainly composed of secondary organic products.”

The evidence is weak, and the authors should specify “global radiation” and cite papers that observed the similar diurnal trends of such a NOA factor, if available.

Reply:

We made major revision of the NOA part (now its name is changed to MNOA). Although the diurnal variation of NOA is very likely to be attributed to the secondary formation, it is hardly to find a NOA factors with similar diurnal pattern in previous studies. Sun et al., (2011) reported a NOA factor related to secondary transformation of amines from marine and industry sources but the factor showed a noon peak, different from our case. Due to the limited amount of AMS measurements in MBL, the comparison of specific marine factor become difficult so we can only make the speculation. New sentences started in Page 12 Line 3 “...and the diurnal variation of MNOA shows a broad afternoon peak with maximum at 1600UTC (Figure 4), similar to that of the amines-related secondary factor in the New York City despite maximum at noon time (Sun et al., 2011).” Besides, “global radiation” was defined in Page 9 Line 8 “... with global radiation (the sum of the direct solar radiation and diffuse radiation).”. We have to use the global radiation because the solar radiation was not derived during the cruises.

Page 13, Line 20: The authors suggested “This can be useful for better estimation of marine DMS related SOA both in field measurements and in models”. However, MSA as a fraction of SOA can vary largely and different from time to time (especially between summer and winter). In addition, MOA in this case might not be equivalent to SOA.

Reply:

We agree that MSA as a fraction of SOA may vary between different seasons, especially between summer and winter in which our measurements did not cover. Considering that AMS-PMF is an often-used method for distinguishing SOA and few other methods can provide more robust estimation of SOA, we think our result can at

least provide a hint on, not the whole marine SOA, but the SOA formed from DMS-oxidation in the measuring seasons, especially in spring. Of course, MSA cannot trace the portion of SOA which is formed from other pathways, e.g. secondary formation from gaseous amines. According to referee's comments and the explanation above, we revised the sentences in manuscript to be more cautious, emphasizing the season (spring) and SOA portion (DMS-related SOA) in/for which the MSA and scaling factor 1.79 can be applicable. The new sentence is now in Page 11 Lines 26: "We therefore infer that the relation between MSA and its concomitant (DMS-related) SOA is roughly stable over the Atlantic, and suggest to estimate MOOA mass concentration as the product of the MSA concentration multiplying the factor of 1.79." Further analysis of data from other oceans/seasons are needed in future to examine this correlation coefficient.

3. For better presentation quality and reading experience, the English language and scientific writing in this paper can be more precise and largely improved. Just give a few examples: Page 11, Line 9: "These S/C ratios derived from the PMF analysis tool contain however certain estimation uncertainties and have therefore to be used with caution." This seems to be a grammatically wrong sentence.

Reply:

Thank you very much for the comments. The authors of this paper have tried best to improve the language. We apologize that we did not make it be edited by a professional person/company due to very tight schedule of the author(s). We also noticed that a basic language correction and smoothing procedure would be provided by the journal as the last step of publication. Hope that would be helpful.

The sentence in Page 11, Line 9 is removed when re-organizing the text. This sentence is now changed to: "Note that the S/C ratios derived from the PMF analysis tool have to be used with caution because of calculation uncertainties (Aiken et al., 2007), but they can still provide indication on significance of sulfur when calculated with the same



tool among the factors from the same dataset.” (Page 10 Line 7)

Page 12, Line 24: “The minimum of the diurnal variation ( $0.04 \mu\text{g m}^{-3}$ ) appears around 09:00, probably linking to the increase of mixing layer in morning.” This sentence needs to be re-phrased and also supported with references.

Reply:

Based on the re-analysis of the MOOA diurnal variation, we think it is insufficient to attribute the minimum to the dilution effect of the rising boundary layer. Because the drop of the MOOA concentration was not found at the similar time point during the MOOA-dominating period. So, this sentence is removed.

Page 10, Line 27: I think it is more precise to say “57 hours” rather than “about 2 consecutive days”, unless there was an interruption.

Reply:

Done.

Page 13, Line 6: “the this OA component”. Despite the grammatical error and lack of references, “OA component” was vague in the context.

Reply:

Thanks for reminding this. We reorganized this paragraph and deleted the mentioned sentence.

Page 17, Line 17: In this paragraph, the author said “still questionable” and then “This suggests... could be not correlated”. This led to confusion due to the inappropriate English or logical expression.

Reply:

We apologize for the unclear causality here. We improved the sentences as: “This does not conflict with the speculation that the MHOA is related with marine primary

emissions, because the mass fraction of organics in the sea spray aerosol was found to be size-dependent: increasing with the decreasing particle size (Gantt et al., 2011; Quinn et al., 2015). The enrichment factor of organic compounds, i.e. the ratio between organic carbon in sea spray aerosol and that in sea water, is also largely influenced by the particle size (Quinn et al., 2015). In addition, the transfer of organic matters from seawater to the particles is chemoselective, more complicated than the inorganic sea salt (Schmitt-Kopplin et al., 2012)".

Other technical and specific comments to be addressed:

1. Generally when discussing seasonality, the difference between "spring & autumn" might not be as distinct as that between "summer & winter", in term of many factors such as meteorological parameters and marine bioactivity. Besides the "spring vs. autumn" comparison, the authors may also want to look into "spring/autumn vs. tropic". In addition, their measurements on board was changing with time and location at the same time, so this will be different from those studies took place at a ground site over seasons. I wonder if the authors would like to make some comments on these.

Reply:

Thanks for the comments. Yes, the meteorological parameters such as temperatures and RH were not very different between spring and autumn as between summer and winter. We added the description on tropical case and compared the species mass concentrations in spring, autumn and tropic (Session 3.1.2). Although no big difference was found between spring and autumn for organics, sea salt, nitrate and so on, sulfate showed very discrepant average or median mass concentration in spring and autumn, maybe related to different biological activities. This may suggest even with the similar temperatures and RH, the seasonal events such as biological activities may still influence the aerosol chemical composition.

Considering the comparability between the mobile platform and stationary site, we admit there could be big difference caused by marine biomass distribution, e.g. more

dense phytoplankton group near the coastal region than the remote ocean. But because of very limited amount of the mobile measurements over the ocean, it is quite difficult to find records for the similar regions. So we collected the aerosol chemical composition in several regions covering the ship tracks, and checked if they were comparable to our results. In future it would be helpful to have more information of submicron aerosols over the ocean based on satellite data.

2. The authors should try to clarify the influences from the “open oceans”, “marine”, and “coastal” when interpreting results in the discussions, even though the boundaries might be blurry. For example, on Page 11, Line 30, the author stated “The S/C ratio of the MOA factor is also over twice that of marine factor observed in Paris (0.013, Crippa et al., 2013b), implying a stronger influence from marine phytoplankton on aerosol particles over the ocean than those in the coast city.”, but actually the abundance of phytoplankton can be much higher in the coastal areas. See [https://earthobservatory.nasa.gov/GlobalMaps/view.php?d1=MY1DMM\\_CHLORA](https://earthobservatory.nasa.gov/GlobalMaps/view.php?d1=MY1DMM_CHLORA)

Reply:

Thanks for the comments and website link. Due to the limited on-board measurements, we did not find the S/C ratios from offshore sites or open oceans in previous studies. So the S/C ratio from coastal measurements in Paris was used as a reference for comparison. We noticed that the comparison is not sufficient to support the conclusion of “stronger influence ... over the ocean than those in the coast city”, and the S/C ratio should be used with caution because of calculation uncertainties. So this conclusion is removed and we only compare the S/C ratios among the OA factors in this study. Now in Page 10, Line 6: “This leads to a high S/C ratio (0.030), which is 10 to 30 times higher than that of other factors (Figure 3).” Nevertheless, it is important to stress that organosulfates with biogenic sources may not correlate with chlorophyll a level, as the former is produced from the secondary pathway and the latter is the indicator of the primary biogenic mass (Huang et al., 2017).

3. The authors are suggested to add discussions for organosulfates, since they can make a considerable contribution to continental SOA masses at certain locations, and also derived from the same biogenic precursors over the oceans. For example, how is this class of compounds measured using AMS? Was it included in organics or sulfate, or neither?

Reply:

Thanks a lot for the suggestion. Organosulfates (except MSA) are also one of the important components of the marine SOA transformed from the precursors such as isoprene, monoterpenes and so on (Claeys et al., 2010; Surratt et al., 2007). We added small discussion on the organosulfates in the revised manuscript to stress the existence of organosulfates (Page 10 Line 28): “it is well-known that isoprene and monoterpenes oxidation lead also to the formation of organosulfate compounds (Claeys et al., 2010; Fu et al., 2011; Iinuma et al., 2007; Surratt et al., 2008; Surratt et al., 2007), which can contribute to the  $C_xS_y^+$  fragments observed in the MOOA factor.” We hesitated to discuss more about the organosulfates in this paper because a parallel paper focusing on the organosulfates is in preparation, which included detailed analysis on a sub-dataset of Polarstern measurements. The paragraph below may answer referee’s questions:

Our MOA (now changed to MOOA) mainly includes MSA fragments (as shown in Figure 1), while the contribution of organosulfates to MOOA may be tiny as found in previous marine study (Claeys et al., 2010). Using AMS, the fragments of organosulfate (e.g. MSA) are recognized as sulfate and organics (Figure 1). The quantification of organosulfates requires the laboratory calibrations using standard chemicals of known organosulfates (Huang et al., 2015; Huang et al., 2017).

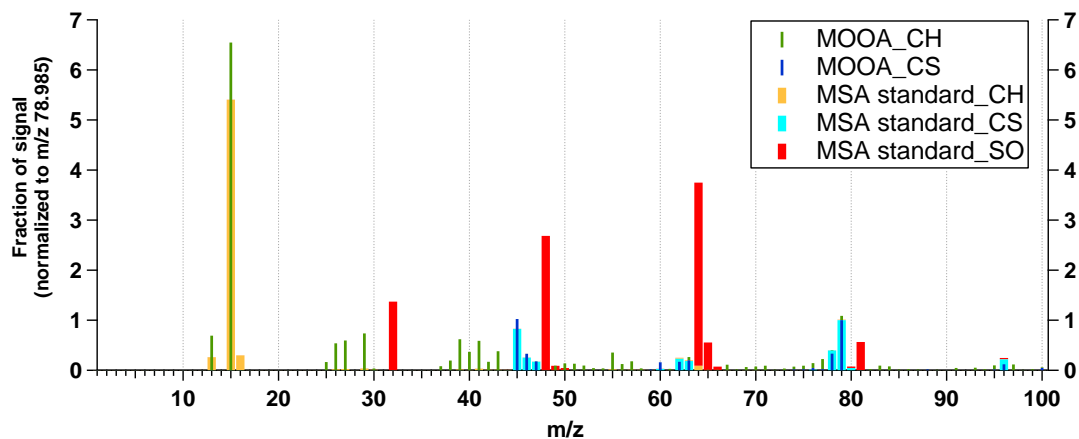


Figure 1 Mass spectra of MOOA factor (CH and CS ions) and pure MSA (CH, CS and SO ions)

4. Last but not least, the authors should revise the manuscript carefully by their own. Just give a few examples: 1) Page 12, Line 34: “Figure 4” – should this be Figure 5?

Reply:

Thanks for the detailed comments! In original sentence “Figure 4” was mentioned for “the average case”. In order to be more precise, the sentence is changed to “The diurnal pattern for this specific period (Figure 6b), with minimum of  $0.11 \mu\text{g m}^{-3}$  at 07:00 and maximum of  $0.25 \mu\text{g m}^{-3}$  at 16:00, is more noticeable than the average case (Figure 4b).” (Page 11 Line 6).

2) Acronym: define before use. For example, “SOA” was not defined but used in the abstract; “OA” was firstly defined on Page 10, Line 30 in the main text; “biomass burning” was defined but not used in many places.

Reply:

We checked the manuscript and corrected the use of acronyms. The definition of acronyms was added to the abstract: Page 1 Line 17 for OA, Page 2 Line 2 for SOA. Biomass burning (BB) was defined in Page 12 Line 22 and the abbreviation is mainly

used in the section 3.2.5 Combustion oxygenated organic aerosol (Comb-OOA), e.g. Page 15 Line 2, “the average BB organic aerosols”, Line 15 “ some often used BB tracers”.

3) Please be consistent when using terms such as “fPeak” or “fpeak”, “C<sub>x</sub>H<sub>y</sub>O” or “C<sub>x</sub>H<sub>y</sub>O<sup>+</sup>”. 4) Please be consistent about adding a “\_” between numerical values and their units. 5) Please specify “C<sub>x</sub>S<sub>j</sub>+” on Page 12, Line 21.

Reply:

We went through the text and uniform the terms: e.g. fPeak, C<sub>x</sub>H<sub>y</sub>O<sup>+</sup> and C<sub>x</sub>S<sub>y</sub><sup>+</sup>. We also uniform the format of values and units: put a blank between them.

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