ACP-2019-976-RC1

The manuscript of Triesch et al. focuses on the measurements of free amino acids (FAA) in different marine compartments (underlayer water samples, sea surface microlayer, size segregated aerosol samples at different heights, cloud water) at the Cape Verde Atmospheric observatory (CVAO) and at the Mt. Verde during September/October 2017. Further, through the case study authors discusses the possible transfer of specific FAA from the ocean to the atmosphere up to the cloud level. This work for the first time aims to provide a detail chemical analyses of FAA, of both the sea surface layers as well as of atmospheric samples (size segregated aerosols and cloud water), thus representing a promising approach to contribute to the fundamental state-of-the-art knowledge on the linkage between the ocean and atmosphere. In addition, this work reports on unique marine and atmospheric organic matter data from remote tropical areas, being rare in comparison to continental/coastal ones. Thus, my opinion is that this manuscript presents valuable data sets and after some revision, it will be definitely worth to publish.

We thank the reviewer for the careful examination of the manuscript and the supporting information. In the following, please find a point-by-point response to the questions and concerns. All references to the manuscript (e.g. page and line numbers) listed in our replies refer to the clean version of the manuscript (without track changes).

Major comments

R#1-1) The authors should comment and discuss that some specific FAA were not detected in particular compartments possibly due to their high LOQ (in particular matrices) and the fact that maybe not enough material was available for their analysis. This is particularly relevant for the selected hydrophilic FAA in comparison to other FAA groups and should be considered when discussing the selective transfer of FAA groups from the ocean to the atmosphere.

We agree with the reviewer's comment that a more detailed discussion of the LOQ of the individual analytes is needed and revised several parts of the manuscript accordingly.

In the SI (Table S1), we listed the LOQ of each FAA in the respective matrices (seawater, cloud water, aerosol particles). In the manuscript, we referred to these LOQs in a number of different places as described in the following:

In section “2.2.1 Seawater analyses” (page 6, line 20-23), we added: “The LOQs were mostly below 10 nmol L^{-1}, however, GABA and Met exhibited LOQs with 24.2 nmol L^{-1} and 16.8 nmol L^{-1}, respectively (due to high blank values). A quantification of some FAAs in seawater, mainly in the ULW with its generally lower FAA concentrations compared to the SML, is therefore partly limited.”

In addition, we mentioned that “The limit of quantification (LOQ) of the individual FAAs in seawater samples is in good agreement with the FAA analysis in seawater samples (e.g. Kuznetsova et al. (2004)) and listed in Table S1” (page 6, line 19-20)

In section “2.2.2 Aerosol particle filter analyses” (page 7, line 6-8), we added: “Although a variance in LOQs between the individual FAAs is apparent, FAAs with relatively high LOQs (39.5 pg m^{-3}) on aerosol particles such as Ala, GABA, Asp in submicron and supermicron aerosol particles could be quantified (as discussed in section 3.2 and 3.4).”

In section “2.2.3 Cloud water analyses” (page 7, line 31-33), we added “Since the LOQs of the FAAs in cloud water are below 0.3 ng m^{-3} and often below 0.06 ng m^{-3}, a limitation of the FAA composition in cloud water due to the LOQs is rather unlikely despite the variance of FAA concentrations (11.2-489.9 ng m^{-3}) in cloud water (section 3.3).”

This possible restriction of the results by the LOQ was also discussed in the results and discussion section in the individual sections on seawater (section 3.1), aerosol particles (section 3.2), cloud water (section 3.3) and concerted measurements (section 3.4).

In section “3.1 Seawater samples” (page 10, line 1-3), we added “As discussed in section 2.2.1, GABA and Met have the highest LOQs of the analytical method used here, which may be one reason why these two analytes could not be quantified in the seawater samples (ULW and SML).”
In section “3.4.1 Hydrophilic amino acids” (page 18, line 19/20), we added “Although some limitations of GABA quantification (Table S1) in seawater exist, a relatively major abundance of it in seawater would not be restricted due to the high FAA concentrations.”
In section “3.4.3 Aromatic amino acids” (page 21 ,line 1-3), we added “Assuming that Phe and Tyr were contributing to ∑FAA in a very small fraction as reported in Barbaro et al. (2011), their concentrations would be below the detection limit and could thus probably not be detected.”

R#1-2) The experiments have been done carefully and the quality of data is good. Authors comprehensively presented the experimental approach and obtained data within the manuscript. Important data are also shown within the SI material. However, some SI Tables are not easily comparable to each other. For example, in the present form Table S9 does not follow the Table S8 format. Comparing Tables S8 and S10, I am wondering why data for all size fractions are presented within Table S10 and only submicron and supermicron classification is done within the Table S8. I suggest to reorganize and to uniform SI Tables to follow the main text clearly and to enable the data comparison more easily.

We thank the reviewer for his positive judgement and his comment. We agree regarding the SI Tables. In order to simplify the comparability of the individual information in the SI, the SI tables (Table S2, S7, S8, S10 and S11) have been standardized in the distinction between submicron and supermicron aerosol particles. This provides a better overview of the measurement data and allow a clearer and easier to understand discussion.

R#1-3) Some sentences/paragraphs require major rephrasing. I found the reading of several statements rather unclear and I proposed some changes accordingly. However, my main concern lies in the sentence style used throughout the manuscript. I believe that the paper should be thoroughly edited.

Following this comment, the manuscript and supporting information has been carefully revised by a professional English-speaking person. The main focus was on the comprehensibility of the sentences/paragraphs. In this context, the specific and technical comments were also implemented.

R#1-4) Once introduced, abbreviations should be used further throughout the text.

We agree with the reviewer's comments regarding the abbreviations. Some of these are taken up again and explained in more detail in the following section "specific and technical comments" for abbreviations such as CVAO, MV and FAA. When revising the manuscript and the Supporting Information, care was taken to ensure that abbreviations (e.g. CVAO, MV, FAA) were used consistently after their introduction. A detailed overview can be found in the change tracking of both scripts attached.

Specific and technical comments

***R#1-5)* Page 2**
L23 Skip …also..; it is confusing considering the previous paragraph

Due to the revision of the introduction and also in line with the suggestions from reviewer 2, we concentrated on the FAA from the beginning and deleted general parts (as the one mentioned by the reviewer) from the manuscript.

***R#1-6)* Page 3**
L7-9 please rewrite;…into consideration *for the accurate prediction of marine organic matter transfer to the atmosphere* (van Pinxteren et al., 2017).

As mentioned above, due to the revision of the introduction and also following the suggestions from reviewer 2, this part was deleted from the manuscript.

***R#1-7)* Page 4**
L1-3 Repetition with the L27-34; I suggest to skip it or rewrite the overall paragraph to avoid repetition.

We agree with the reviewer's comment and have removed the detailed information on the sampling sites of the campaign in the introduction and concentrated on the different sampling approaches for seawater, aerosol particles and cloud water. The text now reads as follows in the Introduction (page 3, line 31 – page 4, line 5): "So, the aim of the present study is to investigate the occurrence of FAA in the marine environment regarding all important compartments; i.e. the ULW, the SML, the aerosol particles and finally cloud water in the remote tropical North Atlantic Ocean at the Cape Verde Atmospheric Observatory (CVAO). Their abundance, origin and possible transfer from the seawater as well as their transport within the atmosphere are studied in particular. Therefore, the FAA are measured on a molecular level and divided into hydrophilic (glutamic acid, aspartic, GABA), neutral (serine, glycine, threonine, proline, tyrosine) and hydrophobic compounds (alanine, valine, phenylalanine, isoleucine, leucine) according to their hydropathy index. Especially the similarities and differences between the amino acid composition in submicron (0.05-1.2 µm) and supermicron (1.2-10 µm) aerosol particles are elucidated. Finally, the potential of individual FAA as proxies or tracers for specific sources of aerosol particles and cloud water in the tropical marine environment is outlined."

R#1-8) L14-15 Unclear/confusing sentence, please rewrite

We agree with the comments of the reviewer and have reworded the sentence and also added a figure of the sampling locations based on the comments of reviewer 2. It now reads (page 4, line 16-19): "During this campaign, concerted measurements were performed including the sampling of size-segregated aerosol particles at the CVAO and seawater sampling at the ocean site (~16°53ˈ17ˈN, ~24°54ˈ25ˈE). The location was carefully chosen with minimal influence of the island and located in wind direction to the CVAO as shown in Fig. S1."

R#1-9) L15 The abbreviations for the sampling sites (CVAO and MV stations) were introduced within the Introduction, thus please use it further throughout the text (e.g. .P4 L30, P5 L6, P12 L30…)

We agree with the reviewer's comment and following the introduction we now use the abbreviations CVAO and MV.

R#1-10) L19: I suggest avoiding abbreviations within the title (*Seawater sampling*). Types of samples could be specify within the following paragraph

Following the advice of the reviewer, we have changed the title of section 2.1.1 to "Seawater sampling" (page 4, line 23). As noted by the reviewer, the two collected seawater sample types (SML and ULW) are described in more detail in this section.

R#1-11) L20: Please correct ..were taken *from* a fishing boat…

The sentence (page 4, line 24) was changed to “The seawater samples were taken from a fishing boat, starting from Bahia das Gatas, São Vicente.”

R#1-12) L22 and L23: Skip -pre-cleaned- as it is stated in L25-26 that all sampling material was pre -cleaned

Following this comment, we have omitted the additional description of the sample bottles by "pre-cleaned". The text is now written (page 4, line 25-28) as “The surface films adhered to the surface of the glass plate and were removed with Teflon wipers directly into a bottle. This glass plate approach is described in detail by Cunliffe (2014). The ULW was sampled in a depth of 1 m into a plastic bottle fitted on a telescopic rod. To avoid influences from the SML, the bottles were opened underwater at the intended sampling depth.”

Page 5

R#1-13) L16 I suggest to use the FAA abbreviation throughout the text

As mentioned in comment R#1-4, we agree to the continuous use of the abbreviation “FAA” and have integrated it in the manuscript and in the Supporting Information.

R#1-14) L22 Add…include *determination of* glycine (Gly)…

Following the comment of the reviewer, we included “determination of”. It is now written (page 6, line 1-4) as “Besides, the FAA analysis includes the determination of glycine (Gly), L-alanine (Ala), L-serine (Ser), L-glutamic acid (Glu), L-threonine (Thr), L-proline (Pro), L-tyrosine (Tyr), L-valine (Val), L-phenylalanine (Phe), L-aspartic acid (Asp), L-isoleucine (Ile), L-leucine (Leu), L-methionine (Met), L-glutamine (Gln) and γ-aminobutyric acid (GABA) (purity ≥ 99 %, Sigma-Aldrich, St. Louis, Missouri, USA).”

R#1-15) L34 Unclear sentence part (..in 0.1 min back to 5 % B and…); please rewrite

We agree with the comment of the reviewer and rewrote this sentence part (page 6, line 12-14). Now it reads: “The flow rate of the eluent was 0.3 mL min 1 and the eluent gradient program was 5 % B for 1 min, 5 % B to 100 % B in 16 min, 100 % B for 2 min constant, in 0.1 min from 100 % B to 5 % B and the 5 % B was then kept constant for 3.9 min.”

R#1-16) L34 Please correct: …This *analytics OR analytical procedure* can be used…

The sentence was corrected to “This analytical procedure can be used for amines, too, as described in van Pinxteren et al. (2019).” (page 6, line 14/15)

Page 6

R#1-17) L2-5 Unclear/confusing sentence, please rewrite

We agree with the comment of the reviewer and rewrote this sentence (page 6, line 15/16), which now reads: “Since no chiral column was used in the UHPLC separation, we cannot differentiate between L- and D- amino acids in our ambient samples.”

R#1-18) L6 Introduce the LOQ abbreviation here instead within L24

Following the comment, we introduced the LOQ abbreviation on page 6, line 19/20. It is now written as: “The limit of quantification (LOQ) of the individual FAAs in seawater samples is in good agreement with the FAA analysis in seawater samples (e.g. Kuznetsova et al. (2004)) and listed in Table S1.”

Moreover, we used only the LOQ abbreviation on page 7, line 4-6: "The LOQs of the individual FAAs in aerosol particle samples are listed in Table S1 and are in good agreement with the sensitivity of other analytical methods for FAAs in aerosol particles (e.g. Matsumoto and Uematsu (2005))."

R#1-19) L13 Please add the method used for the trace metal determination

The method for the trace metal determination was described in more detail on page 7, line 9-11: "The analysis of mineral dust tracers on nucleopore foils sampled with the Berner impactor was performed with the Total Reflection X-Ray Fluorescence S2 PICOFOX (Bruker AXS, Berlin, Germany) equipped with a Mo-X-ray source on polished quartz substrates as can be seen in Fomba et al. (2013)."

R#1-20) L13 Remove a space before -total-

We agree with the comment of the reviewer and removed the space before -total-. On page 6, line 27-29 it is written now: "The aqueous particle extracts were divided into aliquots for the analysis of water-soluble organic carbon (WSOC)/total dissolved nitrogen (TDN), inorganic ions and amino acids."

R#1-21) L23 Rewrite to be clear that data obtained for the aerosol samples were blank corrected

Following the comment, we rewrote this sentence. Now it reads: All values presented here for aerosol particle samples are field blank corrected." (page 7, line 4)

***R#1-22)* Page 7**
Please explain the calculation of EF_{SML} based on LOQ/2 estimation (Figure 1) in the case of the missing ULW data within the 2.2.4 Section.

To better describe this procedure, we added in section "2.2.4 Enrichment factors" an explanation for the calculation of EF_{SML} based on LOQ/2 in the case of missing ULW data. Now it reads (page 8, line 11-14): "The FAA concentration in the ULW was assumed to be based on the concentration (LOQ/2) of individual amino acids for seawater samples from the same campaign day when individual FAA could be quantified in the SML samples, but not in the corresponding ULW ones due to FAA values below the LOQs (listed in Table S1). For the calculation of this estimated EFSML, specially marked in the following, the concentration 25.2 nmol L^{-1} was used for $c(analyte)_{ULW}$ in equation (1)."

R#1-23) L28 I suggest to name the EF_{aer} as the -aerosol enrichment factor- instead of the -enrichment factor aerosol- (see accordingly the Fig 5 caption) 3

Following the comment of the reviewer we changed the name of EF_{aer} to aerosol enrichment factor throughout the manuscript.

***R#1-24)* Page 8**
Authors should discuss if the observed variability of FFA concentrations in seawater samples (especially in the SML) was comparable to the variability of the overall DOC and/or TDN pools. It seems to me that high variability of DOC and/or TDN content actually caused the variability of their FAA constituents.

To put the FAA data in context with the DOC/TDN data, we included the DOC/TDN data in Figure 1 and we evaluated a possible connection between FAA variability in seawater (ULW and SML) and DOC and TDN variability in seawater (ULW and SML). However, no statistical relevant correlation/link between FAA and DOC or TDN in terms of variability was found. The study of the correlation of variability (FAA and DOC/TDN) was included in the revised manuscript (page 10, line 12-14) as follows: “Nevertheless, the variance of the ∑FAA concentrations in the SML or ULW observed here could neither be explained by the variance of DOC or TDN values, nor by wind speed and chl-*a* concentrations (see Fig. 1, Table S2, S5), since no correlation between these parameters and the concentration or enrichment of FAA was found.”

R#1-25) In the context of further FAA transfer discussion, it would be useful to follow the FFA classification into hydrophilic, neutral and hydrophobic groups as done for the atmospheric samples.

We agree with the reviewer's comment and have extended the FAA's illustrations (Fig. 1, 2, 3, 4) and discussion in the different marine compartments not only to the individual amino acids but also to the amino acid groups.
In section “3.1 Seawater samples” (page 10, line 4-5) the text now reads: “Looking at the percentage composition within the ULW (10.1 % hydrophilic, 57.0 % neutral, 32.8 % hydrophobic) and the SML (10.6 % hydrophilic, 61.7 % neutral, 27.7 % hydrophobic), the values are similar to each other.”
In section “3.3 Cloud water samples” the sentence “In terms of the hydropathy classification, the first part of the campaign (27/09/2017-5/10/2017) was dominated by neutral FAAs, whereas a sudden increase of the hydrophilic FAAs was observed in its second part (06/10/2017-08/10/2017).” was added on page 16, line 18 – page 17, line 2.

Page 9
R#1-26) L2 Add …Mediterranean Sea *of* 0.07-0.60 µmol L-1…

We agree with the reviewer's comment and added ‘of’. Now it reads (page 11, line 3-5): “Reinthaler et al. (2008) considered concentrations of dissolved FAA of 0.02-0.13 µmol L^{-1} (ULW) and of 0.43-11.58 µmol L^{-1} (SML) in the subtropical Atlantic Ocean as well as values of 0.07-0.60 µmol L^{-1} (ULW) and of 0.77-3.76 µmol L^{-1} (SML) in the western Mediterranean Sea.”

R#1-27) L4-6 Add …. in the SML or in the ULW *since* no correlation between…

The reviewer's comment was implemented in the new sentence. It now reads (page 10 line 12-14): ”Nevertheless, the variance of the ∑FAA concentrations in the SML or ULW observed here could neither be explained by the variance of DOC or TDN values, nor by wind speed and chl-*a* concentrations (see Fig. 1, Table S2, S5), since no correlation between these parameters and the concentration or enrichment of FAA was found.”

R#1-28) L23-24 The sentence should be supplemented to indicate the particular oceanic regions with the comparable FFA levels as at the CVAO.

We agree with the comment and have specified the region of study accordingly.
In section “2.1 Study area” the following sentences (page 4, line 13-16) were inserted: “In accordance with the classification of Longhurst (2007), the ocean around the Cape Verde Islands belongs to the region “North Atlantic Tropical Gyral Province (NATR)”, which is described as the region with the lowest surface chlorophyll in the North Atlantic Ocean having a greater annual variability than seasonality.”
In section “3.1 seawater samples” the discussion (page 10, line 32- page 11, line 7) reads as follows: “Altogether, it can be concluded that there is some variability within the FAA concentration in the SML and in the ULW, with a clear trend of its strong enrichment in the SML. The fact that the FAA concentrations were in accordance with the ones measured at the same location in 11/2013 (0.64 µmol L^{-1}, Table S4), supports the suggestion that the FAA concentrations reported here can be considered representative of the NATR region as part of the North Atlantic Ocean. These

concentrations are generally similar comparing them to FAA concentrations in other marine regions (Kuznetsova and Lee, 2002;Reinthaler et al., 2008). Reinthaler et al. (2008) considered concentrations of dissolved FAA of 0.02-0.13 µmol L^{-1} (ULW) and of 0.43-11.58 µmol L^{-1} (SML) in the subtropical Atlantic Ocean as well as values of 0.07-0.60 µmol L^{-1} (ULW) and of 0.77-3.76 µmol L^{-1} (SML) in the western Mediterranean Sea. Consequently, the FAA concentrations in the NATR region, with its very low surface chlorophyll and a greater annual variability than seasonality, are in the same order of magnitude compared to other marine regions (i.e. subtropical Atlantic and western Mediterranean Sea (Reinthaler et al., 2008))."

Page 10
R#1-29) L16 Add …of dissolved FAA (*PM_{10}*; sum of B1-5)…

This sentence was omitted in the revision of the manuscript. The FAA's concentration discussion is now more focused on the distinction between submicron and supermicron aerosol particles and reads as follows (page 12, line 3-7): "In the submicron aerosol particles, the concentration of ∑FAA was between 1.3 ng m^{-3} (1/10/2017) and 6.3 ng m^{-3} (7/10/2017). Whilst the concentration ∑FAA varied between 0.2 ng m^{-3} (6/10/2017) and 1.4 ng m^{-3} (22/09/2017) in the supermicron size range, the highest atmospheric concentrations of ∑FAA were found in the submicron aerosol particles (mean of 3.2 ng m^{-3}) compared to the supermicron ones (mean of 0.6 ng m^{-3})."

R#1-30) L17-21 Please indicate the PM size fraction citing the Matsumoto and Uematsu (2005) as well as Barbaro et al, 2015

Following the comment of the reviewer we defined the PM size fraction of both studies, Matsumoto and Uematsu (2005) and Barbaro et al, 2015, in more detail. Now it reads (page 12, line 23-28): "Matsumoto and Uematsu (2005) found averaged total concentrations of dissolved FAA with 4.5 ng m^{-3} on aerosol particles (average of < 2.5 µm and > 2.5 µm) in the western North Pacific Ocean. Moreover, Wedyan and Preston (2008) observed an average concentration of dissolved FAA of 2.5 ng m^{-3} on total suspended particles (TSP) during a transect ship cruise in the Atlantic Ocean. For Antarctic aerosol particles, the observed mean total FAA concentration on size-segregated aerosol particle samples (< 0.49-10 µm) at the Mario Zucchelli Station was 4.6 ng m^{-3} (Barbaro et al., 2015)."

Page 12
R#1-31) Please explain why would the low percentage of hydrophobic FAA and higher percentages of hydrophilic FAA point to the local oceanic origin of FAA in aerosol size fractions. As marked previously, it would be useful to refer to the seawater FAA classification in this study.

This part was thoroughly revised.
In the Introduction (page 3, line 17-20), we stated "This divides them into hydrophilic, neutral and hydrophobic amino acids as discussed in Barbaro et al. (2015) for FAA in Antarctic aerosol particles. They also observed that hydrophilic FAA in the Antarctic were predominant in locally produced marine aerosol particles, while hydrophobic amino acids prevailed in aerosol particles collected at the continental station."
We intensively discussed the single FAA contributing to the hydrophilic fraction and the text now reads (page 14, line 21-27): "Following this hydropathy classification, the submicron aerosol particles consisted on average of 5 % hydrophobic, 15 % hydrophilic and 80 % neutral amino acids, while the supermicron aerosol particles contained on average only 7 % hydrophobic and 93 % neutral amino acids (Table S7). During the campaign, an increase in the contribution of hydrophilic amino acids was observed with a maximum of 55 % on 7/10/2017. Barbaro et al. (2015) reported that hydrophilic components were predominant (60 %) in locally produced marine Antarctic aerosol particles, whereas hydrophobic compounds were rather dominate aerosol particles collected at the continental station (23 % and 27 %). According to the conclusions by Barbaro et al. (2015), the

relatively high content of hydrophilic FAA found here points at least at some influence of local oceanic sources."

R#1-32) L31-32 Unnecessary sentence, repetition

Following the comment of the reviewer, we removed that sentence.

Page 13

R#1-33) L6-10 The PM mass concentrations of size segregated samples at the MV were substantially lower than those obtain at the CVAO. Thus, it could be expected that the levels of Na and MSA constituents will be accordingly lower at MV in comparison to CVAO. For the sake of comparison of aerosol tracer levels at different stations, it is more reasonable to consider the contribution of Na and MSA to the PM mass (of corresponding size fraction) instead of comparing the concentration levels.

We strongly revised this part and following the suggestions of the reviewer, we included and discussed ratios of FAAs to other aerosol particle constituents (PM, Na^+, MSA, WSOC) of the corresponding size fractions regarding CVAO and MV. The section (page 15, line 23-32) now reads: "The particles at the MV exhibited lower particle masses, as well as lower concentrations of the aerosol particle constituents. The decrease in concentrations of $\sum$FAA, PM, sodium, MSA and WSOC was reduced by a factor of three to four regarding the submicron aerosol particles. However, no uniform depletion ratio between their concentration at the CVAO and the MV was found for the supermicron aerosol particles (Table S11). While the PM of the supermicron particles was reduced by a factor of four at the MV (similar to the submicron aerosol particles), sodium and WSOC were depleted more strongly (factor of 11-12) compared to their respective concentrations at the CVAO. This suggests that the submicron particles were rather uniformly affected and depleted, likely by cloud processes, while the supermicron particles were influenced by clouds, and potentially other sources, in a non-uniform way. Nevertheless, the abundance of the marine tracers (sodium, MSA), together with the presence of FAA in the aerosol particles (which mainly had a similar composition compared to the oceanic and ground-based particulate FAA) indicated an oceanic contribution to the aerosol particles at cloud level."

R#1-34) L24-27 Authors should comment on the fact that high LOQ of particular FAA could resulted in their variabilities or selective determination in cloud water samples.

We agree with this comment and have added on page 7, line 31-33 a statement to a possible limitation of the FAA composition in cloud water due to the LOQ.
It reads now: "Since the LOQs of the FAAs in cloud water are below 0.3 ng m^{-3} and often below 0.06 ng m^{-3}, a limitation of the FAA composition in cloud water due to the LOQs is rather unlikely despite the variance of FAA concentrations (11.2-489.9 ng m^{-3}) in cloud water (section 3.3)."

R#1-35) L28-32 to P14 L4-8 This paragraph deals with the possible dominant origin of FAA in the cloud water collected at the MV as within the 3.4.5. Section. I suggest skipping this paragraph 4 and combining the cloud water source discussion within the 3.4.5 section to avoid the repetition.

We agree with the reviewer and included the entire cloud water discussion in section "3.3 Cloud water samples". Section 3.4.5 has been deleted and section "3.4.4 Transfer of amino acids from the ocean into the atmosphere" now contains only some sentences on the enrichment factor in cloud water, which reads now (page 22, line 20-24): "Applying the concept of the enrichment factor to cloud water and calculating the EF_{CW} (Eq. (2)), the $EF_{CW(\sum FAA)} = 4 \cdot 10^3$ (based on SML) and $1 \cdot 10^4$ (based on ULW) could be determined. As mentioned in section 3.3, several atmospheric processing (aging), oceanic transfer and biogenic-driven processes might contribute to this high enrichment and need to be addressed in future studies. The high FAA concentrations and enrichments might have implications on OM processing through clouds and are worth further studying."

R#1-36) L33-34 Please specify the study location of Gioda et al. (2009)

Following the comment of the reviewer we specified the study location of Gioda et al., (2009). Now the text reads (page 16, line 11-13): "The concentrations of cloud water sulfate (average: 2.9 µg m^{-3}, Table S12) and sodium were higher than in cloud water samples, collected at East Peak in Puerto Rico, which can be seen in Gioda et al. (2009)."

Page 15

R#1-37) The authors should comment and discuss that some specific FAA were not detected in particular compartments possibly due to their high LOQ (in all matrices) and/or the fact that not enough material was collected for their analysis. This is particularly relevant for selected hydrophilic FAA in comparison to other FAA classes/groups and should be considered when discussing the selective transfer of FAA groups from the ocean to the atmosphere.

We agree with the reviewer and carefully discussed LOQ restrictions in several parts of the manuscript as follows: In the experimental part section "2.2. Analyses" an estimation was given for which amino acids due to the LOQ a restriction of the sensitivity in the marine compartment would be possible and thus no quantitative statement on the analyte would be possible.

This possible restriction of the results by the LOQs was also discussed in the results and discussion section in the individual sections on seawater (section 3.1), aerosol particles (section 3.2), cloud water (section 3.3) and concerted measurements (section 3.4). The detailed response and citations from the manuscript can be found under comment R#1-1.

Page 17

R#1-38) Although beyond the scope of this paper, I suggest discussing the potential connection of hydrophilic, hydrophobic and neutral FFA to CCN activity.

We included on page 2, line 27-30: "Due to their structure and hygroscopic properties, amino acids can act as both ice-forming particles (INP) (Wolber and Warren, 1989;Szyrmer and Zawadzki, 1997;Pandey et al., 2016;Kanji et al., 2017) as well as cloud condensation nuclei (CCN) (Kristensson et al., 2010) in the atmosphere when amino acids such as arginine and asparagine can exist as metastable droplets instead of solid particles at low relative humidity; this showed a laboratory study (Chan et al., 2005)."

However, we are not aware of a study that investigates amino acids within their hydrophilic, hydrophobic and neutral characteristics. As no CCN studies with regards to amino acids were performed here, we would like to not stress this topic too much in the manuscript.

Page 18

R#1-39) L11-13 Please correct: For the calculated EF_{aer}, it should be noted that no further FAA formation or degradation pathways on the aerosol particles are considered, including biological or photochemical atmospheric reactions, *and* a possible transport from other than marine sources *is excluded*.

We agree with the reviewer's note and have incorporated his correction comments for page 21. In the course of re-structuring parts of the manuscript according to the suggestions of reviewer 2, this part was changed to: "A quantitative metric for comparing compounds in the ocean and in the atmosphere is the EF_{aer} (Eq. (2)). The concept is mainly applied to closed systems (e.g. Quinn et al. (2015), Rastelli et al. (2017)) because FAA formation or degradation pathways on the aerosol particles including biological or photochemical atmospheric reactions, and possible transport from other than marine sources are excluded in this parameter." (page 21, line 9-12)

R#1-40) L14 remove -as-

In the course of re-structuring to address the comments of reviewer 2, this sentence was changed and reads now (page 21, line 13-15): "The averaged EF_{aer} of ∑FAA in the individual Berner stages of the case study at the CVAO based on SML and ULW concentrations are shown in Fig. 5."

Page 17

R#1-41) L7-9 Please correct: Previous studies showed that organic material ejected into the atmosphere during bubble bursting, *results* in the formation of sea spray aerosol particles containing similar organic…..

We agree with the reviewer's comment and corrected the sentence on page 22, line 10-11. Now it reads: "Previous studies have shown that OM ejected into the atmosphere during bubble bursting, results in the formation of sea spray aerosol particles containing OM similar to SML (Russell et al. (2010);Cunliffe et al. (2013) and references therein)."

Page 19

R#1-42) L18 I suggest to change the title to -Origins of FFA in cloud water-, to be consistent with previous section

Taking into account the review comment R1#-35, the section 3.4.5 was removed.

SI material

R#1-43) Page 2, L17 ..and *is* discussed in more…

We agree with the reviewer's comment and corrected the sentence in the SI (page 3, line 5-6) to "It was generally low but increased during the campaign from 0.1 µg L^{-1} to 0.6 µg L^{-1} and is discussed in more detail by van Pinxteren et al., (2020)."

Additional changes performed by the authors

Due to the comments by reviewer 2 several parts of the manuscript were changed and adopted to the reviewer´s suggestions. This affected the following parts:

- The introduction: here the focus was more clearly placed on the FAA
- Discussion about the amino acid groups (hydrophilic, neutral and hydrophobic) in all marine compartments (seawater, aerosol particles and cloud water)
- Discussion about the concerted measurements of FAA in marine compartments
- Discussion about the aerosol enrichment factor
- Discussion and conclusion in general: stronger focus on the novelty value of the main findings shown here in the context of previous studies

When discussing the mean lifetime τ of individual amino acids (section 3.4 and Table S13), the unit of τ was changed from days (d) to hours (h).

In addition, the acknowledgement was also revised to thank the people from the OSCM. The added sentence is now as follows: "We further acknowledge the professional support provided by the Ocean Science Centre Mindelo (OSCM) and the Instituto do Mar (IMar)" (page 24, line 3-4)
The measured data were published on PANGAEA. The data availability statement was therefore updated and reads as follows: "Data availability. The data are available through the World Data Centre PANGAEA under the following link: https://doi.pangaea.de/10.1594/PANGAEA.914220." (page 23, line 26/27)

The previous citation of van Pinxteren (submitted 2019) was updated to van Pinxteren et al. (2020) in the revised manuscript and supporting information.

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