

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).

General Comments

R#2-1 a) This paper describes the measurement of free amino acids in seawater (microlayer and underlying water), aerosol, and cloud water at Cape Verde Island. The types and abundances of individual amino acids present in different sample types are compared. Enrichment factors are calculated for amino acid concentrations between the microlayer and underlying water, and between the water and aerosol. The contribution the amino acids make to the pools of organic carbon and total dissolved nitrogen in water and aerosol are presented. The work largely presents an inventory of amino acids for this location and the given sample types.

We have strongly revised the manuscript and added a lot of additional interpretation which represents clearly more than an inventory of amino acids and is explained in more detail in R#2-2 point 4. We agree that the implications were not well enough elaborated. From the revised results, we derived the atmospheric implication that simple parameters alone cannot describe the abundance and enrichment of FAA in the diverse marine environments. This work shows that, for a proper representation of FAA in oceanic/atmospheric models, their drivers need to be better understood and more studies are needed to unravel their complex composition.

R#2-1 b) The concerted measurement of amino acids in underlying seawater, microlayer, aerosol, and cloud water are novel. The authors also mention that these are the first reported free amino acids concentrations for marine clouds. Enrichment factors between seawater and aerosol are useful for comparison but I urge the authors to elaborate a little on the caveats of their use in an open system, namely that it is unlikely that a high fraction of the aerosol (and cloud droplets) measured came from the location of the measured seawater (and even less likely they emerged from those waters when the water was sampled). Concentrations in the atmosphere are also subject to ageing. This differs from other work on sea spray aerosol in closed systems where all particles are emitted from the contained source waters and minimal ageing occurs on the nascent particles (at least upon collection).

We thank the reviewer for his comments. We agree with the reviewer that the caveats of the concept of enrichment factor between seawater and aerosol were not treated well enough in the manuscript. It is true that the concept of the aerosol enrichment factor originally originates from controlled tank experiments and therefore does not take into account e.g. ageing processes or long-range transport processes of the aerosol particles, which does not completely correspond to the real conditions of field investigations. However, having pointed this out, we believe that the EF_{aer} is still a useful metric for comparison purposes. In recent studies, enrichment factors have been calculated from open systems and therefore provide a basis for comparative studies (e.g. Russell et al. (2010) or van Pinxteren et al. (2017)). In the revised version of our manuscript, we have underlined the uncertainties related to this concept and explained that the EF_{aer} might be useful for comparison purposes, but should be treated with caution.

The same applies to the enrichment factor between cloud water and seawater, which was calculated in the same way. Again, in the revised version, we point out that this is simplified approach, which only considers a possible transfer from the ocean to the aerosol particles and cloud water (neglecting

e.g. atmospheric processing). To illustrate the EF_{CW} , we included the values (with respect to the SML and the ULW, respectively) in Figure 5 in the revised version.

As a result, we have very carefully addressed these points in the following parts of the revised manuscript: In section 3.4.4 it is written now (page 21, line 9-13): “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. However, for comparison purposes, it might be useful to calculate the EF_{aer} also from open systems as done e.g. by Russell et al. (2010) or van Pinxteren et al. (2017).”

In addition, we have revised the abstract and the conclusion accordingly: The abstract (page 1, line 29-31) now reads: “Considering solely ocean-atmosphere transfer and neglecting atmospheric processing, high FAA enrichment factors were found in both aerosol particles in the submicron range ($EF_{aer(\Sigma FAA)}: 2 \cdot 10^3 - 6 \cdot 10^3$) and medium enrichment factors in the supermicron range ($EF_{aer(\Sigma FAA)}: 1 \cdot 10^1 - 3 \cdot 10^1$).”

The conclusion (page 23, line 10-14) is adopted as follows: “In a simplified approach, considering only a possible transfer from the ocean onto the aerosol particles and cloud water (neglecting e.g. atmospheric processing), the aerosol enrichment factor was calculated. A high FAA enrichment in the submicron aerosol particles of $EF_{aer(\Sigma FAA)}: 2 \cdot 10^1 - 6 \cdot 10^3$ and a medium enrichment on supermicron aerosol particles $EF_{aer(\Sigma FAA)}: 1 \cdot 10^1 - 3 \cdot 10^1$ were observed. Applying the same concept to cloud water, an enrichment of $4 \cdot 10^3 - 1 \cdot 10^4$ was obtained.”

Besides the concept of the enrichment factor, the revised manuscript deals with possible atmospheric processes/ aging, which are discussed in sections 3.4.1, 3.4.2 and 3.4.3 when discussing the individual amino acids or amino acid classes.

To address the representativeness of the aerosol particle measurements, we added the information about the aerosol particle measurements at the CVAO on the 30 m tower, which represent the conditions in the open ocean (and not just the surf zone), as follows (page 4, line 33 – page 5, line 2): “Size-segregated aerosol particles were sampled using five stage Berner-type impactors (Hauke, Gmunden, Austria) at the top of a 30 m sampling tower at the CVAO since this location best represents the conditions above the ocean pursuant to previous studies. The internal boundary layer (IBL), which can form when air passes a surface with changing roughness (i.e. the transfer from open water to island) is mainly beneath 30 m (Niedermeier et al., 2014).”

R#2-2) This work could be strengthened by: 1. elaborating on the significance of amino acids in the ocean and atmosphere and how the scientific community benefits from their inventory. 2. investigating the drivers of their observations by further relationships amongst their datasets and incorporating other datasets, e.g. Did variations in measured aerosol volume drive variations in amino acid concentrations? Did primary productivity (via remotely sensed chlorophyll-a) influence FAA concentrations in water and air? 3. further exploration of commonalities among and differences between their amino acid data across different sample types. 4. what implications can be drawn from the analyses accomplished and those I suggest? Are there certain ratios of amino acids in seawater that hold constant in seawater samples but not in the aerosol and clouds? What could cause this? 5. better connecting to past work. How do the observations compare not just to the values reported in other work but to the conclusions drawn in other research? How do drivers of amino acid concentrations differ between the ocean and atmosphere? What are the surface activities of the amino acids and can they predict their transfer in sea spray aerosol?

We thank the reviewer for this comment and the suggestions to improve the manuscript. We have addressed the points raised by the reviewer as shown in the following:

1. elaborating on the significance of amino acids in the ocean and atmosphere and how the scientific community benefits from their inventory.

1) *Significance of amino acids*: Amino acids are important in the oceans and in the atmosphere for a number of reasons. Due to their chemical structure, amino acids contribute to both dissolved organic nitrogen (DON) and dissolved organic carbon (DOC) and are therefore important biologically available sources of nitrogen and carbon. Especially in oligotrophic regions, such as the NATR region investigated here, the amino acids are an important source of nutrients for bacteria and other microorganisms. Besides the biogenic formation of amino acids in seawater and probably also on aerosol particles (e.g. GABA, see R#2-31), amino acids (as parts of proteins) also play an important role as ice-forming particles (INP) (Wolber and Warren, 1989; Szyrmer and Zawadzki, 1997; Pandey et al., 2016; Kanji et al., 2017) or cloud condensation nuclei (CCN) (Chan et al., 2005; Kristensson et al., 2010) in the marine environment. Besides abiotic and biotic conversion processes of amino acids, secondary organic aerosol particles (SOA) can be formed by the reaction of glyoxal with amino acids (Haan et al., 2009). These are important reasons why amino acids in the marine environment have been studied both in seawater and on the aerosol particles in different regions.

With our concerted FAA measurements in all marine compartments (seawater (ULW, SML), size-segregated aerosol particles and cloud water), it was possible not only to investigate the individual compartments (section 3.1, 3.2 and 3.3), but also to focus on the individual FAA and amino acid groups to identify similarities and differences. These are discussed in detail in the comments R#2-2 3), R#2-2 4), R#2-21, R#2-27. Furthermore, for the first time the FAA concentrations and compositions in cloud water were determined (section 3.3) and compared with the other two compartments (seawater and aerosol particles) in section 3.4.

In the revised version, we stronger elaborated the significance of the amino acids, as outlined in the Introduction: “Amino acids, either free (FAA) or in combined form (CAA), contribute to the global nitrogen and carbon cycle and to the atmosphere-biosphere nutrient cycle (Zhang and Anastasio, 2003; Wedyan and Preston, 2008). They are produced in the ocean and are reported to be in the upper layer of the ocean, the sea surface microlayer (SML) (Kuznetsova et al., 2004; Reinthaler et al., 2008; van Pinxteren et al., 2012; Engel and Galgani, 2016). The SML, as the direct interface between the ocean and the atmosphere, may play an important role as a source of organic matter (OM) in aerosol particles within the marine environment (Cunliffe et al., 2013; Engel et al., 2017; Wurl et al., 2017). Specific organic groups of compounds, including nitrogenous OM (Engel and Galgani, 2016) can be strongly enriched in the SML. From the ocean, amino acids as part of the class of proteinaceous compounds can be transferred into the atmosphere via bubble bursting (Kuznetsova et al., 2005; Rastelli et al., 2017). These proteinaceous compounds are often analyzed as sum parameter ‘proteins’ using an analytical staining method with Coomassie blue developed by Bradford (1976) and often applied in previous studies (Gutiérrez-Castillo et al., 2005; Mandalakis et al., 2011; Rastelli et al., 2017). Despite their attribution to proteins the FAAs are better utilizable forms of nitrogen instead of proteins for an aquatic organism such as phytoplankton and bacteria (Antia et al., 1991; McGregor and Anastasio, 2001).” (page 2, line 14-26)

Furthermore, we have added the following text in the introduction about the significance of amino acids in the atmosphere (page 2, lines 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).”

Benefits: Although FAA measurements in the marine atmosphere have been available so far, there is a lack of measurements considering both the abundance and molecular composition of amino acids simultaneously in the different marine compartments, especially in the tropical Atlantic Ocean. The measurements performed here, together with the interpretation of the data, will provide better insights into the FAA abundance, origins and possible transfer from the seawater and their transport within the atmosphere 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.

We would like to mention that the concerted FAA measurements performed here have already been included in a discussion paper published at “Biogeoscience” as “Free AA concentrations ... recently quantified in cloud droplets ... on the Cape Verde islands” (Jaber et al., 2020).

2. investigating the drivers of their observations by further relationships amongst their datasets and incorporating other datasets, e.g. Did variations in measured aerosol volume drive variations in amino acid concentrations? Did primary productivity (via remotely sensed chlorophyll-a) influence FAA concentrations in water and air?

2) We agree that the incorporation of other data sets was partly missing in the manuscript. Consequently, we included additional available parameters with the aim to explain the variation of FAA concentrations.

For seawater, DOC and TDN measurements (in ULW and SML), chl-*a* concentrations (as an indicator for primary productivity) and wind speed were included to investigate the observed variance of FAA concentrations in seawater. However, no statistically significant correlations could be found (in more detail in R#2-5).

For the aerosol particles, wind speed, wind direction, the particulate mass (PM) of the aerosol particles and the chl-*a* concentrations in the seawater were considered to describe the variance of the atmospheric FAA concentrations. However, again no statistically relevant correlations could be found (detailed information in R#2-6).

The surface activity of the individual amino acids was also considered in the revised version. The octanol-water partition coefficient (K_{OW}), the topological polar surface area (TPSA) and the density were included (Table S9). However, we found that these simple physico-chemical parameters could not explain the variance of the FAA by statistically relevant correlations. In addition to the parameters representing the surface activity, the mean lifetime τ of the aerosol particles (Table S13) was also taken into account. However, no statistically relevant correlations were found here either. For a detailed answer to the investigations on surface activity and mean lifetime we would like to refer to the comment R#2-33.

3. further exploration of commonalities among and differences between their amino acid data across different sample types.

3) In the revised manuscript version, a clearer focus is put on the comparison of amino acid profiles (in terms of individual amino acids and amino acid groups) in the different marine compartments. After the introduction of the individual marine compartments, seawater (section 3.1), aerosol particles (section 3.2) and cloud water (section 3.3), the composition of the amino acid profiles was examined in more detail in section 3.4. A distinction was made between hydrophilic (section 3.4.1), neutral and hydrophobic (section 3.4.2) and aromatic amino acids (section 3.4.3).

The main results are summarized in the Conclusion (page 22, line 29-34): “The similarities between the FAA composition in the seawater (SML) and on the submicron aerosol particle samples, as described in section 3.4, indicated that a certain FAA contribution, in particular the hydrophilic amino acids Asp and Glu in the submicron aerosol particles at the CVAO, was probably caused by sea spray and might be transferred up to cloud level. The neutral and hydrophobic amino acids were also present in all marine compartments, suggesting some interconnections. Stable amino acids like

Gly are often reported as long-range tracers, but their abundance in seawater and marine air masses prevailing during the sampling period suggest an (additional) oceanic source.”

For a detailed overview of the individual discussion of the amino acid profiles (in the individual compartments and the cross-compartment comparison in the marine environment), we would like to refer to the comments R#2-20, R#2-21, R#2-22, R#2-24 and R#2-27.

4. what implications can be drawn the analyses accomplished and those I suggest? Are there certain ratios of amino acids in seawater that hold constant in seawater samples but not in the aerosol and clouds? What could cause this?

The main points (summarized in the revised conclusion of the present study) are:

- A certain FAA contribution, particularly the hydrophilic amino acids Asp and Glu in the submicron aerosol particles at the CVAO, was probably caused by sea spray and might be transferred up to cloud level.
- Stable amino acids like Gly are often reported as long-range tracers, but their abundance in seawater and the marine air masses prevailing during the sampling period suggest an (additional) oceanic source.
- Indications for biological production of amino acids on the aerosol particles (GABA) were observed, supporting recent finding of a high active enzymatic activity on marine aerosol particles.
- The high FAA concentrations and enrichments in cloud water, which have been reported here for the first time. Their composition, together with the high concentrations of inorganic marine tracers (sodium, MSA), indicate at least to some extent to an oceanic transfer and biogenic formation that remains subject to future work.

From these findings, together with the now added additional interpretations to our data, we derived the atmospheric implications (see answer to R#2-1 a) and added the following in the Abstract and in the Conclusion: ”Finally, the varying composition of the FAA in the different matrices shows that their abundance and ocean-atmosphere transfer are influenced by additional biotic and abiotic formation and degradation processes. Simple physico-chemical parameters (e.g. surface activity) are not sufficient to describe the concentration and enrichments of the FAA in the marine environment. For a precise representation in organic matter (OM) transfer models, further studies are needed to unravel their drivers and understand their composition.” (page 2, line 3-8) and “Altogether, the varying composition of FAAs in the different matrices shows that their abundance and their enrichments in the SML and their atmospheric transfer are not determined by single environmental drivers (e.g. wind speed) and/or simple physico-chemical parameters (e.g. surface activity). The ocean-atmosphere transfer of FAAs is influenced by biotic and abiotic formation and degradation processes. Further studies are required to unravel their drivers and understand their complex composition that, finally, have to be considered in OM transfer models.” (page 23, line 17-23)

For more details, we would like to refer to our answer to the referee comment R#2-3.

These results and implications were derived from studying the ratios via the percentage composition of the FAA in the individual compartments together with other information (e.g. environmental parameters and physico-chemical properties). For a more detailed discussion of the amino acid ratios/ profiles, we would like to refer to our answers to the Review comments R#2-21 R#2-26 and R#2-27, R#2-31.

5. better connecting to past work. How do the observations compare not just to the values reported in other work but to the conclusions drawn in other research? How do drivers of amino acid concentrations differ between the ocean and atmosphere? What are the surface activities of the amino acids and can they predict their transfer in sea spray aerosol?

We agree that the connection of our work to previous studies was not strong enough. In the revised manuscript version, we have significantly extended the comparison of our observations to the conclusions drawn in earlier studies. We have also outlined the uncertainties of sources and drivers of amino acids in the marine environment and connected our findings to literature studies at several parts of the manuscript as follows:

In the Introduction, we have included the following (page 3, line 3-14): "Based on a cluster and factor analysis, Scalabrin et al. (2012) suggested two possible sources for the amino acids in the ultrafine Arctic aerosol particles. First, the authors mentioned the regional development (isoleucine, leucine, threonine) and long-range transport (glycine) of amino acids from marine areas; secondly, the influence of local sources such as of marine primary production (proline, valine, serine, tyrosine, glutamic acid). A different approach of Mashayekhy Rad et al. (2019) investigated the atmospheric proteinogenic aerosol particles in the Arctic and attributed them to different sources based among others on the reactivity of the distinct amino acids. The authors differentiated here between long-range transport (glycine), terrestrial and marine aerosol particles (proline, valine, serine, tyrosine) and coastal and marine phytoplankton and bacteria (isoleucine, leucine and threonine) as important sources for amino acids (Mashayekhy Rad et al., 2019). In fact, previous studies have assigned individual amino acids to specific marine biogenic sources and used them as biomarkers. Hammer and Kattner (1986) reported correlations between aspartic acid, diatoms and zooplankton in seawater. GABA (γ -aminobutyric acid) was referred to as an indicator for the microbiological decomposition of OM (Dauwe et al., 1999; Engel et al., 2018) and is probably used as a microbiological proxy in aerosol particles."

In section 3.1, we carefully revised the section on FAA enrichment in the SML and it reads now as follows (page 10, line 16-30): "The results of the individual FAA concentrations in seawater (ULW, SML) and their EF_{SML} , listed in Table S3, show clear differences between the individual amino acids and the amino acid classes. The most highly enriched amino acids in the SML are the neutral ones with values of up to 203 compared to the hydrophilic (EF_{SML} : 2-98) and hydrophobic (EF_{SML} : 1-96) amino acids. This may be related to the fact that Ser, Thr and Gly as part of the neutral amino acids, are main components of cell wall proteins (Hecky et al., 1973). The direct release of FAAs through cell lysis and the associated destruction of the cell wall can thus explain the increased enrichment of neutral amino acids in the SML. Our study confirmed that the SML is often non-uniformly enriched with FAAs as outlined from previous observations (Kuznetsova and Lee, 2002; Reinthaler et al., 2008; van Pinxteren et al., 2012; Engel and Galgani, 2016). Different factors, such as the transport of FAA from the ULW to the SML, the in-situ production by an extracellular hydrolysis of CAA or a direct release of FAA by cell lysis probably cause the observed enrichment of FAA in the SML. Kuznetsova and Lee (2002) showed that the rapid extracellular hydrolysis of CAA in the SML was not the cause of the non-uniformly enrichment in SML. Moreover, they suggested that the intracellular pools of organisms rich in DFAA and DCAA compared to seawater can be leached out by stressed microorganisms, resulting in the release of DFAA which in turn influences the pools of both DFAA and DCAA in seawater. Based on previous studies, the transportation and releasing mechanisms seem most likely to be the reasons for the observed enrichment of FAA."

And in section 3.2.1, we revised the discussion as follows (page 14, line 21-27): "Following this hydrophathy 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.”

As mentioned above (R#2-2, point 2), we included a variety of environmental factors and also considered substance-specific properties as well as the mean lifetime τ to explain the amino acid concentrations in seawater or on the aerosol particles and to predict the transfer. However, no statistically relevant correlations could be found for all additionally investigated parameters. Therefore, the drivers of amino acid concentrations between ocean and atmosphere could not be distinguished.

R#2-3) Ultimately I would like the authors to demonstrate that they have done more than make some measurements and that we have gained new insight into the ocean-atmosphere system beyond an inventory of amino acids. The manuscript needs to set up what was done on top of our current understanding of sea spray aerosol formation and the transfer of different compounds/-classes into sea spray aerosol. By building on past research the present work is prepared to more clearly communicate its findings and to also contribute new knowledge to the field.

Based on the new measurement data obtained in our work, we have now added many more additional interpretations to our data. Our novel approach of the concerted measurements (ULW, SML, aerosol particles and cloud water) – which is clearly more than ‘some measurements - together with the analysis at molecular level provided a rich data set of the ambient amino acids. These measurements allowed to study similarities and differences of FAA in the various compartments and indicated that a certain FAA contribution, in particular the hydrophilic amino acids Asp and Glu in the submicron aerosol particles at the CVAO, was probably caused by sea spray and might be transferred up to cloud level. We showed that the neutral and hydrophobic amino acids were also present in all marine compartments, suggesting some interconnections. Stable amino acids like Gly are often reported as long-range tracers, but their abundance in seawater and the marine air masses prevailing during the sampling period suggest an (additional) oceanic source. The oceanic link is supported by a high atmospheric concentration of ocean-derived compounds (sodium, MSA), a high fractional residence time of the air masses above water and a low-to-medium impact of other non-marine sources (based e.g. on the mass concentration of trace metals). In addition, we could derive some indications for the biological production of amino acids on the aerosol particles (GABA), supporting the recent finding of a high active enzymatic activity on marine aerosol particles. We found that aromatic amino acids are either not transferred from the ocean into the atmosphere or react very quickly, in any case they are present in low concentrations close to the LOQ. By distinguishing between submicron and supermicron aerosol particles, differences in the chemical composition of these aerosol particle size classes could be identified, which show a much higher complexity of the FAA composition in the submicron aerosol particles. FAA were present in the size range for aerosol particles associated with CCN activity and cloud water, and might be connected to CCN activity due to their hygroscopicity and soluble character, but this effect was not investigated here.

Regarding the enrichment factor (see our answer to the comment R#2-1 b).

These aspects are summarized in the conclusions of the revised version: “In a simplified approach, considering only a possible transfer from the ocean onto the aerosol particles and cloud water (neglecting e.g. atmospheric processing), the aerosol enrichment factor was calculated. A high FAA enrichment in the submicron aerosol particles of $EF_{\text{aer}(\Sigma\text{FAA})}: 2 \cdot 10^1 - 6 \cdot 10^3$ and a medium enrichment on supermicron aerosol particles $EF_{\text{aer}(\Sigma\text{FAA})}: 1 \cdot 10^1 - 3 \cdot 10^1$ were observed. Applying the same concept to cloud water, an enrichment of $4 \cdot 10^3 - 1 \cdot 10^4$ was obtained. The high FAA concentrations ($11.2 - 489.9 \text{ ng m}^{-3}$) and enrichments in cloud water were reported here for the first time. Their composition, together with the high concentrations of inorganic marine tracers (sodium, MSA),

indicate at least to some extent an oceanic transfer and biogenic formation that remains subject to future work. Altogether, the varying composition of FAAs in the different matrices shows that their abundance and their enrichments in the SML and their atmospheric transfer are not determined by single environmental drivers (e.g. wind speed) and/or simple physico-chemical parameters (e.g. surface activity). The ocean-atmosphere transfer of FAAs is influenced by biotic and abiotic formation and degradation processes. Further studies are required to unravel their drivers and understand their complex composition that, finally, have to be considered in OM transfer models. To the best of our knowledge, this study was the first that simultaneously analyzed the FAA in all marine compartments - seawater including the ULW and the SML, size-segregated aerosol particles and cloud water – in such detail to obtain indications on their sources and interconnections.” (page 23, line 10-24).

Altogether, we believe that the results mentioned above provide new insight into the ocean-atmosphere system beyond an inventory of amino acids.

R#2-4) Introduction. page 3. line 22. Barbaro et al. (2015) investigated FAA in size-segregated Antarctic aerosol particles to gain information about FAA as possible tracers of primary biological production in Antarctic aerosol particles line 24. Although there are several studies in different marine regions, there is a lack of ambient measurements of FAA simultaneously in seawater and in size-segregated aerosol particles in the tropical Atlantic Ocean It would strengthen the justification for the work if it was stated why we must know this information specifically for the Atlantic Ocean and atmosphere.

As stated correctly by the reviewer, there are several studies of amino acids in marine samples. However, source attributions of amino acids are still not clear and might vary between different marine locations. In addition, there is mostly a lack of measurements that regard the abundance and molecular composition of amino acids simultaneously in marine compartments - in seawater and in the atmosphere – especially in the tropical Atlantic Ocean. Such studies are, however, crucial to learn more about sources and fate of the amino acids in the ambient marine environment. Besides, cloud water studies of amino acids are lacking. However, such studies are needed to investigate if these important compounds (see answer to the comment R#2-2) are transferred or maybe produced in marine clouds.

We referred to these points more clearly in the introduction and reads as follows (page 3, line 22-30): “Despite several studies of FAAs also conducted in the marine environment, there is still a huge uncertainty to the question whether FAAs are of marine origin or not. Matsumoto and Uematsu (2005) showed that the long-range transport of land-derived sources largely contributes to the amino acid concentration in the North Pacific. On the other hand, based on a positive correlation between amino acids in seawater and the atmosphere, Wedyan and Preston (2008) pointed out the particulate amino acids in the Southern Ocean to be of marine origin. These findings are likely due to regional varying source strengths, given different meteorological and biological conditions, which require further measurements in distinct marine regions necessary. Unfortunately, measurements are lacking that regard the abundance and molecular composition of amino acids in both seawater and size-segregated aerosol particles, especially in the tropical Atlantic Ocean.”

The region investigated here, the NATR region around the Cape Verde Islands (see R#2-5), is an interesting but rarely studied oligotrophic region. The region of study is of huge interest as it is home to a remote marine time-series observatory with low anthropogenic influences where particles during the campaign were predominantly of marine origin. Therefore, our studies provide a better understanding of the FAA in such a region on a molecular level, in order to be able to describe their sources and fate in the marine environment more precisely.

The main statements of this study concerning amino acids in the atmosphere in this marine region in view of the present state of knowledge and the new main findings of this study are summarized in more detail in answer R#2-8. For the general importance of the amino acids, please see our answer to the referee comment R#2-2.

R#2-5) There is little effort made to investigate the drivers of amino acids data for the ULW and SML. The data are not plotted against other measurements. We are merely offered (page 9, line 19): "On the basis of previous studies, the transportation and the releasing mechanisms seems to be most likely for the observed enrichment of FAA." At this point, we were not given a strong motivation for the measurements and we haven't learned anything from them. Then it is explained that the data are in agreement with other datasets and then this one dataset is extrapolated to the entire North Atlantic Ocean. I wouldn't extrapolate data from one location to the entire North Atlantic Ocean.

We agree that not sufficient effort was made to investigate the drivers of amino acids data for the ULW and SML and have consequently included correlations to environmental parameters in the revised version. However, correlations of the FAA data with either wind speed, chl-*a*, DOC or TDN could not explain the variance in FAA concentrations or enrichments. That was, however, in agreement to results of previous studies.

The transportation and releasing mechanisms is discussed in section 3.1. We found high enrichments of FAA in the SML that we considered interesting. The environmental parameters could not explain the observed FAA enrichment in the SML of neither the individual nor the amino acid groups. However, we could observe that the neutral amino acids in particular are more abundant than the hydrophilic or hydrophobic ones. This may be related to the fact that neutral amino acids are the main components of cell wall proteins and are directly released by cell lysis, as we concluded by comparing our findings to results from the literature.

The motivation for the measurements are summarized in the revised introduction, please see our answer to the referee comments R#2-2 and R#2-8.

Several changes according to these aspects were done in the revised manuscript. For a better visualization, we have included the temporal variation of the respective additional parameters (wind speed, chl-*a*, DOC and TDN) in Figure 1. The discussion of the correlations to environmental parameters was added and reads (page 10, line 12-14): "Nevertheless, the variance of the \sum 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." For explanations of the observed enrichment of the FAA in the SML we want to refer to the referee comment R#2-2 point 5).

R#2-5_1) Then it is explained that the data are in agreement with other datasets and then this one dataset is extrapolated to the entire North Atlantic Ocean. I wouldn't extrapolate data from one location to the entire North Atlantic Ocean.

We agree that we might not extrapolate the local data to the entire North Atlantic Ocean, as it appeared in the first version of the manuscript. In the revised version we have subdivided the marine regions in more detail according to Longhurst (2007). Thus, in the revised manuscript version we refer to the "North Atlantic Tropical Gyral Province (NATR)" region (introduced by Longhurst (2007)). This region is defined as follows in the manuscript (page 4, line 13-16): "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."

We compared our measurements with previous own FAA measurements at the same location (data from 2013, shown here for the first time in Table S4) and the good agreement allowed us to conclude that FAA concentrations shown in this study can be considered as representative for the NATR region surrounding the Cape Verde Islands.

Moreover, we compared the FAA concentrations obtained here to published FAA data from the subtropical Atlantic Ocean the western Mediterranean Sea and concluded that "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 (Reinthal et al., 2008)).” (page 11, line 5-7).

R#2-6) Similar to the seawater measurements, the atmospheric research would be stronger if it explored the drivers of the amino acid data. Particularly lacking from the aerosol work are aerosol sizing measurements. In their absence, we do not know if the variability in amino acid concentrations in the air are driven by changes in particle concentrations. Similarly, amino acid concentrations in the air could be related to wind direction.

Following the comment of the reviewer, and similar to the seawater results, we have tested to correlate the particulate FAA concentrations with wind speed, wind direction, chl-*a* data in seawater as well as the particulate mass concentrations of the aerosol particle samples to explain the variance of amino acid concentrations in the aerosol particle samples. However, statistically relevant correlations could not be found. In the manuscript the text now reads (page 12, line 10-12): “Correlations between the Σ FAA concentrations of the size-segregated aerosol particles (considered as submicron, supermicron and PM₁₀) and the wind speed were not observed for here reported data (Fig. 2, Table S2).”

and on page 12, line 18/19: “No significant correlation could be observed between the Σ FAA concentration of size-segregated aerosol particle samples (submicron, supermicron and PM₁₀) and the chl-*a* concentration in seawater.”

and on page 14, line 13-15: “However, the reasons for the high concentration of hydrophilic FAAs within these respective sampling days remain unclear, since no change in the environmental parameters determined (e.g. wind speed, wind direction, chl-*a* concentration, Fig. 2a) was observed.”

For a better visualization, we have included the temporal variation of the respective additional parameters (wind speed, wind direction and chl-*a*) in Figure 2.

Other:

R#2-7) Did particle type vary across the different aerosol sampling periods?

We have included the discussion of several meteorological parameters and back trajectories as well as particulate mass concentrations and included the new small chapter “*First indications of aerosol particle origin*”. From these discussed measurements we could not observe changes in the type of particles across the different aerosol sampling periods. In the revised manuscript it reads now as follows: “To obtain a first indication of the particle origin, that might help to explain the differences in the particle composition concerning amino acids, the particles were associated with the origin of the air masses and with marine and dust tracers. Overall, the CVAO station experienced north-easterly trade winds during this campaign, which are typical for this season within this region (Fomba et al., 2014; van Pinxteren et al., 2020). According to physical and chemical specifications such as the air mass origins, particulate MSA concentrations and MSA/sulfate ratios as well as particulate mass concentrations of dust tracers, aerosol particles predominantly of marine origin with low to medium dust influences were observed. It has to be noted that dust generally influences the supermicron particles to a larger extent than the submicron particles (Fomba et al., 2013). Further information on the classification of the air masses are given in the overview paper of this campaign (van Pinxteren et al., 2020).” (page 11, line 25-33)

R#2-8) If amino acids have already been measured in remote marine air/aerosol, how does the present manuscript advance our knowledge?

Although some studies on amino acids in marine aerosol particles have been published so far, the source of the amino acid as well as their transfer and fate is still not clear. Especially with regard to the connection between ocean and atmosphere, the gap in the simultaneous investigation of both marine compartments is problematic with regard to FAA. We have briefly summarized the current

state of knowledge on this subject in the revised introduction. This now reads as follows (page 3, line 21-30): “Although the study and characterization of amino acids are of paramount importance for atmospheric scientists, the true role and the fate of amino acids in the atmosphere are still poorly understood (Matos et al., 2016). Despite several studies of FAAs also conducted in the marine environment, there is still a huge uncertainty to the question whether FAAs are of marine origin or not. Matsumoto and Uematsu (2005) showed that the long-range transport of land-derived sources largely contributes to the amino acid concentration in the North Pacific. On the other hand, based on a positive correlation between amino acids in seawater and the atmosphere, Wedyan and Preston (2008) pointed out the particulate amino acids in the Southern Ocean to be of marine origin. These findings are likely due to regional varying source strengths, given different meteorological and biological conditions, which require further measurements in distinct marine regions necessary. Unfortunately, measurements are lacking that regard the abundance and molecular composition of amino acids in both seawater and size-segregated aerosol particles, especially in the tropical Atlantic Ocean.”

We wanted to investigate the already mentioned knowledge gap about the ocean-atmosphere relationship more closely through our methodological approach and the associated FAA analysis, and to introduce new aspects or support previous assumptions of other studies in this marine region as well. To this end, we have summarized the main objectives of our work in the revised introduction as follows (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.”

The main findings that are beyond the state of knowledge are summarized in the conclusion and have been outlined in our answer to the referee comment R#2-3.

Specific Comments

R#2-9) Abstract. page 1. line 20. “The total concentration (PM10) was between 1.8–6.8 $\mu\text{g m}^{-3}$ and tended to increase during the campaign.” Perhaps instead of “during the campaign”, give the time period or relate to a potential/suspected driver of the observation.

Following the comment of the reviewer the added the time period of the campaign in the Abstract and it reads now (page 1, line 12-14): “Measurements of free amino acids (FAA) in the marine environment to elucidate their transfer from the ocean into the atmosphere to marine aerosol particles and to clouds were performed at the MarParCloud campaign at the Cape Verde islands in autumn 2017.”

Introduction.
page 2.

R#2-10) lines 8-12. The marine nitrogen cycle is alluded to but vaguely. "(T)he bulk DON pool CAN BECOME bioavailable" or "IS bioavailable"? What is remineralized nitrogen? Perhaps get straight to proteniaceous compounds and amino acids as being large members of the DON pool.

Due to the revision of the introduction and also following the suggestions from the reviewer's, we focused on the FAA right from the beginning and therefore general parts (as the one mentioned by the reviewer) were deleted from the manuscript.

R#2-11)line 21. much more work has been done on amino acids as ice nucleating entities (and as antifreeze entities) than just Szyrmer and Zawadzki, 1997 and should be acknowledged because it gives importance to this work.

We agree with the comment of the reviewer. We have added more references in the reference to the INP ability of amino acids. It reads now (page2, 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)."

R#2-12)line 23. "Amino acids are also present and have been described in the marine environment." This has already been established. The first 3 sentences of this paragraph feel like we are going backwards. In the previous paragraph we went from the ocean to the atmosphere and now we are back to describing amino acids in the ocean.

We agree with the comment of the reviewer and have removed the noted sentences in the course of restructuring the introduction. With the revised introduction, the focus should now be more clearly on amino acids and the structure of the introduction should also be more coherent with the revision (as outlined in the answer to the referee comment R#2-2).

R#2-13)line 28. (Engel et al., 2017) lacking other citations for the microlayer's importance in the ocean and sea spray aerosol.

Following the comment of the reviewer, we added other citations in this sentence. Now it reads (page 2, line 17-19): "The SML, as the direct interface between the ocean and the atmosphere, may play an important role as a source of organic matter (OM) in aerosol particles within the marine environment (Cunliffe et al., 2013;Engel et al., 2017;Wurl et al., 2017)."

page 3.

R#2-14) line 7. "However, chl-a concentration solely does not adequately describe the complete spectrum of biological activity (Quinn et al., 2014)" should reference a paper on marine microbiology, like Azam and Malfatti. 2007. Microbial Structuring of Marine Ecosystems. DOI: 10.1038/nrmicro1747

We agree with the comment of the reviewer and have removed the noted sentences in the course of restructuring the introduction. With the revised introduction, the focus should now be more clearly on amino acids and the structure of the introduction should also be more coherent with the revision (as stated in the answer to the referee comment R#2-2).

page 4.

R#2-15) line 13. "During this campaign, concerted measurements were performed including sampling of sizesegregated aerosol particles at the CVAO and seawater sampling at the ocean site (16.53°N, 24.54°W). The location was carefully chosen with minimal influence of the island and located in wind direction to the CVAO" This second sentence is crucial to the rationale of the study and should be demonstrated. I'm surprised to not see a figure referenced here.

We agree with the comment of the reviewer and added a Figure S1 in the SI. Figure S1 shows an overview of the sampling locations MV, CVAO and seawater sampling site now and additionally the prevailing wind direction was added. In the manuscript, now it reads (page 4, line 13-19): “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. 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°53'17'N, ~24°54'25'E). The location was carefully chosen with minimal influence of the island and located in wind direction to the CVAO as shown in Fig. S1.”

In addition, we would like to point out that a general introduction to the campaign and the setting is given by the (now published) article by van Pinxteren et al., 2020 that we refereed to (for example: “van Pinxteren et al. (2020) provide further details on the MarParCloud campaign.”, page 4, line 20/21).

R#2-16) line 20. "The seawater samples were taken with a fishing boat, starting from Bahia das Gatas, São Vicente." A study site map with water and aerosol locations and winds during sampling periods would be good to "connect the dots". Figure S1 does not do this.

We agree with this comment and the previous comment (R#2-15) of the reviewer and added Fig. S1 as an overview of the sampling locations MV, CVAO and seawater sampling site in the SI.

page 6.

R#2-17) line 2. "Since no chiral column was used in the UHPLC separation, it is possible that not only L-amino acids, which were used as the standard, were quantified, but that the here presented concentrations were possibly quantified as the sum of the L- and D-amino acids." So why not simply report them as L- and D-amino acids instead of reporting them as L-amino acids but acknowledging that is not accurate?

We thank the reviewer for this comment and have changed the sentence so that it now reads as follows: “Since no chiral column was used in the UHPLC separation, we cannot differentiate between L- and D- amino acids in our ambient samples” (page 6, line 15/16).

Furthermore, the following manuscript does not differentiate between D- and L-amino acids.

R#2-18) 2.2.4 Enrichment Factors

For calculating enrichment factors (EFs) between aerosol and the SML, I wonder if it would be more appropriate to first calculate the aerosol-ULW EF using equation 2 and then apply the SML-ULW EF (EF_SML) from equation 1 for that analyte. So there would be EFs for aerosol relative to the underlying water and relative to the SML and they would differ by the EF_SML (Eq.1). Ideally this would resolve the current inconsistency in invoking the Na⁺ concentration of the SML when comparing to aerosol to SML (Eq. 2) but not when comparing SML to ULW. The way it stands now, if you calculate aerosol EFs relative to the ULW and the SML they won't differ by the EF_SML and instead will differ by an EF_SML calculated using equation 2.

As mentioned above (answer to the reviewer comment R#2-1b), we strongly revised the here applied concept of the calculation of the EF_{aer} and pointed out the relating caveats. We used the EF_{aer} for comparison purposes and in this regard, we included the EF_{aer} based on ULW (besides the EF_{aer} based on SML). The Na⁺ concentration in the ULW and SML was very similar (12.45 g L⁻¹ in the ULW and 12.53 g L⁻¹ in the SML), therefore differences in the EF_{aer} (related to the SML and to the ULW, respectively) are mainly due to the different concentrations, i.e. the high SML enrichments of the FAAs.

To calculate the EF_{aer} (Equation 2), we followed previous studies. In these studies, the enrichment of a substance was calculated by using on the aerosol particles the concentration of the substance in relation to the sodium concentration in relation to the substance concentration and sodium concentrations present in seawater. This was done, for example, under controlled conditions as described in Rastelli et al. (2017), but also in ambient studies such as Russell et al. (2010) and van Pinxteren et al. (2017).

page 10.

R#2-19) line 1. This sentence is confusing: "In the study of Reinthaler et al. (2008) DFAA contributed with ~12 % to DOC and with ~30 % to dissolved organic nitrogen (DON) in the SML of the Atlantic ocean and the western Mediterranean Sea." Is it DFAA in seawater accounting for ~12% of DOC (in seawater), and DFAA in SML accounting for ~30% of DON in the SML?

We agree with the reviewer and have reworded the sentence (page 11, line 17-19) as follows: "In the SML of the Atlantic Ocean and the western Mediterranean Sea, the DFAA contributed with ~12 % of the DOC and ~30 % of the dissolved organic nitrogen (DON) (Reinthaler et al., 2008)."

R#2-20) line 28. The characterization of FAA into the hydrophilic, hydrophobic, and neutral classes is nice. What might be the drivers and implications?

The FAA's division into hydrophilic, hydrophobic and neutral amino acid classes was used to ensure better comparability with previous studies. Often different analytical methods are used to study the FAA in different marine compartments, mostly seawater or aerosol particles, and often different individual FAA standards are used. In order to be able to make statements about the FAA with similar physico-chemical properties, the classification of the amino acids is based on the 'hydropathy index'. This classification was used, for example, in previous studies to classify the FAA results in the aerosol particles at the CVAO. This can be read as follows in the revised manuscript (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."

Moreover, the classification within this study was used to investigate the FAA composition in the different marine compartments (section 3.1, 3.2 and 3.3) and also to compare the compartments with each other. This is done after subdividing the amino acid classes in sections "3.4.1 Hydrophilic amino acids" and "3.4.2 Neutral and hydrophobic amino acids".

page 11

R#2-21) line 11. "However, the presence of Glu, Asp and GABA as part of the hydrophilic species in the submicron aerosol particles (on 22/09/2017, 4/10/2017, 6/10/2017, 7/10/2017) strongly indicated a local oceanic origin." If amino acids can indicate aerosol type or source, this should be central to this work, explained in the introduction, and examined for each sample. Also, how did the amino acid profiles of the different sample types (and individual samples) compare? Were the same relative abundances consistent across the ULW, SML, and different aerosol size classes? Are all the amino acids measured commonly found in the ocean and are they exclusive to the ocean?

There is still a high uncertainty about source attributions of FAA in the marine atmosphere. In the revised manuscript, we addressed this issue in several parts in the revised manuscript as outlined in the following. In the introduction we listed the results concerning the attribution of FAAs to distinct sources from other studies (detailed changes are shown in our response to comment R#2-8) and concluded: “Despite several studies of FAAs also conducted in the marine environment, there is still a huge uncertainty to the question whether FAAs are of marine origin or not. Matsumoto and Uematsu (2005) showed that the long-range transport of land-derived sources largely contributes to the amino acid concentration in the North Pacific. On the other hand, based on a positive correlation between amino acids in seawater and the atmosphere, Wedyan and Preston (2008) pointed out the particulate amino acids in the Southern Ocean to be of marine origin. These findings are likely due to regional varying source strengths, given different meteorological and biological conditions, which require further measurements in distinct marine regions necessary. Unfortunately, measurements are lacking that regard the abundance and molecular composition of amino acids in both seawater and size-segregated aerosol particles, especially in the tropical Atlantic Ocean.” (page 3, line 22-30)

In our work we performed a proper comparison of the FAA in the different matrices and concluded: “The high complexity of FAA observed in seawater was also found in the aerosol particles as well as in cloud water, and generally shows a high similarity between FAA in the different compartments. All marine compartments contained Gly, Ser, Glu and Ala as dominant species, i.e. representatives of the hydrophilic, neutral and hydrophobic groups. However, the percentage contribution of the individual FAAs to the \sum FAA varies within the different compartments.” (page 18, line 3-7)

The individual FAA are discussed in section 3.4.1, 3.4.2 and 3.4.3.

Important findings from our work show:

“Consequently, the usage of the major FAAs as chemical biomarkers seems to be restricted to some extent due to their lack of source-specificity” (page 14, line 9/10)

Still, we could conclude from our investigations that “...a certain FAA contribution, in particular the hydrophilic amino acids Asp and Glu in the submicron aerosol particles at the CVAO, was probably caused by sea spray and might be transferred up to cloud level. The neutral and hydrophobic amino acids were also present in all marine compartments, suggesting some interconnections. Stable amino acids like Gly are often reported as long-range tracers, but their abundance in seawater and marine air masses prevailing during the sampling period suggest an (additional) oceanic source.” (page 22, line 30-34)

This is in agreement with results from Barbaro et al. (2015). “Barbaro et al. (2015) reported that hydrophilic components were predominant (60 %) in locally produced marine Antarctic aerosol particles, whereas hydrophobic compounds were rather dominant 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.” (page 14, line 24-27)

Furthermore, we found “In addition, some indications for the biological production of amino acids on the aerosol particles (GABA) were observed, supporting the recent finding of a high active enzymatic activity on marine aerosol particles. Aromatic amino acids are either not transferred from the ocean into the atmosphere or react very quickly; in any case, they are present only in small concentrations close to the LOQ.” (page 23, line 3-6)

We think that the results summarized here, together with the reported high FAA concentrations in cloud water (see Comment R#2-27), are new and interesting and help to gain better insights into sources and transfer of FAA to the marine environment.

page 12

R#2-22) line 2. "Considering the amino acid classifications from Barbaro et al. (2015)), it can be concluded that the submicron aerosol particles with low averaged percentage of hydrophobic species (5 %) and higher percentages of hydrophilic species (4-55 %, mean

of 15 %) could have local oceanic origin." Similar to previous comment. The different amino acid characterizations: hydrophathy index of Kyte & Doolittle 1982; Pommie et al. 2004; and Barbaro et al. 2005 should be explained early on and would add value to the present work.

Following the reviewers' suggestion, we have described the concept of the hydrophathy index already in the introduction. It reads now (page 3, line 14-18): "Grouping amino acids as regards their physico-chemical properties ('hydrophathy' index (Kyte and Doolittle, 1982)) allows different studies to better compare them what Pommie et al. (2004) suggested pursuant to the partition coefficient between water and ethanol. This divides them into hydrophilic, neutral and hydrophobic amino acids as discussed in Barbaro et al. (2015) for FAA in Antarctic aerosol particles."

And on page 3, line 34 – page 4, line 2: "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 hydrophathy index."

Furthermore, this classification of amino acids, introduced in the introduction, is taken up in the discussions of the individual marine compartments (section 3.1, 3.2 and 3.3) and in the comparison of the marine compartments with regard to the FAA classifications (section 3.4.1, 3.4.2, 3.4.3).

R#2-23) line 5. "This is supported by a predominant marine origin of the aerosol particles according to the air masses history, particulate MSA concentrations and MSA/sulfate ratios and particulate concentrations of dust tracers (Table S8)." It would be nice to lay this out because we make it this far into the manuscript wondering about the presence of aerosols from land in these samples. I would characterize the air masses early on. Do the back trajectories differ for the samples collected at 2 different elevations? And which back trajectories were used? Hysplit or Flexpart? Is this mentioned in the Experimental section?

We thank the Reviewer for his comments regarding the missing discussion of the aerosol particle origins and the air mass characterization in the manuscript.

Information about the calculation of the backward trajectories and the validity of those for both sampling stations (CVAO and MV) was added in the experimental part, section 2.2.2 (page 7, line 12-17) and it reads now: "Back trajectory analyses provided information regarding the origins of air masses. Seven-day back trajectories were calculated on an hourly basis within the sampling intervals, using the NOAA HYSPLIT model (HYbrid Single-Particle Lagrangian Integrated Trajectory, <http://www.arl.noaa.gov/ready/hysplit4.html>, 26.11.16) in the ensemble mode at an arrival height of 500 m ± 200 m (van Pinxteren et al., 2010); van Pinxteren et al. (2020) provide more details. The calculated backward trajectories are representative for both aerosol particle sampling stations (CVAO and MV)."

Moreover, a short characterization of the air masses was added at the beginning of section 3.2.1, which reads now (page 11, line 24-33):

"First indications of aerosol particle origin

To obtain a first indication of the particle origin, that might help to explain the differences in the particle composition concerning amino acids, the particles were associated with the origin of the air masses and with marine and dust tracers. Overall, the CVAO station experienced north-easterly trade winds during this campaign, which are typical for this season within this region (Fomba et al., 2014; van Pinxteren et al., 2020). According to physical and chemical specifications such as the air mass origins, particulate MSA concentrations and MSA/sulfate ratios as well as particulate mass concentrations of dust tracers, aerosol particles predominantly of marine origin with low to medium dust influences were observed. It has to be noted that dust generally influences the supermicron particles to a larger extent than the submicron particles (Fomba et al., 2013). Further information on the classification of the air masses are given in the overview paper of this campaign (van Pinxteren et al., 2020)."

R#2-24) line 7. "The higher complexity in the FAA composition on the submicron aerosol particles could only be determined because the analytical method applied here is able to quantify the individual molecular FAA species." Yes, that is good, and I would encourage the authors to leverage this resolution in their data. See previous comments regarding the comparison of amino acid "profiles" for different sample types and what do the presence of each amino acid tell us.

We thank the reviewer for this comment.

In the revised manuscript version, a clearer focus has been placed on the comparison of amino acid profiles (in terms of individual amino acids and amino acid groups). Above all, this comparison is made between the individual marine compartments in section 3.4.1, 3.4.2 and 3.4.3. After introduction of the individual compartments and their FAA specifications in the individual section 3.1 (seawater), 3.2 (aerosol particles) and 3.3 (cloud water). For detailed answers to the amino acid profiles, please refer to the comments R#2-2, R#2-20, R#2-21, R#2-22 and R#2-27.

R#2-25) line 10. "The composition of FAA on the size-segregated aerosol particle samples with focus on the comparison of the submicron with the supermicron aerosol particles as well as the comparison of aerosol composition with the seawater composition will be discussed more detailed in section 3.4." Please simplify this sentence.

During the revision of the manuscript, this reference to section 3.4 was removed.

R#2-26)3.2.2 Size-segregated aerosol particles at the mountain station (MV) This section reports particulate matter (PM) masses for both the MV and CVAO stations. The Experimental section only reports particle volume measured for cloud water sampling at Mount Verde, and not at CVAO, with liquid water content (LWC) somehow derived. I am in favor of reporting what is measured directly. Here, is particle mass derived from the particle volume measurements or from the mass of filter samples (at CVAO) and cloud water (at MV) recovered? I assume there was a particle volume monitor at both CVAO and MV. I encourage plotting all particle volume data against other particle measurements like FAA. Does FAA abundance track particle volume?

We thank the reviewer for this comment. The measurement technique of the PM was added in the section 2.2.2 and it reads now (page 7, line 11-12): "The particulate mass (PM) of the aerosol particle samples was determined by weighing the filter before and after sampling (van Pinxteren et al., 2015)."

We have explicitly outlined all parameters that were measured in the aerosol particles at both, the CVAO and the MV (e.g. PM, WSOC, Na⁺ and MSA) in the Table S11 in the SI and referred to this Table in the manuscript.

We also refer to the online particle size distributions (PSND), which were also taken at both sampling stations (CVAO and MV) during the campaign. In the revised manuscript it reads as follows (page 15, line 16-19): "Additional online measurements of particle size number distributions (PSND) at the CVAO and the MV, described in Gong et al. (2020) were in good agreement with one another during cloud-free times. This indicated that, for cloud-free conditions, the aerosol particles measured at ground level represented the aerosol particles at cloud level, i.e. the aerosol particles within the marine boundary layer were well mixed."

Moreover, in the manuscript, in section 3.2.2 we compared the chemical composition of the aerosol particles at the CVAO and at the MV. It reads now (page 15, line 23-32): "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.”

page 13

R#2-27) line 24. "In cloud water samples with $\Sigma\text{FAA} < 65 \text{ ng m}^{-3}$, usually Gly was dominant followed by Ser. Cloud water samples with $\Sigma\text{FAA} > 290 \text{ ng m}^{-3}$ showed a higher complexity in the FAA composition, especially towards the end of the campaign, including the appearance of Asp." Did the relative abundances of the FAA vary, indicating different FAA profiles, or were they similar, indicating a consistent FAA profile? What was the profile of hydrophobic, hydrophilic, and neutral amino acids and how did that compare to the aerosol samples?

We thank the reviewer for this comment. Not only the FAA concentrations in cloud water varied during the campaign, but also the composition of these. Samples with a total FAA concentration of $< 65 \text{ ng m}^{-3}$ showed a low variance in composition, because Gly was dominant, followed by Ser. A higher complexity of the FAA composition could be found at FAA concentrations in cloud water with $> 290 \text{ ng m}^{-3}$. Besides Gly and Ser, Ala and Asp played an important role and Thr, Leu and Ile also contributed to the FAA complexity. Thus, it is not possible to speak about a uniform FAA profile over the whole campaign, but a certain variance was observed.

Looking at the amino acid classes in the cloud water, it can be seen that the neutral FAA were dominant in the first part of the campaign (27/09/2017-05/10/2017) and towards the end of the campaign (06/10/2017-08/10/2017) the proportion of hydrophilic amino acids increased significantly. Similar to the observations in the submicron aerosol particles at CVAO. However, here above all GABA was higher concentrated FAA in submicron aerosol particles at the CVAO. But GABA, for example, was not detected in cloud water.

A detailed comparison of the individual amino acids/amino acid classes in the individual compartments, also with regard to aerosol particles and cloud water, can be found in sections 3.4.1, 3.4.2 and 3.4.3.

In order to clarify and discuss the composition of the individual amino acids and amino acid groups in the cloud water sample, Figure 3 was extended and the hydrophilic, neutral and hydrophobic amino acid groups were clearly pointed out. The variability of FAA composition in cloud water of the individual amino acids and amino acid classes were addressed as follows in the revised manuscript (page 16, line 16 – page 17, line 4): “In cloud water samples with $\Sigma\text{FAA} < 65 \text{ ng m}^{-3}$, Gly was usually dominant, followed by Ser. However, cloud water samples with $\Sigma\text{FAA} > 290 \text{ ng m}^{-3}$ showed a higher complexity in FAA composition, including the concentrations of Asp and Ala. Other abundant FAA were Thr, Leu and Ile. In terms of the hydrophobicity 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). Comparative studies on the FAA composition of cloud water in the marine environment are lacking, but especially in the second part of the campaign, it pointed to a local marine (biogenic) influence.”

R#2-28) Were the sodium, sulfate, and MSA measurements made on cloud water also made on the aerosol samples? Other than FAA (in section 3.4) which other data are common to the two sample types - aerosol and cloud water - that would allow us to compare them?

Sodium, sulfate and MSA measurements were made for both, cloud water and aerosol particle samples. The results of the measurements of inorganic ions in cloud water are listed in Table S12 and are discussed in sections 3.3 and 3.4. For aerosol particles at the CVAO, the results of inorganic ion measurements are listed in Table S8 and are discussed in section 3.2. For the comparison

between the aerosols of both stations (CVAO and MV) regarding inorganic ions and PM, the results are listed in Table S11 and are also discussed in section 3.2.

In the manuscript we addressed this topic in section 3.3. It reads now (page 16, line 9-13): “The inorganic marine tracers in cloud water (Na^+ : $5.7 \mu\text{g m}^{-3}$, MSA: 25.1 ng m^{-3} , Table S12) were also present in higher concentrations compared to the aerosol particle samples at the CVAO (submicron: Na^+ : 72.3 ng m^{-3} , MSA: 6.0 ng m^{-3}) and the MV (submicron: Na^+ : 17.0 ng m^{-3} , MSA: 1.8 ng m^{-3} , Table S11). The concentrations of cloud water sulfate (average: $2.9 \mu\text{g m}^{-3}$, 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).”

and on page 17, line 9-12: “The presence of the marine tracers (sodium, MSA) in cloud water supports a coupling to oceanic sources. In addition, the majority of low-level clouds were formed over the ocean and ocean-derived components are expected to have some influence on cloud formation (van Pinxteren et al., 2020). Nevertheless, contributions from the desert and other non-marine sources cannot be excluded.”

And on page 2, line 2-3: “The abundance of inorganic marine tracers (sodium, methane-sulfonic acid) in cloud water suggests an influence of oceanic sources on marine clouds.”

R#2-29) page 15

line 10. "the reactivity/ mean life time _ of the amino acids" Please explain.

A definition of the mean life time was added in the manuscript. Now it reads: “The mean life time τ of the individual amino acids depends on the pH-dependent rate constant k and the OH radical concentration of the different atmospheric scenarios (SI, Eq. (3)).” (page 18, line 11-13)

Additionally in the SI (page 17) the equation for the calculation as well as a short discussion about the mean lifetime was added.

R#2-30) line 22. "The mean lifetime _ of Glu (remote aerosol case: 0.02 d" Thats 29 minutes.

Is that considered long?

Comparing the mean lifetime τ of the individual amino acids in the remote aerosol case (valid for conditions at the CVAO) listed in Table S13, it can be shown that amino acids with a mean lifetime τ of 1.20 h (Ala) and 0.48 h (Gly, Glu) have a comparatively longest lifetime τ . Whereas some amino acids have a much shorter mean lifetime with e.g. 0.007 h (Tyr) or 0.014 h (Phe). The estimation method for the mean lifetime is discussed in detail in the SI on page 17. Thus, it can be concluded that when comparing FAA under remote aerosol conditions, FAA with e.g. a mean lifetime of 0.48 h have a longer mean lifetime than most other FAA.

We have integrated the discussion of mean lifetime τ into the discussion about Gly (τ : 0.48 h), because Gly is often used as a long-range tracer with comparatively high stability (within the FAA). This can be read as follows in the revised manuscript (page 20, line 10-16): “Compared to other amino acids, Gly and Ser have a very low atmospheric reactivity (McGregor and Anastasio, 2001) and therefore a higher mean lifetime τ (Gly: 0.48 h, Ser: 0.24 h; remote aerosol case, Table S13). Due to its atmospheric stability, Gly is proposed as an indicator for long-range transport (Barbaro et al. (2015) and references therein) and has a very low atmospheric reactivity (McGregor and Anastasio, 2001). However, our results clearly show that Gly and Ser are also present in seawater to a high extend, likely resulting from the siliceous exoskeleton of diatom cell walls (e.g. Hecky et al. (1973)). Hence, besides long-range transport, a transfer from the ocean via bubble bursting might be an additional likely source of the stable, long-lived FAA in the atmosphere.”

R#2-31) line 30. "The presence of GABA on the submicron aerosol particles pointed out that (marine) microorganisms were present on the aerosol particles and produced GABA via microbiological decarboxylation of Glu." Until the authors demonstrate that GABA cannot exist outside of a (marine) microorganism, this statement is unfounded. The opposite is a safe assumption: that any compound produced by marine microorganisms will also be found in the seawater, either by active release by the living microorganism

or via release of the dead microorganism (residence times will vary).

We have carefully revised the discussion on hydrophilic FAA, including GABA. Our interpretation approach is based on previous studies regarding GABA in the marine environment (Dauwe et al., 1999; Engel et al., 2018) as already introduced in the Introduction (page 3, line 13/14). They reported GABA as an indicator for microbiological OM degradation and GABA can therefore be considered as a microbiological proxy in aerosol particles in the marine environment. Thereby GABA can be actively released by microorganisms on the aerosol particles or passively, e.g. by the death of the microorganisms. Based on these previous studies, the finding that the air masses were mainly marine (indicating non-marine sources of minor importance) and the high GABA concentrations on the submicron aerosol particles at CVAO, we have addressed the discussion in the revised manuscript as follows (page 18, line 17-33): “A conspicuous finding is the high concentration of GABA, which is present exclusively in the submicron aerosol particles (B1 and B2: 0.05-0.42 μm) at the CVAO. Despite the relatively high LOQ of GABA in seawater (Table S1), a major abundance of GABA in seawater would be detectable. GABA is a metabolic product of the decarboxylation of Glu, which has been detected in all marine compartments. Furthermore, it can be produced by microorganisms (Dhakal et al., 2012) and is considered as an indicator for the microbiological decomposition of OM (Dauwe et al., 1999; Engel et al., 2018). The abundance of GABA on the submicron aerosol particles suggests that (marine) microorganisms were present on the aerosol particles and likely produced GABA via microbiological decarboxylation of Glu. Microbial processes on marine particles have recently been reported by Malfatti et al. (2019). The authors observed a diverse array of microbial enzymes transferred from the ocean into the atmosphere with an even higher activity on the particles compared to seawater. On this basis, they hypothesized that active enzymes can dynamically influence the composition of marine aerosol particles after ejecting from the ocean. The high GABA concentrations on the aerosol particles reported here are well in line with this hypothesis. Interestingly, GABA was not detected in cloud water samples, although bacteria were found during the campaign in cloud water (van Pinxteren et al., 2020) whose presence has been reported in the literature (Jardine, 2009; Vaitilingom et al., 2013; Jiaxian et al., 2019). It remains speculative whether GABA was degraded in cloud water despite its rather long lifetime (remote cloud case: 28.8 h, Table S13) or whether it was not produced by the bacteria in cloud water.”

R#2-32) page 17

line 2. "the presence of bacteria in cloud waters has been reported in the literature (Jiixian et al., 2019)." Microorganisms have been documented in the air since at least Darwin's HMS Beagle voyages so a few more citations here would be appropriate. A few of note:

Jardine, B. Between the Beagle and the barnacle: Darwin's microscopy, 1837-1854. *Stud. Hist. Philos. Sci. Part A* 40, 382–395 (2009).

Salisbury, J. H. On the Cause of Intermittent and Remittent Fevers. *Am. J. Med. Sci.* 51–75 (1866).

M. Vaitilingom et al., Potential impact of microbial activity on the oxidant capacity and organic carbon budget in clouds. *Proceedings of the National Academy of Sciences of the United States of America.* 110, 559–564 (2013).

Following the comment of the reviewer we included more citations regarding bacteria in cloud water as recommended. Now it reads (page 18, line 29-31): “Interestingly, GABA was not detected in cloud water samples, although bacteria were found during the campaign in cloud water (van Pinxteren et al., 2020) whose presence has been reported in the literature (Jardine, 2009; Vaitilingom et al., 2013; Jiixian et al., 2019).”

R#2-33) 3.4.2 Neutral and hydrophobic amino acids How do the surface activities vary across the different amino acids? Does abundance in aerosol correlate to surface activity? Or a combination of surface activity and reactivity/lifetime?

We thank the reviewer for his comment. To consider the surface activity of each individual amino acid, the K_{OW} , the TPSA and the density as listed in Table S9 were considered. These parameters were used to study the variation of the atmospheric FAA concentration on the aerosol particles. However, these simple physico-chemical parameters could not explain this variance by statistically relevant correlations.

In addition to the surface activity parameters, the mean lifetime τ of the aerosol particles (Table S13) was also considered, as proposed, to study the variance of the FAA concentrations on the aerosol particles. However, no statistically relevant correlations were found here either. But for example, Gly, with a long mean lifetime τ , occurs dominantly on the aerosol particles. Aromatic amino acids with a shorter mean lifetime τ , were not found on the aerosol particles.

Therefore, neither the surface activity parameters nor the mean lifetime τ of the individual amino acids are suitable to explain the variance of FAA concentrations on the aerosol particles in the marine region investigated here by statistically relevant correlations.

These points were addressed as follows in the revised manuscript (page 14, line 15-18): "In addition, we considered further FAA physico-chemical parameters such as the octanol-water partition coefficient (K_{OW}), the topological polar surface area (TPSA), which describes the surface activity, and the density (Table S9) to describe the concentration changes. However, no statistically relevant correlations between the FAA concentration or composition and physico-chemical parameters were found here either."

page 18.

R#2-34) line 13. "a possible transport from other than marine sources is included in this parameter." The language is (not on purpose) vague and should clearly state that the waters measured and used for the enrichment factors have not been demonstrated to be the source of the aerosols measured.

As mentioned above (response to comment R#2-1b) we have carefully revised the discussion of the EF_{aer} , showed the uncertainties related to calculating the EF_{aer} in an open system.

R#2-35) line 16. "Regarding the transfer of OM from the ocean into ambient aerosol particles, solely organic carbon as a sum parameter has been regarded to date and no distinction of single organic matter classes for ambient measurements has been performed."

Although not written altogether clearly, this statement seems to state that compound classes nor compounds have been resolved from ambient aerosol. This is false as there is a number of studies that have accomplished this. See:

Molecular diversity of sea spray aerosol particles: Impact of ocean biology on particle composition and hygroscopicity RE Cochran, O Laskina, JV Trueblood, AD Estillore, HS Morris, ... Chem 2 (5), 655-667

Quinn, P. K., Collins, D. B., Grassian, V. H., Prather, K. A., & Bates, T. S. (2015). Chemistry and Related Properties of Freshly Emitted Sea Spray Aerosol. Chemical Reviews. American Chemical Society. <https://doi.org/10.1021/cr500713g>

As part of the restructuring of the discussion part related to the EF_{aer} (review comment R#2-1b), section 3.4.4, this sentence was removed from the manuscript

R#2-36) Figure 5. Please include in the figure the size range for each Berner stage.

Following the comment of the reviewer we included in Figure 5 the size range of each Berner stage. Figure 5 can be found on page 21.

R#2-37) page 19.

line 7. "Previous studies showed that organic material ejected into the atmosphere during bubble bursting, resulting in sea spray aerosol particles containing similar organic material to that of the SML (Russell et al. (2010); Cunliffe et al. (2013) and references

therein)." This - the basics of sea spray aerosol formation - need to be brought up later and the investigation of hydrophilic/-phobic amino acids in different particle types needs to be established in this context.

We thank the reviewer for his comment. The current state of knowledge on primary marine aerosol formation by the bubble bursting process (incl. jet and film droplets) was briefly summarized in section 3.4.4, since this section deals with a possible transfer of FAA from the ocean into the atmosphere.

It should be noted that our ambient investigations cannot provide detailed mechanistic investigations of the bubble transfer. For this purpose, tank experiments under controlled conditions are necessary (that are currently performed in our group).

However, due to the FAA composition in the SML and on the submicron aerosol particles and the similar percentage contribution of FAA to DOC and WSOC in these two compartments, we suggested that film droplets contributed to the transfer of FAA.

The summary of the state of knowledge on primary marine aerosol formation via bubble bursting and the proposed interpretation of our observation is now included on page 22, line 10-19, and reads as follows in the revised manuscript: "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). Especially the film droplets have been reported to be enriched in OM and are suggested to transfer OM from the SML onto submicron aerosol particles (Wilson et al., 2015). The supermicron aerosol particles tend to form from the larger jet droplets and thus represent the ULW composition (Blanchard, 1975; Wilson et al., 2015). We cannot derive mechanistic transfer characterizations from the ambient measurements performed here. Nevertheless, the constant FAA enrichment in the SML together with the strong FAA enrichment in the submicron aerosol particles strongly suggest that film droplets form the submicron particles. However, Wang et al. (2017) showed that jet drops (which transfer OM from the ULW) also have the potential to contribute significantly to the formation of submicron sea spray aerosol particles, so, jet droplets can also contribute to FAA formation."

R#2-38) line 29. "In situ-formation of FAA in cloud water, maybe due to biogenic formation or enzymatic degradation of proteins, selective enrichment processes as well as pH dependent chemical reactions might be potential sources." site Malfatti, F., Lee, C., Tinta, T., Pendergraft, M. A., Celussi, M., Zhou, Y., : : : Prather, K. A. (2019). Detection of Active Microbial Enzymes in Nascent Sea Spray Aerosol: Implications for Atmospheric Chemistry and Climate. *Environmental Science and Technology Letters*, 6(3), 171–177. <https://doi.org/10.1021/acs.estlett.8b00699>

We thank the reviewer for pointing out this interesting paper. In fact, the results, suggesting microbial activity on aerosol particles, shown by the transfer of enzymes, fits well with the observations made here and might explain the abundance of GABA on the aerosol particles. Please also note our reply to R#2-31.

We included this reference in the revised manuscript in the following context:

"The abundance of GABA on the submicron aerosol particles suggests that (marine) microorganisms were present on the aerosol particles and likely produced GABA via microbiological decarboxylation of Glu. Microbial processes on marine particles have recently been reported by Malfatti et al. (2019)." (page 18, line 23-25)

and "Altogether, the in-situ formation of FAA in cloud water by chemical processes in the cloud or by atmospheric biogenic formation or enzymatic degradation of proteins, as proposed by Malfatti et al. (2019), as well as by selective enrichment processes and pH dependent chemical reactions might be potential sources." (page 17, line 14-17)

Technical Corrections

Introduction.

R#2-39) page 2. line 8. "surface global ocean" to "global surface ocean" line 16. "utilizable sources of nitrogen" to "utilizable FORMS of nitrogen"

Due to the revision of the Introduction and also following the suggestions from reviewer 2, we focused stronger on the FAA from the beginning and therefore general parts (as the one mentioned by the reviewer) were deleted from the manuscript.

On page 2, line 24-26 the suggested correction was implemented. Now it reads: "Despite their attribution to proteins the FAAs are better utilizable forms of nitrogen instead of proteins for an aquatic organism such as phytoplankton and bacteria (Antia et al., 1991;McGregor and Anastasio, 2001)."

R#2-40) page 3. line 31. "underline seawater" to "underlying seawater"

Following the commentary of reviewer 1 (R#1-4), we have used the abbreviations in the manuscript continuously after the introduction: This sentence (page 3, line 31-32) reads now: "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)."

R#2-41) page 4. line 4. "as proxies or tracer" to "as proxies or tracerS"

Following the comment of the reviewer we changed the sentence on page 4, line 4/5. And it reads now: "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#2-42) In the Experimental section perhaps change "analytics" to "analyses"

We agree with the comment of the reviewer and changed in the experimental section "analytics" to analyses, e.g. page 5, line 25/26: "2.2 Analyses" and "2.2.1 Seawater analyses"; page 6, line 25 "2.2.2 Aerosol particle filter analyses" and page 7, line 19: "2.2.3 Cloud water analyses".

R#2-43) 2.2.1 Seawater sample analytics Was the standard addition method applied to samples to assess for recovery efficiency of the entire process?

The standard addition method was also used to evaluate the recovery efficiency in the development of desalination methods and analytical measurement methods. The recovery efficiency stated here is based on the fact that a defined concentration of amino acids was added to a seawater sample prepared from milli-Q water using sea salt standard (Sigma-Aldrich, Germany) and this sample was treated using the same procedure as the real seawater samples. The FAA concentration in the seawater sample was then determined after sample preparation. To consider the recovery efficiency, the percentage of the measured concentrations to the added FAA concentration was calculated. The recovery rate thus determined is consistent with tests using standard addition methods and proved to be less complex and more practical.

R#2-44) page 6. line 23. "All here presented values" to "All values presented here" line 29.

Following the comment of the reviewer we changed the sentence on page 7, line 4 to "All values presented here for aerosol particle samples are field blank corrected."

R#2-45) "The cloud water samples were operated the same as seawater samples". change "operated" to "handled" or "processed".

We agree with the reviewer's comment and changed the sentence on page 7, line 20/21 to “The cloud water samples were processed the same as seawater samples for the analysis of DOC/TDN and inorganic ions (section 2.2.1).”

R#2-46) line 31. "syringe filters filters" to "syringe filters" or "syringe tip filters"

We agree with the reviewer's comment and changed the sentence to “After the filtration with 0.2 μm syringe filters (Acrodisc-GHP; 25 mm, Pall Corporation, New York, USA), an aliquot of the prepared cloud water was derivatized based on the AccQ-TagTM precolumn derivatization method (Waters, Eschborn, Germany).” (page 7, line 22-24)

R#2-47) page 10. line 12. Remove "It is obvious that" as it is confusing language.

Following the comment of the reviewer we removed “it is obvious that”. Now it reads (page 12, line 4-7): “Whilst the concentration ΣFAA 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 ΣFAA 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#2-48) line 14-15.

Reword. Use "neither" instead of "both".

Following the comment of the reviewer we changed “both” to “neither”. Now it reads (page 12, line 33/34): “ ΣFAA included all investigated amino acids (listed in 2.2.1) except for Met and Gln, analytes which were neither detected in the size-segregated aerosol particle samples.”

R#2-49) page 12 line 32. "aerosol particles" is redundant. aerosols are particles. say just "aerosol" or "particle", here and elsewhere.

Since the term "aerosol" refers to the entirety of a gas with the particles suspended in it, we would prefer to use the term "aerosol particles" in the manuscript and in the SI. The term “aerosol particle” is also common in other publications, for example (Cochran et al., 2017; Forestieri et al., 2016; Frossard et al., 2019; Koulouri et al., 2008).

R#2-50) page 17. line 7. "Neutral amino acids" to "acids"

We changed the typo. Now it reads (page 20, line 8/9): “Neutral amino acids were generally the amino acid group with the highest concentration in all investigated marine compartments, accounting for more than 50% of the FAA total (Fig. 4a-d).”

R#2-51) line 10. "A further explanation approach" remove "approach"

Following the comment of the reviewer we restructured the sentence. Now it reads (page 20, line 15/16): “Hence, besides long-range transport, a transfer from the ocean via bubble bursting might be an additional likely source of the stable, long-lived FAA in the atmosphere.”

Additional changes performed by the authors

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).

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 23, line 25-26)

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 14/15)

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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