

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

### **Major comments**

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

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

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

Once introduced, abbreviations should be used further throughout the text.

### **Specific and technical comments**

#### **Page 2**

L23 Skip ...also..; it is confusing considering the previous paragraph

#### **Page 3**

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

#### **Page 4**

L1-3 Repetition with the L27-34; I suggest to skip it or rewrite the overall paragraph to avoid repetition.

L14-15 Unclear/confusing sentence, please rewrite

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

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

L20: Please correct ..were taken *from* a fishing boat...

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

#### **Page 5**

L16 I suggest to use the FAA abbreviation throughout the text

L22 Add...include *determination of glycine* (Gly)...

L34 Unclear sentence part (...in 0.1 min back to 5 % B and...); please rewrite

L34 Please correct: ...This *analytics OR analytical procedure* can be used...

#### **Page 6**

L2-5 Unclear/confusing sentence, please rewrite

L6 Introduce the LOQ abbreviation here instead within L24

L13 Please add the method used for the trace metal determination

L13 Remove a space before -total-

L23 Rewrite to be clear that data obtained for the aerosol samples were blank corrected

#### **Page 7**

Please explain the calculation of  $EF_{SML}$  based on  $LOQ/2$  estimation (Figure 1) in the case of the missing ULW data within the 2.2.4 Section.

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

## Page 8

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

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

## Page 9

L2 Add ...Mediterranean Sea *of* 0.07-0.60  $\mu\text{mol L}^{-1}$ ...

L4-6 Add .... in the SML or in the ULW *since* no correlation between...

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

## Page 10

L16 Add ...of dissolved FAA ( $PM_{10}$ ; sum of B1-5)...

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

## Page 12

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

L31-32 Unnecessary sentence, repetition

## Page 13

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

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

L28-32 to P14 L4-8 This paragraph deals with the possible dominant origin of FAA in the cloud water collected at the MV as within the 3.4.5. Section. I suggest skipping this paragraph

and combining the cloud water source discussion within the 3.4.5. Section to avoid the repetition.

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

### **Page 15**

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

### **Page 17**

Although beyond the scope of this paper, I suggest discussing the potential connection of hydrophilic, hydrophobic and neutral FFA to CCN activity.

### **Page 18**

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

L14 remove -as-

### **Page 17**

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

### **Page 19**

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

### **SI material**

Page 2, L17 ..and *is* discussed in more...