Replies to Referee 2:

The manuscript presents an overview of the MarParCloud campaign at Cape Verde Islands in Sept – Oct 2017. Several interesting new scientific findings are reported in brief or just mentioned, but the main scope here is to present a synopsis of all oceanographic and atmospheric observations that have been carried out during the field campaign. If the Authors' intention was to give a flavour of a very multidisciplinary experiment, I think this emerges quite clearly from the paper. I have only two major remarks about the science (see my major comments below). Besides, since most of the results are object of specific papers in preparation, my remarks are mainly on the quality of the presentation.

We thank the Reviewer for the evaluation and the constructive comments. Replies to the specific Referee's comments are provided below in red and new parts included in the manuscript are marked in *italics*:

Major comments:

1. The Abstract is rather descriptive but it ends up with a strong statement: "from a perspective of particle number concentrations, marine contributions to both CCN and INP are rather limited". I think that the Authors here should make an effort to a) be more quantitative (or refer to other papers either published or in preparation) and b) make sure that there is enough information in the Abstract as well as in the main body of the manuscript to support such conclusion. I have not found any sections in the paper dealing with CCN, just a brief mention at line 560 focusing on the proportion of CCN at low supersaturations accounted for by coarse particles, but what about the estimated contribution by primary marine aerosols?

We agree that the CCN results and the respective marine contributions were not discussed in sufficient detail and we extended this part. Additional information on CCN was added to Chapter 5.1.1., including also a new Figure (Fig. 9) with CCN number concentrations for a range of super-saturations and for dust and marine air masses. In addition, the fraction of sea spray particles in all CCN and in all particles is now given. The respective text in the second paragraph of Chapter 5.1.1. now reads as follows:

"N_{CCN} at different supersaturations were compared during dust and marine periods, as shown in Figure 9. During dust periods, the aerosol particles show a great enhancement in Aitken, accumulation and coarse mode number concentrations, such that overall N_{CCN} increases distinctly. N_{CCN} at a supersaturation of 0.30% (proxy for the supersaturation encountered in clouds present during the campaign) during the strongest observed dust periods is about 2.5 times higher than that during marine periods. As suggested by Modini, et al. (2015), Wex, et al. (2016) and Quinn, et al (2017), the coarse mode aerosol particles can be attributed to sea spray aerosol (SSA) in a marine environment. In these studies, the fraction of sea spray aerosol was determined based on three-modal fits from which the particle number concentrations in the different modes were determined. A similar analysis was done for this study. During marine periods, SSA accounted for about 3.7% of CCN number concentrations at 0.30% supersaturation and for 1.1% to 4.4% of N_{total} (total particle number concentration). The hygroscopicity parameter kappa (κ) averaged 0.28, suggesting the presence of OM in the particles."

In addition, in order to be more quantitative, we included results from a study of free amino acids in all marine compartments (seawater, aerosol particles, cloud water) that was very preliminary at the time of writing the first version of the current manuscript but are currently more advanced and published at ACPD (Triesch, et al. 2020) within this SI. We referred to the results that are discussed in the separate paper but included the main findings in the overview paper in section 5.7.1 and it reads: *" A more comprehensive set of samples was analysed for FAA on molecular level as important organic nitrogen- containing compounds (Triesch, et al. 2020). The FAA, likely resulting from the ocean, were strongly enriched in the submicron aerosol particles ($EF_{aer\ (FAA)}$ 10^2-10^4) and to a lower extent enriched in the supermicron aerosol particles ($EF_{aer\ (FAA)}$ 10^1). The cloud water contained the FAA in significantly higher concentrations compared to their respective seawater concentrations and they were enriched by a factor of $4 \cdot 10^3$ compared to the SML. These high concentrations cannot be currently explained and possible sources such as biogenic formation or enzymatic degradation of proteins, selective enrichment processes or pH dependent chemical reactions are subject to future work. The presence of high concentrations of FAA in submicron aerosol particles and in cloud water together with the presence of inorganic marine tracers (sodium, methane sulfonic acid) point to an influence of oceanic sources on the local clouds (Triesch, et al. 2020)."*

This further shows a (qualitative) link between ocean-derived compounds being transferred to the atmosphere up to cloud level. In the abstract, such information had been added before but are specified now and it reads "*Organic nitrogen compounds (free amino acids) were enriched by several orders of magnitude in submicron aerosol particles and in cloud water compared to seawater*."

With these modifications (and the upcoming ones about INP, see 2.) we included more solid facts in the manuscript and we think there is now enough information in the abstract as well as in the main body of the manuscript to support our conclusion. Finally, we want to point out that this manuscript is intended to provide an overview about the MarParCloud campaign and give a basis and orientation to the single papers on the specific topics of MarParCloud that are partly under revision and partly being currently finalized.

2. There is instead a section about INP (5.7.4) providing a short summary of the study of Gong et al. (2019b) and concluding that primary marine INP should be four orders of magnitude more abundant to account for the ambient INP concentrations measured in cloud and aerosol samples. If this is the basis for the final statement included in the Abstract, I suggest to report it along with the main hypothesis made by the Authors who assume "INP not enriched or altered during the production of sea spray" from the SML (lines 966 – 967), which is a strong assumption, in my opinion. Otherwise, more supporting information can be extracted from Gong et al..

We agree with the referee that a more comprehensive discussion about the INP findings is needed and thus more details and explanations on the INP analysis were added as follows:

In the main text (section 5.7.4.):

"N_{INP} in PM_1 were generally lower than those in PM10 and, furthermore, N_{INP} in PM_1 at CVAO did not show elevated N_{INP} at warm temperatures, in contrast to N_{INP} in PM_{10}. These elevated concentrations

in PM_{10} decreased upon heating the samples, clearly pointing to a biogenic origin of these INP. Therefore, ice active particles in general and biologically active INP in particular were mainly present in the supermicron particles, and particles in this size range are not suggested to undergo strong enrichment of OM during oceanic transfer via bubble bursting (Quinn et al., 2015 and refs. therein). "

Furthermore, we pointed out that INP are assumingly not enriched or altered in the supermicron mode during the production of sea spray as follows:

"Assuming sea salt and the INP to be similarly distributed in both sea and cloud water (i.e., assuming that INP would not be enriched or altered during the production of *supermicron* sea spray *particles*), N_{INP} is at least four orders of magnitude higher than what would be expected if all airborne INP would originate from sea spray."

We performed changes and clarifications concerning the INP measurements in the abstract:

"However, INP measurements indicated also a significant contribution of other non-marine sources to the local INP concentration, *as (biologically active) INP were mainly present in supermicron aerosol particles that are not suggested to undergo strong enrichment during ocean-atmosphere transfer."*

And in the conclusion:

"However*, based on the findings that (biologically active) INP were mainly present in supermicron aerosol particles that are not suggested to undergo strong enrichment during ocean-atmosphere transfer as well as the INP abundance in seawater and in cloud water*, other non-marine sources most likely significantly contributed to the local INP concentration."

Specific comments:

1. Three of the seven research questions (page 5) should be retuned:

a. Question #1: specify what are the metrics of interest (number, mass, CCN, etc.).

We included the specific metrics of interest and it now reads:

- To what extent is seawater a source of OM on aerosol particles (regarding number, mass, chemical composition, CCN and INP concentration) and in cloud water?

b. Question #2: do "OM groups" mean source contributions or chemically-defined classes?

We mean chemically defined classes and changed it to:

- What are the important chemically-defined OM groups (proteins, lipids, carbohydrates - as sum parameters and on molecular level) in oceanic surface films, aerosol particles and cloud water and how are they linked?

c. Question #3: this reads like a rhetorical question; it should be restructured into something like "What are the main biological and physical factors responsible for the occurrence and accumulation of OM etc."

We changed question #3 to:

- What are the main biological and physical factors responsible for the occurrence and accumulation of OM in the surface film and in other marine compartments (aerosol particles, cloud water)?

2. Line 225: Hg cannot be considered a good “example for trace metals”, as it exhibits unique chemical properties.

We agree that this sentence was not well phased and replaced it with: “Ocean surface mercury (Hg) associated with OM was investigated.”

3. Line 226: “pigments [...] were captured..”, I do not think the verb is appropriate.

We agree and replaced “captured” by “analysed”.

4. Line 227: “...(DMS), VOCs..”; as DMS is a VOC, this should be better put as “..(DMS), other VOCs..”.

We changed the sentence as suggested.

5. Line 439: “These issues will be further analysed”, does it mean elsewhere in the paper or in a future publication? Please specify.

We specified it and it now reads: “These issues will be analysed in further studies. “

6. Section 4.1.4: What is the relevance/representativeness of the five cloud scenes?

The different cloud times have shown to affect the in-cloud time of an air parcel that in turn affects in-cloud chemical processes (e.g. Lelieveld and Crutzen, 1991). For example, stratocumulus clouds is the time an air parcel spents in a cloud higher (at low mixing) compared to the in-cloud time of an air parcel in cumuli clouds. Moreover, it has been shown that the formation of MSA is enhanced when strong in-cloud processing occurs. We have added these considerations and therefore the relevance together with the respective references in a concluding sentence in chapter 4.1.4 and it now reads:

“The different cloud scenes reflect typical situations observed in conditions with either weaker or stronger winds. The average in-cloud time of an air parcel might depend on cloud type and cloud cover that in turn impacts in-cloud chemical processes (e.g. Lelieveld and Crutzen, 1991), such as the formation of methane-sulfonic acid and other organic acids (Hoffmann et al., 2016 und Chen et al., 2018). Future studies will relate the chemical composition of the aerosol particles and cloud water to the cloud scenes and their respective oxidation capacity. However, the rather coarse horizontal resolution of the satellite sensor and the missing information about time-resolved vertical profiles of thermodynamics and cloud condensate limits a further detailed characterization of these low-level cloud fields and their formation processes. A synergistic combination with ground-based in-situ and remote sensing measurements would be highly beneficial for future investigations *to elucidate how cloud chemistry might be different for the varying cloud scenes depending on horizontal cloud patterns and vertical cloud structures.”*

7. Lines 478 – 480: “A synergic combination with ground-based in-situ and remote sensing measurements would be highly beneficial for future investigations”. The sense of this sentence is rather obscure (“beneficial” for what? For which of the seven re-search questions listed in the introduction??), please clarify.

As mentioned above, we have changed the statement and added: “A synergistic combination with ground-based in-situ and remote sensing measurements would be highly beneficial for future

investigations *to elucidate how cloud chemistry might be different for the varying cloud scenes depending on horizontal cloud patterns and vertical cloud structures."*

The cloud types implicitly belong to these two (revised) research questions:

- To what extent is seawater a source of OM to aerosol particles (regarding number, mass, chemical composition, CCN and INP concentration) and in cloud water?

- What are the important chemically-defined OM groups (proteins, lipids, carbohydrates - as sum parameters and on molecular level) in oceanic surface films, aerosol particles and cloud water and how are they linked?

Although studies of in-cloud chemistry in relation to the cloud scenes were not the aim of the campaign, the differentiation between the different cloud scenes provides a first step for such analysis in the future and are in our opinion worth to be shown here as auxiliary information.

8. Section 4.2. I suggest to report here only the concentration levels of the main indexes of biological productivity (chlorophyll concentrations) and their spatial distribution (sub-section 4.2.2 is ok), while I would postpone the discussion about pigment distribution to section 5.4.

We thank the referee for this suggestion. We understood that the order of presentation may lead to some confusion and have re-structured this part (following also the suggestions of reviewer #1). In the revised version, we mentioned the chl-a concentrations in section 4.2. and shifted the discussion of the pigment results to section 5.4.1. To underline that pigments as well as bacteria were analysed we:

i) renamed section 4.2.1 to "Pigment and bacteria concentration in seawater"

ii) included this information in section 4.2.1 and it now reads " *Chl-a concentrations varied between 0.11 µg L^{-1} and 0.6 µg L^{-1}, and are more thoroughly discussed together with the pigment composition in section 5.4.1. Moreover, as other but phytoplankton organisms can contribute to the OM pool, bacterial abundance was analysed in the SML and bulk water samples and these data are reported in section 5.7.3*."

iii) concluded section 5.4.1. with: " First analyses show that the DOC concentrations were not directly linked to the increasing chl-*a* concentrations, however their relation to single pigments, *to the microbial abundance*, to the background dust concentrations and finally to wind speed and solar radiation will be further resolved to elucidate potential biological and meteorological controls on the concentration and enrichment of DOC."

Due to the inclusion of the pigment discussion in chapter 5.4.1. (that therefore became very long) we have added another subsection for the comparison of DOC data from the two sampling techniques (5.4.2 DOC concentrations: manual glass plate vs. MarParCat sampling) in 5.4.2.

9. Line 535: satellite fluorescence measurements. Which satellite?

They were achieved from the MODIS-Terra satellite, this is stated in the Figure caption and we also added this information in the text.

10. Section 5.1.1: see major comments.

We included CCN information and discussion as described above in 1).

11. Lines 602-603: "..suggested an ocean influence on cloud water". Please, be more precise here. The data show a cloud water composition dominated by seasalt with little quantities of other solutes: this is simply the effect of the larger scavenging efficiency of coarse particles with respect to submicron ones, but it is worth reminding that this picture is "mass-based" (all cloud drops coalesce into one single sample inside the CASCC) while in terms of number concentrations (how many cloud drops originated from marine sources), it is difficult to tell solely on the basis of the data shown in Fig.11.

We thank the reviewer for these comments and fully agree that the mass vs. number discussion was not clearly addressed. We accordingly included the suggestions in Chapter 5.1.2 and it now reads:

"These compounds were also observed in the coarse fraction of the aerosol particles, *suggesting* that the coarse mode particles served as efficient CCN and were efficiently transferred to the cloud water. *To emphasize, these chemical analyses are based on mass, but the control of the cloud droplet number concentration comes from CCN number concentrations, including all particles with sizes of roughly above 100 nm. As larger particles contribute more to the total mass, chemical bulk measurements give no information about a direct influence of sea spray particles on cloud droplet concentrations, but it can show that the chemical composition is consistent with an (expected) oceanic influence on cloud water."*

And at the end of the paragraph:

"In summary, cloud water chemical composition seemed to be *dominated* by coarse mode aerosol particle composition, and the presence of inorganic marine tracers (sodium, methane sulfonic acid) *shows that material from the ocean is transported to the atmosphere where it can become immersed in cloud droplets. More detailed investigations on the chemical composition, including comparison of constituents from submicron aerosol particles and the SML with the cloud water composition are planned."*

Another aspect that suggests an influence of marine-derived particles on cloud processes is the finding of TEP (particles that are clearly of marine origin), in submicron aerosol particles. While these are first results, the occurrence of these ocean-derived compounds in very small particles might be related to cloud processes that will be investigated in future studies.

We have mentioned this in chapter 5.7.2 and underlined it more clearly:

"Interestingly, a major part of TEP seems to be located in the sub-micron aerosol particles (Fig. 19). Sub-micron aerosol particles represent the longest living aerosol particle fraction and have a high probability to reach cloud level and *to contribute to cloud formation* and the occurrence of TEP in cloud water, which strongly underlines a possible vertical transport of these ocean-derived compounds."

Finally, we included the particle mass/number issue in the conclusion as follows:

"We clearly see a link between the ocean and the atmosphere as (i) the particles measured at the surface are well mixed within the marine boundary layer up to cloud level and (ii) ocean-derived compounds can be found in the (*submicron*) aerosol particles at mountain height and in the cloud

water. The organic measurements will be implemented in a new source function for the oceanic emission of OM. From a perspective of particle number concentrations, the marine contributions to both CCN and INP are, however, rather limited. *These findings underline that further in depth studies differentiating between the aerosol number and aerosol mass are strongly required."*

12. Line 644: ".. by southern Hemisphere": it is not clearly shown in Figure S1.

Fig. S1 shows the air mass back trajectories and from the plots, it seems that the air masses partly passed the Southern Hemisphere. However, this is subjective information and from the wind direction plot, this was not confirmed. We agree with the reviewer that "by the southern Hemisphere" could not clearly been shown and therefore we have revised this part. It now reads: "High DMS concentrations on September 19^{th} – 20^{th} occurred when air originated predominantly from the Mauritanian upwelling region (Figure SI1) and on September 26^{th} and 27^{th}." As furthermore stated, "These elevated concentrations will be linked to the phytoplankton composition …" to elucidate further biological connections.

13. Section 5.3.2: The results about HONO look so preliminary that I am not sure they deserve a dedicated section. They can be reduced to a short paragraph at the end of the previous section about trace gases.

We agree and included the HONO discussion in the trace gas section 5.3.

14. Line 850: Enrichment factors for DOM are reported. Please specify the concentration unit used for DOM (organic carbon, organic nitrogen or UV absorption?).

The DOM comprises the sum of the single DOM fractions (in µg/L) and the EF is the (dimensionless) ratio between the DOC/DOM in the SML and the ULW. We added this information in chapter 5.7.1 and it now reads: *"The DOM concentrations were derived from the sum of the individual compound groups (in µg L^{-1})* and the EFs for DOM varied from 0.83 to 1.46, which agreed very well to the DOC measurements described in section 5.4.1."

15. Lines 987 – 988: ".. a daily variation of the number of particles formed was observed (but from a very limited set of samples, n = 3) probably related to the daily sampling conditions. To explain these observations, two different hypothesis can be postulated..." Actually, it is not clear at all what observations the Authors are referring to, because no data are shown but simply a "daily variation" is observed and only for three samples. Similarly to the HONO case, the impression is that the state of the analysis of the Go:PAM dataset is just too preliminary to be discussed in a dedicated section.

We thank the reviewer for this comment. In fact, the results were very preliminary at the time of writing the first version of the current paper. However, the data interpretation has strongly proceeded in the meantime and the data are included and discussed in a separate paper by Zabalegui, et al., 2019. In the revised version, we refered to this paper (that is also published within this SI) and shortly summarized the main findings and it reads now: "A subset of the collected SML samples were investigated within the Go:PAM *and showed that particles were formed when these samples were exposed to actinic irradiation. These particles resulted most likely from the reaction of ozone with gaseous products that were released from the SML as shown recently (Ciuraru et al. 2015)*

and the results obtained herein are explained in more detail in a separate paper by Zabalegui, et al., 2019. "

16. Section 5.9.1. Please provide a short description of COSMO.

We included a short description of COSMO and it now reads: "*COSMO is a compressible and non-hydrostatic meteorological model and the current weather forecast model of the German Weather Service. The numerical calculation of the weather forecast is achieved by using information of the underlying orography and land-use, as well as boundary data of all meteorological fields. The needed boundary and initial fields will be derived from re-analysis-data and/or input parameters from coarse-resolved weather model data."*

17. Section 5.9.2. Please provide a short description of MUSCAT.

We included a short description of MUSCAT and it now reads: *"The new emission scheme will be implemented to the aerosol chemical transport model MUSCAT (Multi-Scale Chemistry Aerosol Transport). MUSCAT is able to treat atmospheric transport and chemical transformation of different traces gases as well as particle properties. In addition to advection and turbulent diffusion, sedimentation, dry and wet deposition through the transport processes are considered, too. MUSCAT is coupled with COSMO that provides MUSCAT with all needed meteorological fields (Wolke, et al. 2004). The multiscale model system COSMO-MUSCAT will be used further to validate the emission scheme of OM via small and meso-scale simulations. "*

18. The graphical quality of the figures must be improved.

The quality of the Figures appears partly poor due to the "copy and paste" procedure to the pdf document. We will provide the graphics in highest resolution and will upload them as a separate file.

19. Figure S4: there is something wrong in this figure

We thank the reviewer for this note and corrected the Figure S4. It now clearly shows the cloud events (according to the method of Gong e al. 2019a).

Additional information: Data availability: We uploaded out data on World Data Centre PANGAEA (https:\\ww.pangaea.de/) and included the respective DOIs in the revised manuscript.