

Referee 2:

In the paper the authors report GLY and MGLY concentrations on Atlantic aerosol particles and the seawater. This is an interesting work for atmospheric scientists since GLY and MGLY play important roles in atmospheric chemistry. There are two issues which the authors may pay more attention to revising. Firstly, the scientific significance of the paper is not expressed in the introduction. We cannot know why the authors conducted this campaign work. If just to know the concentration of GLY and MGLY in the seawater and in sea ambient air, the significance is not enough.

Response: *We agree to the reviewer that the aim of the work was not sufficiently expressed. Therefore, we have changed the last part of the introduction and it now reads:*

“As the open ocean and marine interactions are suggested to be an important source for the alpha-dicarbonyls in the atmosphere and because of the few available field measurements in the marine area the aim of this work was to analyze GLY and MGLY in marine samples from the Atlantic Ocean. Firstly the easy-to-use approach of PFBHA derivatisation combined with solvent extraction and GC-MS analysis was validated for the seawater and aerosol samples and secondly the method was applied to the field samples collected during a transatlantic Polarstern cruise in spring 2011. In seawater the focus was on the SML, as it is the direct interface between ocean and atmosphere. Connections of GLY/MGLY in the SML and on aerosol particles to environmental parameters such as global radiation, temperature, distance to the coastline and biological activity should help to find hints regarding the production of these compounds and a connection between these carbonyls in the ocean and in the atmosphere.”

Secondly, the conclusion based on the correlations between dicarbonyl concentrations in aerosol and in seawater is doubtful or at least not plausible since the sampling sites were different. So the section of '3.3.3' may be deleted or rewritten.

Response: *The sampling of seawater (SML and bulkwater) and aerosol particles was indeed partly co-located, probably this was not clearly worked out in the manuscript. We now underlined this fact by the re-phrased sentence:*

“The green crosses present the sampling points where oceanic water sampling and clean marine aerosol sampling was co-located, meaning that SML samples were taken in the midterm of aerosol sampling.” (chapter 2.1)

and

“Within the Polarstern Cruise there were six sampling dates when seawater and clean marine aerosol particles sampling was co-located, meaning that SML sampling was in the midterm of the 24 hours-aerosol sampling time.” (chapter 3.3.3).

As in the literature the question of a sea-air transfer of GLY was raised, we wanted to show the correlation between the two carbonyls in the SML and in the aerosol particles. We agree that a direct correlation cannot be concluded and the dataset is limited, these results can give a first basis for further studies regarding air-sea interaction of the alpha dicarbonyls.

The following are some detailed comments. Line 23-24 page 15302, why not the interaction of MGLY?

Response: *We mentioned the hints for interactions of both GLY and of MGLY between ocean and atmosphere. However we put an emphasis on GLY because the correlation was slightly more pronounced for GLY as described in the text (chapter 3.3.3). At the moment we cannot state reasons for the stronger correlation of GLY compared to MGLY. This might be due to the limited data set and will be subject of further investigations.*

Line 3 page 15303, this reference is old and cannot imply the current situation.

Response: *We replaced this reference with an actual review paper from Cunliffe et al. (2013) that focuses on the sea surface microlayer and also covers ocean – atmosphere interactions.*

Line 6-15 page 15305, the environmental significance of the paper is not clearly expressed.

Response: *This part is changed now (see first comment) and the significance of this work is now more clearly expressed at the end of the introduction part.*

3.1 and 3.2 sections can be put into experimental section other than 'results and discussion'.

Response: One part of this research work was verifying the suitability of the PFBHA derivatisation-solvent extraction – GC/MS for the two alpha dicarbonyls in complex real samples such as seawater and aerosol samples. Therefore, we feel that the validation procedure, such as testing the influence of matrix effects by the standard addition method, is an important part of the results. Also the comparison of the method with other studies, regarding for example the limits of detection, might be more suitable in the “results and discussion” part than in the experimental.

Line 17 page 15311, give the complete name for the abbreviated name of ‘chl a’.

Response: The abbreviation “Chl-a” is explained and used throughout the manuscript.

Section of ‘GLY/MGLY concentrations on aerosol particles’ page 15313, the authors are suggested to use a table to list the aerosol GLY and MGLY concentrations other than text description.

Response: A Table with the concentrations of GLY and MGLY is added in the Supplementary material, furthermore the concentrations are mentioned in the text (chapter 3.3.2).

Line 17 page 15314, give the completed name of DOC.

Response: The term “DOC” – dissolved organic carbon – is explained by the first use and then the abbreviation DOC is used through the text.

Line 7-8 page 15316, Can the authors explain how the biological activity act as a sink for GLY and MGLY?

Response: The role of microorganism regarding carbonyl compounds in seawater was summarized by Zhou and Mopper (1997) that we cited in the manuscript. They found that microorganism take up carbonyls under oxic conditions and release them under anaerobic conditions. Since the sea surface is well oxygenated Zhou and Mopper concluded that anaerobic activity is inhibited and therefore they assume that microbial activity probably represents a net sink rather than a net source for carbonyl compounds in the sea surface microlayer.

In our study we don’t see a correlation between the alpha dicarbonyls and chl-a in seawater.

However, we see a negative correlation between chl-a and the concentrations of the alpha dicarbonyls (and oxalate) in the aerosol particles.

As there are strong interaction of GLY and MGLY between gas phase and the aerosol phase, this could be pointing to the fact that at higher biological activity the gas phase chemistry is changed in the direction that the alpha dicarbonyls are less produced or stronger degraded. This point was also raised by Referee 1 and is now added in the manuscript and reads:

“In contrast to seawater, where no connection to Chl-a was found, a negative correlation of GLY and MGLY to Chl-a was observed for GLY ($R^2 = 0.54$, Fig. S24) and less pronounced for MGLY ($R^2 = 0.28$, $n=6$, Fig. S25). Also oxalic acid on aerosol particles showed a slightly negative correlation to Chl-a ($R^2 = 0.28$, $n=6$, Fig. S26). Miyazaki et al. (2010) also observed increased concentrations for GLY on marine aerosol particles in times of lower biological activity, while MGLY concentrations were hardly affected. One possible explanation for the diminished concentrations of GLY and MGLY in the aerosol particles without changes in seawater concentrations could be changes in the gas phase chemistry depending on biological activity. Biological activity could affect the concentrations of the carbonyls and their precursors in the gas phase leading to a reduced production and/or stronger degradation of the alpha dicarbonyls and affecting their partitioning between gas and aerosol phase. However due to the limited dataset and the lack of gas phase measurements clearly more research is needed on this subject.”

Line 15-16 page 15316, this sentence is confusing in grammar and rewrite it.

Response: We rephrased this sentence to “Within the Polarstern Cruise there were six sampling dates when seawater and clean marine aerosol particles sampling was co-located, meaning that SML sampling was in the midterm of the 24 hours aerosol sampling time. These sampling dates are illustrated with a green cross in Figure 1.”

Line 18-21 page 15316, the scientific significances of correlation between α -dicarbonyls in the SML and in aerosol are doubtful.

Line 7-8 page 15317, the conclusion is doubtful based on the above phenomenon

Response: We agree that it is uncertain to conclude a direct correlation between our compounds in the SML and aerosol only based on the same trend of the measured concentrations.

However, as we do see this slight correlation between our compounds in the SML and in the atmosphere we slightly suggested a possible hint for interaction between the SML and in the atmosphere. Though we pointed out that no mechanisms can be concluded and definitely more research is needed on this subject.

As mentioned earlier, in the revised version we stronger underlined the fact that aerosol and SML sampling were co-located regarding the data where we found these correlations.

Line 26-28 page 15316, this sentence looks useless in the context.

Response: We think that the information that bubble bursting processes are not limited to surface active compounds but are also found for non-surface active organics it important in this context as GLY and MGLY are rather non-surface active and bubble bursting could be a transfer way of these carbonyls to the atmosphere as well.

Table 1, what is the 'local sampling time'? for seawater sampling or for aerosol sampling? More detailed sampling time should be given in the table.

Response: Table 1 lists the parameter for seawater sampling. This information is now added in the Table heading.

Table 2, the caption is too simply to readers. More detailed information on description of data should be added.

Response: The caption of Table 2 is extended, the detailed information on how the data were achieved can be found in the text (chapter 3.2)

Table 3, the abbreviated names are confusing to the readers and the completed names should be given.

Response: We added the complete names in the heading of Table 3.

Fig 4, the symbols such as 'Ox' and 'CHL A' are not consistent with the description in text and unit such as C is not right.

Response: We changed "CHL A" to "chl-a" and we introduced this abbreviation and used it through the text. Also we changed "ox" to oxalate" and "c" to concentration".

Moreover the authors changed "aerosols" to "aerosol particles" and "POLARSTERN" to "Polarstern" throughout the text.

In line 132 a typing error was corrected.