

### **Author's response:**

#### ***On the impacts of phytoplankton-derived organic matter on the properties of the primary marine aerosol – Part 2: Composition, hygroscopicity and cloud condensation activity***

Atmos. Chem. Phys. Discuss., 10, 26157–26205, 2010

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We thank the reviewers for their comments on the submitted manuscript. Below we have embedded detailed responses (A) to the reviews, with the reviewers' statements (R) indicated in italics.

It should be clarified that the manuscript under revision is a shortened version of the paper submitted initially, as requested by the editor and one of the reviewers of the manuscript. The only modifications with respect to the initial version is that the methods section has been shortened, adding references to previous work for a more detailed description, while only the main plots have been included in the body of the manuscript. Some of the main concerns raised by the reviewers are related to information that was removed from the initial version of the paper in order to shorten the manuscript. This information will be added back to the paper for clarification.

#### **Reviewer 1:**

*R1- "My major issue with the paper is how the concentrations of organic material in the experiments described here relate to what is present in the real world. The authors should give a sense of typical concentrations of exudates in sea water so the reader can understand the significance of the results using concentrations on the order of 512  $\mu\text{M}$ ."*

A-This aspect was actually addressed in the initial version of the paper, but was later removed in order to shorten the manuscript. The samples were prepared in a concentration from 175 to 512  $\mu\text{M}$  OC<0.2  $\mu\text{m}$  (table 1 of the manuscript), which is in the range of concentrations found in high biological activity areas, as described in part-1 of the manuscript (Fuentes et al., 2010b), where references to previous literature and data from field measurements are provided. Only for the experiments with *E.huxleyi* and *Phaeocystis* exudate, unrealistically high concentrations above 900  $\mu\text{M}$  were also used, to follow the trends of the effect of the organic matter on the particle behaviour and explore the possibility of organic mass fraction saturation. This explanation will be added in the methods section for clarification.

*R1- "Atmospheric sea spray" is not an appropriate term for the aerosol sampled by O'Dowd et al. The term implies that it is only sea spray aerosol and does not recognize that it most likely was chemically modified by secondary processes after being emitted from the ocean."*

A-The term "atmospheric sea spray" will be replaced by "atmospheric marine aerosol".

*R1- "Section 2: A figure showing a flow chart of sample preparation for the various aqueous media (artificial seawater, natural seawater, etc.) would help clarify the methods used for the reader."*

A-Because of the simplicity of the methods, we do not think it is necessary to include a figure schematic to explain how the samples were obtained. The artificial seawater was prepared by simply dissolving the required solutes in deionised water and the

seawater samples were prepared by dilution of natural seawater with artificial seawater to obtain the desired organic matter concentration. This will be briefly summarised in the methods section, including a reference to previous publications, for a more detailed description.

*R1- "The correlation between chl-a and particle organic fraction in O'Dowd et al. (2008) is quite poor ( $r^2 = 0.25$ ). It is likely that the small degree of variance accounted for by chl-a is due to primary emissions of sea spray collected on the filter. The remaining variance is most likely due to secondary processing incorporating organics that may or may not be of marine origin."*

A- This observation will be taken into account in the description of the results by O'Dowd et al. (2008).

*R1- "Why was a 0.2  $\mu\text{m}$  filter used? Because this is how seawater dissolved organic matter is usually defined?"*

A- "A filter pore size of 0.2  $\mu\text{m}$  was used in order to remove the existing bacteria, and avoid biodegradation of the organic matter released by the marine algae during the aerosol experiments. The organic size pool below 0.2  $\mu\text{m}$  does contain colloidal and dissolved organic matter, as described in Verdugo et al. (2004). We remark that the term "dissolved organic fraction" (DOC) applied to the size range <0.5-0.7  $\mu\text{m}$  is misleading (Verdugo et al., 2004), since the term DOC is defined as a function of the size, and not the solubility of the material separated. This explanation was originally included in the first version of the manuscript and will be added in the final version of the paper."

*R1- "Most of the organic fraction of the dry particles is dominated by sea salt: : :." This statement doesn't make sense since "sea salt" is typically taken to mean inorganic."*

A- The authors mean here that the inorganic fraction dominates over the organic fraction, or, in other words, the particle composition is mainly made up of sea salt. The sentence will be restructured in order to clarify.

Suggestions on minor corrections by reviewer 1 will be addressed.

## **Reviewer 2:**

*R2- "The main problem I have with the paper is that it makes general conclusions about phytoplankton-derived organic matter impacts on the marine aerosol cloud condensation activity after essentially filtering out majority of organic matter via ultra-filtration (0.2 $\mu\text{m}$  membrane filter) if there was any significant water insoluble particulate or colloidal matter present. Therefore, the title and abstract should clearly state that the studied effects were due to mainly dissolved organic matter. [...] Ultra-filtration through 0.2 $\mu\text{m}$  filter is normally used to separate insoluble organic matter (particulate organic matter, colloidal organic matter, viruses, bacteria or any live material). Therefore, I expect that authors either provide direct evidence that significant amount of insoluble material was present in their proxies or avoid using term "colloids" or "insoluble material" throughout the paper. I strongly suggest emphasizing in the abstract and possibly the title that studied impacts were of mainly soluble OM."*

A- We strongly disagree with the observations of the reviewer, which seem to contradict previous literature on the classification of the marine organic matter as a function of size and solubility. A filter pore size of 0.2  $\mu\text{m}$  was used in order to remove the bacteria existing in the seawater samples and avoid biodegradation of the organic matter during the aerosol experiments. Organic colloidal matter does exist in the size range  $<0.2 \mu\text{m}$ , where it is defined as “organic microgels” (Verdugo et al., 2004), coexisting with truly dissolved organic matter. We argue that ultra-filtration is a standard method used to separate the insoluble from soluble organic matter. Filtration is a method used to separate components as a function of the size, not as function of the solubility. We remark that the term “dissolved organic fraction” (DOC), frequently applied to the organic pool below 0.5-0.7  $\mu\text{m}$  is misleading, since the term DOC is defined as a function of the size (Verdugo et al., 2004), and not the solubility of the material separated, thus it is inappropriate for defining the organic carbon pool in this size range. The fact that there exist colloidal microgels in our samples is also supported by experimental evidence, as indicated by the high molar mass (1.37-2.37 kDa) found from our Langmuir isotherms measurements. By using a filter of 0.2  $\mu\text{m}$  pore size, it is the organic macrogel fraction with molar mass above 2 kDa plus algal biomass, that were removed (Verdugo et al., 2004), while the microgel fraction remained in our samples. In addition, it is suggested that aggregation of marine colloidal matter  $<0.2 \mu\text{m}$  is an important mechanism for the formation of aggregates in the supermicron size range (Wells and Goldberg, 1993). If the colloidal fraction is considered to be insoluble, then it follows that there is insoluble material in our organic samples, even if this is a fraction of the total insoluble organic matter in seawater.

We clearly defined at the beginning of the abstract the size range of the organic matter we worked with. We will add a description on the nature of the organic matter in the samples in the final version of paper, as explained above and state which organic fractions were removed by filtration. We have discussed the results of our study in a balanced way, stating throughout the manuscript that the effect of the colloidal organic pool  $>0.2 \mu\text{m}$  on the particle behaviour should be determined experimentally and compared with the results of our study.

*R2- “Little attention is given to filtration method as I already argued. It would be appropriate to justify OM concentrations used in the experiments. How 512 $\mu\text{M}$  concentration compares to the real world? 512 $\mu\text{M}$  would convert to  $\sim 6\text{mg/l}$  concentration which is unheard off in anthropogenically unperturbed environment. Considering a largely dissolved material in the filtered proxies, 512 $\mu\text{M}$  concentration would be even more staggering.”*

A- Description of the filtration method was explained in the initial version of the manuscript. Part of this description was removed in the shortened version of the paper. The samples were prepared in a concentration from 175 to 512  $\mu\text{M}$  OC $<0.2 \mu\text{m}$  (table 1 of the manuscript), which is in the range of concentrations for this organic size pool in high biological activity areas, as described in part-1 of the paper (Fuentes et al., 2010b), where references to previous literature and data from field measurements are provided. Only for the experiments with *E.huxleyi* and *Phaeocystis* exudate, unrealistically high concentrations above 900  $\mu\text{M}$  were also used, to follow the trends of the effect of the organic matter on the particle behaviour and explore the possibility of organic fraction saturation. This explanation will be added in the methods section for clarification.

*R2- "There was little effect when monolayer method was applied. However, one could argue that absence of the effect could be due to wrong monolayer approach. For example, monolayer is possibly forming at every air/water interface of rising bubbles, thus proving against single monolayer approach."*

A- The monolayer experiments were actually performed to test whether the existence of a single monolayer on the water surface is sufficient to enrich the bubbles reaching the surface and the subsequent generated particles, in comparison with the case of absorption of organics in the air/water interface of rising bubbles (bulk mixing method). The little effect observed in the monolayer experiments could be either due to a quick removal of the monolayer during bubbling, as this monolayer was not replenished, or to a low organic enrichment of the bubbles and particles generated from a single monolayer on the water surface. The higher enrichment obtained in the bulk mixing experiments could indicate that the mechanism by which bubbles and particles would be substantially enriched in organic matter would be the absorption of organics on the air/water interface of rising bubbles, rather than by the only existence of an organic monolayer on the water surface. The results in this work are however inconclusive, as further work is necessary in order to evaluate the time scale for the removal of organic monolayers by bubble bursting. This discussion will be included in the revised version of the manuscript.

*R2- "I have a problem with Figure 5 as it is presented and discussed. This plot was already introduced in Part 1 paper, which lies on a dubious assumption that 80% of OM is present in the 0.2um filtrate (again only supported by reference and not by the measurement). Reverse the assumption that only 20% of OM is left after ultra-filtration and the plot will easily match O'Dowd/Facchini/Keene experiments."*

A- The reviewer makes here a wrong interpretation of the procedure presented in the manuscript and previous publication. The relationship between OC and Chl-a concentration was modelled on the assumption that the organic pool below 0.2  $\mu\text{m}$  accounts for 80% of the marine organic matter (Kepkay, 2000). This assumption affects only the x coordinate in Figure 5, i.e., the inferred Chl-a values, and not the particle organic mass fraction composition (y coordinate), since the particle composition was derived from experimental measurements. The only possibility for our data to match O'Dowd et al.'s (2008) experiments would be shifting our dataset towards the left on the x axis, i.e. towards lower Chl-a concentrations. If our data matched O'Dowd et al. experiments in this manner, then we would have values of Chl-a concentrations of 0.2  $\mu\text{g/L}$ , characteristic of low biological activity areas, for OC concentrations above 300  $\mu\text{M}$  (OC concentration used in the experiments). The resulting OC-Chla relationship would therefore not be representative of reality, where the concentration of OC in low biological activity areas is on the order of 50-80  $\mu\text{M}$ . Hence, this line of thought leads to a non-realistic conclusion.

The literature comparison made by the reviewer, concerning the primary particle organic enrichment, does not seem to be only impartial. Objectively, we should also add to the discussion the results of the work by Sellegri et al. (2008), where experiments on the hygroscopic growth of primary aerosol generated from unfiltered natural seawater, resulted in hygroscopic growth suppressions of 10%, which is on the order of findings in our study. It is therefore not clear that the lower organic enrichment in our experiments with respect to Facchini et al. is exclusively due to the removal of the organic pool  $>0.2 \mu\text{m}$ . Differences with respect to other works can also be due to the organic matter concentration in the sample seawater and the aerosol generation technique. One of the main findings of the present work is that the

seawater organic concentration and the type of algal exudate affect the composition of the generated particles; hence, this finding could partially explain the diverging results between different studies. This discussion has already been included in the paper.

*R2- "Comparison with Sellegri 2006 and Modini 2009 is only valid when considering that in both studies OM was dominated by DOC: Sellegri used artificial fully soluble organic compound SDS while Modini et al. admitted that the OM in the seawater could have been dominated by significant river run-off (inevitably containing lots of DOC)."*

A- The reference to Sellegri et al. is actually Sellegri et al., 2008, "Role of the volatile fraction of submicron marine aerosol on its hygroscopic properties", (notice that there is an erroneous reference in our references list). Sellegri et al.'s (2008) conducted hygroscopicity measurements with unfiltered seawater from North Atlantic waters, thus, containing organic matter from the whole organic pool size spectra. We remark that the term DOC used by the reviewer for the organic size range  $<0.2 \mu\text{m}$  is misleading and we are reluctant to use this ambiguous term in our paper. As explained above, the DOC pool contains organic matter  $<0.5\text{-}0.7$  microns (Verdugo et al., 2004), and includes colloidal organic compounds of low solubility and truly dissolved organic matter. A more appropriate definition of the organic matter in seawater should be made by defining the size range, when solubility measurements are not available. We believe that it is appropriate to mention in the manuscript that the seawater samples in Modini et al. (2009) could possibly be affected by river run-off, which can affect the organic content of the samples. We are, however, reluctant to state that the samples in Modini et al. (2009) were dominated by soluble organic matter, as no measurements of solubility were provided by these authors.

*R2- "There was no truly hydrophobic material (at least there is no direct evidence provided), therefore, I would suggest using term "less hydrophilic"."*

A- This reviewer's statement makes hardly any sense. The surface tension suppression observed in the pendant drop measurements presented in section 3.5 provide direct experimental evidence of the presence of hydrophobic material in the exudate samples, while the Langmuir isotherms in section 3.4 prove that hydrophobic material could be extracted from the seawater, as evidenced by the formation of an hydrophobic film on the surface of the Langmuir trough. The fact that the organic pool studied represents a fraction of the total organic spectra does not imply that it does not exhibit hydrophobic properties. This is demonstrated by the surface suppression measurements provided in the paper.

*R2- "The whole story of this paper is conceptually built around dissolved organic carbon which is particularly evident in this chapter. The whole theoretical framework used in this paper can not accommodate particulate nor colloidal organic matter of largely insoluble material."*

A- Again, we argue here that there is not colloidal matter in the samples employed. As explained above, the fact that there is colloidal organic matter in the studied size fraction is supported by previous literature (Verdugo et al., 2004) and also by our experimental results, as reflected by the large molar mass (1.37-2.37 kDa) derived for the hydrophobic fraction extracted from the organic samples.

*R2- "In their Part 1 paper authors demonstrated that the main particle size which was largely enhanced by primary production using various proxies was around 40nm. This is very different not only from O'Dowd/Facchini observations as stated in this*

*and Part 1 paper, but also those of Ellison et al. (1999, JGR) who also proposed a theoretical model for the observed insoluble/hydrophobic material. The observed particle size in Ellison paper was around 200nm, quite similar to the accumulation mode size in O'Dowd/Facchini/Keene papers. I argue that the main reason for such discrepancy are not various temperature, wind speed effects or secondary processes invoked by authors in Part 1 paper, but particulate and colloidal material which was filtered out by ultra-filtration."*

A- We argue against this distorting interpretation of what is presented in part-1 of the paper and the inadequate literature comparison made by the reviewer. We also find inappropriate that the reviewer includes objections to aspects of the part-1 of the paper that are independent of what it is presented on part-2 of the study:

- The analysis by the reviewer distorts the conclusions stated in part-1 of this study. In Fuentes et al. (2010b) it was concluded that there was an enhancement of the particle production below 100 nm, and not particularly of the particle size at 40 nm, as the reviewer mentions, due the organic surfactants.
- We argue that our results of size distribution can be directly compared for validation with atmospheric measurements by O'Dowd et al. (2004), as the atmospheric measurements are subject to secondary processes and environmental factors (wind speed, temperature), which are not reproduced in the laboratory simulations. We strongly disagree with the statement that the effect of temperature, wind speed and secondary processes exert no significant effect on the shape and properties of the atmospheric size distribution. In fact, claiming that environmental factors do not affect the particle production contradicts experimental evidence (Martensson et al., 2003; Sellegri et al., 2006). The reviewer's statement is also contradictory with experiments in Fuentes et al. (2010b), where similar effects on the particle size distribution <450 nm were observed when using unfiltered natural seawater (thus containing both the organic macro- and microgel colloidal fractions plus algal cells) and when using seawater proxies filtered for OC<0.2 µm.
- We argue that the atmospheric size distribution has a unique profile, represented by the distribution reported by O'Dowd et al. (2004), as suggested by the reviewer and that 200 nm is the size representing the submicron atmospheric marine aerosol distribution. Different shapes of atmospheric marine aerosol size distributions have been reported in different studies, where the modal sizes have been shown to be highly variable depending on the measurement period (Sellegri et al., 2006). The comparison by the reviewer with the study of Ellison et al. (1999) is definitely distorted. Ellison et al. (1999) did not "observe" any particle diameter; what these authors did is to choose for their model a mean particle size for the marine aerosol of 200 nm, which was the modal size found in atmospheric measurements in ACE-1 (Brechtel et al., 1981). As mentioned above, there is high variability in the modal sizes in atmospheric measurements as shown in Sellegri et al. (2006), where values can vary from 40 nm to 100-200 nm.
- Comparison with Facchini et al. (2008) is not adequate in the context of particle size distributions measurements. Facchini et al. (2008) did not provide any particle size distributions measurements, but only analysis of the particle organic mass fraction as a function of the particle size.
- Keene et al. (2007) produced their primary aerosol with glass frits, which is a different system to the one used in the present work and that does not represent the physics of formation of bubbles plumes in real conditions. It has been demonstrated in Fuentes et al. (2010a) and Sellegri et al. (2003) that

the laboratory mechanism for primary aerosol formation affects the resulting particle size distribution shape, thus explaining that different dominant modal sizes are found in different works. In addition, we have demonstrated in the present and previous publications that the effect on the particle size distribution shape and particle properties depend on the organic matter content and the exudate type in the seawater. Thus, it is expected to observe divergence between different studies because of the different nature and concentration of the organic matter in the source seawater employed.

*R2- "According to authors approach organic material is forming a film with an area of 119.54 cm<sup>2</sup> and mass of 24µg. Assuming OM density of 1.4g/cm<sup>3</sup> that converts to a film thickness 1.5nm. According to the thermodynamic model of Oppo et al. (1999, Marine Chemistry) only particles as small as 1.5nm would be 100% organic assuming such film thickness. It seems that Oppo model suits this paper perfectly with very thin films producing particles in the size range of 40-100nm with little organics. It must be colloidal and particulate organic material which would be capable of producing larger particles highly enriched in OM which remains to be proven."*

A- This is another incorrect interpretation by the reviewer of the experimental procedures described in the paper. It is not said in the manuscript, as the reviewer states, that 24 µg of organics in our samples form a monolayer with an area of 119.54 cm<sup>2</sup>. In the paper it is mentioned that the virial analysis employed is based on the assumption that a monolayer of 24 µg of marine hydrophobic matter compressed to a surface area of 119.54 cm<sup>2</sup> leads to a surface pressure of 20 Nm/m, as adopted by Pogorzelsku and Kogut (2003). This is not data derived from our samples, as the reviewer seems to believe, but a relationship derived from measurements by Barger and Means (1985) with a number of large molar mass organic compounds contained in marine organic matter. A surface pressure of 20 Nm/m for marine organics is characteristic of condensed liquid phase film (results in this work and Pogorzelsku and Kogut, 2003), thus it is expected that the relationship between mass and area provided by Barger and Means (1985) is not for a densely packed film. Thus, the area 119.54 cm<sup>2</sup> would not be valid to calculate the monolayer thickness. To calculate the monolayer thickness it would be required to determine the area of a condensed monolayer with all the molecules vertically tilted. Consequently, the reviewer's calculation of the film thickness and subsequent analysis are not valid.

The statement by the reviewer that larger organic matter leads to thicker organic monolayers and consequently to larger particles, seems rather speculative. There is no experimental evidence relating film thickness with particle fluxes production. We provided evidence in Fuentes et al. (2010b) that organic matter >1 micron does not lead to the production of more of the larger particles in the particle size range <450 nm. We recognize, however, that the effect of supermicron organic matter on the submicron aerosol properties and the particle size range above 450 nm has not been evaluated in our study. For this reason, we have stated throughout the paper that the effect of the supermicron organic matter on the particle composition and behaviour should be determined experimentally and compared with the results in the present study.

*R2- "P26173, line 8. Could it be that the lower end values obtained here are again due to the filtering effect?"*

As explained in the paper, the derived values are within the range found for marine organics films, presenting typical values of surface films collected in areas away from

the influence of anthropogenic emissions. Obviously, the removal of organic matter  $>0.2 \mu\text{m}$  from the samples by filtration must affect the results of molar mass and limiting area estimated using the virial state equation approach.

*R2- "P26175, line 23. The statement ": : :are in the order of molar mass and hygroscopic growth of biopolymers present in the marine exudate" needs evidence. Where was it measured?"*

A- This paragraph by the authors is making reference to the molar mass values determined in our study and those of Pogorzelski and Kogut (2003) and the hygroscopic growth measured for oxygenated organic compounds in the marine exudate. We agree that a citation should be included here. This will be added to the final version of the manuscript.

*R2- "P26177. Is the low sensitivity to OM density and GF due to low OM content in the studied particle sizes or series of assumptions used? Low sensitivity needs better discussion of why and how that can be true."*

A- The sensitivity analysis presented was conducted for the organic volume fraction range in our study. We believe that an analysis of the sensitivity to these assumptions for compositions above those found in our work would not provide any additional useful information for the interpretation of the data presented, however we will clarify in the text that the sensitivity results are only valid for the range of organic volume fractions obtained in the study.

*R2- "P26177. I am quite puzzled about the dominance of the linear Raoult term over the non-linear Kelvin term. To me this is only possible in relatively small particles produced by specific DOC. For larger particles Kelvin term should dominate as demonstrated by Dusek et al. (2006, Science)."*

A2- We disagree with this statement. As explained in the paper, the replacement of low molar mass sea salt by high molar mass exudate leads to a substantial reduction in the number of solute moles and then, to a dominance of the solute effect (Raoult) over the curvature effect (Kelvin) in the Kohler equation. Similar effect has been found in Li et al. (1998) with experiments with SDS/NaCl particles and with mixed organic macromolecules/inorganic compound particles (Moore et al., 2008; Kristensson, 2010). In addition, contrary to the reviewer's last statement, it is generally known that the curvature effect (Kelvin effect) becomes more important the smaller the particle size.

*R2- "P26180, lines 1-3. I would argue that while being questionable it is not unlikely that primary marine OM are responsible for the increased cloudiness. Methodological constraint of this study using ultra-filtration does not allow such a general conclusion and, therefore, should be rewritten in a more balanced way."*

A2- We believe that we have discussed this aspect in a balanced way with the following paragraph, following the CCN experiments description. (p.26179, line 20)  
"In the present study marine organic matter  $<0.2 \mu\text{m}$  has been used; thus, further research is needed in order to determine whether the observed behaviour extrapolates to large colloids in the size range above  $>0.2 \mu\text{m}$ , or indeed, whether such colloids play a role in particles of the smaller sizes where organics are most substantially enriched."

The paragraph the reviewer refers to in p. 26179-26180 includes some theoretical results on how it is, however, questionable, that large size OM can induce an



enhancement of the CCN activity. For a molar mass >2kDa and organic mass fraction of 50%, the surface tension at the point of activation should be as low as 55 mN/m to reduce the critical supersaturation, which is a very low value for the diluted conditions expected at the point of activation.

*R2-“P26181, lines 22-30. Was the effect small just because of little organics present in the particles?”*

A- The part of the text indicated by the reviewer does not deal with any “effects” but with the deviation on the prediction of kappa values from CCN and hygroscopicity measurements, which is found to be <5%.

*R2- “I am confused by the circular argument of this study: the observed growth factor and activation effects are due to small amount of organic matter, which was indirectly estimated from growth factor measurements. How else can it be?”*

A- We find this reasoning by the reviewer largely confusing. The hygroscopic growth of the particles is determined by its chemical composition. We have derived the particle composition from the hygroscopic growth measurements but the hygroscopicity and CCN measurements are independent of any inferred compositions.

#### **Minor comments**

*R2- “P26159, line 18 (also next page line 4). The term “atmospheric seaspray” is new and possibly requires small introduction.”*

A- The term seaspray will be replaced by the term “atmospheric marine aerosol”, as was requested by reviewer 1.

*R2- “P26166, line 19-20. Species names should be properly spelled and in italic.”*

A- The names of the species are properly spelled. The names will be written in italic.

*R2- “P26170, line 7. O’Dowd 2008 paper deals with primary not secondary organics. Wrong style or please explain.2*

A- O’Dowd et al. 2008 paper deals with atmospheric marine aerosol measurements, thus potentially including both primary and secondary organics. The atmospheric measurements in O’Dowd et al. 2008 are based on Mace Head measurements from Cavalli et al. (2004).

*R2- “P26171, line 16. “higher effect” should be replace with “stronger effect”.”*

A- The sentence will be modified.

*R2-“P26173, line 5. should be “regression coefficients above 0.87”. Also please state significance level, e.g. “P<0.01” or similar. Coefficients themselves mean nothing until statistical significance is stated which depends on the number of points/observations constraining regressions.”*

A-The significance level will be added.

R2-“P26174, line 12. Keene et al. 2007 measured soluble OC, not WIOC which dominated in Facchini 2008 study. Please explain if not a miss-print?”

A-The misprint will be corrected.

R2-“P26175, line 18. “represents and undissolved” should be “an” instead of “and”.2

A- The typo will be corrected.

R2-“Same page, line 21. “on study” should possibly be “studied”.

P26177, line 23. “: : :due to dominance of the Raoult: : :”

A-The sentence will be modified.

R2-“P26178, line 1. I believe “organic fraction” should be “mass” otherwise doesn't make sense.”

A-“Mass” will be added to the sentence

## References

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