

### **REVIEWER COMMENTS IN ITALICS:**

*I cannot support publication of this manuscript in ACP in its current form. There are many issues raised in the first round of comments that have not been addressed. I still have major questions about the uncertainties of the measurements (concentrations, gradients, K and fluxes), the data analytical approaches employed (particularly those concerning WIOM), and consequently, the strength of the papers conclusions. I have tried to restrict these further comments to the authors responses to my first set of comments and the additional material that has been added to the revised manuscript (e.g. abstract and section 3.5). However, now that I've looked at the Stull (1988) reference, which was missing from the ACPD paper, there are also some questions about the gradient transport theory underlying this work that I believe should also be addressed.*

*Gradient transport theory:*

*- In the chosen parameterisation for K, please clarify in the text what value was used for the timescale, t.*

### **AUTHORS COMMENTS IN NORMAL TEXT:**

The eddy covariance micro-meteorological system was operated at 10Hz and the eddy flux data were processed to 30min average time resolution to capture the smallest and the largest relevant eddies. The standard deviation of the vertical wind velocity over those timescales was used to calculate K. This approach follows from diffusivity fundamentals (e.g. Reible, 1998) where characteristic vertical transport distance is  $\sigma = \sqrt{2Kzt}$  and after rearranging for the vertical wind speed becomes  $\sigma = \sqrt{2Kz/t}$ . Time becomes irrelevant as it was a unit time (1s). The text has been updated accordingly.

*- The turbulent transport coefficient, K, is different for different variables. E.g. K for heat and moisture transport is a factor of 1.35 times greater than the K for momentum transport under neutral conditions (Stull, 1988). Please clarify which K is considered to apply here.*

*- A number of other parameterisations exist for calculating K. With the sonic anemometer data it should be possible to use some of these other parameterisations. This is another source of uncertainty that may need to be represented in the uncertainty of the reported fluxes. To know this, please indicate how sensitive the calculated fluxes are to the chosen parameterisation of K. This is important because the true fluxes of any of the chemical species are not available to validate these measurements (the sea salt fluxes compared against in Fig. 8 and discussed in section 3.3 each have their own set of problems). Nor were they available in the 2 earlier studies of Ceburnis et al., (2008) and Valiulis et al., (2002), which relied on the same theory and parameterisation of K. I.e. this approach for calculating fluxes has not been rigorously validated yet.*

The two comments above will be treated as one in our response as they relate to the same issue. Further, given that the general reason for rejection is based on what we regard as the two above un-informed points, we will respond with a generic response. First we consider the

request for this additional work as beyond the scope of the paper; and second, the source of the information upon which the review bases their remarks has been selectively quoted in a seemingly poorly informed manner.

K values can be derived from flux measurements in only two ways: directly from the measurements of the standard deviation of the vertical wind velocity (our approach) OR when simultaneously measuring eddy covariance flux and the gradient of a parameter of interest, e.g. VOCs (Goldstein et al., 1996). Therefore, reviewer’s notion to use eddy covariance measurements for parameterising different K values is unfounded.

The reviewer refers to *Stull* and to differences in the turbulent transfer coefficient depending on which parameter is referred to in terms of flux. For the record, *Stull* states “*Sometimes, different K values are associated with different variables. A subscript “m” is used for momentum, resulting in  $K_m$  as the eddy viscosity. For heat and moisture, we will use  $K_H$  and  $K_E$  for the respective eddy diffusivities. There is some experimental evidence to suggest that for statically neutral conditions:*

$$K_H=K_E=1.34K_m.$$

*It is not clear why  $K_m$  should be smaller than the other K values. Perhaps pressure-correlation effects contaminated the measurements upon which the aforementioned equation was based.”*

In a nutshell, there is no fundamental reason why different K values should be different given that eddy diffusivity exceeds any specific molecular or particle diffusivity by many orders of magnitude (Figure S1).

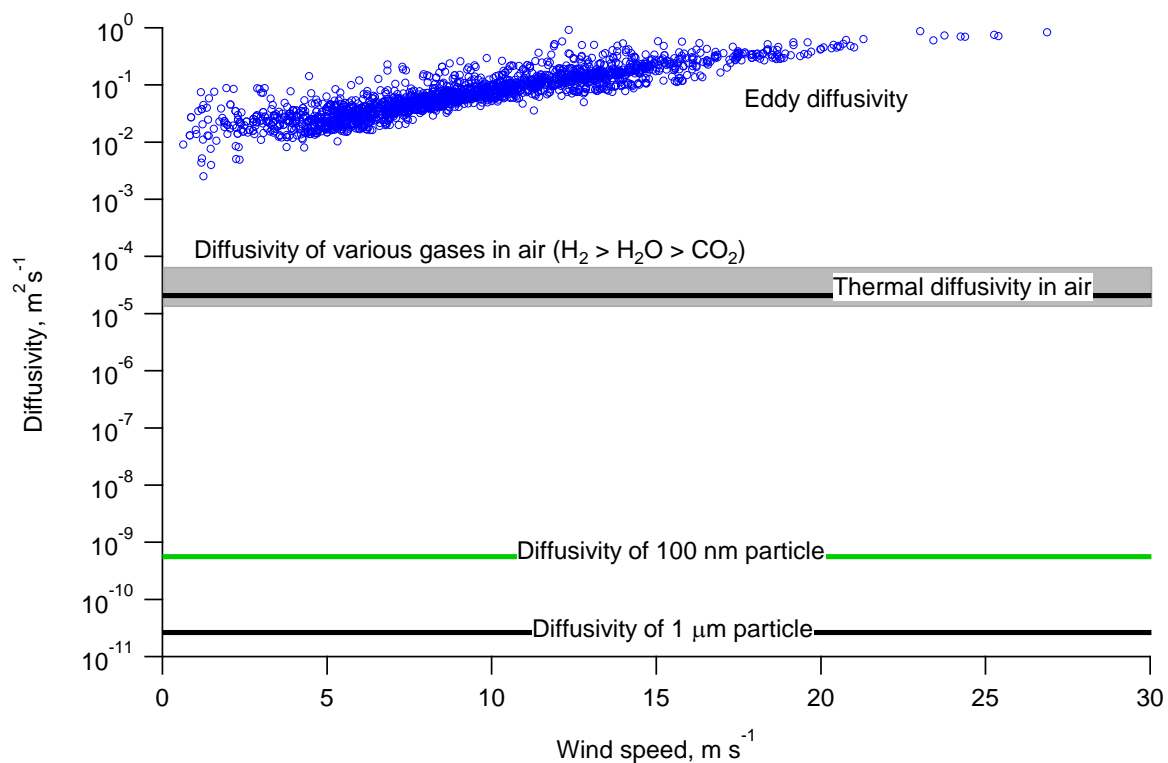


Figure S1. Summary of diffusivity values and the relationship between eddy diffusivity and horizontal wind speed.

Moreover, the reviewer requests this extraordinary exercise because, as the reviewer states, “*This is important because the true fluxes of any of the chemical species are not available to validate these measurements*”. With all due respect, if we had true fluxes why would we be making these flux measurements and, how do we get true fluxes? We refrain to responding to any subsequent comments relating to this issue as we do not feel they are raised on a sufficiently sound foundation to respond further too, let alone to have a paper rejected on.

We trust that the subsequent and more specific responses to the reviewer comments are sufficient to have the manuscript deemed suitable for publication.

*Responses to the authors responses:*

- *Line 288: If this interpretation is based not on the actual range of values for the height of the internal boundary layer reported by Norton et al., (2006) but only on their statement that the effect is smaller at higher levels, then clarify this here.*

The following sentence has been modified. “Norton et al. (2006) showed that the internal boundary layer was typically limited to below 10m and never propagated to the top of the tower in marine sector, consequently, having small effect on our measurements at 10 m and 30m.”

- *Section 2.5. The equations for calculating propagated errors are much clearer now, but the concentration uncertainty values are still not reported. What are the uncertainties for the normalised concentrations presented in Fig. 3? How do these compare to the variances? I disagree with the authors response. There is a decision to be made about whether the error bars in Fig. 3 represent the accuracy (uncertainty) or precision (variance) of the measurements. The error bars should reflect as best as possible the confidence one can place in these measurements. If the uncertainty is large this needs to be represented by the error bars in the Figure (especially considering that large uncertainty is the authors stated reason for the mixed nss-SO<sub>4</sub> profile). Although the sea salt profile seems clear, given the questionable assumption of stationarity over the many hours required for sampling I am still far from convinced about the accuracy of the NH<sub>4</sub>, nss-SO<sub>4</sub>, WSON, MSA, WSOM and WIOM gradients.*

We now added a Table S1 for Suppl Material with the analytical uncertainties of all chemical species. It is quite possible there is a miscommunication problem. Fig 3 is only a summary of concentration measurements. As all individual profiles were averaged for presentation they are presented with a respective standard deviation. Fig. 3 is only for visualisation purposes to classify the profiles. When it comes to calculating fluxes they are calculated from individual profiles using Table S1 and respective propagated errors are calculated by formulas listed in chapter 2.5. We cannot be more transparent here.

The corresponding sentence is modified: “The uncertainty of individual concentrations (C) (provided in Table S1) and the gradient (G) was calculated by the following Equations:”

New sentence was added: “The normalised averaged concentration profiles allowed classification and categorisation of the profiles, but the normalised data were not used for calculating gradients and fluxes.”

- *Line 380: How exactly were the nitrate and oxalate profiles 'distorted'. And 'systematically'. Need to be more specific.*

Please refer to continued sentence (lines 379-381 in the previous version) “i.e. the concentrations of oxalate and nitrate significantly diverged from the sea salt one at the lowest sampling height of 3 meters while following the sea salt profile above 10 meters”. The shape of the profiles was repeatedly (synonymous to systematically) distorted. Respected sentences were modified accordingly.

- *Line 396-419: This discussion still fails to mention wind speed, a factor which is later shown to explain as much variance in WIOM fractions as Chl a concentrations. Therefore, this discussion is not only speculative but also inconsistent with results presented later in the paper. It needs to be modified or removed.*

The effect of wind speed on OM fractional contribution to sea spray is very much an open question and more importantly has not been quantified due to the opposite seasonality of wind speed and chlorophyll and cannot be resolved by multivariate analysis. It can only be done on independent variables whereas wind speed and chlorophyll are seasonally related. For example, the effect of wind speed could be elucidated if very low OM fractional contributions were often observed during summer or high fractional contributions during winter which was never the case with very few exceptions (only two).

One sentence has been added to mention the effect of wind speed.

- *Line 434: The ambiguity supposedly responsible for the unexpected nss-SO<sub>4</sub> profile is not reflected in the error bars in Fig. 3. As things stands the main text says don't trust the nss-SO<sub>4</sub> profile, but the small error bars in Fig. 3 invite the opposite interpretation. This is one reason I suspect the error bars in Fig. 3 should represent the measurement uncertainties and not variances, to give a better sense of exactly how much confidence we can place in these measurements.*

We would like to make several points which in combination should level the argument. As it was pointed out earlier the Fig.3 is for visualisation purposes. Data used to generate Fig. 3 are not utilised in a quantitative manner further in the study. Error bars in Fig. 3 may look small, but the averaged profile itself is hardly showing any pattern (if we were to blow up horizontal scale the errors would become visually larger). In the first review round we demonstrated that excluding winter sulphate, sulphate errors and profiles would become tighter defined, but neither us nor the reviewer likes arbitrarily excluding data. Lastly, we provide Table S1 where winter sulphate concentration uncertainty is much larger than during summer.

- Line 444: The definition of the calculated ammonium displayed in Fig. 4 is still not specific enough in the main text. Does 'full neutralisation' mean all the sulfate and nitrate was neutralised?

Thanks for pointing this out. "Full neutralisation" was meant by sulphate only and considering that nitrate was more likely to be neutralised by sodium (causing chloride depletion) in the marine boundary layer.

The sentence was clarified accordingly.

- Line 501: The SSS-oxalate relationship is not similar to the SSS-nitrate relationship in Fig. 5 so this discussion needs modifying or removing. In general, it is not ok to simply select a subset of points that would follow a positive linear relationship and conclude that all of the data does. If certain points are excluded from the analysis, sound reasons need to be presented for doing so. The SSS-oxalate relationship looks far more similar to the SSS-MSA relationship.

A confusing reference to the similarity with nitrate was removed and the section is now discussed without referencing to "partial linearity".

- Line 550 (also applies to lines 314- 317): This is still not clear. Please indicate the actual values that were used for the relative uncertainties in G and K in the error propagation calculations. I am troubled by the authors response and the statement added to line 316 that  $\Delta K$  will be small because it is an average of hundreds of values. To give an accurate sense of the uncertainties in the calculated fluxes,  $\Delta K$  should reflect the large variance in K over the long sampling times (which can be seen for example in Fig. 1). K and therefore the calculated fluxes are likely to display substantial diurnal variation. Uncertainty in K and therefore flux will also arise due to the choice of parameterisation used to calculate K (as mentioned above). Since each F is an average over a number of days and nights the calculated  $\Delta F$  needs to reflect the variance and uncertainty in K. I suspect the true uncertainties in F are much greater than currently reported.

We disagree with the reviewer on this interpretation. The calculated fluxes represent many hours and, therefore, we have to use an average value and the precision of the average. The concentrations (and the gradient) were averaged by the continuous sampling over many hours while K had to be averaged according to statistical rules – precision of average K was represented by the standard deviation of the average. Very large or very small K values in the respected ranges would likely be accompanied by different concentration gradients (and fluxes) which we could not resolve. It would be incorrect using parameters of inconsistent uncertainties in the same equation. If we take the averaged gradient and any random  $K_z$  value over the sample duration that would imply that the gradient is constant over time while  $K_z$  is changing which, obviously, is incorrect.

Regarding diurnal variation of K we did not observe any statistically significant diurnal variation neither in summer nor in winter as presented in Figure S2. K was also tightly related to wind speed (Fig. 1&2) which (and the lack of diurnal variation) is typical in wind shear dominated turbulent environment like marine sector at Mace Head, contrary to often buoyancy dominated turbulent environment overland.

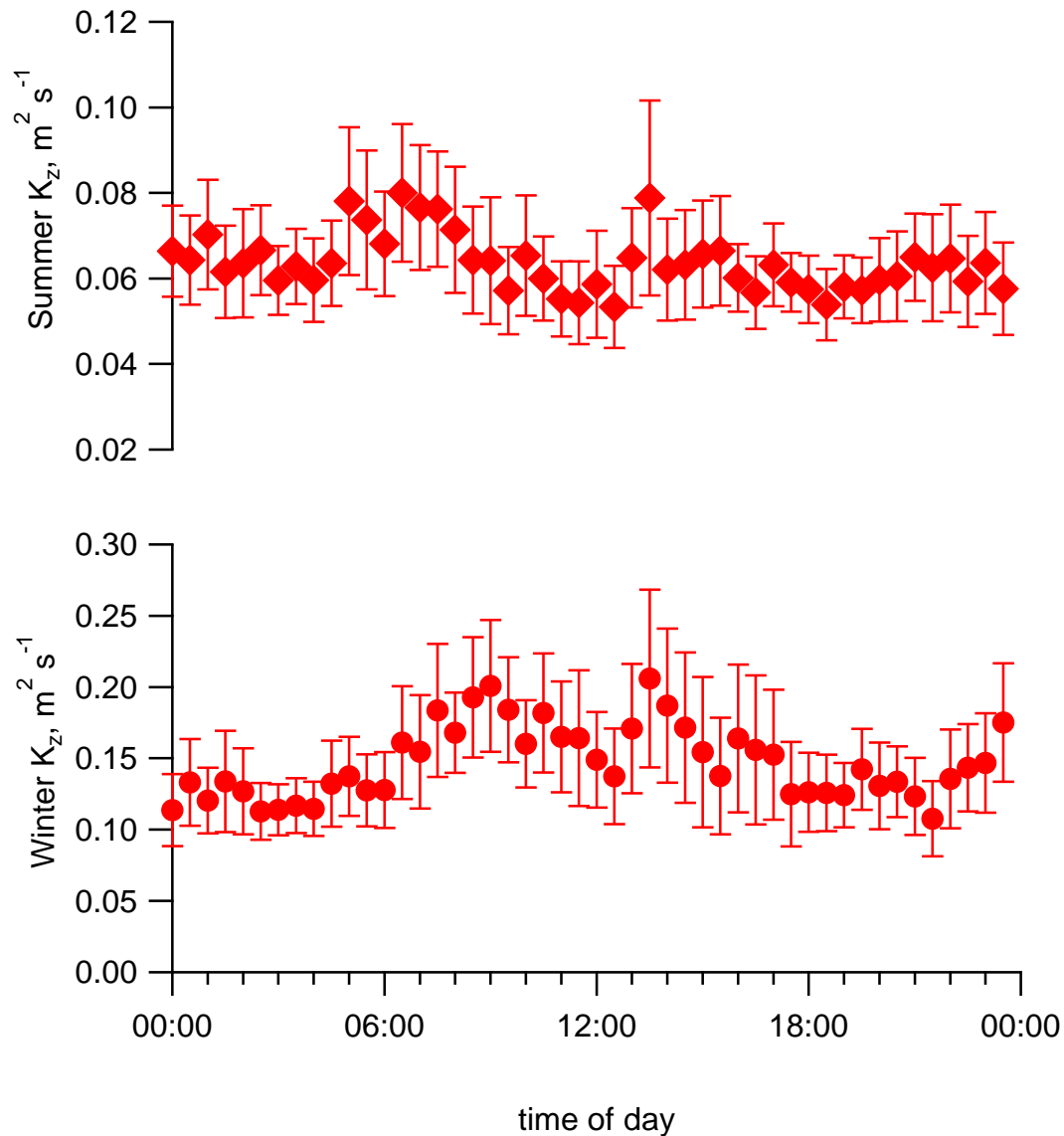


Figure S2. Diurnal variation of  $K_z$  during summer (June-July) and winter (December-February). Data used in the graph represent gradient samples collected in respective seasons.

- Line 589: My original comment hasn't been addressed. Since the relationship is only fitted to the positive flux points the wind speed range the fitted relationship is valid over needs to be reported (this should also be done for the other fitted relationships in the paper). The negative flux points are still not included in Fig. 7. The discussion here is qualitative and vague and does not provide a strong enough basis for suggesting that the relationship between WIOM flux and wind speed would be linear and not some other function (e.g. power law). The phrase "...best fitted to the line" needs to be made more precise.

The only negative fluxes obtained were of WIOM. These are now included in the Fig 7. It was not possible to fit WIOM relationship to any other functional form otherwise the uncertainties of the coefficients were an order of magnitude larger than the values themselves. So the linear fit was considered the best.

- Line 609: *I agree with the authors decision to remove the physically-unsupported WSOM-wind speed parameterisation from Fig. 7. The problem is, the parameterisation is still discussed in the text and included in Table 3. If no confidence can be placed in the parameterisation than it should be removed from the paper entirely. A side point, the argument that the opposing signs of the WIOM and WSOM flux dependencies on wind speed suggests that a 'significant' fraction of WSOM is processed WIOM is not clear. It should be further expanded on or removed.*

The discussion section of unsupported WSOM-wind speed parameterisation is removed including Table 3.

- Line 708: *If the authors contend "...that a fraction of measured WSOM was associated with sea spray" then it is a simple exercise to check whether the OMss relationships with wind speed and Chl a concentrations would hold if it is assumed that, say, 25% or 50% of measured WSOM contributes to OMss.*

Indeed, the relationships were held with 25% and 50% of measured WSOM contributing to OMss. The variances even marginally increased (e.g. from 0.58 to 0.60 or from 0.57 to 0.61). That does not mean, however, that e.g. 50% of WSOM is processed WIOM, but rather that WSOM being largely secondary is also related to chlorophyll-a. Moreover, WIOM is more strongly correlating to chlorophyll-a than WSOM (0.61 vs 0.51). In conclusion, we rather do not extend beyond conclusions made by Decesari et al. (2011) about processed primary OM based on highly resolved chemical composition with respect to correlations and variances.

- Lines 710 - 721: *The fact that there is correlation between Chl a concentrations and wind speed (coincidental or not) is precisely the reason why a multivariate analysis is required to answer the questions asked here. I do not accept the authors response that "...multivariate analysis won't solve the condudrum which can only be elucidated." As it stands, the conclusion that the OMss dependence on wind speed is significant but weaker than the OMss-chlorophyll relationship is completely unjustified. No evidence is presented to conclude one factor is more important than the other. This conclusion should be removed or supported by further analysis.*

We disagree with the reviewer. Correlation between the wind speed and chlorophyll-a has no scientific meaning and, therefore, coincidental (seasonal). Coincidental variance has to be taken into account and the conclusion about the weaker OMss relationship with the wind speed is justified. Otherwise, one should explain the meaning of wind speed and chlorophyll-a covariance which we cannot.

*New additions to the manuscript:*

- *Abstract lines 16-21: Sentence needs updating. Only chemical gradients, and not fluxes, are presented for the secondary components.*

Done

- *Abstract line 27: What is the meaning of the sentence the "...observed seasonal pattern of sea salt production was mainly driven by wind action with the tentative effect of marine OM"? Sea salt fluxes are only examined as a function of wind speed in the main text. What is the 'effect of marine OM'?*

The effect of marine OM to modify fractional contributions of sea salt and WIOM in sea spray has not yet been quantified, but can be inferred.

The sentence was modified to "The observed seasonal pattern of sea salt production was mainly driven by wind action in addition to yet unquantified effect of marine OM modifying fractional contributions of SS and OM in sea spray".

- *Table 1: Include headings for the columns*

Done

- *Line 742: Please support this statement by providing, for example, the average sea salt concentrations in summer and winter.*

Done. Sea salt concentrations and respective fluxes were generally the largest in winter ( $0.2\text{-}0.85 \mu\text{g m}^{-3}$  and  $0.9\text{-}2.5 \text{ng m}^{-2} \text{s}^{-1}$ , respectively) and the smallest in summer ( $0.07\text{-}0.5 \mu\text{g m}^{-3}$  and  $0.1\text{-}0.6 \text{ng m}^{-2} \text{s}^{-1}$ , respectively) which was mainly due to the wind pattern over the North East Atlantic.

- *Line 749-758: I have tried hard but simply cannot follow this sea salt replacement argument. I suspect there is confusion between the terms 'sea salt' and 'sea spray'. Please confirm if some uses of the term 'sea salt' are actually referring to 'sea spray'. In any case, the argument is vague and needs to be outlined more precisely. What do 'somewhat diminished' and 'stronger sea salt and wind speed power law relationship compared to pure sea salt relationship' mean exactly? And how does this provide evidence of a sea salt replacement effect? I doubt these flux results are accurate enough to be able to detect such an effect.*

The terms "sea salt" and "sea spray" were not confused. The concept of sea salt replacement has been introduced by Oppo et al. and further supported and developed by observations at Mace Head. If we start with a pure sea water droplet (be it film or jet drop) then after evaporation of water pure sea salt particle is born. If, however, the sea water droplet starts as a mixture of sea water and insoluble (often colloidal) OM, then after evaporation of water some of the sea salt got replaced by OM, hence the term fractional OMss. During winter the OM in sea water is almost negligible, but during summer it is far more abundant, especially its colloidal form (Facchini et al. 2008). The gradient flux method relies on concentration gradient which is affected by the sea salt replacement effect.

We agree that the effect may not be proven by the flux-gradient method, but the observations are consistent with each other. We have added a cautionary statement considering the uncertainty of the method and clarified the text.

"However, considering the uncertainties of the flux-gradient method, a quantitative proof of the aforementioned effect was not possible."



- Line 762: Please support this statement by providing, for example, the average WIOM concentrations in summer and winter.

Done. The observed WIOM concentrations during summer were 0.1 - 0.44  $\mu\text{g m}^{-3}$  versus 0.06 - 0.19  $\mu\text{g m}^{-3}$  during winter.

- Line 764-768: How is this possible? The fluxes are directly proportional to the chemical gradients (Eq. 1). Shouldn't both quantities display similar seasonal variations?

When all things equal then fluxes are directly proportional to gradients. However, in case of WIOM we have varying biological activity in the flux footprint area which is breaking direct proportionality. Scenario 1: little biological activity in the footprint area producing small gradient, but the wind and corresponding turbulence can be low or high resulting in low to moderate flux at best. Scenario 2: high biological activity and respective WIOM concentrations in the flux footprint area and either low or high wind speed will result in moderate to high flux. Consequently, a moderate WIOM flux can be a result of low and high wind speed with varying biological activity in the flux footprint area. Moreover, biological activity in the footprint area may be different from the biological activity in the North East Atlantic region due to general patchiness of phytoplankton blooms.

- Line 782: Why can the Yoon et al., (2007) study, which is based on satellite Chl a concentrations, be used to investigate the link between the sign of the WIOM gradient (production or removal) and coastal Chl a concentrations, but the same thing can't be done directly with this dataset because satellite Chl a concentrations aren't accurate enough?

Yoon et al. studied relationship between concentrations and chlorophyll-a concentrations in a large region of North East Atlantic while the fluxes depend on biological activity closer to the coast, in the flux footprint area where chlorophyll-a concentrations cannot be estimated correctly due to poor resolution of satellite images and large errors at coastal interfaces.

- Line 784 (and Lines 145-152): Please state explicitly the 'open ocean' region that Chl a concentrations used in this paper were calculated over? What percentage of this region did the flux footprint region take up?

Open ocean region was 10x10deg or roughly 1000x1000km upfront of Mace Head as in Rinaldi et al. (2013). This has been added to the text in chapter 3.4. Flux footprint area (10x10km at best) would constitute <0.0001 fraction of the above region.

- Line 787: Where is it shown that "...WIOM fluxes were dependant on biological activity in the flux footprint area (0.2-10 km)"?

Lines 769-778 when discussing WIOM fluxes clustering with respect to seasons.

- Line 790: How is this conclusion arrived at? Specifically, how does it follow from the results presented and discussed here that the sea spray source function presented by Ovadnevaite et al., (2013) can be justifiably combined with the primary OM parameterisation of Rinaldi et al., (2013)? This conclusion should be further supported or removed.

We decided to remove Lines 790-796 from the text as the consideration in question does not arise from the results of this study.

Table S1. Measurement uncertainties of concentration profiles for individual chemical species.

ID sample	Height	NH4	NO3	WSON	WSOC	WIOC	Na	SO4	dma	dea	Oxa	MSA	Nss SO4
MH100408	3	6%	5%	92%	16%	11%	5%	5%	<DL	<DL	5%	5%	6%
MH100408	10	5%	5%	61%	13%	10%	5%	5%	<DL	<DL	5%	5%	6%
MH100408	30	5%	5%	55%	9%	9%	5%	5%	9%	15%	5%	5%	5%
MH240408	3	8%	5%	49%	10%	12%	5%	5%	<DL	<DL	5%	5%	6%
MH240408	10	5%	5%	27%	8%	10%	5%	5%	<DL	<DL	5%	5%	6%
MH240408	30	5%	29%	77%	6%	9%	5%	5%	9%	15%	5%	5%	5%
MH290408	3	29%	5%	<DL	49%	26%	5%	5%	<DL	<DL	<DL	5%	6%
MH290408	10	32%	5%	<DL	22%	29%	5%	5%	<DL	<DL	<DL	5%	6%
MH290408	30	13%	24%	10%	7%	15%	5%	5%	9%	15%	<DL	5%	5%
MH270508	3	5%	5%	<DL	8%	12%	5%	5%	<DL	<DL	5%	5%	6%
MH270508	10	5%	8%	41%	8%	10%	5%	5%	<DL	<DL	10%	5%	5%
MH270508	30	5%	<DL	125%	7%	10%	5%	5%	9%	15%	<DL	5%	5%
MH250608	3	20%	5%	34%	7%	15%	5%	5%	9%	<DL	5%	5%	7%
MH250608	10	5%	11%	77%	6%	11%	5%	5%	<DL	<DL	5%	5%	5%
MH250608	30	5%	<DL	<DL	5%	11%	5%	5%	9%	15%	<DL	5%	5%
MH300608	3	<DL	5%	<DL	22%	22%	5%	5%	<DL	<DL	5%	5%	6%
MH300608	10	<DL	5%	105%	19%	15%	5%	5%	<DL	<DL	5%	5%	6%
MH300608	30	7%	7%	15%	13%	13%	5%	5%	9%	15%	5%	5%	5%
MH140708	3	5%	32%	28%	6%	10%	5%	5%	9%	15%	5%	5%	5%
MH140708	10	5%	<DL	15%	5%	12%	5%	5%	<DL	<DL	5%	5%	5%
MH140708	30	5%	<DL	28%	5%	13%	5%	5%	9%	15%	5%	5%	5%
MH220808	3	5%	5%	23%	5%	19%	5%	5%	9%	15%	5%	5%	5%
MH220808	10	5%	5%	13%	5%	24%	5%	5%	<DL	<DL	5%	5%	5%
MH220808	30	5%	41%	63%	5%	21%	5%	5%	9%	15%	5%	5%	5%
MH080908	3	54%	5%	19%	11%	12%	5%	5%	<DL	15%	5%	5%	7%
MH080908	10	85%	5%	8%	7%	10%	5%	5%	<DL	<DL	5%	5%	6%
MH080908	30	5%	5%	18%	8%	12%	5%	5%	9%	15%	5%	5%	6%
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MH111208	3	17%	8%	<DL	38%	13%	5%	5%	<DL	<DL	7%	27%	9%
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MH140109	3	23%	5%	<DL	23%	15%	5%	5%	<DL	<DL	10%	<DL	21%
MH140109	10	<DL	8%	<DL	13%	14%	5%	5%	9%	15%	11%	<DL	16%
MH140109	30	<DL	7%	52%	11%	25%	5%	5%	9%	15%	<DL	<DL	13%
MH250209	3	5%	5%	100%	10%	12%	5%	5%	<DL	<DL	5%	5%	7%
MH250209	10	5%	5%	12%	5%	14%	5%	5%	<DL	<DL	6%	5%	6%
MH250209	30	5%	5%	678%	8%	13%	5%	5%	9%	15%	5%	5%	6%
MH040309	3	5%	5%	<DL	<DL	5%	5%	5%	<DL	<DL	7%	5%	14%
MH040309	10	5%	5%	<DL	24%	13%	5%	5%	<DL	<DL	<DL	5%	12%
MH040309	30	6%	6%	40%	26%	17%	5%	5%	9%	15%	<DL	6%	7%
MH050509	3	5%	5%	33%	10%	11%	5%	5%	<DL	<DL	16%	5%	6%
MH050509	10	5%	5%	10%	6%	13%	5%	5%	<DL	<DL	9%	5%	6%
MH050509	30	5%	12%	23%	6%	19%	5%	5%	9%	15%	8%	5%	5%

Goldstein, A. H., Fan, S. M., Goulden, M. L., Munger, J. W., and Wofsy, S. C.: Emissions of ethene, propene, and 1-butene by a midlatitude forest, *Journal of Geophysical Research: Atmospheres*, 101, 9149-9157, 10.1029/96JD00334, 1996.