REVIEWER 1

The manuscript summarizes results from previously published papers (e.g., Ceburnis et al., 2011; Decesari et al., 2011; Kunz et al., 2002) and concludes, based on these earlier results, that the clean air sampling criteria in use at Mace Head ensures that anthropogenic and coastal influences on aerosol are minimized. Hence, aerosols sampled with the clean air criteria are primarily marine in origin. This conclusion is reasonable and, at the same time, not particularly new. Especially since the data are presented in very general terms. The reader is left with the sense that, on average, 80% of the OM sampled at Mace Head is of marine origin during clean air sector sampling. This perfunctory analysis does little to address the anthropogenic/combustion derived OM in terms of variation and quantification of concentration. Based on published data, BC is persistently present at Mace Head - even during periods that fit the clean air sampling criteria (e.g., see the organic plume reported by Ovadnevaite et al., 2011). There must be an organic component that accompanies the non-negligible BC. How does the concentration of anthropogenic/combustion OM vary with meteorological conditions during periods of "clean air"? Answering this question instead of assuming the anthropogenic/combustion OM component is unimportant for the interpretation of Mace Head results, would strengthen the paper. Additional concerns are listed below.

We thank the reviewer for the acknowledgement that the conclusions are reasonable. The conclusion may not be new, but has been disputed, hence the motivation for the paper. The variability of BC or more precisely equivalent BC (EBC) can be and has been studied at high resolution with typical concentrations of 10-20 ng m-3 occasionally reaching but never exceeding 50 ng m-3 observed at Mace Head in clean maritime air masses. It must be stressed that EBC significantly overestimates BC concentration by up to 50% or more due to reasons discussed below. There is indeed an OM component that accompanies BC concentration, but we have only three samples where the anthropogenic fraction of OM was unambiguously determined by isotope analysis. Ceburnis et al. (2011) reported (Table 2 of Ceburnis et al. (2011)) that anthropogenic OM varied in the range of 13-24% or 24-79 ng m-3 in clean marine air masses and the variability could not be linked to specific air masses (Figure 1 of Ceburnis et al. (2011)).

p. 7312, line 8: "...thereby leading to artificially high values to aerosol parameters...". What does this mean?

Parameters changed to concentrations

p. 7312, lines 9 – 10: Has the objection really been a ": : :.dominance over or drowning out of a natural marine aerosol signal" or, rather, that the contribution of non-marine sources to the aerosol has not been adequately quantified (or acknowledged)?

Yes, the former.

p. 7312, lines 14 – 17: Are these regression results for the "clean air sampling criteria" or do they include all data?

The regression results are indeed presented for the clean air sampling criteria only. It is now noted in the abstract.

p. 7314, lines 27 – 29: Good question. The answer should be informed by an estimate of the fraction of OM that is of non-marine origin. That estimate is never quantified in this paper.

This statement is not correct. The quantitative estimate of the contribution of non-marine sources at Mace Head based on carbon isotopes is given by Ceburnis et al. (2011) and used to defend the argument that clean marine air is dominating over non-marine advections under strictly controlled sampling conditions. Similar results are seen in HNMR analysis.

p. 7315: Line 8: Define "clean sector".

It is now changed to "marine sector" which is 190-300 deg sector.

p. 7316, line 27: 700 1/cm3 is several factors higher than the particle concentrations observed in remote regions of the Southern, Pacific, and Arctic Oceans. Why was such a high concentration used to define baseline conditions?

This is not necesssarily true. See O'Dowd, CD, B Davison, JA Lowe, MH Smith, RM Harrison, and CN Hewitt, Biogenic sulphur emissions and inferred sulphate CCN concentrations in and around Antarctica, J. *Geophys. Res.,* 102, 12,839-12,854, 1997.

Further, the threshold value of 700 particles cm-3 is based on long term observations of the marine sector particle concentration measurements along with BC as reported in Cooke et al. (1997). Therefore, particle concentration limit is tightly related to BC concentration of <50 ng m-3 at 95% probability. It must be stressed that BC concentration of less than 50 ng m-3 typically is 10-20 ng m-3 as documented by Cooke et al. (1997). The lower particle number limit value however desirable would exclude many air masses, laden with either sea spray particles or secondary aerosol particles produced in the boundary layer (O'Dowd et al., 2010). For instance, in stormy conditions as outlined in Ovadnevaite et al. (2012) the number of submicron sea spray particles can reach 700 particles cm-3 and above when BC concentration is less than 10 ng m-3. This issue is further addressed in response to questions raised by Reviewer 2.

Cooke, W. F., S. G. Jennings, and T. G. Spain (1997), Black carbon measurements at Mace Head, 1989– 1996, J. Geophys. Res., 102, 25,339–25,346.
Ovadnevaite, J., Ceburnis, D., Canagaratna, M., Berresheim, H., Bialek, J., Martucci, G., Worsnop, D. R., and O'Dowd, C.: On the effect of wind speed on submicron sea salt mass concentration and source fluxes, J. Geophys. Res., 117, D16201, doi:10.1029/2011JD017379, 2012.
O'Dowd, C., Monahan, C., and Dall'Osto, M. On the occurrence of open ocean particle production and growth events, Geophys. Res. Lett., 37, L19805, doi:10.1029/2010GL044679, 2010.

p. 7317, lines 1 – 2: BC concentrations of 50 ng/m3 inferred from an aethalometer measurements are also high for baseline conditions. Why was this limit chosen? Also, the aethalometer actually measures the change in transmission through filter paper. An assumed mass absorption efficiency is used to convert transmission (or absorbance) to BC mass concentration. Was a uniform mass absorption efficiency assumed? Whatwas it? What is the uncertainty associated with the derived BC mass concentrations?

The BC value of 50ng/m3 is the limit value and should not be confused with the actual values. The typical values in sectored marine air masses are typically 10-20 ng m-3 (Cooke et al., 1997; Junker et al., 2006). The mass attenuation coefficient of aethalometer was 16.8 m2 g-1.

Junker et al. (2006) indicated that thermo-optical method suggested a much higher value (35 m2 g-1) in clean marine sector samples which would result in even lower threshold BC value of 24 ng m-3 and the typical clean marine sector values of only 5-10 ng m-3 which is then comparable to BC values from the pristine environments. In summary it suggests, that BC values reported for MH are most likely systematically biased high, again strengthening our argument.

This issue is further addressed in response to questions raised by Reviewer 2.

Junker, C., Jennings, S. G., and Cachier, H.: Aerosol light absorption in the North Atlantic: trends and seasonal characteristics during the period 1989 to 2003, Atmos. Chem. Phys., 6, 1913–1925, doi:10.5194/acp-6-1913-2006, 2006.

p. 7317, last paragraphs: Are there examples of Savoie et al. being used to "define more recent experiments in terms of source apportionment"? Provide references.

The word "define" should be replace by "refute" and the sentence reads "Consequently, the results from that experiment simply cannot be used to <u>refute</u> more recent experiments in terms of source apportionment".

p. 7319, lines 3 – 4: What does "TOA" stand for? What region of the Pacific was sampled? What does "VOCALS" stand for? What "Pacific waters" are being referred to here? How was "clean marine air" screened for?

It should have been spelt TAO – Tropical Atmosphere Ocean, in the tropical Pacific region of 8 deg North – 8 deg South. VOCALS - VAMOS Ocean-Cloud-Land-Study, in the southeast Pacific region of 15-30 deg South. Clean air conditions were defined as BC <4.5 ng m-3 and CO<61 ppb..

p. 7319, lines 5 - 9: Very confusing. Based on Figure 3, the off-line and AMS data that are referred to are from Mace Head. This should be made clear in the text and the figure caption.

Has been clarified that AMS and off-line data refer to Mace Head.

p. 7319: Many typos to be fixed.

Fixed.

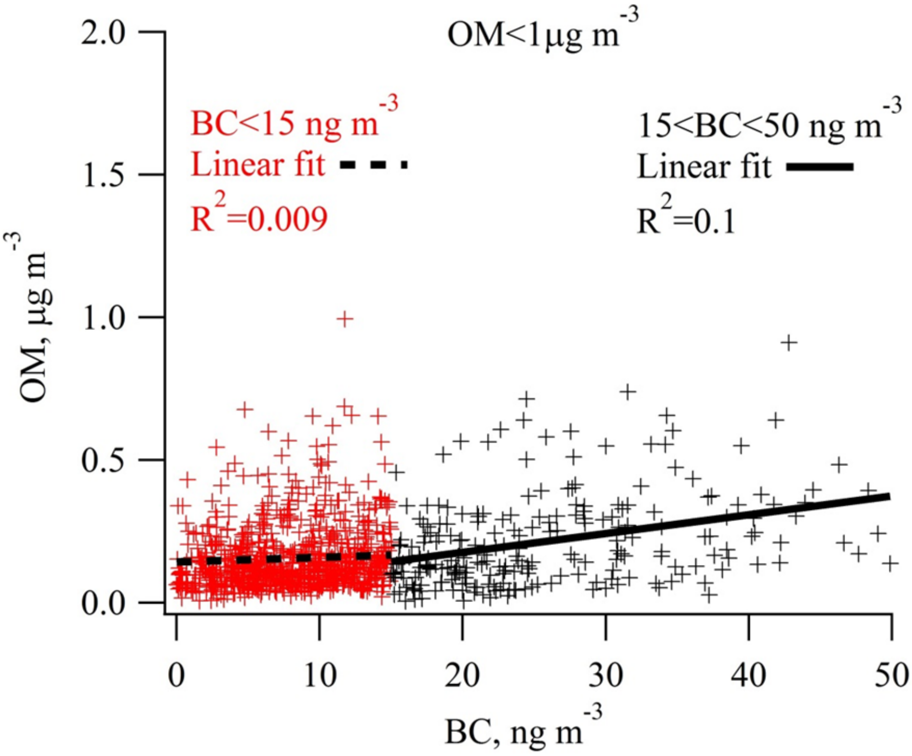
p. 7319, line 17: Should be changed to ": : :the OM reported as marine OM at Mace Head is DOMINATED BY CLEAN MARINE OM AEROSOL." Given that equivalent BC mass concentrations up to 30 or 40 ng/m3 were measured during the "marine primary organic aerosol plume" reported by Ovadnevaite et al. (2011), some fraction of the measured OM had to have a combustion origin.

Done, but also, it must be noted that Ovadnevaite et al. (2011) demonstrated short spikes of 20-30 ng m-3, not 30-40 ng m-3. Slightly elevated values of EBC (equivalent BC) can be a result of absorbing organic matter which has been suggested in a number of papers (e.g. Cappa et al., 2008).

Cappa, C. D., D. A. Lack, J. B. Burkholder, and A. R. Ravishankara (2008), Bias in filter-based aerosol light absorption measurements due to organic aerosol loading: Evidence from laboratory measurements, Aerosol Sci. Technol., 42, 1022–1032, doi:10.1080/02786820802389285.

Figure 3: Were the linear regressions calculated assuming no error in an independent variable? If so, it is difficult to assess the results of the linear fit given that the BC mass derived from the aethalometer, especially at lower concentrations, most likely has a fairly high uncertainty. Also, in the two left-most panels, there appears to be one outlier with high OM and low BC concentrations. If this outlier is removed and both variables are assumed to have errors, what does the fit look like? In the two middle panels there again appears to be two populations, one with high OM concentrations and one with low OM concentrations. What happens to the fit if these are treated separately? It is difficult to see the correlation between the low OM and BC concentrations because of the scale used on they y-axis. Similarly, in the two far-right panels, there appear to be two populations. What happens to the fit if they are treated separately?

We do not understand how the random uncertainty of BC measurement can affect the presence or absence of the correlation. Moreover, there is no scientific justification in removing a part of the dataset. It sounds like someone would know in advance that the correlation between BC and OM must exist and it is a matter of data treatment to demonstrate it. Nevertheless, we attempted the suggestion to remove the high OM concentration data which once again resulted in no-correlation between OM and BC. A weak relationship, however not statistically significant, most likely is a result of OM absorption as suggested by Cappa et al. (2008) and supported by the inadequate mass attenuation coefficient suggested by Junker et al. (2006) and discussed above.



p. 7319, line 18: change to “ratio”

Done

p. 7319, line 21: change to “additional”

Done

p. 7319, lines 27 – 28: Change to “: : :is DOMINATED BY NATURAL MARINE OM”.

Done

p. 7320: The results of Lin et al. (2012) are not relevant to Mace Head. As the manuscript points out, air masses sampled at 36N are not impacted by the same meteorology as those sampled at Mace Head. If anything, given the non-negligible fraction of anthropogenic SO4 found by Lin et al. (2012), these results weaken the main argument of the paper. I would remove this discussion.

The other reviewer considers this study as important, but needs better context and information.

Figure 4. Sample number is meaningless to the reader. Longitude should be plotted on the x-axis. In any case, all samples except one indicate a significant contribution from anthropogenic sources.

We have no right to modify a Figure under copyright. We clarified Longitude in Figure caption.

p. 7321: The results of Ceburnis et al. (2011) are summarized here and the reader is told that, during clean air sampling, 80% of the carbonaceous aerosol was marine. How does the concentration of non-marine OM vary with changing transport regimes? How does the non-marine OM correlate with measured BC? Also – there is no reference to Figure 5 in the text.

Ceburnis et al. (2011) reported carbon isotope analysis of the total carbon, therefore, BC mass, however, small, was part of the result, i.e. 20% of non-marine carbon was OC+BC. The referee’s request is not clear – “changing transport regimes?” the issue of marine OM v BC is discussed above. The source apportionment is for the specific Mace Head marine sector scenario and associated transport regimes.

Reference to Figure 5 is added.

Figure 6: What is displayed along the x-axis? Does each bar correspond to one sample? If so, it appears that almost every sample collected from the marine sector contains some anthropogenic influence. Indeed, the Descesari et al. results appear to agree with those of Ceburnis et al. that about 20% of the OM at Mace Head has a combustion origin under the clean air sampling criteria. Under clean air sampling criteria, anthropogenic/combustion OM may not “drown out” the marine OM, but it should be quantified and acknowledged in all results reported from Mace Head.

The individual samples are displayed along the x-axis. The sampling period is imbedded in sample notation. Figure caption is clarified.

p. 7322, line 4: change to “revealed”

Done

Figure 8. The figure shown does not correspond to the figure caption.

Figure caption is corrected to “HR-ToF-AMS mass spectra (major organic family CH and CHO only) of marine organic plume detected on the 15th–16th August 2009 at Mace head, Ireland”.

p. 7322, last paragraph: First sentence needs to be fixed.

Corrected

Figure 9 caption: Right panel appears to be only for the period of the cruise, not for two seasons as is stated in the caption.

Figure caption was clarified by insertion”(b)”.

p. 7325, first paragraph: Should be Coe et al. (2006).

Corrected

p. 7326 - 7327: Many typos to correct.

Done

p. 7328, line 26 – p. 7329, line 2: Under the clean air sampling criteria, marine OM appears to dominate the total OM mass measured at Mace Head. The presence of BC, however, indicates that there is an anthropogenic/combustion component of the OM. What fraction is this component under the different meteorological conditions that prevail during the clean air sampling criteria?

Considering several lines of evidence the fraction of anthropogenic OM component at Mace Head under the clean air sampling criteria is 10-20% depending on the air masses and meteorological conditions during sampling. That includes carbon isotope method, AMS fingerprint, HNMR all of which correspond to low BC concentrations.

p. 7329, lines 6 – 8: A decrease in anthropogenic SO4 sampled at Mace Head does not necessarily imply a decrease in anthropogenic/combustion OM.

Accepted, we never claim so. The decrease of sulphate and the reasoning behind should be considered alongside source apportionment using carbon isotopes (Ceburnis et al., 2011) as anthropogenic particles are fully internally mixed after the 3000 km transport over the ocean.

REVIEWER 2

For the past several decades, Mace Head has been a key measurement site for assessing marine aerosol and gases transported from the North Atlantic. However, as pointed out in this paper, there have been concerns regarding issues of the long range transport of pollution at the site and its influence on assessing "marine" properties as well as the contributions of enhanced coastal influences relative to open ocean values. A paper directly addressing these concerns is certainly warranted and of interest.

The title suggests this will be the "go to" paper for readers to satisfy any concerns that have been raised over Mace Head data. However, this paper needs greater rigor if it is to live up to this expectation. Perhaps understandably, the paper is written from the point of view of one who is largely satisfied with Mace Head data. Certainly MH data and resulting papers have helped highlight the significance of a marine OM source. However, in reading this paper I get a sense that some relevant and nagging issues have been "papered over". Some specific concerns are identified below.

In summary, I think this is potentially an important paper but it needs to be more convincing. The authors need to make a greater effort to address issues such as those identified here.

The paper also appears to have been written in haste and there are also an excessive number of places plagued by poor grammar and incorrect spelling etc. I will not comment on this here but significant editing for this is necessary.

Text is tidied up now.

P7314, Lines 29-: In order to answer this question, the authors need to demonstrate that major experiments carried out there over the years (and resulting papers) employed sufficiently robust criteria to ensure their results were not compromised or biased by the problems noted by others (and in this paper).

The entire paper was devoted to demonstrating that the major experiments carried out over the years either did or did not employ sufficiently robust criteria to ensure the validity of conclusions made in this and other papers.

P7315: This is rather a cursory description of the facility. Exactly what heights have been used on the tower over the years? Inlets and plumbing used? What standard equipment has been in place and are archived data sets available to examine etc. What is the evolution of sampling strategy and criteria over the years and why? Is their archived data available, etc?

More details on sampling have been added to the section with regard to inlets, sampling heights and equipment. Some archived data are, indeed, available upon request and many researchers have used our data in their papers contributing to over 400 papers published over 20 years mentioning Mace Head facility.

Pg 7316 Lines20-to next page L5: Describes "new" sampling criteria employed since 2001. These are said to include: Wind direction (already noted as not reliable)

The sentence was rephrased "... not only wind speed, but also..."

Total Number < 700 cm-3 - Need a clear discussion of why this number was chosen. Numerous experiments in Atlantic, Pacific etc. in past two decades using these instruments find clean aged marine concentrations are typically well below 700 cm-3 and excursions to these values can often indicate pollution. Conversely, very clean scavenged air is often associated with nucleation with total numbers far larger than 700 cm-3. Hence cleanest air can also be excluded? How about at least a long term scatterplot of number vs BC when number is less than 700cc and there is no evidence of nucleation(there have been a number of studies of nucleation at the site).

This question was already raised by Reviewer 1 (and addressed above). In addition, some of the clean air can, indeed, be excluded, but the conservative approach is useful in avoiding significant contamination of samples by anthropogenic air masses. Exclusion of some clean air is certainly not a problem if the integrity of the signal is preserved.
The choice of CN and BC thresholds are somewhat arbitrary; however, the choice 700 CN m-3 is consistent with studies in the remote Southern Hemisphere, for example, O'Dowd et al., (1997) found that total CN in pristine air below 53 deg S over the South Atlantic and Weddell Sea, Antarctica, while generally less than 500 cm-3, often reached 700 cm-3 in clean air outside new particle production events. Further, Dall'Osto et al, (2010) found the CN frequency distribution was bimodal, with a background mode concentration of ~700 cm-3. For BC, O'Dowd et al., (1993) found NE Atlantic background marine air masses comprised BC mass concentrations between 10-35 ng m-3 while the lowest BC mass in modified marine air was observed to be 208 ng m-3. Cooke et al., (1997) provided statistical evidence that marine air masses with CN concentrations less than 700 cm-3 were associated with BC concentrations less than 75 ng m-3. Based on these studies, it seemed reasonable to select the more strict threshold of 50 ng m-3. The threshold seems to be justified when the correlation between CN number and BC mass concentration is analysed. For a typical OM plume event, shown in Figure 2, lasting about one day, BC mass ranged from 4-27 ng m-3 for CN less than 700 cm-3 with no correlation being evident (R^2=0.05). For an extended period from 1st January – 31st May, 2009, comprising of 995 hours of data that meet the criteria of CN less than 700 cm-3, wind direction 190-300°, and non-recirculating marine-sector air mass back trajectories, no correlation is observed either (R^2=0.06). Regarding the extended case, 1.6% of the data records reside outside the 50 ng m-3 threshold; however, the contaminated samples amount to a negligible percentage of data in this category which are removed in any event for exceeding the 50 ng m-3 threshold. The lack of correlation between CN and BC below the 50 ng m-3 threshold suggests a robust set of criteria for marine background air mass sampling.

New figure 2

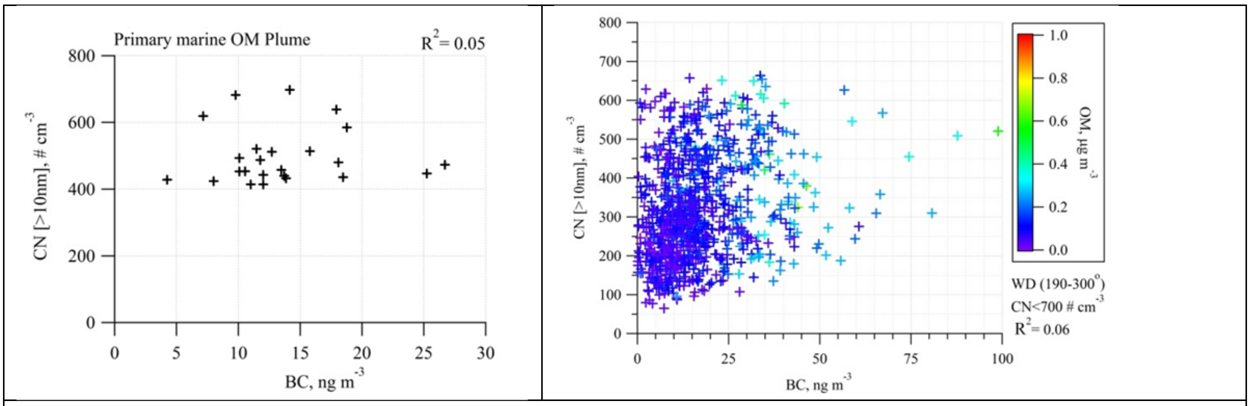


Figure 2. (left) scatter plot of CN versus BC for organic sea-spray event lasting approximately

30 hours. (right) scatter plot of CN versus BC for 5 month extended (1st January to 31st May, 2009) period of combined wind direction (190-300 deg) and CN (<700 cm-3) concentration sector control.

BC less than 50 ng m-3. Where did this criteria come from. How does it compare to values from other clean marine sites etc.? Is there a relation between excursions in number and BC below these criteria when recent nucleation is not evident?

Please refer to the above question and the answer to the Reviewer 1 question.

P7316: "More sophisticated sampling" is mention but not clearly described. For example, in the Cavelli paper referenced here that discusses OC measurements they state their Berner impactor was placed only 3 meters above the surface and 50 m from the shoreline (Fig. 1d). How many other papers used this sampling location? This is also much lower than altitudes mentioned later for other instrument in different papers. It is impossible to believe that aerosol at this altitude was not impacted by coastal SSA and emissions produced at the nearby shoreline, not to mention the offshore island influences mentions below. Anyone who has lived by the shoreline knows plumes from surf like Fig 1d are often visible to the naked eye. It would seem likely that some effects of these coastal aerosol influences are expressed in the data. For example, offshore number concentrations tend to be slowly varying but coastal breaking waves can induce spikes of 50cc over tens of seconds and should be evident in low tower data a substantial part of the time.

Only 6 samples were reported in Cavalli et al. (2004) (refer to Table 1). From then on all sampling was moved to 10m level to avoid the implicated surf-zone emissions. Indeed, some spikes of particles can occur over the time scale of seconds, but those numbers are small against the typical sea spray concentrations of several hundred particles per cm3. As already discussed by Ceburnis et al. (2008), particle flux footprint (when sampled at 10 m height) is several hundred meters from the sampling point (and therefore encompasses a fraction of emissions from the surf zone), but the concentration footprint, i.e. the footprint of the entire aerosol population sampled on the filter or the impactor for many hours, is several tens or even hundreds of kilometres.

This brings up an important issue. The question is to what extent coastal sources might influence the interpretation or conclusion of a given paper. I feel that early on (maybe in the MH facility section) there needs to be some summary table of the history of measurement locations and criteria used for sampling at MH and which ones were employed for what measurements in the papers referenced in the text (and other papers if possible). Elsewhere, the paper indicates claims no coastal influences present at 7m and above (P7327 L11) while others show an influence. This Table and discussion could be part of the initial "Facility" section.

We agree with the reviewer and the proposed Table was added to the paper.

The paper would benefit from a clear presentation/summary of height resolved comparisons for measurements over the MH tower that have presumably been done during various "clean sector" conditions, other than the Coe reference that was supposedly made under anomalous more polluted conditions.

The requested information is included in the above Table.

Morover, the referenced Cavelli paper is one of many from MH that employ back trajectories over the ocean. Such trajectories (say over 4 days) are mentioned as a clean air criteria in several other papers. However, many of these trajectories shown in Cavelli were in the free troposphere 4 days earlier and suggest entrainment of FT air upwind of MC is an expected influence (eg, seasonal boreal biomass burning over Canada know to regularly advect over the North Atlantic) on boundary layer aerosol. Such air is usually transported without scavenging in the FT such that a 4 day trajectory should never be used as a criteria for clean marine air. My intent is not to single out the Cavalli paper but I feel these are just a few of the issues that should be addressed, explored and communicated if this is to be a "go to" paper for MH.

Back-trajectory analysis was used as a supportive, but not the definitive argument in many papers, including Cavalli et al. (2004). Following the reviewers argument, back trajectories should never be used in any context or purpose as there will always be reasons not to trust them. We did not over interpret backward trajectories, only discussed them as suggested by NOAA. In case of Cavalli et al. and our many other papers, they are indicative of clean marine air masses which have spent at least 4 days over the ocean and the last 48hours in a turbulent boundary layer, facilitating wet deposition of pre-existing aerosol particles and replenishment with nascent sea spray aerosol particles.

P7317, Lines7–: Reference to Fig. 2 and caption argues a 1:1 relation in plot indicates data is "true marine". Actually, it just says when conditions meet the more recent criteria then the Savoie sulfate data and the new data will agree, as they must. A "truly marine trajectory" does not indicate it is only marine air or aerosol.

Figure 2 does indicate that the new sectoring system is far superior to the system employed by Savoie et al. in terms of sampling the least perturbed marine air masses. However, the results of this comparison should be considered together with other lines of evidence (carbon isotopes, AMS, HNMR), all of which are presented later in the text.

P7318 Top: Arguments are made that anthropogenic sulfate has been significantly reduced at MH since the earlier Savoie study so that now it is less likely influenced by anthropogenic aerosol. This remains unclear as the criteria have also changed and the total sulfate was earlier argued to be recirculated European pollution that should be exclude from consideration of "so called" clean marine conditions. What change has there been in this clean marine sulfate. This argument needs to be made more robust. While improvements over the Savioe criteria are clear, the question remains "to what extent does the MH sampling protocol ensure MH data effectively excludes pollution (local or long range) or coastal effects such that clean marine air is ensured.

As our clean sector sampling protocol did not change over more than a decade (started in 2001), initial comparison with Savoie et al. system (operated intermittently from 1989 to 2004) does give a clear idea of the amount of re-circulated pollution. The decreases of sulphur emissions all over the Northern Hemisphere have decreased anthropogenic sulphate concentrations and consequently the fraction of non-marine sulphate over the North Atlantic, thereby, increasing the fraction of marine sulphate. Without isotope analysis, one cannot be more quantitative.

P7318: Section 3.2 Organic mass– As this paper focus is on MH data, it seems odd and out of place to start the discussion of MH OM with a protracted discussion of results for the clean

Pacific (Shank et al.). I expected a summary of the findings from MH and some discussion of their pertinent issues affecting them before trying to make comparisons to Pacific data.

The reference to Pacific Ocean has been moved. The section has been restructured.

Lines 25–: The authors point out the Shank et al. paper claims to only describes the submicron OC mass. Elevated OM from marine sources is recognized in Shank et al. but the issue they raise is over the 2008 O'Dowd et al. paper and their Figure2 where they also report the submicron %OM vs. chlorophyll. Shank et al. argue that their regional mean submicron data falls on the lower envelope of the O'Dowd data and the resulting dependency goes through the origin, leading them to suggest a possible non-marine source contribution to the O'Dowd data. This would appear to be the more relevant concern for this this paper.

This is indeed the motivation of our defence – there are no rational grounds for Shank et al to conclude that because we see higher OM over a different ocean that the OM is non-marine.

P7319 Fig 3 data: The authors seem to miss the point here. Even assuming occasions with large marine sources of OM, the question remains as to how much a continental source of OM (known to exist) might contribute to OM measurements at MH in general and not just isolated events evident in Fig 3(middle).

We presented several lines of evidence in support of our claim that under strict sampling criteria the overwhelming amount OM at Mace Head is of marine origin. The results from AMS study are just first in line followed by other evidence from carbon isotope analysis, HNMR, etc. We are not necessarily concerned with the general OM measurements at Mace Head but more so with OM in marine sector sampling.

Fig. 3 (top middle) shows two distinct groupings of data that would suggest separate analysis is needed. [BTW-The 3 years of Yoon AMS data are indicated as 1hr data but this seems unlikely as that would suggest about 60 hours of the high OM data. Are data points averages over days? weeks? or ??] Instead of separating these out, they appear to be grouped together to generate correlations. Without doing more sophisticated stratification (preferable) just eliminating the highest OM grouping should be done to explore what the vast majority of the MH data has to say. This should be shown and the OM scale expanded to 0-1 ug/m3 with a fit. Presumably other variables are available that might enhance stratification and understanding.

Figure 3 Top left graph is clearly indicated as "OM versus BC for off-line sampling (~70 h over 1 week per point) from Yoon et al. (2007)". It was ~70 hours over 1 week period. Berner impactor data were purposely separated from high time resolution AMS data to demonstrate the absence of correlation in both Berner and AMS data sets. The 3 years from the AMS is from this study; the high OM data are indeed 1 hour, see Ovadnevaite et al., 2011. Regarding stratification of AMS data, please refer to the same Reviewer 1 comment.

Here the Shank et al. plot of AMS OM vs. BC(their Fig 5c) would appear to extrapolate to a value of about 0.6 ug m-3 OM at 40 ng m-3 BC expected to be due to combustion sources. This value is close to the black line value fit [Fig. 3 (top middle)] through most of the MH data at this concentration. If this does represents a combustion source at MH then the density of data points shown in this three year data set indicates this source might dominate typical OM values at MH. This is an issue of direct relevance to this paper and to extrapolation of MH data to other regions.

We strongly believe that the extrapolation of the results from one study to another is scientifically unjust and biased as the context between the studies is very different. Moreover, following the reviewers argument, we would like to point out that at BC concentration of 40 ng m-3 Shank et al. reported OM concentrations in the range of 0-1.5 ug m-3 (Figure 5a). Hence, picking the extrapolated value of 0.6ug/m3 based on filtered data (Figure 5c) has no scientific value as it does not take into account measurements of elevated BC values associated with very low OM values. The organic matter concentration can be either extremely low or very high in polluted air masses – where does that lead us? It was not our intention to discuss every number in Shank et al. paper if that same paper did not imply that Mace Head data were significantly affected by anthropogenic particulate matter based on highly questionable extrapolation.

Similar concerns over Fig 3 (bottom middle). This may also need log scale for clarity. What is the fit to the lower group of values for BC>5 ng m-3? Presumably most of the data averages around the value of 28 reported in Table 1 and lying between the Bates values for NE USA of about 20-34.

It is true that the Mace Head ratio based on the long-term impactor measurements falls in between the Bates values for NE USA. However, contrary to Ceburnis et al. (2011) study, Bates et al. did not perform as comprehensive quantification of fractional contribution of non-marine OM or as detailed characterisation of it as in our studies. It remained unknown whether OM measured in Bates et al. study originated from fossil fuel or continental biogenic sources or some fraction from marine sources. Bates et al. have concluded that the high OM concentrations could have been due to secondary aerosol formation over land. Table 1 data suggest that high OM/BC ratios are associated with biogenic sources (continental or marine), contrary to a low ratio of anthropogenic OM. However, no further conclusions can be drawn what the ratio alone can tell us.

P7320, Lin Study and Fig. 4: This figure is important, even though only one cruise, as it clearly shows the pronounced influence of anthropogenic sources all across the Atlantic. It would be helpful to add some information on wind speeds and direction. Presumably both black carbon and anthropogenic OC is also present, as would be expected from the referenced Bates et al. 2005 showing high OC fraction in this continental air off the east coast of the USA. Table 1 also shows the MH north Atlantic plumes straddle these values at 28 under non OM plume conditions, consistent with the Bates et al. measurements. This appears to indicate a persistent OM contribution at MH from continental sources. This should be discussed and accounted for more directly. While this paper argues more generally that 80% of the clean carbonaceous material is marine, it is not clear how variable this fraction is or what drives the variability. As the title implies, this is the paper should address this.

The study of Lin et al. did not present OC or BC values, they only dealt with sulphur isotopes, so we cannot discuss the requested information. Their study tells us that the contribution of anthropogenic sources across the Atlantic can be variable, especially when sampling is unsectored, with the fractional anthropogenic contribution ranging from 10-80%. It was exactly for this reason that we employed several lines of evidence trying to prove the existence and quantify the contribution of marine sources to OM content observed at Mace Head, instead of discussing OC/BC variation only which presents highly conflicting picture. The

isotope method, HNMR or AMS methods are far superior quantitatively to the OC/BC ratio alone.

Pg7322: Coastal Sources and Artifacts I think logically this section should be presented much earlier in paper as it is fundamentally linked to the site itself.

This suggestion we do not agree with – the sector sampling is more fundamentally linked – it's a perspective issue.

Lines10-12: The authors argue that Lidar data that shows surf spray plumes from upwind islands are visible and readily mixed and dispersed vertical contributes little to the MH measurements. However, the referenced Kunz et al. abstract says– " Using the lidar in the scanning mode allowed 2-D profiling over a spatial scale of 10 km, revealing significant primary aerosol plumes produced by breaking waves, particularly in the surf zone, and at high wind speeds also at open sea. The initial plume heights were some tens of meters, and evolved to hundreds of meters while transported over only a few kilometers from the source. The plumes were traceable to distances of more than 10 km downwind from the source. "

The Kunz figures 6 and 7 clearly show pronounced island sourced plumes of SSA and their enhancement over the open ocean signal. Examples should be included in this paper as part of the motivation for this discussion. Although many factors lead to the choice of a measurement site, ideally one might hope for a site less impacted by such plumes when trying to assess open-ocean conditions.

While the indicated Kunz plumes will vary with environmental conditions, they will certainly contribute to clean sector aerosol under most conditions even if they dilute and mix into background to become no longer visible. Over these distances their influence should be mixed up high enough to impact all altitudes on the MH tower. What needs to be clarified is to what extent these plumes and their SSA and, presumably coastal OC typically enhanced in most coastal regions, contribute to MH measurements, particularly when strong marine offshore OC sources are not active (ie. more typical conditions).

The most relevant and important quote from the Kunz paper is quoted in the manuscript: *"The influence of the background renders it difficult to quantify the effect of locally generated plumes on the concentrations at Mace Head in relation to wind direction and wind speed. In particular because during transport from the islands to the Mace Head station, vertical dispersion causes dilution of the primary aerosol, which brings the concentrations close to background levels. Hence the influence on the concentrations measured at Mace Head is relatively small, if any. An attempt to quantify this effect from direct measurements of aerosol concentrations at Mace Head, based on differences in concentrations between selected wind directions, showed no clear correlation. Also there was no obvious concentration enhancement when the wind was from the directions of the islands. At low wind speed, the concentrations of super-micron aerosol particles at Mace Head exhibited variability of not more than a factor of 2, and less in elevated wind speeds, although this cannot be directly linked to the offshore plumes. The concentrations of sub-micron aerosol particles are not affected [Kleefeld et al., 2002], in good agreement with other studies of surf aerosols [de Leeuw et al., 2000]."*

We feel this could not be more clear.

Lines12—: The offshore increase from a white cap threshold of 4 m/s (arguable) to 8 m/s represents a quadrupling in the drag coefficient and expected SSA production.

***Not sure what the reviewer's point is here, but clearly it strongly supports the author's arguments if the whitecap threshold is 4 m/m. please refer to* Callaghan, A., G. de Leeuw, L. Cohen, C.D. O'Dowd, Oceanic Whitecap Coverage: Measurements from the North Atlantic, *Geophys. Res. Letts.*, doi:10.1029/2008GL036165, 2008.**

The onset for whitecapping during the MAP cruise, was 3.7 m/s.

While higher offshore winds must eventually make these island plumes less distinguishable against the offshore source it is not logically consistent that they would have a "minimal impact". They are certainly still present and their relative influence will be strongly wind speed dependent. For example, suppose coastal OM linked to SSA production were at times say an order of magnitude higher in coastal breaking waves than in offshore waters. The possibility that the plume lidar backscatter approaches offshore values would not imply the coastal contribution to offshore OM would be minimal. Morever, the referenced Kunz paper actually shows pronounced island plume influences evident at 8 m/s (their Fig 6d). I think it is appropriate, if not essential, that one or two panels from this Kunz figure be included and discussed in this paper. The data and discussion in the Kunz paper indicate plume enhancements are often significant for many kilometers downwind of the islands and for 10's of meters in the vertical even if they dilute by vertical mixing by the time they reach MH.

The reviewer's comments are plausible individually, but breakdown in the collective argument, e.g. the onshore-offshore experiments suggest that an order of magnitude higher onshore concentrations are not plausible!

We disagree with the reviewers suggestion to include figures from Kunz et al. the reviewer also seems to misinterpret these figures – the correct text interpretation is quoted below in a non-selective manner.

Quote " Figure 6c shows results that were obtained in NW wind with a speed of 3 m s_1. The results presented in Figure 6d were obtained on JD 178 of 1999 (27 June) in westerly wind with a speed of 8 m s_1. The attenuated atmospheric volume backscatter coefficients inside the plumes are similar to previously observed values, but the ''background'' attenuated atmospheric volume backscatter coefficients are higher due to the surface production of sea spray aerosol by breaking wind waves. The value of the attenuated atmospheric volume backscatter coefficient inside the plume at 500 m from the island is about a factor of 2.5 enhanced with respect to the background. At 4 km downwind from the source the attenuated atmospheric volume backscatter coefficients is reduced to half this value. Another feature observed in Figure 6d are plumes originating from breaking wind waves, visible as very narrow and slightly lighter coloured lines. They are visible over several kilometres".

Lines 25–: What is the reason for this narrowing of the WS at MH. Is a local land induced topographic effect that would have little influence on SSA offshore production or is a larger scale coastal affect that can influence the fetch and production offshore? What impact does this have in trying to relate MH data to offshore data under similar wind speeds?

There is no reason per se. Don't we all know the distribution of wind fields are inhomogeneous – otherwise no flow. We would be worried if the distributions where the same.

Further, the Celtic Explorer cruise path was intentionally directed into an extreme summer storm gusting 34 m s-1, hence, the project experienced the full strength and range of open ocean winds. The Mace Head station, on the other hand, was on the southern boundary of cyclonic circulation and, therefore, only experienced a fraction of the wind speed range. The local topographical features have little effect on the wind speed measured at Mace Head versus the open ocean wind upfront of the station as was demonstrated by Gantt et al. (2011).

Gantt, B., Meskhidze, N., Facchini, M. C., Rinaldi, M., Ceburnis, D., and O'Dowd, C. D.: Wind speed dependent size-resolved parameterization for the organic mass fraction of sea spray aerosol, Atmos. Chem. Phys., 11, 8777–8790, doi:10.5194/acp-11-8777-2011, 2011.

P7324 Referring to coastal sources and Coe measurements at 7m and 22m it is stated that: L6 "While some differences were seen in number concentrations. . .. ." What differences? what sizes? Discuss data.

See response to next comment.

L10 It is also stated that there was no evidence of surface perturbing particles at 7m. But what about the 3m used in the Cavalli and other measurements. Also the Coe paper goes on to say most of the measurement period was not clean and that——— It is difficult to establish whether the organic material in the mode seen at large particles by O'Dowd et al. (2004) as the ultraclean conditions observed by O'Dowd et al. Were not observed during NAMBLEX.

The OM seen by O'Dowd et al is more so seen in the small particle mode – also, while the overall conditions in Coe et al were not 'clean', there were sufficient clean sector periods to support the conclusions! Please note that we have no intention of transcribing text from all relevant papers, rather we cross-reference; however, we do quote in this open access review from Coe et al., 'The size resolved data show that there is little evidence of systematic enhancement of particles at the 7m sampling level, though, a few local sources of pollution enhance the Aitken mode concentration for brief periods at the 7m level but not at 22 m. Even particles in the 1 to 3 µm range showed no evidence of the surface layer perturbing the concentration of particles at 7m above ground level. Figure 4 shows a scatter plot of the submicron sulphate and organic mass as measured by the AMS at the 7 and 22m level. There is no systematic difference in either the sulphate or the organic concentration sampled at the two levels. A few periods show the enhancement of organic mass at 7 m, this correlates with the enhanced 50 nm particle number concentration'.

Moreover, the 2008 Ceburnis reference in discussing tower gradients says: ". . .The difference in concentration (or gradient) between 3 and 10 m reaches 90% of it's value at 1170 m from the coast line, while the difference between 10 and 30 meters reaches 90% of its value at 4840 m (Figure 1). Emissions from greater distances have minimal contribution to the flux footprint and gradient profile in terms of upward fluxes, but clearly, such emissions can influence the absolute concentration which in turn influences the deposition flux magnitude." While this appears to confirm an influence of the upwind islands and intervening waters it does not clarify a near coastal source.

Indeed, the quoted statements from Ceburnis et al. do not exclude an influence of the upwind islands as it does not confirm it either. However, Figure 1 in Ceburnis et al. does show that "surf zone emissions at a range of 80–180 m from the tower could have contributed up to 20% of the difference in concentration between the 3 m and 10 m levels and <5% between 10 m and 30 m level".

Overall, the gradient method used at Mace Head revealed that the concentration gradient (not the concentration itself) is composed of emissions up to 5 km distance from the measurement location. All sources within that distance, starting from surf zone finishing with distant islands can contribute to the concentration gradient, but the exact contribution of individual sources cannot be deduced from the gradient itself. Only additional estimates or experiments reveal the strength of those sources.

Not shown are profiles of concentrations or simple size-resolved data at various tower altitudes that spek to this under under some representative conditions. The 2008 Ceburnis et al. Fig 2 would be worth introducing here before discussing footprints etc. The greatly enhanced sea-salt evident in this figure below 10m on the tower speaks for itself. I suggest the authors include this figure and discuss, particularly with regard to papers using 3m data mentioned earlier.

The greatly enhanced sea salt evident in Figure 2 of Ceburnis et al. study tells nothing in terms of contribution from surf-zone emissions. Sea salt must be enhanced at 3 m level as the sea salt is produced exactly from the surface. Therefore, without being able to quantify the surf-zone emissions precisely, the gradient profile data at 3 meters was not used to quantify fluxes. Only data from 10 and 30 meters were used to calculate fluxes.

P7326: The gradient discussion needs more details provided to support the claims made.

This comment becomes redundant due to above answers.

P7327, Lines3–: “40% higher sea-salt mass” at MH seems to contradict argument of minimal impact of coastal/island influences claimed earlier.

Indeed, 40% higher sea salt in supermicron sizes may suggest an influence of surf zones emission in that size range, however, “no discernable differences for submicron sea-salt is observed”. The whole argument of our paper is to demonstrate that <u>submicron</u> OM originates predominantly from marine sources.

Lines 8-9: Not clear what data is being discussed. Paragraph starts talking about ship CE (2006) with winds typically much higher than MH (Fig. 9). Then talks about de Leeuw data (2000) and enhanced mass arising from winds at MH 1.8 time higher than ship. Please take the time and text to discuss more completely and rewrite for clarity.

We believe the text is sufficiently clear – the reviewer is combining separate and distinct statements from different parts of the manuscript and arriving at the wrong conclusion.

Lines 10– “. . .no discernable enhancement. . .” contradicts 50% increase evident in above mentioned 2008 Ceburnis plot. Please explain.

This comment was already answered with respect to Ceburnis et al. study

Fig 11 : While superficially the three measurements shown in Fig. 11 look similar, because they are expressed as a % sea-salt a direct comparison is not clear. For example, earlier the %OM in coarse sizes at MH were mentioned as being 50% of the total. Here the coarse Celtic explorer % appear substantially less than the MH values. Although this offshore comparison can be problematic, can this be shown more clearly as % of the total OM present in each size class?

This is also confounded by dramatically higher winds (that should produce dramatic increases in sea-salt) on the Celtic Explorer (Fig 9) where even effective sampling can be questionable. One worries about and apples and oranges comparison.

There is no contradiction in Figure 11. The coarse fraction OM indeed contributed to about 50% of the total OM mass. However, fractional OM contribution in sea spray is very small in absolute value due to overwhelming contribution of sea salt in the supermicron range. The caption of the Figure 11 clarified.

P7328, Lines 8-16: Not clear why this point is made. I am not sure who is surprised that the Shank et al. OM data is at the low end of the values reported by O'Dowd et al?

Perhaps nobody is surprised; nevertheless, it seems that quite a cohort of the community is surprised about the OM levels over the NE Atlantic, hence the comment.

P7329: Many of these summary points are incomplete for some of the reasons pointed out above and do not directly address the question posed by the title.

The summary points underpin the answer to the title question; the summary points are complete and are the statement of fact!

REVIEWER 3

This paper was motivated by Savoie et al. (2002) and Shank et al. (2012) who inferred that Mace Head, a coastal monitoring site, may be unrepresentative of aerosol at lower latitudes, that it may be frequently impacted by continental sources and that it may also be frequently impacted by aerosol emission from the local surfzone. This is an important debate, and it needs reconciliation. For example, if it is concluded that Mace Head samples aerosol which is unrepresentative of the marine background, the utility of the Mace Head record will be diminished and the implication for aerosol-climate studies will be significant.

The submicron organic mass fraction is the focus O'Dowd et al. The summary provided by Shank et al., in their Table 1, with Mace Head's Org/SO4=3.5 (larger than all other table entries), with Mace Head's CO=130 ppbv (about a factor of two above the marine background), and with Mace Head's Org=0.9 ug/m-3 (a factor of three larger than all other entries), are all exemplary of the debate. What are we to make of this?

First there is the point made by O'Dowd et al. on p. 7318. We are told that Shank et al. (2012) misinterpreted the Mace Head measurement of Org. Apparently, Shank et al. did not acknowledge that 50% of the reported Org mass (Mace Head) resides at supermicron size. This is relevant because in Table 1 (O'Dowd et al.) we see that the OM/BC ratio at Amsterdam Island exceeds that at Mace Head. Apparently, both averages (Mace Head and Amsterdam Island) correspond to integration over the sub- and super-micron size range.

In their conclusion, O'Dowd et al. explain the disparate OM/BC ratios (Shank et al. vs Mace Head) in terms of marine productivity. Apparently the marine OM source is more dominant in the north Atlantic, compared to a weaker marine OM source in the tropical and subtropical

Pacific. Also, OM and BC in the tropical Pacific can derive from continental biomass combustion. A case for the latter is made in Figure 9 of Shank et al.

Overall, I am satisfied with the explanation provided by O'Dowd et al. Yet, in my opinion, three things are missing. 1) How were the Mace Head BC measurements made at Mace Head?, 2) What is the BC uncertainty and thus the OM/BC uncertainty, particularly at low BC values (again at Mace Head)?, and 3) The Mace Head CO values (Table 1 of Shank et al) are rather large. How does 130 ppbv square with the claim that Mace Head frequently samples air that comes directly off the North Atlantic?

We would like to thank the reviewer for clearly summarising outstanding major issues.

1) ***BC measurements were continuously performed at Mace Head by measuring particle absorption and, therefore, represent equivalent BC (EBC) concentration. Unfortunately, there is no established uncertainty of EBC measurements as the "standard" BC measurement does not exist. The uncertainty, however, largely depends on mass attenuation coefficient which was discussed answering other Reviewer's comments. It has to be noted, though, that EBC measurements would more likely overestimate rather than underestimate BC concentration in clean marine air masses due to numerous absorbing species (dust, organic matter) and multiple scattering off the filter in absorption photometers.***
2) ***CO values may look large, but cannot be directly used to assess the aerosol phase. CO is mainly destructed by photochemical processes which are not very efficient in the cloudy and stormy North Atlantic environment while the particles are mainly removed by wet deposition and in-cloud scavenging both of which are very efficient in the cloudy and stormy North Atlantic. Moreover, Shank et al. has demonstrated that at relatively low CO concentration OM concentrations can be anything from nearly zero to very high suggesting that it is impossible to quantify anthropogenic source contribution based on CO measurements only (Figure 5, Shank et al.)***

Specific Comments:

Figure 10 – The text says that measurements were made over a 32 m gradient, but the figure caption says that gradients were evaluated 3 m, 10 m and 22 m heights. Which is it? Also, what is used to normalize the value on the abscissa of Figure 10? Also, from Figure 10 it seems there are two regimes for WIOM (i.e., figure captions "production" and "removal"). If two regimes are indeed the case, we need to know the frequency of occurrence of the two regimes, so that we can conclude regarding the relative importance of surface source versus the source aloft. Regarding the conclusion that WIOM is "…not a surfzone or coastal artifact." I don't think this is evident from Figure 10, and hence, I think that the distinction between with is shown in Figure 10 and what is inferred in Ceburnis et al. (2008) needs to be much more carefully delineated.

Figure caption has been corrected. The gradient measurements involved an extension of the 22 m tower sampling point to 30 m. The frequency of occurrence of both regimes requires a very expensive long-term experiment beyond the scope of this study. The source of WIOM is not aloft, but it can be upwind surface derived – there is an important distinction.

P.7318/L.28 – Classically, isn’t nss-sulfate is derived from a _total_ sulfate measurement and a Na measurement?

Indeed, nss sulphate is derived from a total sulphate concentration and Na concentration, both of which are large numbers and, therefore, introducing large errors.

Use of the word “artefact”. In my view, it is important to say whether, or not, the surfzone is a significant _ source_ for mass measurements made on the tower. If it is a significant source, that does not condemn the Mace Head measurements, but it does cause concern, especially if they are used to extrapolate to the broader north Atlantic region.

I don’t think “artefact” is the correct word. Here is how a scientific dictionary defines: artifact also artefact (är'tə-făkt')

1) An object produced or shaped by human craft, especially a tool, weapon, or ornament of archaeological or historical interest.

2) An artificial product or effect observed in a natural system, especially one introduced by the technology used in scientific investigation or by experimental error.

Corrected

In my opinion there is disconnect between the discussion in the text (Re: the “unique marine organic aerosol hydrocarbon fingerprint) and what is shown in Figure 8. The former suggests carbon-bond unsaturation, compared to a “refined hydrocarbon”, but the latter suggest oxygenation. Also, I wonder if the “□” concept (p.7322) needs a reference.

There is no disconnect- just new concepts to accept! The text refers to the hydrocarbon part of the total mass spectrum presented in Figure 8, which is unique and different from anthropogenic hydrocarbons. If the reviewer implies that oxygenation would diminish the input from class CnH2n+1, then classes CnH2n-1 and CnH2n-3 should be absent as well, yet they are very prominent (m/z: 27, 41, 55 as well as 39, 53, 67), therefore, the absence of CnH2n+1 would rather point to the different hydrocarbon source and not oxygenation.

Reference added to the manuscript: (McLafferty, 1993); McLafferty, F. W., and F. Turecek: Interpretation of Mass Spectra, 4 ed., Univ. Sci. Books, Mill Valley, Calif., 371 pp., 1993.

P.7325 – Here the flux footprint and concentration footprint are defined. However, the sentence that introduces the definition says “…concepts of the flux footprint and the concentration.” I will admit being uninformed about footprints, but I will hazard a guess that the upwind fetch contributing to the concentration signal could be comparable to the width of the entire north Atlantic basin. Yet, O’Dowd et al. state that the concentration footprint is (only) 10 to 100 times the flux footprint. The latter is stated to be ~1 km, so the former is estimated to be <100 km. That is surprising.

The concept is, indeed, correct. The flux footprint, i.e. most probable distance at which the emissions are contributing to the flux at a particular level – in this case was ~1km. The entire gradient footprint, i.e. the distance at which the emissions form the entire gradient, was ~5 km. The concentration footprint, i.e. emissions responsible for the composition of the entire

aerosol population sampled at the sampling location was likely 100 times larger which is in the order of several hundred kilometres. The emissions from further distances can still be discernable, but depend on the dry&wet deposition and in-cloud scavenging. Theoretically, the entire North Atlantic basin can contribute to the observed concentration, including the North American continent; however, the most distant emission will contribute the least. Carbon isotope analysis has revealed that continental North American sources can only contribute up to 20% of total carbon observed at Mace Head, supporting the above concept. The other concept has it that it takes about 2 days to fill the entire boundary layer with sea spray particles (Ovadnevaite et al. 2012) again limiting the distance to half the North Atlantic since it takes about 4 days for the air masses to cross the North Atlantic (Cavalli et al., 2004; Ceburnis et al., 2011)

P.7327 – Should this phrase be omitted: “that resulted in 3-10 times higher concentration”?

This argument is presented for the reason that if Mace Head measurements are significantly affected by surf zone emissions than the flux derived from the measurements at Mace Head should produce unrealistically high fluxes, but it was exactly opposite.

P.7327 – O’Dowd say that winds are stronger at Mace Head, but that seems to contradict Figure 9b.

Clarified for that specific case

Shank et al. and O’Dowd et al define different thresholds for whitecapping (7 and 4 m/s respectively). What is the origin of this disparity?

O’Dowd et al. never defined threshold for white capping as 7 m s-1, but rather 3.7 m s-1.

Callaghan, A., G. de Leeuw, L. Cohen, C.D. O’Dowd, Oceanic Whitecap Coverage: Measurements from the North Atlantic, Geophys. Res. Letts., doi:10.1029/2008GL036165, 2008.

Figure 11 – “Size Range” should be either “Diameter Range” or “Radius Range”. Are the particles dried before sampling?

Particles were not dried as they are presented in Figure 11.

Editorial:

Figure 5 is not referenced in the text

Done.

The Figure 8 caption does not match what is shown in the figure.

Caption is corrected.

Spelling: “athropogenic”, “MAP ruise”, “occurance”, “intermitted”, “rockey”, “haracterisation”, “centred”, “revealled”, “overage”, “heigth”, “orrganic”, “whas”

Typographic: “criteria along”, “values to aerosol”, “two addition reasons”, “OM/BC ration”, “which can be”, “in a good”, “coastal enhancement of artefacts”, “include addition southern hemisphere”, “WIOM is the clean marine air”, “cannot exclusively”

Spelling and typos are corrected

Acronym definition: GAW, TOA

Acronyms are defined