

Interactive comment on “Do anthropogenic or coastal aerosol sources impact on a clean marine aerosol signature at Mace Head?” by C. O’Dowd et al.

Anonymous Referee #2

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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

C2348

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.

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).

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?

Pg 7316 Lines 20- to next page L5: Describes “new” sampling criteria employed since 2001. These are said to include: Wind direction (already noted as not reliable)

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

C2349

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 700 cc and there is no evidence of nucleation (there have been a number of studies of nucleation at the site).

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?

P7316: "More sophisticated sampling" is mentioned 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 mentioned 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.

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.

C2350

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.

Moreover, 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 known 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 Cavelli 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.

P7317, Lines 7-: 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.

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 excluded 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 Savoie 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.

C2351

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.

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 Figure 2 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.

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).

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.

C2352

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.

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.

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.

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.

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

C2353

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).

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.

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

C2354

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. Moreover, 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.

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?

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.

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.

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

C2355

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.

Not shown are profiles of concentrations or simple size-resolved data at various tower altitudes that speak to this 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.

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

P7327, Lines 3–: “40% higher sea-salt mass” at MH seems to contradict argument of minimal impact of coastal/island influences claimed earlier.

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.

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

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

C2356

the Celtic Explorer (Fig 9) where even effective sampling can be questionable. One worries about apples and oranges comparison.

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?

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.

Interactive comment on Atmos. Chem. Phys. Discuss., 13, 7311, 2013.

C2357