

O'Dowd et al., Atmos. Chem. Phys. Discuss., “Do anthropogenic or coastal aerosol sources impact on a clean marine aerosol signature at Mace Head?”

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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/m³ (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?

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 what is shown in Figure 10 and what is inferred in Ceburnis et al. (2008) needs to be much more carefully delineated.

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

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-fäkt')

- 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.

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 “ Δ ” concept (p.7322) needs a reference.

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.

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

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

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?

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

Editorial:

Figure 5 is not referenced in the text

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

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”

Acronym definition: GAW, TOA