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Atmospheric Chemistry and Physics Discussions

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 #1

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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, aerosol 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 pub-

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lished 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.

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

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)?

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

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.

p. 7315: Line 8: Define "clean 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?

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? What

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was it? What is the uncertainty associated with the derived BC mass concentrations?

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.

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?

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.

p. 7319: Many typos to be 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.

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?

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p. 7319, line 18: change to “ratio”

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

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

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.

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.

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.

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 clear 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.

p. 7322, line 4: change to “revealed”

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

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

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.

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

p. 7326 - 7327: Many typos to correct.

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?

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

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