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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Quantification of the carbonaceous matter origin in submicron marine aerosol particles by dual carbon isotope analysis" *by* D. Ceburnis et al.

Anonymous Referee #1

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General comments:

The manuscript presents interesting results from a study of sources to organic marine aerosols at Mace Head, Ireland. The use of both the 13C and 14C provides valuable information on sources to carbonaceous marine aerosols.

The study is thus important, but unfortunately the manuscript is not as well written as one could expect. There are several examples of un-specific and confusing sentences. I also suggest that some of the native English-speaking authors perform a careful check of the text, in order to improve it. It was quite surprising to find out from the supplemental material that the manuscript had already been in review for PNAS,

given the present quality of the manuscript.

The introduction lacks a thorough introduction to carbon-isotopes and their use in atmospheric studies. Such a section is now hidden in the experimental section (page 2754) and I suggest to move relevant parts to the introduction (leaving experimental details in the experimental section). Also state typical delta 13C values in terrestrial and marine environments.

The use of 13C as a tracer warrants a more in-depth discussion, since it is well-known that isotope fractionation effects occur during atmospheric processing. This can be due to differences between 12C and 13C in e.g. reaction rate constants, and condensation and volatilization processes in the atmosphere or on the sampling filter. This should be discussed in further details in the introduction.

It is very surprising that the marine samples contain 0% continental non-fossil carbon, given that most (continental) studies find more than 50% non-fossil carbon in non-urban areas (as recently reviewed by Hodzic et al., ACP, 2010). This finding is so un-expected that it should be supported by other measurements or at least back-trajectories (using for example the freely available HYSPLIT model), before the manuscript can be accepted. This was actually also suggested by one of the reviewers of the PNAS submitted version of the manuscript ("Simple markers such as elemental tracers and meteorological conditions are given no quantitative representation in this work.") The 14C results should be compared with previous measurements.

Specific comments

The title: The wording "dual carbon isotope analysis" will not be clear to most readers, and I suggest to be more specific and just use 13C and 14C instead. Is it necessary to state both aerosol and particles in the title?

Affiliations: Please state all affiliations at the same level of detail - affiliations 3, 5, 6, and 7 are too short.

Page 2751 line 22-23: " Organic matter has been observed, to different degrees, in marine aerosol particles for many decades" - be more specific and avoid the use of vague statements such as "to different degrees" and "for many decades".

P2752 L7: Define MSA before using.

P2752 L15: unclear statement: inconclusive due to varying pollution degree - please clarify.

P2752 L16-18: The sentence seems out of context.

P2752 L23: How can D<1.5 micrometer aerosols be submicron?

P2753 L11: I do not agree with the postulate that the marine samples were likely the cleanest possible to obtain in the Northern Hemisphere. There are more remote areas in e.g. Greenland and Canada. Please rephrase.

P2754 L4: What do you mean by "the samples were treated to carbon isotope analysis"? The sentence should be rephrased.

P2754 L5: Remove "fuel" in "continental non-fossil fuel carbon". There are many other sources to non-fossil carbon, so state that those mentioned are some of the primary sources.

P2754: The discussion of isotopic fractionation (of primarily 13C?) should be more in-depth as stated above. Why should condensation favor the lighter isotope? Why do the authors assume that a "dominant primary source of marine organic aerosol" should not be affected by isotope fractionation? Could isotope fractionation occur during evaporation of compounds from primary aerosols or aerosol phase reactions or as a sampling artefact? The discussion should reflect the associated uncertainties.

P2754 L17-19: Please find a more suitable reference for this general statement.

P2754 L27: Should start with "Measurements of TC concentrations..." Has TC been defined?

P2755 L4-5: Which analysis protocol was applied?

P2755 L15: The sentence is unclear. I suggest rephrasing into something like this: blanks were measured and were also estimated from regression analysis.

P2756 L16-20 and 21-26: These lines should be rewritten using better English grammar and clarity.

P2757 L17: How do you define continental sources? As all non-fossil sources, whether natural or man-made? If so, then it would be better to state this more clearly.

P2759 L7-10: It is not clear how these other studies support the interpretation.

P2760 L25: It is a very strong statement that the NH marine boundary layer can be exceptionally clean and largely devoid of anthropogenic material. Such a statement should be documented by measurements of e.g. elemental tracers. See also general comments.

P2761: The discussion should reflect that the conclusions are drawn on a limited study of only 6 samples. Are these results in line with previous investigations at the site?

P2761 L25: Should be changed to "Dual carbon isotope.." and "marine and polluted aerosol samples"

P2761 L27-: Should be rephrased to reflect that the conclusions are drawn from a study of 6 samples.

P2762 L7-: The sentence is not clear and should be more descriptive. What are the 30%?

Table 2. The notation delta 14C should be described in the text or footnotes. Are the uncertainties due to counting statistics? What is the overall uncertainty on the results?

Interactive comment on Atmos. Chem. Phys. Discuss., 11, 2749, 2011.