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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 #2

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General comments: This paper uses a dual isotope method for source apportionment, which is an important topic and an interesting approach. However major changes and clarifications are necessary, before this paper can be accepted.

1) The literature on 13C measurements for aerosol source apportionment is not very well reviewed and many important citations are missing. For example, the term dual isotopic characterization and some first applications were already presented by Currie et al., 2000 (references therein). This should obviously be cited. Other examples follow later in the review, but the authors should conduct a thorough literature study and use it for introduction and discussion of results.

2) The methods still need to be discussed in more detail before they are sufficiently clear and can be evaluated.

a. Pg 2755: How was TC converted to CO2 for 13C analysis?

b. Pg 2755, line 15ff: Please describe exactly how field blanks were treated. What was used for C_blank in eq 2?

c. Pg 2756, line 10ff: This is confusing. What was the motivation for calculating marine 13C this way? I assume this does not refer to the blank correction, so please give the exact equation that was used. Was only BC subtracted? What about other fossil sources? Moreover, there are several other studies that give significantly higher delta 13C values for BC closer to -27 ‰ (Huang et al., 2006; Ho et al., 2006) or even higher for biomass burning BC. There are more examples in the literature, which I encourage to authors to look up.

d. Equation 5, more explanation is needed here: Which data sets were used to minimize this equation? The same subset of data that is used later for source apportionment? How would this method perform if the variation in the data was not governed by 3 main sources, but maybe 4 or 5 sources would be important?

3) Contrary to the authors assertion there is evidence of isotopic modification carbonaceous aerosol by photochemical processing (e.g. Wang and Kawamura, 2006 and some more recent studies). The more convincing argument to neglect it, would be the absence of a seasonal variation in polluted 13C values.

4) The estimate of delta 13C of 29‰ for fossil carbon is somewhat in contradiction to most of the literature values that I am aware of, which are in fact closer to -27‰ to 26 ‰Ṡee e.g. Ho et al., 2006 (roadside); Hueng et al., (2006) (tunnel); Widory et al., 2004, Currie et al., 2000. The values estimated here should be compared with a larger number of literature values, (not only the ones cited in this review).

5) This relatively low estimate of fossil delta 13C could also be the reason for larger ma-

rine contributions than in the literature, or the absence of non-fossil continental sources. At least a sensitivity study should be conducted that uses the more commonly measured isotopic value of fossil sources.

6) Along with the other reviewers I am puzzled by the absence of non-fossil continental sources in the marine aerosol. But moreover also by the relatively large contribution of fossil sources to the polluted aerosol compared to the non-fossil continental sources. In various continental locations a much bigger contribution of non-fossil sources has been measured. Is there any hypothesis why this would change with transport over the ocean?

References: Widory, D., S. Roy, Y. L. Moullec, G. Goupil, A. Cocherie, and C. Guerrot (2004), The origin of atmospheric particles in Paris: a view through carbon and lead isotopes, Atmos. Environ., 38, 953-961. Huang, L., et al., Atmos. Environ., 40, 2690-2705, (2006). Wang, H. B., Kawamura, K., J. Geophys. Res., 111, D07304 doi.: 10.1029/2005JD006466, (2006). Currie, L.A., Evolution and multidisciplinary frontiers of 14C aerosol science, Radiocarbon, 42 (1), 115-126, 2000. Ho, K.F., S.C. Lee, J.J. Cao, Y.S. Li, J.C. Chow, J.G. Watson, K. Fung, Variability of organic and elemental carbon, water soluble organic carbon, and isotopes in Hong Kong, Atmos. Chem. Phys., 6, 4569-4576, 2006.

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