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Interactive comment on "Source apportionment of the carbonaceous aerosol in Norway – quantitative estimates based on ¹⁴C, thermal-optical and organic tracer analysis" by K. E. Yttri et al.

Anonymous Referee #3

Received and published: 21 April 2011

The paper is aimed at source apportionment of carbonaceous aerosol in Norway, however, it has a broader appeal in terms of method development to correctly apportion multitude of carbonaceous aerosol sources. The paper should certainly be published in ACP subject to addressing the comments listed below.

My major comment and concern is about LHS method used for source apportionment. There is a general problem with a type of random simulations in the way that given a wide range of input parameters (with their own uncertainty ranges) it will always be possible to find the solution, but how stable and reliable would it be? Does it tell

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anything about the quality of input parameters, measurement accuracy, etc.? I am not convinced that equal assignment of parameters between low and high limits is realistic. If you relax parameters too much, the solution will easily be found, but will it be valuable? There seems to be no validation presented in the paper which would help assessing usefulness of LHS or superiority over e.g. Monte Carlo or any other method. It is not clear at which stage negative contributions are excluded. If at the end for representations purposes only, then it is fine, but if in the process, then LHS must be positively biased. Negative contributions could be very informative. For instance, if contribution range is between -5 and +5% it is very likely 0 and the range is largely due to noise. However, excluding negative contributions, LHS gives an impression that the range is e.g. 0 to 5% and most likely value is 2.5%, hence, discernable from noise. Such considerations must be included in the text to provide readers with a proper justification of the method. The most probable value is the most important. Those most probable values (are they 50th percentiles?) should then be compared with the measured ones to see if there is any systematic bias, e.g. systematically below or above measured values. At the moment there seems to be no validation of the method, just the output. For example, if LHS output is 16% OCbb it should be compared against (OC/LG)bb relationship (especially Norwegian data) and so for other parameters. Or ECbb and ECff should be summed up and compared with the measured EC. At the moment Table 4 and Table S4 suggest to me that EC is systematically overestimated by LHS when compared to measurements, especially during summer. Same could be applied to F14C, worked out from equations and compared with measurements.

Other specific comments.

Do authors have a support for splitting particulate mass at PM1 which certainly does make sense but only when justified?

Reference to Gilardoni ACPD paper in many places of the current paper would be very helpful. It is a very similar study, but by no means undermines the novelty of this paper, primarily due to the fact that Monte Carlo simulations were used in Gilardoni paper. I

would also have suggested vice versa if I were a reviewer of the other paper.

Page 4. Experimental methods. Site description should be more informative, especially when aimed at generalizations of typical Norwegian conditions.

Page 9. Estimation of TCbb. It is not clear how TC/LG relationships in PM2.5 or PM10 are adapted for PM1. Why TC is used and not OC when LG is certainly a part of OC. This would unnecessarily increase the range and uncertainty in bb source contribution. Also, how those ranges are compatible with other studies, but that is, however, a lesser problem as study is aiming at describing Norwegian conditions and citing previous Yttri et al. paper.

Page 18. Line 576. Consideration of natural sources in Norway without accounting for sea salt, seasalt-sulphate, biogenic sulphate and crustal material in the form of dust can not be complete and even miss-leading. It should clearly be stated that major ions have not been measured in this study and, therefore, estimation of natural sources is limited. The whole chapter is somewhat misleading as it should be named "mass closure of PCM" and not PM where other inorganic species should be measured as well to aim for mass closure.

Interactive comment on Atmos. Chem. Phys. Discuss., 11, 7375, 2011.

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