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Atmospheric Chemistry and Physics Discussions

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.

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General response on source-apportionment methodology and LHS

The referees have some concerns about the source-apportionment (LHS) methodology, which can be summarised:

i) omission of negative solutions may bias the results

ii) the 'flat' distribution functions used are unrealistic

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iii) a Monte-Carlo methodology might produce different results

These concerns are understandable, but we believe unfounded. Some of these complexities behind our views are outlined below, but it is important to realise that the main purpose of the LHS approach is to show which solutions are consistent with the observations, and the elimination of negative solutions is a key part of this process.

First just a personal example on the background to the LHS methodology, and the deliberate rejection of standard (e.g. gaussian) approaches to the specification of possible emission ratios. One of us (DS) began the LHS approach as part of the CARBOSOL project (Gelencser et al., 2007), partly in response to the fact that it was so hard to decide on best-estimate emission ratios. Indeed, in CARBOSOL we had to widen the allowed range of e.g. OC/EC because the initial best estimate of the emission experts lay outside the range of values first suggested the experts dealing with ambient data. Subsequent requests for "best-estimate" ratios and ranges from colleagues in several studies (Szidat et al.,2009 and this work) have produced similar differences of opinion - there is in fact no "best" estimate for many of the ratios we are dealing with.

Such differences reflect the widely varying ratios found in the literature, and the differing experiences and backgrounds of those involved. These exercises have taught me to approach source-apportionment with a large degree of pragmatism, and the LHS assumptions reflect this.

The LHS is thus not designed to give a 'best'-estimate, it is designed to show which solutions are possible. One of the encouraging and important findings of these LHS studies has been that one can allow very wide ranges of emission ratios, and still end up with quite similar general findings - the impossible combinations are excluded by the requirement that all solutions are positive - or equivalently consistent with observations.

Some complexities of source-apportionment for organic aerosol are also very relevant:

1) Although we have some estimates of precision of some emission rates, we gener-

ally know very little about accuracy. For example, the biomass burning ratio (TC/LG)bb depends on appliances, maintenence standards, type of wood, dampness, and a host of other factors. The average ratio will likely change a lot with time-of-year (from residential to agricultural burning), and with distance from source (e.g. from Elverum-type scale to Artic Smoke scale). This also makes (TC/LG)bb dependent on wind-direction for example, even for the same season and sites. We can roughly take care of the range of such estimates, but we cannot assign a proper statistical distribution.

2) Much of the literature uses different analysis techniques, e.g. to get EC/OC ratios. Again, we can specify a loose range, but cannot assign a proper statistical distribution to this.

The referees say that elimination of negative solutions gives a positive bias. However, the main reason behind negative solutions is not some effect of random noise, but that a combination of factors being tested is not consistent with the amounts of carbon present on the filter. For example, we allow the possibility of very high (TC/LG)bb ratios, as such ratios might arise as a result of loss of LG as an air parcel moves along. On many occassions though, such high ratios will result in an unrealistically high contribution of biomass-burning to both the F14C and OCbb, ECbb components. The measured values of F14C and other compounds will often exclude that such high bb contributions are possible. Indeed, excluding “impossible" solutions is one of the points of the exercise. If we allow negative solutions we are essentially accepting all impossible combinations.

With this in mind, it would be wrong to accept negative solutions to LHS. Such solutions represent not just acceptable errors caused by normal-distribution type problems, but also represent unacceptable solutions.

Considering Monte-Carlo (MC) versus LHS, then this difference is of very minor importance. As discussed in e,g. McKay et al., 1979, LHS provides essentially the same results as MC at significantly less cost. Considering the very significant uncertainties in other aspects of source-apportionment, there would be no advantage in using MC over LHS, and indeed it would be harder to explore the same number of input parameters.

Extra Ref:

McKay, M. D.; Beckman, R. J. & Conover, W. J. A Comparison of Three Methods for Selecting Values of Input Variables in the Analysis of Output from a Computer Code Technometrics, American Statistical Association and American Society for Quality, 1979, 21, pp. 239-245

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