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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Source apportionment of carbonaceous aerosol in southern Sweden" *by* J. Genberg et al.

Anonymous Referee #2

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The manuscript is one in the recent series of papers dealing with 14C-based source apportionment of atmospheric aerosol at various locations. It relies on largely the same methodology as the previous papers do, but applies a slightly different statistical approach to handle the large uncertainties inherent in the measurements and the calculations. Thus the major statements of the manuscript are not really unexpected and can be deduced from the conclusions of similar papers and from considerations of common sense. Nevertheless, given the duration of the study which is quite remarkable among similar 14C-based measurements and the detailed evaluation of data makes it publishable provided that certain aspects of the manuscript improve significantly.

I have three major concerns with the manuscript in its present form:

(1) This manuscript as well as many other papers treats 14C-measurement as some-

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thing decisive and unquestionable in splitting aerosol carbon between fossil and modern carbon. While this potential generally exists, there are many pitfalls associated with drawing conclusions from its results. First, 14C-measurement in source areas might be biased by the presence of specific 14C-sources (not applicable here as sampling took place at a background site). Second, the broad source categories that can be derived with excessive uncertainties from 14C and other macro tracer measurements are oversimplified for the purpose of meaningful source apportionment. There are many source categories listed for human activities which do not fit into the implications provided by the defined categories. Cooking, frying, tyre wear, biofuel components in fuel, etc. are just a few examples that may be falsely categorized into the simplified concept based on their 14C-signature. When defining the basic aerosol classes and when interpreting the result, this issue should be addressed explicitly in order to avoid misunderstanding.

(2)Similarly to earlier studies the representation of semi-volatile OCs has not been improved in the simplified concept of the manuscript. As a rule of thumb, gas phase OC is about 10 times more abundant than particulate OC, thus merely the change of temperature can drive vast amount of OC into the aerosol phase. These adsorbed species are not part of the sampling artefact as they are in equilibrium with the collected particles. They are also not SOA components by definition, since they still are as emitted. This should also affect both the OCff/ECff and OCbb/ECbb ratios in different seasons. Whereas this fact is explicitly acknowledged in the manuscript (Page 13592 Line 29), a single value is used for each throughout the calculations. This results in very large calculated contribution of OCbio in winter. Although the authors elaborate on the possible reasons for such a large contribution in Section 3.4.3, they forget to mention the most likely case that a large part of it should have been simply assigned to OCbb as a result of the increased partitioning of SVOCs at low temperatures. I would suggest that at least two different ratios (OCff/ECff and OCbb/ECbb) should be used for the measurement data, one for winter and one for summer. Hopefully such seasonal ratios can be deduced from available literature. Accounting for this effect would significantly improve the reliability of the source apportionment.

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(3) The Introduction of the manuscript is very poorly written: reading it, it gives the false illusion that this is more or less the first study of this kind, which is not true. References are randomly selected: sometimes very basic papers are cited, in the same paragraph a 2010 paper is quoted. (e.g bomb effect – 2010, Suess effect - 1955). There are quite a number of incorrect or at least clumsy statements, (e.g. 'EC consists of. . .graphite-like structures', 'EC. . .lead to a warmer climate'. It is stated that '(the 14C) method is complicated with the. . . dilution of atmospheric modern carbon by fossil carbon emitted from combustion of fossil fuels': this is not a complication, this fact makes possible the use of 14C technique in dating and in many environmental applications, including the one presented by the authors!

Minor comments:

Page 13590 Line 1 Replace 'Then' with 'When'

Page 13593 Line 3 'Allthough'

Interactive comment on Atmos. Chem. Phys. Discuss., 11, 13575, 2011.