Interactive comment on “Fossil and non-fossil sources of organic carbon (OC) and elemental carbon (EC) in Göteborg, Sweden” by S. Szidat et al.

Anonymous Referee #1

Received and published: 15 September 2008

In recent years 14C measurements have advanced into the techniques of aerosol source apportionment, allowing unambiguous distinction between fossil and non-fossil carbon on a bulk level. The authors of this manuscript, by relying on their solid expertise gained during previous applications of the technique in the field of aerosol science, attempt to extend its potential by combining it with more conventional aerosol separation techniques. While it would be tempting to obtain radiocarbon data for common aerosol sub-fractions experimentally, the question arises how successful the combination of a rather outdated separation method and a high-tech analytical method could be? Are the radiocarbon results almost as reliable for the major sub-fractions of carbon as for total carbon? To answer these questions, we need to have a closer look at the chemi-

S7072
cal separation methods used for obtaining the major carbonaceous sub-fractions, and their relation to the method of calculation of the major source categories. The Achilles heel of the entire concept is the two-step combustion method used to separate OC and EC. This method is based on a simple thermal method developed by Cachier et al. (1989) admittedly for Diesel soot analysis. The original method was modified substantially in order to minimize its well-know biases such as the premature combustion of soot carbon due to the presence of alkali metals, or char formation from organic carbon during the low-temperature oxidation step. The application of water extraction prior to combustion may reduce both since inorganic ions and char-forming WSOC are both removed. However, the authors try to use this method for PM10 samples dominated by biomass burning, which would possibly give at least highly dubious results for two reasons. First, the use of the simple two-step thermal method was demonstrated to overestimate EC in biomass burning aerosol due to significant charring of not only the water-soluble but also of the water-insoluble organic compounds (Yu et al., 2002; Reid et al., 2005). Secondly, biogenic particles which were shown to make a significant fraction of supermicrometer carbonaceous aerosols, also have a very high tendency for charring (Bauer et al., 2002). These two effects combined yield a mixture of Diesel soot, biomass burning soot and an unspecified but likely significant measurement artifact (char) which the authors consider as total EC. Given the possibly high level of char-forming insoluble OC in the samples, it could strongly bias the EC fraction subjected to radiocarbon analysis. Thus it is very uncertain to which fraction the fMC data are referring to. The consequences of this bias propagate through the entire paper: ECwood is derived directly from radiocarbon analysis of the alleged total EC fraction, OC is obtained by subtraction of EC thus it is also affected; OCwood is derived from known EC/OC emission factors for wood burning (although an alternative method with levoglucosan/OC emission factors is also used, though with levoglucosan concentrations measured in PM2.5 aerosol fraction and not in PM10); and OCbio is determined as a residual term. The authors consider OCbio as secondary organic aerosol only; however, since most measurements were done on PM10 aerosols, the contribution of
primary biogenic aerosols could be non-negligible and should be accounted for. Since most of the conclusions of the paper are based on results that are likely strongly biased by experimental inconsistencies, they cannot be evaluated until the entire concept is revisited.


Interactive comment on Atmos. Chem. Phys. Discuss., 8, 16255, 2008.