

Interactive comment on “A new comprehensive approach to characterizing carbonaceous aerosol with an application to wintertime Fresno, California PM_{2.5}” by P. Herckes et al.

Anonymous Referee #2

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The paper is based on a highly complex and labor-intensive analytical procedure for the bulk characterization of urban aerosol. While the solubility separation and the use of FTIR and ¹³C-NMR is not new at all, there are some original aspects in the manuscript, e.g. digesting the filter off the insoluble residue. There are, however, fundamental problems with the paper which make the results and conclusions highly questionable.

1. The entire study is based on results obtained from a single composite sample claimed to be representative of urban and suburban environment in Fresno. However, the method by which the composite sample was prepared (identical parts from

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all filters) suggests that the sample is strongly biased to a few highly loaded samples (possibly obtained downtown and under conditions of strong inversions) rather than providing mean concentrations for the sampling period and the region.

2. This single composite sample was subjected to a complex multistep analytical procedure in which large volumes of solvents (50 and 100 ml) were applied than removed (what about insoluble organic impurities left behind?), the sample aliquots were oscillating between being dissolved and the dry state (what about sample losses?), passing through large volume ion-exchangers, etc.. It appears that the entire procedure was run without any quality control: no blank experiments are reported (e.g. what happens if blank filters were subjected to the same procedure), no real recovery experiments were carried out (e.g. putting known amounts of aerosol model compounds such as suggested by Fuzzi on a blank filter and run the experiments). What the authors call 'recovery' is not recovery in the analytical sense: they simply measure total (organic) carbon on the filter fractions and relate all recovered carbon mass to that number. It may well be that a large fraction of the aerosol organic compounds were actually lost during the elaborate procedure, but part of the loss is compensated by the mass of preconcentrated organic impurities of the large volumes of solvents (One can easily calculate how much residue may be left behind by evaporating 100 ml of solvents containing only a few tens of ppm insoluble impurities)

3. During the procedure some of the solvents used are very aggressive and some of the conditions applied are very harsh for the 'sensitive' organic macromolecules likely present in the sample. A 'real' recovery study suggested above would have also been helpful to exclude that such chemical changes do not occur.

4. The authors are not consistent in applying the traditional concept of OC/EC in aerosol science. In their 'recovery' calculations they incorrectly relate their measurements to total organic carbon (instead of total carbon), and call the insoluble residue which likely consists of sooty material (elemental or black carbon) 'particulate organic carbon'.

5. In their conclusions the authors sometimes make contradictory statements (remember, there is only one sample): once they explain the relatively low fraction of WSOC by the predominance of flaming combustions in fireplaces (page 8433 line 5), later they also explain the abundance of levoglucosan by wood combustion (page 8436 line 27-29) in spite of the fact that anhydrosugars are primarily released by smoldering and very little in flaming combustion.

Interactive comment on Atmos. Chem. Phys. Discuss., 7, 8423, 2007.

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