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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Comparison of the levels of organic, elemental and inorganic carbon in particulate matter in six urban environments in Europe" *by* M. Sillanpää et al.

Anonymous Referee #2

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General Comments:

The work by M. Sillanpää et al. presents an interesting "monitoring" strategy: sampling on periods in which severe PM pollution events have been observed in various cities and focusing analyses on the carbonaceous fractions, which are likely to have major contributions to the PM health effects and are the most difficult to quantify, due to various sampling and analytical artifacts. A large set of elements have also been quantified, which might give information on possible aerosol sources. However, the level of the discussion is very disappointing, as it remains very much too speculative.

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Full paragraphs indeed start with “most likely explained by”, “most likely due to”, “suggesting that”, “could be explained by e.g. ...” and actually present commonly accepted concepts, without using the data acquired in this work to give evidence of what is stated. This manuscript, as such, only presents and compares data, without any further step to explain the observed differences and similarities. The same applies to more technical considerations (OC found on back filters) or to comparisons with other studies or measurements: no conclusion is derived from the observations. One can wonder if this work represents a significant enough contribution to the atmospheric chemistry research for constituting a stand-alone article.

Specific Comments:

p. 2724, line 25: the fact that the deliquescence point was not reached does not mean that the contribution of water to the filter mass was negligible.

p. 2725, line 22: please specify how the increasing of OC solubility from left to right (i.e. I guess fro OC1 to OC4) was established. Is this rule valid for OCP also?

p. 2726, line 2: according to last paragraph in page 2725, XRF measured the total Ca concentration. So why does Ca2+ appears in page 2726, line 2?

p. 2726, line 9: integrating thermograms to get CO3 concentrations might be an interesting approach. However, readers would certainly like to know and understand how it has been established that (1) CO3 contributes to OC4 only, (2) CO3 peak starts at 210-225 s and ends at 250-275s, and (3) how the CO3 peak start and end times are determined for each thermogram.

p. 2728, line 7-9: it is not clear whether the back filter OC amounts where used to correct the front filter data. Such a correction relies on assumptions that should be specified. If back filter OC data were not used for correcting front filter, it should also be clearly stated and explained. Positive artifacts are believed to depend - among other parameters - on face velocities. Isn’t it surprising that the OC back / OC front filter ratio

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are similar in the coarse and fine fractions, whereas the minor and main flow rates usually differ by 1 order of magnitude in a virtual impactor?

p. 2728, line 24-26 and p. 2729 line 14-19: what does the comparison with the works by Querol et al. and Putaud et al. bring to the discussion? Are the observed differences due to the various sampling and analytical techniques employed, or due to seasonal variations, or meteorology-linked variations?

p. 2728, line 28 - page 2729, line 6: the discussion about method-dependent OC/EC split should be moved up to the "Experimental" section, or down after discussions of EC concentrations.

p. 2729, from line 21: this section compares PM2.5 / PM10 ratios for EC and OC, whereas parag. 2 page 2727 compares PM2.5 / PM2.5-10. More consistency is recommended.

p 2729, line 23-24: there is no logical link between "EC existed mainly in the fine particulate fraction" and "EC is produced only in combustion processes" if a statement about the size of combustion particles is mot included.

p. 2729, line 25 - page 2730, line 2, in line with the general comments: some reasons for explaining the OC size distribution could be confirmed or ruled out by looking at other data. E.g. decreases in fossil fuel combustion for heating should be confirmed by decreases in EC. Can shifts in OC sampling artifacts at least partially explain the observed differences?

p. 2730, line 18: the OC vs. EC correlation and the OC/EC ratio are also consistent with carbonaceous species coming mainly from (light or heavy duty?) Diesel engines. Ships' fuel is very rich in S compared with road vehicles' fuel. Can SO4 concentrations be used to demonstrate the role of ship emissions?

p. 2730, line 26: again, a clear explanation on how CO3 is distinguished from other OC4 components is missing.

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p. 2731, line 6-15: would this discussion still hold considering that XRF leads to Ca and not Ca2+ concentrations?

p. 2731, line 21-24: the reason why the distribution of Ca among the fine (PM2.5) and coarse (PM2.5-10) fractions could not be derived from XRF analyses of the VI samples should be clarified.

p. 2732, line 16-18: can the effect of the filter loads be ruled out to explain the differences in the contribution of the various OC fractions?

p. 2732, line 19-20: is the 33% OC1 fraction observed for Barcelona and Athens significantly higher than the 26-29% observed at the other sites?

p. 2732, 2nd paragraph: these observations simply suggest that as such, the thermal program does not lead to any valuable data regarding the various OCi fractions. This could prevent from seeing any relationship between OC1 (front) and OC1 (back). Such a relationship (if any) could be a useful tool for a crude correction of the OC positive sampling artifact.

p. 2734, paragraph 1: the most striking observation re. PM mass concentrations is certainly the very different PM2.5/PM10 ratios observed at the various sites. Other statements are not clearly demonstrated from data.

p. 2734, paragraph 3: as already commented, the thermal program does not seem to lead to valuable data regarding the 5 OC fractions, which should therefore not be quoted again in the conclusion.

Interactive comment on Atmos. Chem. Phys. Discuss., 5, 2719, 2005.

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