

Interactive comment on “Seasonal variations of the Water Soluble Organic Carbon massfraction of aerosol in two valleys of the French Alps” by J.-L. Jaffrezo et al.

Anonymous Referee #2

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General comments The paper deals with a very important issue of atmospheric chemistry, i.e. the determination and quantification of the fraction of carbon which is "water soluble" (WSOC). A very large number of data is presented, covering a period longer than two years in two different alpine valleys. As far as I know is the only work presenting a so extended database on WSOC. The main scientific goal, however, is not to present a list of numbers but to verify if "The current belief (which) maintains that the WSOC fraction is "low" for primary OC and that it increases with aging of the aerosol, together with the general oxidation state of organic matter", may be verified (or confuted) by the elaboration and interpretation of full seasonal cycles. In view of such an

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aim, the selection of the sampling sites seems to be very appropriate, due both to the difference in the impact of human activities on the air quality (favoured by the closure of TMB) and to the relevance of primary organic emission from natural sources (forests).

Nevertheless there are some points on which I suggest a revision or I need a comment by the authors.

Specific comments 1. Section 2.4. In my opinion, the experimental process which has lead to the selection of the method for WSOC determination does not appear very well described. I understand that the first intention was to use the EC/OC analyzer described in Section 2.3 using the filter after one or more 'extractions' with water, but I do not understand well why the authors have performed this check when has been already verified, in previous experiments (Pertuisot Row 10- 4007) the need of filtering the extract before the analysis. It is clear that filtration is needed because some resuspension of particulate, and of the carbon matter associated with in, can interfere with the WSOC determination. Such mechanical artefact certainly affects also the determination of OC/EC on the filter after the extraction, with the additional complication that, during drying, some of the 'resuspended' material may be redeposited, in a non reproducible way, on the surface of the filter. Hence, in my opinion, all the part describing and discussing the extraction/filtering and subsequent recovery of EC/OC is not relevant to the main goal of the paper and should be removed. In addition the distinction between EC and OC is quite labile and different techniques may lead to different results in their ratio [ME Birch: Analyst, May 1998, Vol. 123 (851-857)]. Hence if, on a hand is true that one of the property of EC is its 'insolubility', it is also true that solubility depends on several factor that is difficult to control during the 'extraction' and that not all the carbon determined as EC is 'insoluble'. This fact add further unclerness to this part of the article. 2. Rather, major details should be given using the data which should be available in the thesis by Pertuisot (1997, which is not easily accessible). For example: how the need to use 10 ml of sample for each determination is linked to the choice of using 17 mL of water; how many replicates are possible? and so on

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3. At Row 01/4010 it is claimed that in the paper by Decesari et al. "no clear seasonal pattern has emerged", while in the cited paper is affirmed: "The WSOC air concentration exhibits a seasonal trend, with an annual periodicity similar to that of TC and aerosol inorganic constituents, and the aerosol samples collected during the summer season show a different chemical composition, compared to the samples of other periods of the year". Hence the authors statement seems much more stronger with respect to the conclusion of the cited paper and I think it must be reconsidered, taking also on account of the subsequent statement (Row 02,03/4010) in which it is said that a distinct seasonal pattern was observed, and similar to the one observed by Decesari.

4. I have some doubt on the significance of Figure 8, which plot DCA vs WSOC. In fact, DCA, given the method use for their determination, are a fraction of WSOC. Hence the plot DCA vs WSOC is confounded, because a certain degree of correlation is forced when DCA are a non negligible part of WSOC (it appears that some times more than 60

5. Another element of confusion in reading the figures is given by the unit of measure used for expressing the concentration. In fact, even if the information is not retained nor on the graph axes neither in Table 3, it is clear (from Rows: 12,15/4007 and Row :11/4009) that the concentration of WSOC/OC/EC is given in $\mu\text{g}/\text{mc}$ of Carbon. On the other hand the concentration of DCA seems to be given in $\mu\text{g}/\text{m}^3$ in mass of substance. In my opinion, it is sounder to compare data given with the same unit. Since it is difficult to estimate the mass of organic substances from the carbon mass, it is appropriate to convert the information on DCA mass concentration in DCA carbon concentration. It easy to realize that the weight of DCA in mass on the whole WSOC (or on the corrected WSOC) it is almost doubled if different units of concentration are used (maybe more, because the mass of C in DCA is around 40

6. Comparing the regression parameters of the correlation OC/K (figure 11) and WSOC/K (Rows 10,11/4016), it seems, if I'm not wrong, that OC and WSOC are quite perfectly correlated! In fact, at both sites, we have almost the same r^2 (0.82 vs 0.81

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at Chamonix; 0.62 vs 0.62 S.Jean) and intercept (3.5 and 3.7) and only a difference in the slope (26.7 vs 62.3 at Chamonix, 6.8 vs 16.2 at S.Jean). That means that plotting OC vs WSOC should give a very smooth line. In addition, one can estimate that OC has been (always) found at both sites at a concentration approx. 2.3 times higher than WSOC. I suggest revising this part of the paper because there is some confusion in data representation or there is some important issue that deserves to be better presented and discussed.

7. A concluding remark, related to the precedent comment, concerns the decision of the authors to present OC and EC data in a different paper, which is referred to as "in preparation". Why the discussion on factors influencing OC concentration is disjoint from the present one? It is regrettable that in this paper a full discussion on the organic matter behaviour and the relationship between OC, EC, and WSOC is, to a certain degree, hindered by such a decision. I think that a more complete information of the distribution of organic matter (which means using only a part of all the EC/OC database, those coupled with WSOC data) will help the scientific relevance of the paper and will allow for more detailed and funded speculation concerning processing, transformation and transport of organic matter. I make this comment also because the authors complain, in the introduction, about the limited number of "ancillary data (E.)" to delineate the respective impacts of sources and aging on the evolution of WSOC concentrations and the mass fraction relative to organic carbon (OC)" (Row 4-6/4001).

Minor comments

Row 20/4005: maybe too many digits in values and errors

Row 20/4005: the term "aliquot" should be used for liquid phase [(ISO 11074-2, 1998)]: "Known amount of a homogeneous material, assumed to be taken with negligible sampling error. The term is usually applied to fluids]. For solid I think the correct term to use is "portion" [(IUPAC, 1990; ISO 11074-2, 1998): "Each of the discrete, identifiable portions of material suitable for removal from a population as a sample or as a portion

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of a sample, and which can be individually considered, examined, tested or combined

Row:12-15/4007 add a 'g' in $\mu\text{C}/\text{mL}$

Row:2/4008 use "portions" instead of "punches"

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