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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Variability of carbonaceous aerosols in remote, rural, urban and industrial environments in Spain: implications for air quality policy" *by* X. Querol et al.

Anonymous Referee #2

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This paper presents a very impressive and interesting data set on carbonaceous aerosol compiled since the last 12 years in 78 locations over Spain. Considering the quality of the dataset and of the results presented in the paper I recommend the publication to ACP.

General comments: 1/ Implications of the results presented here are important for air quality policy issues (as discussed in the manuscript), but also (and mainly, in my opinion) for the scientific community. This kind of dataset allows pointing out the most relevant research activities that need to be conducted in the next years. This aspect of the discussion/conclusion is not developed (or scarcely) in the paper. I will have preferred

a specific section on this point rather than the general discussion on the implication in terms of air quality policy (section 5.1). This last section, mostly discussing the recent WHO report on health effect of BC, should be incorporated in the introduction (see next comment).

2/ Introduction. This section needs to be reorganized. Overall it is too technical and many aspects should moved to the experimental section (description of the methodologies used to measure OC/EC, BC, nmC ; artifacts). From my point of view, the introduction should focus on carbonaceous aerosol (concentrations in different locations in Europe, trends –paper of Pio et al 2011-), health impact (why the WHO report on BC is only discussed in the final section of the paper?), trends of other pollutants (especially PM ; Querol et al, 2008) . In other terms, provide information to the reader in order to contextualize this work in a more precise way.

3/ You clearly state (p6981, line 15) that "The assessment of the comparability between the different thermal protocols used is not the objective of the present work". I agree with that point but as your conclusions can be greatly affected by these methodological issues (sampling artifact included), I suggest that you add a specific section in order to discuss these aspects (in addition to the section 2.5). In the current version of the manuscript, all the elements of comparison (or all the elements available) are discussed but in different part of the manuscript. I think that it will be clearer for the reader to discuss these aspects in one dedicated section. Also, as many methodologies are used to measure OC/EC, BC, nmC, I suggest that you add at the beginning of the experimental part, a specific section discussing of some fundamental aspects of the measurements of these still mysterious fractions. Because, conceptually, OC/EC and BC are mostly defined by the methodology used to measure it. Once again the aim of this comment is to clarify the manuscript. I'm totally aware that these methodological issues are a no end story. . .

4/ Inter annual trends of EC (/BC) are very interesting. Unfortunately no data on the evolution of the vehicular fleet in Spain is shown in the manuscript. Considering the

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statistic provided by the European automobile manufacturer association it seems that the fraction of diesel in Spain increases from 50.6% in 1999 to 70,1% in 2009. Can these trends of EC (/BC) be discussed in the light of more official statistics of the evolution of the vehicular fleet in Spain? Specific comments:

2/ p6973 line 26 : NO2/(OC+EC) 3/ p6974, line 10 “transferred” instead of “formed” 4/ p6974, line 15-19. Be more quantitative 5/ p6975, line 1-5 : Develop this aspect (WHO report etc..). 6/ p6976, line 9-12 : not here 7/ p6976, line 13-16 : develop this section. 8/ P6976, line 18-20. Be more specific. What kind of “state of the art techniques”? what type of organic species? In a more general point of view, is this paragraph useful? 9/ p 6977 , sampling section. Information regarding the site typologies and classification would be more useful here than in S1. 10/ P6979, line 3 : be more specific. What are the LOD of elemental analyzers? 11/p6986, line 19 : 5 μg m-3 12/ p6987 section 3.1 and 3.2 : How are defined : small, midsized and large cities? Can you compare these results with other locations in Europe? From my experience the concentrations of EC and OC seem quite low, especially in urban environnement. 13/ p6990 line 11 : rate of 0.3 yr-1 14/ p6990 line 18 : R2 of 0.99, n=? 15/ p6990 line 22 : μg m-3 16/ p6990 line 26 : Is this assumption corroborated by some trace elements (if available)? 17/ p6991 line 5-15 : Considering the results previously presented, this first paragraph of the discussion is very speculative. For the traffic sites, the ratios OC/EC observed here (1.6-1.7) are too high to state that OC is mainly from primary vehicular exhaust, especially considering a vehicular fleet largely dominated by diesel vehicles. Typically the ratio OC/EC is lower than 0.5 for diesel emissions (for example: Zielinska et al, 2004 or El Haddad et al, 2009). Thus, in the traffic sites discussed in the paper, we can consider that $\sim$75% of the OC is not from diesel exhaust. Considering that 70% of the vehicle fleet is diesel (with respect to the statistic of the European automobile manufacturer association) and EF of gasoline cars, primary gasoline emissions can not explain the OC concentrations observed. 18/ p6991, line 20 : “biomass burning is probably causing an increase of 1μg/m3 in. . .” how this value is estimated? 19/ p 6991, line 25 : 1.0 or 1.6 for the lower OC/EC ratio in traffic sites? 20/ p6993, line 9 : OC/EC

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ratio is also very high for gasoline exhaust (typically 5-10)

Fig 3. Years of works have been necessary to obtain the data presented in this figure. All this work is not emphasized by the figure. Fig 6 : Personal curiosity.. A intense peak of OC and EC is observed in Barcelona during winter 2008. Do you have an explanation of this increase of the concentrations at this period?

Zielinska, B., Sagebiel, J., McDonald, J.D., Whitney, K., Lawson, D.R., 2004. Emission rates and comparative chemical composition from selected in-use diesel and gasoline-fueled vehicles. Journal of the Air & Waste Management Association 9, 1138–1150. El Haddad I., Marchand N., Dron J., Temime-Roussel B., Quivet E., Wortham H., Jaffrezo J.L., Baduel C., Voisin D., Besombes J.L., Gille G. Comprehensive primary particulate organic characterization of vehicular exhaust emissions in France. Atmos. Environ., 43, 6190-6198, 2009.

Please also note the supplement to this comment:
http://www.atmos-chem-phys-discuss.net/13/C2213/2013/acpd-13-C2213-2013-supplement.pdf

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Interactive comment on Atmos. Chem. Phys. Discuss., 13, 6971, 2013.