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## ***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.***

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Received and published: 10 May 2013

Dear Editor,

Please find below my comments on acp-2012-1016.

This manuscript presents a comprehensive phenomenology of non-mineral PM carbonaceous fractions in Spain over the last decade. This subject is of prime interest for the scientific community as well as for decision makers since organic matter is one of the top major PM species and a better knowledge of its main emission sources and (trans-)formation processes in the atmosphere is strongly needed for the elaboration of

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efficient PM reduction action plans. Moreover elemental carbon (EC), which presents much lower concentrations, is gaining more and more attention from decision makers due to its climatic relevance as well as its potential use as an indicator of PM health effect. Authors proposed a clear, concise and well-written manuscript. It also provides valuable information that could be extrapolated elsewhere.

It is to note that the used database is composed of results obtained from very different sites, at different periods and using different analytical approaches. Moreover, the latter analytical methods are known to be subject to various artifacts, some of them probably still to be discovered. This induces large limitations to the study. Nevertheless, authors extensively describe and discuss these limitations. They also propose few insights on possible uncertainty ranges for results presented here, while a solid uncertainty calculation seems hardly feasible in the context of such high and unknown artifact influences. It thus comes that the present manuscript could be considered as a call for standardisation of carbonaceous PM sampling and analysis, which authors do not really insist on. Given these issues, authors decided to mainly discuss inter-annual trends based on nmC (OC+EC) concentrations, which to my opinion sounds very appropriate. They also conclude the manuscript considering the need to develop EBC monitoring. Authors could then be asked to clearly precise their opinion on a what kind of strategy should be chosen for the monitoring of carbonaceous aerosols (if any): for instance, would this strategy rely on collocated nmC and EBC measurements, and then the estimation of OC from the difference of these two parameters (disregarding OC and EC thermo-optical measurements)?

Besides these technical points, data treatments presented are scientifically-sound and well presented. Overall, I would recommend the publication of this manuscript within ACP. However I would have three main concerns, that should be answered before publication:

- the quality of figures is rather low, especially Figure 8 (hazy; X-axis: L = "July" ?; Y-axis not readable).

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- apparent fluctuations within long-term trends (when looked from far away). Interannual trends presented in Figure 6 for long-term datasets globally seem to correspond to two different trends (significant decrease before about 2008, but low decrease, if any, afterwards). This would also be in line with statements repeated in the paper that short-database (<5y) do not show such obvious decreasing trend than longer database. If true, what would be the impact of such a decrease of the decreasing trends on proposed explanations related to the influence of traffic EURO regulations ?

- the ambiguity on the real impact of biomass burning on total organic matter concentrations as presented by authors. Indeed, it is stated P. 6991 L. 5-9 that "The spatial variability of nmC across different atmospheric environments in this study shows that anthropogenic carbonaceous aerosols in Spain within the period 1999–2010 mainly originated from road traffic and in a minor proportion from biomass burning ...". The way this statement can be done would certainly needed to be precised and detailed a little bit more. This statement is also rather vague, as no numerical values are proposed. This issue actually concerns the whole manuscript, which globally point out traffic emissions as the very major sources of nmC in Spain (which I could believe), but also frequently indicate significant influences of biomass burning emissions (e.g. P. 6991 L. 19-22: "biomass burning (domestic, agricultural and forest fires) is probably causing an increase of around  $1 \mu\text{g m}^{-3}$  in the annual nmC mean at regional background sites in northern Spain with respect to the rest of the Spanish territory.": this extra  $1 \mu\text{g/m}^3$  on a yearly basis may represent up to 50% or more during burning periods, isn't?). Could authors please try to precise the impact of biomass burning on carbonaceous matter in Spain ? and more extensively compare this impact to the ones in the rest of Europe? Given the elevated influence reported for biomass burning emissions on air quality within other European countries, these clarifications would seem to remain within the scope of the manuscript, which also attempt to indirectly elucidate major carbonaceous matter sources.

Finally, some specifics comments below:

- Para. 2.2: could be precised as soon as here than Ca and Mg may have different origins than mineral dust aerosols.

- P. 6692: What about the seasonnal variations of non-fossil vs. fossil OC ? Does it tell something about sources ?

- P. 6692: Is cooking expected to be a possible major source at every sites (in particular rural and remote ones) ?

With my Best Regards

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

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