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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.

Anonymous Referee #1

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The manuscript by Querol et al. presents long-term measurements of different fractions/metrics of carbonaceous aerosol associated with different fractions of particulate matter from different monitoring sites using different (and occasionally indirect) analytical methods. Because of the large variability (sampling, analytical) individual measurements are associated with substantially different uncertainties; however, these are not mentioned in the manuscript or supplementary material. The most significant issue of the manuscript is associated with the overall effect of the limitations. The authors outlined the potential limitations, however, they failed to provide how they will bias their findings. At its simplest form, this could be an overestimate/underestimate evaluation (I think the magnitude should also be discussed). For example, if all limitations lead to

C1859

an underestimation, then, the conclusions represent a conservative estimate. This is a critical point because of the many-many limitations of the datasets and the suggestions for air quality policy with data that, afterall, may be not that different. My second concern is associated with the potential implication for air quality policy and the suggestions to include metrics of carbonaceous aerosol. It is not clear whether authors propose to monitor carbonaceous aerosol because of their potential health effects or as an alternative to existing particle mass measurements. If the former, then, at this point, there is no sufficient, statistically-significant and consistent evidence (strong indications, yes) that carbonaceous aerosol are associated with adverse short- and longterm health effects (see the Integrated science assessment on PM2.5 done during the last revision of PM2.5 standards, available at www.epa.gov). The large variability in chemical composition, large sampling/analytical errors are also limiting factors. If the latter, then this is not a valid suggestion because other non-carbonaceous aerosol species showed stronger health effects than carbonaceous (e.g. iron has a stronger potential to form ROS than organic compounds; Ca has a higher risk for respiratory symptoms than BC/EC or OC, etc). With respect to continuous EBC, this is practically continuous measurements of soot carbon (initial air quality measurements before the use of PM10 and PM2.5 mass) which has been rejected in the past because of the inability to account for other types of sources and pollutants as well as changes in traffic emissions (for BC to SOA/NOx-rich emissions). My suggestion here is to exclude this section because it does not rely on the data of the manuscript, it does not include a detailed review of existing health literature (other than one report and one paper) and it is highly speculative. Other comments include: One important limitation is the absence of comparison of EC/BC, OC and nmC measurements to PMx. It is important to know the percentage contribution of these components to mass. Page 6996 lines 5-10. Discussion about the equation OC/EC=aEC^b. I do not understand the practicality of this equation as compared to OC=aEC^b. Why? It is basically $y=ax^{b}$ with y=f(x) so, $f(x)=ax^{b}$ or $z/x=ax^{b}$ which leads to $z=ax^{(b+1)}$.

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C1861