**Atmospheric
Chemistry
and Physics
Discussions**

Interactive comment on "Sources, trends and regional impacts of fine particulate matter in southern Mississippi Valley: significance of emissions from sources in the Gulf of Mexico coast" *by* M.-C. Chalbot et al.

Anonymous Referee #1

Received and published: 21 February 2013

General comments

The paper is mainly focused on source apportionment by Positive Matrix Factorization of PM2.5 sampled in North Little Rock (USA), the topic is interesting, the data set is large and reliable, the paper is well organised, then the paper deserve the publication. However, here below authors can find some suggestion to improve the paper before publication.

Specific comments

My main criticism is related to the choice of parameters (chemical markers or compounds) used for PMF analysis. Although the use of different form of the same elements (or compounds) sometime could give a best source fingerprint, in the paper some parameters are redundant in the PMF analysis. In particular, I understand the use of both K and K+ in the PMF analysis because K (total content) is more related to crustal source, conversely K+ is more specific for biomass burning source (and primary marine). I'm less convinced about the use of both S and SO4, actually S comprise also methanesulfonate (MS) from atmospheric oxidation of biogenic dimethylsulfide, but at this site MS have to be negligible because the SO4/S ratio is higher than 3 (line 13 page 836) likely due to underestimation of S and absence of MS. I disagree to the use of OC1, OC2, OC3 and OC4 together with total OC. The use of more specific fraction of OC makes unnecessary insert total OC in the PMF. Besides, in quantification procedure by PMF all these parameters (K - K+, S-SO4, OC1, OC2, OC3, OC4 –OC), representing more or less the same amount, give an overestimation of the contribution of the source. May be the authors exclude the double parameters from source quantification, but this is not reported in the text; please the authors better explain their approach.

Sampling site and measurements: analytical procedures are simply listed (line 11-14 page 831) and no references to find further information are reported. In particular, what procedures for extraction were used for ICP and ICP-MS analysis? Total or the soluble content is achieved? If soluble content is determined, in which condition? What ICP-MS is used? What resolution is used?

Page 837 formula (9). Why for the calculation of secondary species coefficients are applied to NO3-, NH4+ and SO42-? In my opinion only the secondary species: NO3-, NH4+ and SO42- have to be considered in this specific class of particles. Only NO3-, NH4+ and SO42- have a secondary source from oxidation processes in the atmosphere and/or gas to particle conversion, counter ions have not to be included here. At line 11-13 Authors state that "Nitrate may also be associated with coarse particles

from neutralization of nitric acid with sea salt or calcium carbonate" this is right but Na or Ca have not be included in secondary species even if chemically link to secondary species. Besides, if they are included in "secondary species" they will be considered two times in reconstructed PM. For instance Ca is reported both in "soil" and in "secondary species". Please change this section in accord to these suggestions. Page 837 line 13. I suppose the conversion factor 1.6 is related to OC – OM conversion but is not clear in the sentence put just after nitrate neutralization.

Technical correction

Labels in the figures often are not visible, please use larger size font. Line 12 page 829: "….mass concentrations decreased from 24% to 28%..." I suppose "increases" is the right word. Please specify the acronyms PIXE, ICP, ICP-MS at lines 11-12 and Lat (°N) and Long (°E) at line 4 page 831.