

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 #3

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The manuscript contains important new results and findings, and deserves to be published. Most of the paper has been written carefully. However, the authors need to allow for the comments below, before the final publication can be accepted.

Major comments

On pages 835-836 the authors present the TrMB model. However, it is not totally clear from the manuscript whether dry and wet deposition was really taken into account in this study, and if so, how exactly this was done. They write that the beta factor includes

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all these scavenging processes, but no details are given how this was modeled. The authors should present in more detail what kind of dry and wet deposition modeling was used and how. E.g., in chemical transport models, a very wide range of deposition models are currently used, some more realistic than the others. Whether deposition modeling was included or not, and how exactly, is a crucial issue with respect to the reliability and accuracy of the predicted results. On p. 829 “diesels in ships . . . are subjected to modest emission requirements.” This may be true in the U.S., but certainly not globally. In Europe, it is partly vice versa, e.g., stringent emission limits are currently implemented for shipping in the SECA regions. The authors should define which country /continent they claim the statement is correct for. The same comment applies to conclusions; please revise that statement: “marine traffic and associated emissions of gaseous precursors and particles will grow substantially. . .” In conclusions, it has been written “The annual variability of biomass burning contributions to fine particle mass correlated very well with the burnt area by fires in the US which is directly related to the frequency of El Nino and La Nina events that are modified by climate change.” Later on, “Through this analysis, the effect of events associated with climate change on PM_{2.5} from biomass burning was identified . . .”. In the abstract and later on in the manuscript, the authors discuss the relations of climate change and wild-land fires. Abstract: “The annual variation of biomass burning particles was associated with wildland fires in southeast and northwest US that are sensitive to climate changes.” It has of course been shown in other studies that wildland fires are expected to increase due to the global climate change. However, a ten year period of data from one location is certainly too limited data set for making any judgements on the temporal evolution of climate change, or any of its implications. In addition, wildland fires within one country may be substantially influenced by social factors such as e.g. wildland protection and forestry policies. Throughout this article, it should therefore be made clear that the authors are not trying to make any conclusions regarding climate change, or its influence on the occurrence of wildland fires, based on the data of this study. Regarding the causal relationship of El Nino and La Nina events and the occurrence of wildland fires

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in specific regions of the U.S., concrete evidence should be shown if the authors wish to prove this relation to be correct. There are at least two issues here: 1. how exactly do these events affect the year to year climate in the considered domain ? and 2. How these climatic differences affect the occurrence of wildland fires (considering that there are a lot of confounding factors such as social policies, and the year-to-year variation of weather during the summer season) ? I therefore recommend the authors to rewrite these parts of the manuscript. The source categories (as defined in the article) are partly overlapping, at least 'primary traffic' and 'diesel particles'. Are primary diesel traffic particles part of either, or of both ? These overlaps should be discussed in the paper.

Specific comments

The title is unnecessarily long, it will suffice to write Sources, trends and regional impacts of fine particulate matter in southern Mississippi valley The article also does not really focus on shipping emissions, so the latter part of the title (implications of 2 shipping emissions and SO₂/NO_x emission reductions) should be skipped. The abstract. "The slower decline for NO₃- particles (0.1 μg/m³ per year) was attributed to the spatial variability of NH₃ in Midwest." It is not clear how the longer term trend is caused by spatial variability. The trend should be attributed to e.g. temporal trends of precursor compounds. "Overall, more than 50% of PM_{2.5} and its sources originated from sources outside the state." The authors probably mean '50% of PM_{2.5} and its chemical constituents . . .'. Introduction. p. 830 line 14. Delete 'unique' or define exactly in which respects this would be unique. Methods. p. 832, Define F_{peak}. p. 833. Define α. p. 838. OC₁, . . . OC₄ are not defined. p. 840. What was the quantitative criterion used for statistical significance ? p. 842. "These similarities suggested the robustness of the trajectories regression analysis to determine the spatial distribution of PM_{2.5} mass and source contributions in an urban area." Strictly, these differences only demonstrate that the two alternative statistical procedures produce similar results. The 'robustness' of the whole analysis chain has not been demonstrated by this finding. Please reword.

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Technical corrections

p. 837. Give a reference for the empirical factor in $OM = 1.6 OC$ p. 842. line 10. “Both models”, please define which models these are.

Interactive comment on Atmos. Chem. Phys. Discuss., 13, 827, 2013.

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