Responses to Referee #3 comments
ACP-2012-816

Comment #1: *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 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 that 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.*

Response: The regression coefficient is the product of the transfer coefficients, Q, T and E, describing emissions, dry/wet deposition and entrainment procedures. The deposition may be indirectly computed if emissions and entrainment are known. For example, a region emits X μg/(m^3 hr), the entrainment factor is 1 μg/(m^3 hr) and the regression coefficient is β=0,5, then the deposition losses for the source region on day *i* would be equal to 0.5 μg/(m^3 hr). Alternatively, the values of Q, T and E from a source region may be computed by applying the TrMB model for multiple sites, but this was not the objective of this study. To clarify this issue and provide more information, the statement is updated as follows: "**…β_j (in (μg/(m^3hr)) are the regression coefficients of regions describing the combined outcome of emissions from the area (Q_j), aging and pollutants removed due to gravitational settling, turbulent mix-out, and wet deposition during transport to the receptor site (T_j) and the entrainment of particles from the j-sources to describe the dissociation between the trajectory and the transport (E_j). The units of the Q,T and E factors were in μg/(m^3 hr) within the region…**"

Comment #2: *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. . ."*

Response: We modified the statement as follows: "**The diesel engines in ships operate on fuel that can have extremely high sulfur content and porphyrin-content rich in V and Ni (termed as bunker oil) and they are subjected to modest emission controls in the US and around the world, with the exceptions of passenger ships in Baltic Sea, North Sea and English Channel.**" to highlight that emission controls apply in three regions in northern and western Europe.

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Comment #3: *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 PM2.5 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 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 reconmmend the authors to rewrite these parts of the manuscript.

Response: The goal of this work was not to identify links between climate change and wildfires. Our intentions were: (i) to describe the lack of an annual trend on biomass-related PM2.5 and relate it to the inter-annual variability of wildfires; and (ii) evaluate the results of the trajectories regression analysis by comparing them with the locations of wildfires during the monitoring period. Based on the links between wildfires and climate change (demonstrated by others; references are cited in this manuscript), we indirectly attempted to indicate the possible influence on climate change on air pollution. To clarify this, the statement were modified as follows:

Conclusions: "**The annual variability of biomass burning contributions to fine particle mass correlated very well with the burnt area by fires in the US**" and "**Through this analysis, the effect of wildfires on $PM_{2.5}$ was identified**"

Abstract: "**The annual variation of biomass burning particles was associated with wildland fires in southeast and northwest US**"

Comment #4: *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 operlaps should be discussed in the paper.*

Response: In the analysis of the third factor, we concluded that the "diesel emissions" factors was associated with diesel particles from sources other than vehicles. In the description of the first factor, we also identified that the "primary traffic particles" were associated with both gasoline and diesel vehicles because of the presence of Ni and V in the factor. So, "diesel traffic particles" are part of the "primary traffic particles". This separation was further confirmed by the trajectories regression analysis that showed differences between Factors 1 and 3.

Comment #5: *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 shipping emissions and SO2/NOx emission reductions) should be skipped.*

Response: The title was modified to "**Sources, trends and regional impacts of fine particulate matter in southern Mississippi valley: significance of emissions from sources in the Gulf of Mexico coast.**" We believe that it better describes the content and the conclusions of this work.

Comment #6: *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 PM2.5 and its sources originated from sources outside the state." The authors probably mean '50% of PM2.5 and its chemical constituents . . .'.*

Response: We modified the statements as follows:

- "**The slower decline for NO_3^- particles (0.1 μg/m^3 per year) was attributed to the increasing NH_3 emissions in Midwest.**"
- "**Overall, more than 50% of $PM_{2.5}$ and its components originated from sources outside the state**"

Comment #7: Introduction. p. 830 line 14. Delete 'unique' or define exactly in which respects this would be unique.

Response: It is deleted.

Comment #8: Methods.

- p. 832, Define Fpeak. p. 833. Define alfa.
- 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 PM2.5 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.

Response:

- We added the following statements: "**The F_{peak} value, a user-defined non-zero rotational parameter, controls the subtraction of the profiles from each factor to eliminate the remaining rotational ambiguity by forcing to add one G vector to another and subtract the corresponding F factors from each other and thereby yield realistic solutions.**" and "**The α value, a user-specified variable, defines the distance of outliers ($\alpha\sigma_{ij}$) from the fitted value in order to include them into the analysis.**

$$\left(x_{ij} - \sum_k g_{ik} \cdot f_{kj}\right) \Big/ s_{ij} > \alpha \qquad \textbf{(5)}$$"

- OC1 through OC4 were defined previously (Methods section, Sampling sites and measurements subsection).
- The quantitative criterion was the p-value and the threshold was 0.001. The significant criterion is presented in Table 3. To clarify this, we also included it in the text (Results and Discussion section, Annual trends of fine particles and its sources subsection)
- We modified the statement to show that the good agreement of the models with and without the intercept indicated that contributions from sources within the geographical domain of the study accounted for all of $PM_{2.5}$ mass and its components "**These similarities suggested that $PM_{2.5}$ mass and its components originated from sources within the selected geographical domain.**"

Comment #9:

- 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.

Response:

- The reference Turpin and Lim, 2001 is added in the manuscript.
- It refers to the models with and without the intercept. To clarify this, we deleted the word "both".