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Comment on acp-2021-495

Anonymous Referee #2

Referee comment on "Changes in $PM_{2.5}$ concentrations and their sources in the US from 1990 to 2010" by Ksakousti Skyllakou et al., Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2021-495-RC2, 2021

This manuscript describes the application of a chemical transport model, PMCAMx, to follow the rends in PM2.5 concentrations in the US and their response to emissions changes by simulating three different years: 1990, 2001 and 2010. They use an integrated source apportionment approach, PSAT, to assess the contribution of specific source categories to PM concentrations and exposures.

In general, the manuscript is well written and addresses an important scientific topic: how sources impact pollutant levels and potential exposures. However, at this time there are some significant concerns with the manuscript.

The main concern has to do with evaluation and how it is done and how it is characterized. First, the performance criteria provided are taken from Morris et al., (2005). Those criteria are for assessing daily simulations, not annually-averaged concentrations. One expects much better error statistics (annual biases will not be affected as such). The authors should go back and do a daily evaluation for the applicable periods modeled. This is really a must. Also, there has been updates by the Ramboll team in terms of performance evaluation (e.g., Emery et al., 2017, doi.org/10.1080/10962247.2016.1265027) which should be used. They also make recommendations on considering subdomains in the evaluation. A second major issue is that they do not include California in their analysis, stating: "We have excluded the region of California from this analysis because the coarse resolution used in this application does not allow PMCAMx to capture the significant gradients and high concentrations observed in that area." If that is the case, it can not capture the exposures properly, either, and thus that area should not be included in the analyses. Either California should be included in the evaluation, or it should not be included in the rest of the analyses. Following the recommendations of Emery, their result would suggest California results should be subject to an independent evaluation. Also, the evaluation typically precedes the rest of the results given its importance.

A second concern it their statement "The predicted reductions in sulfate concentrations are less than the reductions in emissions due mainly to the non-linearity of the aqueous-phase conversion of SO2 to sulfate (Seinfeld and Pandis 2016) (Table 1)." While it is true that the formation of sulfate is somewhat non-linear, they have not shown this to be the major reason. This is highlighted by the difference between the reduction in concentrations and exposures, which, all else equal, must be linear given how it is calculated. They state that the difference between the concentration reduction and the exposure reduction as "The change in exposure is a little less, -40% on average due mainly to the location of the major SO2 sources relatively away from urban centers." I think they mean that where the REDUCTIONS occur are mainly away from the urban centers (this is an important difference: if the reductions were uniform in a relative sense, this difference should not occur). Might that also be important in terms of average levels? i.e., if the reductions occur in areas where the conversion is slower, the chemistry can be linear and one gets the result found here. Also, the sentence starting on line 364 is awkward.

The authors have "The major reasons for this behavior are the simultaneous decreases in NOx that have led to increased SOA formation yields and the time required for the formation of this SOA which is often produced away from its sources in high urban density areas. The reasons for this behavior are complex and will be analyzed in detail in future work. However, at least part of the explanation is due to decreases in NOx concentrations over the same period and associated increases in SOA yields." The second sentence seems to suggest that the first sentence is incomplete and will be assessed in the future. This is awkward.

In calculating exposure, the authors should use the census for the years modeled. Population distributions change over time. Maybe have both results (stagnant population and a dynamic one).

The time period modeled is getting rather old. Why not include 2020?

More discussion on how this work compares to other studies that have examined the impact of emissions changes on air quality is in order (e.g., the EPA group, others). Are there any significant differences in their findings? If not, what is the main scientific contribution?

The “,” in line 330 is not needed.