

Interactive comment on “Emission factor ratios, SOA mass yields, and the impact of vehicular emissions on SOA formation” by J. J. Ensberg et al.

Anonymous Referee #2

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This paper provides an excellent and comprehensive analysis of the combined implications of several measurement and modeling studies of Organic aerosol in the Los Angeles basin, a large metropolitan area with a long record of air pollution issues that are related almost entirely, it is thought, to vehicle emissions. What this paper has done that is novel is to use the large number of measurements provided by a recent multi-million dollar study, along with inventory and regulatory data, to assess whether this is true, and if it is, whether the contributions from diesel and gasoline can be quantified. I am a bit disappointed that it has not generated as much online discussion as I had expected (given the offline discussions that I have heard), as that would have allowed

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the authors to expand more on many of the underlying limitations of their analysis.

I support publication of this paper in ACP. The concise style of the paper is great for brevity, but there are a number of minor points that the author could address in a slightly extended revised version (rather than the present Science-like format) that would provide more context and literature comparisons, especially for their somewhat unexplained adoption of the unpublished Zotter result as a starting point.

Suggested starting points for making this paper more complete:

1) I find the abstract organization is a bit awkward, since it first states that the paper will quantify the diesel and gas fractions and then concludes that large fractions of OA may not be from vehicles at all. Since the latter seems to be a prerequisite for the former's answer to be important, I'd suggest rearranging the order to make it clear that while the initial point of the paper had been to separate the contributions of gas and diesel, in the end the paper could not establish the fraction that was from vehicles (in total) well, and hence the uncertainty of the diesel/gas contributions is large and/or can only be quantified relative to each other. 2) I think the most original aspect of this paper is the use of the dual constraints of the matched yields and propagated errors that are implied by the gas/diesel split. Since this is a subtle and new (for this issue) point, some additional explanation could be merited so that the approach may be more easily interpreted and repeated (and cited) by others. 3) While it is clear in the manuscript that the study is for LA, it would be instructive and useful for the authors to provide some comparative information about the extent to which similar conditions may/not exist for other metropolitan areas, or at the very least by including the comparison to identify why these results cannot be extrapolated to other areas for which such detailed information and inventories do not yet exist. 4) I think Referee #1 had an important point, and I think that a response that clarifies this point would improve the paper. Has the uncertainty in the 0.45 days value been explored? 5) Section 3.2.1 – how is a “significant contribution” defined? 6) The authors have used “county” fuel sales; what is the uncertainty associated with this assumption? Are these month totals for June 2010?

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What is the contribution from fuel in non-LA counties? 7) The authors have done a reasonable job of estimating EF uncertainty, but it should be noted that Tables 4-6 are not complete, and there is not "closure" of the compounds for which EF are tabulated and the measured total SV/LV OOA. So the conclusion that SOA yields must be higher than modeled can mean either that compounds are missing (that have non-zero yields) or that the compounds that are included in the models have higher than currently modeled yields. The uncertainty for the missing compounds is more difficult to estimate, especially using tunnel studies not conducted in LA driving conditions. 8) Figure 3. I think the "required" Aggregate SOA Mass Yield is from measurements, right? Specifically, 70% of SVOOA based on Zotter/Hayes? This should be clarified in caption. If so, it would be good to repeat both the assignment and uncertainties mentioned by Hayes/Zotter in determining that SVOOA was "fossil-vehicle SOA". 9) Perhaps other methods for quantifying fossil SOA should be considered, especially if the Zotter paper is, as listed in the references, still not published. I recommend holding publication of this paper until that critical reference is accepted for publication. Moreover, the authors would be more circumspect to include explicit consideration of other approaches for quantifying fossil SOA, given the substantial uncertainty. 10) P. 27795 conclusions "vehicular emissions do not dominate SOA concentrations attributable to anthropogenic fossil activity in Southern California": I think what is meant here (if above interpretation is correct) is that the amount of SVOOA that is fossil is not as large as stated/implied by Hayes et al. (if that paper is interpreted to say that all SVOOA is fossil SOA). I think it is worth clarifying that this conclusion may be specific to the AMS-PMF method of identifying fossil SOA. 11) A similar point is that this approach assumes that emissions from different sources do not interact, i.e. that the emissions of non-vehicle sources do not affect the yields of vehicle sources. This independence underlies the use of single-reactant yields in models, but there is certainly both some laboratory and theoretical evidence to suggest that it may not be sufficient to understand cities as complex as LA. 12) Aren't there also non-fossil sources of CO? How are these accounted? How does this affect the Zotter result?

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