Author's response to reviews

Title: Public Health Impacts of Secondary Particulate formation from Aromatic Hydrocarbons in Gasoline

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Author's response to reviews: see over
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Dear Dr. Forastiere:

Thank you for the opportunity to provide a revised manuscript for MS: 1341270580800656, “Public Health Impacts of Secondary Particulate formation from Aromatic Hydrocarbons in Gasoline.” The responses to comments are provided below; in addition, the revised manuscript has all changes in redline-strikeout. Thank you, best,

Katherine von Stackelberg

Reviewer 1

Line 77: The authors state that SOA yields are lower under high NOx. Yet, aren’t gasoline exhaust emissions released into high NOx atmospheres? What is the SOA potential/implication of this?
Response: The SOA yield is determined by the abundance of NO after the parent aromatic compound has reacted with OH to form an aromatic peroxy radical. For toluene, the time scale for that initial oxidation is about 2 days (ref: Table 5.2 of Seinfeld & Pandis 1998 textbook) at which point the radicals are no longer within the vehicle exhaust plume.

In the CMAQ simulations: How did the authors separate out the aromatic-derived SOA, versus the other OA? Is it tagged in the mechanism? Or is it a sensitivity? How does CMAQ with CB05 keep track of the carbon in the aerosol/PM2.5 fraction? Response: CMAQ provides total anthropogenic SOA as a standard output. With the CB05 mechanism in CMAQ, aromatics are the only precursor for anthropogenic SOA (unlike SAPRC mechanisms, in which alkanes also contribute). Thus, no extra effort was required to separate the aromatic-derived SOA from the standard CMAQ output. The fraction from gasoline is based on source attribution from the SPECIATE database.

Line 68-69: Aromatics contribute to 0.008 – 0.02 ugC/m3 PM2.5; And then Lines 142-143: aromatic hydrocarbons in gasoline contribute, depending on the specific region, approximately 0.1 to 0.45 μg/m3 of PM. Are these two sentences consistent? Response: Yes, they are. In the first case, the units are microgram of carbon per m3 of PM2.5, while in the second case, the results are not normalized for carbon. However, to be consistent, 0.08 (not 0.008) and 0.2 (not 0.02) have been revised to units consistent with how they are referred to later.

Line 157: Low NOx conditions- what is meant by this? In chamber studies, isn’t this represent experiments without any NOx? And do we really reach these pathways in the US? Response: By performing experiments at both high and low NOx limits, the oxidation conditions (initiating oxidant and fate of peroxy radicals) can be maintained relatively constant over the course of the experiment, allowing for the evaluation of the effect of NOx levels in the atmosphere on SOA formation, and so that
these processes may be included in models such as CMAQ (Ng et al. 2007). For instance, compounds like benzene are mainly emitted in source-rich regions; with its slow reactivity, however, benzene can be transported to areas with a lower NOx level before it react substantially, resulting in a higher SOA yield than if it reacted in the immediate vicinity of its sources. By performing experiments at two extreme NOx conditions, the authors were able to obtain SOA yield parameters under high- and low-NOx conditions, allowing for the parameterization of the NOx dependence for atmospheric models, based upon the reactivity of organic peroxy radicals (Ng et al. 2007).

Region-specific random effects – what are the regions chosen, and how were the regions determined? **Response:** The regions are US standard accepted geographic regions based on the US census designations.

Line 166 – Does SPECIATE take into account the annual variation of regional VOCs in gasoline? (e.g., winter versus summer mixes?) **Response:** The National Emissions Inventory (NEI) accounts for geographic and seasonal variations in fuel composition when computing the overall VOC emission rate by county. However, a single speciation profile is applied to all VOC emissions from gasoline engines to obtain emission rates of the individual aromatic compounds.

Is it possible to show the concentration-response function(s) applied in BenMap? **Response:** The US EPA has used the concentration-response functions in BenMap for many years to develop regulatory impact analyses; this would not seem to be the place to show these as they have been specifically discussed and vetted numerous places. All of the functions can be found in the Technical Appendices of the BenMap users manual ([http://www.epa.gov/air/benmap/models/BenMAPAppendicesOct2012.pdf](http://www.epa.gov/air/benmap/models/BenMAPAppendicesOct2012.pdf)). These are cited in the text, and we do not think further explanation is necessary given that these form the basis of all regulatory evaluations and full information can be found at the URL above.

Just to clarify: $7.4M in 2006 dollars (USEPA 2010) is the value of mortality risk? For 1 person? **Response:** That is correct. This is the regulatory value used by US EPA ([http://yosemite.epa.gov/ee/epa/eed.nsf/pages/MortalityRiskValuation.html](http://yosemite.epa.gov/ee/epa/eed.nsf/pages/MortalityRiskValuation.html)) “EPA recommends that the central estimate of $7.4 million ($2006), updated to the year of the analysis, be used in all benefits analyses that seek to quantify mortality risk reduction benefits regardless of the age, income, or other population characteristics of the affected population until revised guidance becomes available.”

Lines 228-299: Is there some estimate of uncertainty associated with the measurement-based estimates of SOA and SOA from aromatics? Since this is not a direct measurement, I am wary that there is not any error associated with these numbers. **Response:** Kleiniedienst et al. (Atmos. Environ. 41: 8288-8300, 2007) estimate the uncertainty to be ± 33% for aromatic SOA. Note that for purposes of the regression between predicted and observed, measurement error in the dependent variable (observations) does not bias the coefficient (slope).

Lines 248-250: Is this reasonable to adjust the aromatic SOA concentrations uniformly across the US? I would expect that the ratio of gasoline-powered aromatic emissions to aromatic emissions from other sources may differ dependent on region. **Response:** We looked into this question as we were developing the analysis and in response to an internal peer-review process that we went through prior to
submitting the manuscript. “Dependent on region” could refer to rural vs. urban regions, or geographic regions across the country. With respect to rural vs. urban, emissions may differ in rural vs. urban areas, but SOA formation is such that impacts are experienced in areas other than where direct emissions occur, and this “travel” so to speak is accounted for by the CMAQ model. After gasoline, the next largest source of aromatics is solvent usage, similar to (f) below and occur predominantly in urban areas, just as with gasoline-powered aromatic emissions (figure taken from Reff et al. 2009). Most major refineries are also in close proximity to urban areas. Finally, VOCs and PM show similar spatial patterns with respect to emissions (Reff et al. 2009).

Table 5: Is it possible to include a definition for “Beta” in the Table header or footer? Response: Beta has been defined (the percentage change in premature mortality associated with a 1 $\mu g/m^3$ increase in PM$_{2.5}$ concentration).
Reviewer 2

Major Compulsory Revisions

Add text that explains the preliminary nature of the quantitative results due to the following issues:

1. Our current representation of national speciated organic emissions and ambient concentrations has important uncertainties. Spatial and chemical variability in emissions, the amount of time needed for PM formation, and varying ambient conditions at different scales all contribute to variability in SOA formation. With 2 orders of magnitude difference in estimate of aromatic contributions to PM2.5 (0.08 to 0.2 in Background paragraph 2), and the possibility that even different types of aromatic sources do not equivalently contribute to SOA formation (similarly to VOC reactivity and ozone), the use of a linear scaling factor to estimate adjusted aromatic SOA concentrations with respect to other sources (the 0.69 multiplier, Results paragraph 2) is a very rough estimation method. **Response:** The aromatic contribution must be taken in the context of the overall PM$_{2.5}$ concentration, e.g., the difference in the proportion of PM$_{2.5}$ contributed by aromatic hydrocarbons is less than two orders of magnitude, particularly since those units are on a carbon basis (e.g., µg C/m$^3$); the difference is actually 0.1 to 0.45 in µg C/m$^3$, and the text has been revised to be consistent. However, it is certainly true that uncertainties exist, and these are discussed further on pp. 11-12.

2. Model performance of aromatic SOA is very poor, as discussed, and while giving the directional contribution and a rough estimate of the quantitative contribution of gasoline aromatics to SOA, the application of a linear regional scaling factor may not represent the actual relationships at the exposure scale. **Response:** However, model performance of aromatic SOA, while poor, has improved substantially. But the point is taken and text has been added on pp. 11-12.

3. Please add mention of the linear (or nonlinear) contribution of aromatics to SOA and PM2.5 formation, the dependence on varying atmospheric physical and chemical conditions, and how these processes may or may not impact the results or conclusion. (Or note that these processes are still only roughly understood.) **Response:** text has been added pp. 11-12.

4. Exposure to road-related sources may be much higher than currently represented by simulation tools. For example, recent near-source and commuter studies have shown significantly higher PM exposure on and near roadways than would be estimated using CMAQ and BenMAP. Also, some studies indicate differences in health impacts from differing PM2.5 compositions. These issues may impact the resulting range of estimates. **Response:** These factors are all suggest that the analysis is more likely to underestimate than overestimate potential impacts as has been noted on p. 12.

Minor Essential Revisions

1. Line 29 (Abstract, methods) should read “… and the results ARE compared...” **Response:** Change made.

2. Line 31 (Abstract, methods) should read “… are used TO calculate premature...” **Response:** Change made.
3. Provide a figure of the CMAQ multiscale grid applied (described in lines 126-129, Methods paragraph 3) and show the different regions adjusted for SOA yields (described on lines 152 – 162, Methods paragraph 6). Response: For this simulation, we used a 12km grid over the contiguous U.S. The map below provides the grid with axes denoting the number of 12km grid cells in the west-to-east and south-to-north directions. However, we do not believe such a figure is necessary to include in the paper itself as this information is available from the CMAQ user’s manual.

The regions adjusted for SOA yields are simply the US standard accepted geographic regions based on US census designations (http://www.eia.gov/emeu/mecs/mecs2002/census.html) and a figure should not be necessary for this.

4. X-axis of Figure 3 is very hard to read. Looks like you can overlay the 2 parts and/or increase all the font sizes. Response: This figure is generated from an online program from the National Toxicology Program and the user has limited flexibility in formatting. However, an attempt has been made to improve the readability of the figure.

5. Clarify lines 310-316 (Discussion paragraph 1) Looks like both points are due to decreases in other PM contributing sources, but the cause of line 313 regarding CSAPR isn’t clear. Response: If SO\textsubscript{2} emissions drop by 50%, sulfate will become a smaller fraction of PM\textsubscript{2.5}, and hence other sources will become more important, particularly since SOA precursors are not expected to decrease. See text at p. 11.
Discretionary Revisions

1. Why was measured 2,3-dihydroxy-4… used to derive SOA (line 148 to 150, Methods paragraph 6) rather than another compound? Is it a very strong indicator or the best from available data? Response: This is the standard compound used as a tracer for toluene.

2. Line 259-262 (Results paragraph 3) – I think it should be 0.20 and higher (not 0.22). Response: Change made.

3. Why are Texas and California discussed specifically (lines 291-292, Results paragraph 5)? Response: These two states have the highest population across all states; therefore, given the public health focus of this paper, it seemed relevant to discuss them particularly. (http://en.wikipedia.org/wiki/List_of_U.S._states_and_territories_by_population)

4. The grid resolution may be somewhat coarse for this analysis. Some discussion of resolution impacts on results might be worthwhile. Response: text revised at pages 11-12.

5. Line 347 (Discussion paragraph 5) – “… with different TRANSPORTATION, FUEL, AND INFRASTRUCTURE DESIGN options, …” Response: Change made.