

Liu et al. describe photochemical oxidation experiments of gasoline vehicle exhaust with and without SO₂ gas. This manuscript is technically sound, provides detailed methodology, and addressed a highly relevant topic in the field of atmospheric chemistry. I recommend its acceptance for publication in *Atmospheric Chemistry and Physics* pending an adequate response to the comments listed below.

General comments:

- In this reviewer’s opinion, this paper could use some copy-editing for language. While the text is comprehensible at present, it is, in some cases, awkwardly worded.

Specific comments:

- Page 23620, line 8: What molecular weight was assumed for hydrocarbons? Was this a weighted-average based on the gas-phase analyses?
- Page 23620, line 17: It may be worth stating the reference from which the authors are obtaining the rate constant for the toluene + OH radical reaction, since this appears to be known a priori (or at least the assumed value).
- Page 23621, line 16021-page 23622, line 7: It may be useful to provide examples of which compounds may be participating in this chemistry (e.g., ethene, 1,2-butadiene, 2-methyl-propene, cyclohexene). Given that the authors have input these species to the MCM, this should be relatively straightforward, and it could be as simple as saying XX% straight alkenes, YY% branched alkenes, and ZZ% cycloalkenes.
 - Another comment related to the MCM work: Given that alkenes are only ~10-20% of the total VOCs, how might these reactions be biased due to neglecting the presence of alkanes, aromatics, etc.? This answer may be that there is no bias, due to the constraints placed on OH, SO₂, O₃, and NO₂, but perhaps the authors should explicitly state this. Maybe this could be validated by re-running the MCM simulations including aromatics in addition to alkenes to assess any differences in model outputs.
- Page 23623, lines 3-8: Is this how the authors are accounting for collection efficiency? Presumably, this approach biases the concentrations of OA, sulfate, ammonium, and nitrate high because LDGVs also emit black carbon (Forestieri et al., 2013; Kamboures et al., 2013; May et al., 2014). In the cited Gordon et al. (2014) reference, those authors do account for black carbon. An approach such as the Middlebrook et al. (2012) composition-dependent CE may be necessary.
- Page 23623, line 23-page 23624, line 6: Previously, the authors state that NMHCs, NO_x, and average OH are different than typical urban conditions. How similar is the injected SO₂ concentration (~150 ppb) to urban or urban-downwind conditions? Could these discrepancies resolve the large differences between chamber observations and field observations? Due to potential differences in these conditions, it may be useful to normalize the sulfate production rate (e.g., $\mu\text{g}\cdot\text{m}^{-3}\cdot\text{sulfate}\cdot\text{hr}^{-1}\cdot\text{ppt}\cdot\text{OH}^{-1}$ or similar) to explore any biases. If the differences still exist, this may enhance the argument related to the role of Creigee intermediates.
- Page 23625, lines 10-17: Given that there is routine analysis for calcium, magnesium, potassium, and sodium in motor vehicle exhaust (California Air Resources Board, 2011), how might the presence of these additional cations affect the H⁺ calculations? I do not think this will change the authors conclusions, but it may be worth noting that the reported values of H⁺ can be considered an upper bound.
- Figures 1-4: Presumably, particle concentrations are wall-loss-corrected. It may be worth explicitly stating this in the caption.
- Figure 5: I would recommend using the same y-axis scale for both figures. This can facilitate direct comparisons between the time series of sulfate and SOA production rates.

- Figure 6: Maybe this is discussed in the text, and I have missed it, but why is there a large difference between the total loss rate and the sum of sCI and OH oxidation for vehicle II? The results for this vehicle are more drastic than the two others. Could the authors use the results from He et al. (2014) to estimate surface reactions on particles for the present study?
- Figure 7b: It may be worthwhile to color the markers in this figure to be consistent with the bar colors in Figure 7a in order to distinguish between “with SO₂” and “w/o SO₂”. This should be obvious to the reader, but it will really drive the point home.
- Figure 9: If I understand this correctly, these figures are discrete points corresponding to data selected from Figure 5? I would recommend some clarification of this in the text. Furthermore, can the authors explain why the slopes are vastly different (i.e., ~4 vs. ~1 vs. ~3)? Given that the initial conditions were largely similar (i.e., Table 2), why might the results be the way that they are? Could this be a function of alkene abundance, or might some other factor play a role?
- Figure 10: Do the authors have sufficient signal in the AMS results to calculate an initial H:C and O:C? These may be uncertain because POA concentrations were low. Based on the figure, it appears that the six data points have a slope of -1 and a y-intercept of roughly 1.8, so I am curious for this reason.

References

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