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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Ultrafine Particulate Matter Source Contributions across the Continental United States" *by* Melissa A. Venecek et al.

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Predicted Ultrafine Particulate Matter Source Contribution across the Continental United States during Summer Time Air Pollution Events Venecek et al. 2018

ACP Anonymous Review – Author Response

Reviewer #1

1.) First, they choose 39 cities? What was the basis for the choice of cities? They appear to be most of the large cities, but it would still be good to know why those were chosen and others were not (fundamentally, did they choose the largest 39 cities, and

if not, why were others excluded? Having a research rational is important.).

Response: The cities selected for analysis are the largest urban regions across the United States that experienced 1-hr ozone concentrations above the level of 70 ppb in 2010. Many of these same cities have been analyzed in previous studies about urban air pollution throughout the continental US (Carter 1994, Carter 2007, Venecek et al 2018a and Venecek et al 2018b). These cities also form the basis for the ozone formation potential scales for VOCs developed by Carter (1994). These points have been clarified on Page 5, Line110-119 of the revised manuscript.

2.) The next very, very important issue is that they chose very limited time chunks in each of the cities, some overlapping, but a very odd collection. What motivated such a choice? If it was model-performance driven, than any statement about good model performance is not so relevant, or at least should be taken very carefully and explained, as the model performance metrics have not been designed for allowing the ability to choose specific locations and times when the model is performing adequately. (This would be akin to letting epidemiologists to go back and choose periods that specifically do or do not find associations. That would be viewed as bad practice and not acceptable.) It is my understanding, and I have checked this with colleagues, that the typical modeling with a state-of-the-science model like CAMx (which is also a regional chemical transport model, similar to UCD), it is applied over the domain over a chosen period, and then you look at all of the results for a model evaluation. (When applied in SIPS, both the location and time period are predetermined.) Thus, what is appears is done here, but not stated, is that it was applied over the continental US (or some similar domain) for some period(s), again not explicitly stated. The manuscript should state what was the actual modeling period used, or did they choose the specific simulation dates a priori for each city and just simulate those? This must be stated if it is the case. If that is indeed, the case, was the model started on the beginning date chosen, or was there a period allowed for the initial conditions to be minimized?

Response: The simulation dates were selected to capture the peak photochemical

air pollution episode at each location in the year 2010 identified by the measured peak ozone concentrations during that year. Each domain was simulated for one week with 3 days spin up and 4 days analysis such that the peak photochemical episode occurred on the last day of simulation. All simulation dates are stated in Table 1. Simulation dates were selected in regional clusters to focus on photochemical episodes driven by regional stagnation leading to the concentration of emissions from routine sources rather than extreme events driven by factors such as wildfires. The simulation dates therefore overlap for many cities within the same region. A figure has been added to the manuscript to illustrate how the dates overlap (figure 1 page 6).

We appreciate the reviewers concern that the episodes should be selected without regard to model performance criteria since this would indeed bias the findings. As described above, the simulation periods were selected using other independent criteria. These points have been clarified on page 5, lines 123-133 of the revised manuscript.

3.) Also, might the authors better justify choosing episodes beginning in March or October. The former seems a bit early, the latter a bit late. Were those the peak episodes that year?

Response: These were the peak ozone episodes that aligned in the south east/south United States. A recent ozone maximum incremental reactivity scale paper (Venecek et al 2018a) also utilized these dates and the average 1-hr max O3 can be found in the EPA AQS Data Mart.

4.) Are we to take a 3-4 day period of the year to represent the source impacts for the whole year? I would expect a rather different set of sources in the winter than the summer. This whole area is not explored or discussed.

Response: The simulated periods capture the maximum photochemical (peak) episodes across the entire US. The results therefore provide source apportionment of ultrafine particles during the peak photochemical period. The title of the manuscript has been changed to more clearly emphasize the focus of the paper (line 1).

Future studies will expand on the analysis to calculate source contributions for an entire year but this analysis is beyond the scope of the current paper.

5.) Why choose 2010 if UFP measurements are available other years? They might look at the recent Pandis paper (https://www.atmos-chem-phys.net/18/13639/2018/) doing a similar exercise over Europe. The choice of just peak ozone events may very well bias the assessment of source impacts on PM0.1 and PM2.5. Why not choose peak PM2.5 events?

Response: There are no consistent measurements of PM0.1 during any year at the majority of the locations simulated in the current study and so the choice of 2010 as a base year seems reasonable in order to leverage the large amount of background work that went into setting up the model episodes and verifying the model results in a related study (Venecek et al. 2018a).

We agree that simulating a full year with combustion for winter heating will lead to different source contributions for PM0.1. An expanded future study will consider a broader range of dates, but this analysis is beyond the scope of this initial study.

6.) The model performance part is also rather opaque and requires references. First the model performance should be brought in to the main manuscript, e.g., as done in most modeling papers, showing overall performance across the entire domain and modeling period (not just selected locations and periods). See Simon et al., (2014) for the metrics typically provided and a more complete discussion of model performance evaluation

Response: All model performance statistics have been brought into the main manuscript and a full comparison has been carried out between all predicted and measured gas and particle phase species (page 11, lines 233-242). Figure 3 and 4 have been added to show all FB and FE for all available monitors with lat/lon location available in the supporting information. Table 2 illustrates the percent of monitors within the entire modeling domain that meet US EPA criteria for 5 specific pollutants measured

throughout the EPA AQS datamart: CO, SO2, NO2, Ozone and PM2.5. As a quick summary of the new analysis, over 95% of the predictions compared to measurements across the entire US domain meet the EPA criteria.

7.) The working of the paragraph beginning on line 220 is also rather strange, it says "": : :MFB values lower than 0.15 and MFE values lower than 0.35 are considered the goal or "excellent" in model performance." Then they go on to say they do not meet them. First, I don't think I have seen EPA have "excellent" as a performance description associated with those levels. I was looking for a citation here (a citation to the specific EPA evaluation metrics is required, as well as adherence to the terminologies used. My understanding is that the current EPA guidance is found at https://www3.epa.gov/ttn/scram/guidance/guide/Draft_O3- PM-RH_Modeling_Guidance-2014.pdf, and looking through that document, I don't see them use the term excellent in terms of performance associated with any metric. (Indeed, a search of that document for the term "excellent" found only two occurrences, one in terms of protocol, another in terms of conceptual model.)

Response: The language on page 12, line 238 in the main manuscript describing model performance has been revised based on EPA guidance.

8.) That said, if the model does not meet the specific guidance levels, what does it meet? If the guidance was developed for regional scale modeling, without allowing selection by location and time, what does that imply here?). Second, if you do not meet them, what does that mean? Unlike the authors, I took the evaluation as not "building confidence in the accuracy of the model results: : :", but left me questioning it. I would very much recommend that the authors follow the EPA guidance (or other recent articles, e.g., those by the AQMEII initiative or Ramboll: Emery et al.) in terms of how to conduct, and report, model performance. Having now looked at the Ramboll study (JAWMA,doi/full/10.1080/10962247.2016.1265027), their ozone and PM performance do notmeet the "goal", nor fully meet the recommended levels for "criteria. One could also follow the approaches recommended by the AQMEII initiative.

Response: We apologize that the original version of the manuscript did not better emphasize the excellent model performance. Over 95% of the model predictions across the continental U.S. meet EPA criteria building confidence in the models predictions. The Reviewer's concerns about proper model evaluation are appreciated and we have strengthened the description of this aspect of the manuscript to give the readers a more complete view of model performance across all pollutants using all metrics recommended by Simon et al.

9.) Thinking more holistically, the proper approach here would be to apply the model to a whole year, or, if that is computationally infeasible (which should be stated), be applied to one month periods in each season, and the results from each of those months be given. If they only want to consider the peak photochemically-active periods, they should choose a three month period (or more, preferably) that will capture events in all the cities for that year.

Response: The reviewer is requesting a different study than the one that we performed. The focus of this current manuscript is the study of PM0.1 during the peak summer photochemical period across the United States in 2010. We have updated the title to reflect this focus "Predicted Ultrafine Particulate Matter Source Contribution across the Continental United States during Peak Summer Air Pollution Events". The following major conclusions of the paper will not change when an entire summer time period is simulated: (i) the majority of the PM0.1 is dominated by primary emissions; (ii) natural gas combustion is a major source of PM0.1 even though it makes minor contributions to PM2.5; (iii) there is significant variability in PM0.1 concentrations and source contributions between cities reflecting the different emissions in each city.

We agree that studies capturing seasonal averages and annual averages will be the next step now that this initial study on peak photochemical events has been completed. These studies will be the topics of future papers.

10.) I was a bit puzzled by the explanation given for Fig. 1. A 4 km grid is pretty fine,

and the mobile sources in a typical urban area like LA are pretty ubiquitous. There are a number of monitors in LA: do any of them not go to zero at night? If not, that might suggest a different issue The mismatch in the evening needs a bit more discussion and justification.

Response: The original manuscript did not incorporate the data from all 19 O3 monitors in the region around Los Angeles. The measurement data in Figure 1 (now Figure S1 in the revised manuscript) has been updated to reflect all available stations.

An error in the model wind fields was corrected in the revised version of the manuscript. This error had caused the winds in each row to advance by one column, effectively moving the winds over the Pacific Ocean over land for coastal California cities such as Los Angeles. The same error was corrected in all domains, but the effects were less severe at inland locations where winds were more uniform. All of the model results throughout the revised paper now reflect correct wind fields (all simulations were rerun).

The net result of the changes summarized above produce measured and predicted ozone concentrations that decrease to $\sim$zero during the evening hours. We thank the reviewer for pointing out the strange behavior in the original manuscript.

11.) They commit to providing the outdoor exposure fields. They should also provide the model and its inputs. I assume this is journal policy, but the authors should likewise commit

Response: The model itself and input data are available to collaborators through direct email request to the corresponding author. A statement to this effect has been added in the data availability section (page 27 line 466-468).

12.) A main conclusion of the paper is that natural gas is the main contributor to population-weighted exposure. This is a rather unique result and certainly requires more justification and discussion in light of the work that has shown via careful experiments that mobile sources and air craft are major sources of natural gas. Where is the

empirical evidence of natural gas being a main contributor and can they show that they have captured the contribution of those other two sources? Their explanation of why Lane et al., or Posner and Pandis is not sufficient to argue that the current results are reliable. How well does the current study capture the spatial dynamics found by the groups from USC (Sioutas), Harvard (Spengler) and UW (e.g., Atmospheric Environment Volume 139, August 2016, Pages 20-29) which tend to point the finger at mobile sources and aircraft, so much so, the latter claim that ultrafine particle counts can be used as a tracer for aircraft turbine emissions. Given the lack of empirical evidence, compounded with their not having done any evaluation of the ultrafine results against observations nationally, the speculative nature of this section suggests it should be removed, or couched in very different terms (i.e., noting the limitations, with a statement of the speculative nature). The question should be asked if there is sufficient evidence to support controls on a source based on the current analysis

Response: A recent study by Yu et al (2018) utilized the UCD-CIT CTM and compared predicted PM0.1 source contribution to PM0.1 CMB results using molecular markers (Xue et al 2018a) at multiple sites across California. The predictions from the UCD/CIT model were in good agreement with the CMB results for PM0.1 OC from gasoline (mobile), diesel (mobile), wood burning, meat cooking and “other sources”. This comparison builds confidence in the accuracy of the regional UFP source predictions from the UCD/CIT model and the ability to properly represent contributions from mobile sources to regional PM0.1 concentrations.

The PM0.1 “other” category in the molecular marker calculation summarized by Xue et al. (2018) is composed of unresolved sources, but the UCD-CIT model at the core of the current manuscript can identify these sources. Major sources of the unresolved material identified by the UCD-CIT model include non-residential natural gas, aircraft and other source that were not tagged. The UCD-CIT model found that natural gas combustion is a significant source of PM0.1 OC in San Pablo, East Oakland, central Los Angeles, and Fresno where the predictions for contributions from mobile sources

were in good agreement with the CMB results.

Direct measurements of particle volatility in natural gas combustion exhaust made by Xue et al. (2018b) suggest that 70% of the natural gas combustion exhaust particles from residential sources (stoves and water heaters) evaporate when they are diluted in the atmosphere. Direct measurements indicated that particles emitted from engines operating on natural gas did not evaporate even at extremely high dilution ratios. The original version of the current manuscript specified that 70% of the particles from residential natural gas combustion sources would evaporate when diluted in the atmosphere, but it was assumed that particles emitted from commercial and industrial sources would not evaporate. Further review of typical commercial natural gas sources for space heating, water heating, etc suggested that these sources may be similar to residential natural gas combustion sources. Therefore, the model simulations in the revised manuscript were rerun while treating both residential and commercial natural gas combustion particles as semi-volatile (70% evaporation). The predicted contribution to PM0.1 from natural gas combustion particles decreases from 54% (original manuscript) to 33% (revised manuscript). Natural gas combustion particles are still important, but slightly less dominant in this revised treatment.

In summary, the current study uses all available empirical evidence to test and verify the predictions of natural gas combustion contributions to PM0.1 concentrations. The comprehensive comparisons to CMB studies in California show that the model calculations properly account for mobile source and food cooking contributions to PM0.1. The results across the rest of the US vary from location to location but are in general agreement with the relative important of mobile sources vs. other sources. We look forward to future datasets that perform PM0.1 CMB studies across the entire US, but do not believe that the findings from the current study should be delayed until those additional measurements have been completed.

13.) The performance evaluation example (e.g., in reference to the EPA goal/excellence criteria) is not the only place where a citation is needed. They state

that a number of other locations have PM0.1 levels above 2 ug/m3 (line 418). Then they go on to say there were sharper gradients in the observations. This would seem to contradict their findings/interpretation. More discussion needed

Response: The locations with PM0.1 greater than 2 μg m-3 were identified in the current study based on predictions from the UCD-CIT model. Likewise, the conclusion that sharper gradients were predicted PM0.1 vs. PM2.5 concentrations is based on UCD-CIT model predictions from the current study, not observations. These concluding statements summarize the findings of the model predictions, they do not seek to compare the model predictions to previous studies (previous sections of the paper are devoted to model performance evaluation). These statements do not contradict the findings or interpretation of the paper. We have clarified the sentence by adding the phrase “In the current study, predicted . . .” on line 447 of the revised manuscript.

14.) Again, they should compare their findings more directly to observations. In the summary, they state their analysis was for “peak photochemical periods during the year 2010.” That is a rather strange way to characterize periods during which, in Atlanta, the 8-hr maximum ozone shown reached only about 50 ppb; I visually averaged, the actual value would be useful) in Cincinnati, about 60, in Los Angeles about 90, in New York about 85 ppb. I am not sure about the other locations, but I think the design value for LA in 2010 was about 120. In Cincinnati, it was about 0.079 (https://www3.epa.gov/airquality/greenbook/hbtcw.html), In Atlanta, about 80 ppb. The chosen periods would not appear to be “peak photochemical smog” periods

Response: The locations and dates correspond to periods when measured 1-hr ozone exceeded 70 ppb across the major geographical regions (south, south east, east, west etc.) in 2010. Figure 1 has been added to the manuscript to illustrate the ozone concentrations on the selected days. All monitor information (site lat/lon) can be found in the supporting information and obtained from the EPA AQS Datamart.

Note that the design value in 2010 is based on ozone measurements from 2008, 2009,

and 2010, with measured values from earlier years typically dominating the statistic during this time period. We believe that the episodes analyzed in the current study represent the peak air pollution events in the major US cities that are driven by routine emissions combined with stagnant meteorology. The revised manuscript thoroughly compares all available measurements of air pollution during the air pollution events.

15.) How different would Fig. 8 be if you simply used the local emissions? It is not apparent what this analysis adds and how it might be useful beyond simply using the inventoried sources

Response: The model application incorporates all major processes (emissions, transport, deposition, chemical reaction), which removes uncertainty of just using a local emission analysis. Given that this information is available, the authors are confused by the reviewers request to use an inferior analysis based only on the emissions inventory. Also note that Figure 8 has been moved to SI in response to another reviewer comment.

16.) Line 425 uses the word "consistent" after saying that the model could not resolve the observed mobile source peaks. While one can see what they may be trying to say, it should be said differently, and more precisely. One could easily have said, experiments have found peak ultrafine particle levels were tied to on-road mobile sources and aircraft emissions, though those three sources combined account for only 22%, and are thus inconsistent with the results here that identify natural gas combustion

Response: Changed text on page 27 line 454-458 to "On-road gasoline and diesel vehicles contributed on average 14% to regional PM0.1 even though peak contributions within 0.3 km of the roadway were not resolved by the 4 km grid cells. This is consistent with other studies that have found an exponential decrease in ultrafine particle concentrations outside of major roadways (Wang et al. 2011) due to the sharp gradient of PM0.1."

Minor:

1.) First line: should be "concentrations" Response: This has been updated in the text – page 2 line 12

2.) L 156: Missing ")" Response: Corrected.

3.) 4 km stated in both of the first lines of the Abstract.

Response: This has been updated in the text – page 2 line 15

4.) Fig. 2. Are these the individual, daily values for each site? If so, why are there not more circles? A bit more information in the figure caption would be useful.

Response: Speciated PM2.5 measurements are 24hr averages taken every 3 days or every 6 days depending on the city. Spin up days were not included in the comparison. All available comparison days were included in the analysis. The authors believe there are a sufficient number of data points (N>50) to properly evaluate the model performance.

5.) Figure 3. What do they mean by "air pollution event"? Is the event simply when they compared their results to the observations? It would be good to know when, and where, the maximums occurred.

Response: See figure 1 and response to previous comments describing criteria for selecting the regional maximum photochemical periods at each city in 2010.

âĂČ Reviewer #2

1. Page 2, lines 22-23. I would suggest removing "As expected." This minimizes the work, as if it were expected, why bother? Response: "As expected" has been removed on page 2 line 21-22 and page 13 line 319

2. In general, the paper could deal with some cleaning up of language, punctuation, etc. Examples Page 2, lines 31-32, use : : : for commercial use Page 3, line 57, should this be low birth weight Page 3, line 64, word national is not necessary, as monitors in the continental US are specified Page 4, lines 80 and 100 (and elsewhere) US or

U.S.? On page 22, line 410, United States is written out. In the SI, page 14, line 118, states is not capitalized. Page 4, line 85, add 'to' between exposure and ultrafine Page 7, line 156, missing a closing parenthesis Page 9, line 194 (and elsewhere including Fig 1 and Fig S2 caption), vs. not vs Page 17, lines 337-340 – Chang et al. (2004) measured: : : Add (2007) to Lane et al. In the SI, Figure S3 should appear after Table S2, as it is cited after Table S2 in the main text. SI, page 14, line 111, Figures compare, not compares

Response: The issues noted by the Reviewer above have been corrected and marked with yellow highlight in the main manuscript

3. In the abstract (Page 2, lines 35-37) and on page 20, lines 364-366), 'higher' and 'lower' ratios should be quantified. Is there a cutoff to determine higher versus lower based on the scatter plot shown in Figure 7?

Response: "Higher" ratio PM0.1/PM2.5 is anything higher than 0.10 and lower is anything lower than 0.05. Text has been added to the main manuscript on page 2 line 35

4. Page 3, line 60. I assume this should be surface area to volume ratio, not just surface area?

Response: This has been updated in the main manuscript page 3 line 61

5. Page 5, Table 1. Please provide more information about why these 39 cities were selected. Was it the availability of observations? Was it the number of O3 days above 70 ppb? As an example, why Charlotte and not Raleigh, NC? Or why Tulsa and not Oklahoma City, OK? Why were Pittsburgh and Chicago not included?

Response: The cities were selected based on the largest population centers across the US that experienced peak 1-hr ozone concentrations greater than 70 ppb in the year 2010. The locations generally correspond to previous studies that also looked at urban regions throughout the continental US (Carter 1994, Carter 2007, Venecek

2018a, Venecek 2018b). Some large population centers did not exceed the 70 ppb threshold and therefore were not included in the analysis. In general, the cities selected for analysis capture a cross section of urban populations across the US reflecting the diversity of emission sources.

6. Page 7, line 141. Please justify why nucleation is not considered. This is in line with a later comment about fraction of PM0.1 that is secondary versus primary.

Response: All of the simulations in the current study were rerun using nucleation based on the ternary nucleation (TN) mechanisms involving H2SO4-H2O-ammonia (NH3) (Napari et al, 2002). This mechanism has been applied in California with good agreement found between predicted and measured PM0.1 and N7 (Yu et al 2018). PM0.1 mass and source contributions in the current study did not change with the addition of nucleation, further confirming the conclusion that PM0.1 is driven by primary source contributions rather than nucleation.

8. Page 8, line 190. Even though it appears that secondary material is not a huge contribution to PM0.1, it would be appropriate to say 'also emit ultrafine particles and their precursors'

Response: Text has been added to the main manuscript page 11 line 226

11. Page 11, line 262. I recognize that the focus of this work is summer. However, would it be appropriate to highlight that the biomass contribution might be different in winter when wood burning for home heating could be a prevalent source of PM0.1 in colder regions?

Response: A sentence has been added at line 292-293 stating that wood combustion will make larger PM0.1 contributions during winter.

7. Section 2.3 and page 8, line 188. Please provide more information about monitors used and the comparison to model output. It says 'measurements averaged' – does this mean multiple monitors were used? Or was a single monitor compared to the model

output for the grid cell in which it resides? For cities with multiple monitors with a grid cell/domain, if multiple are used, it would be appropriate to include that information (perhaps in the SI).

Response: The model performance statistics for the re-generated simulations including nucleation have been updated. All monitors within a CBSA were compared to predictions. Figure 3 and 4 illustrate the MFB and MFE for all comparisons for all available gas and particle phase species (NO2, SO2, CO, O3 and PM2.5). All monitor information across the entire modeling domain has been added to the Supporting Information

9. Page 9. In reviewing Table S2, it appears that only one city does not meet the MFE for O3? If that is the case, it should be more specific on lines 199-200. It would be appropriate to provide the average O3 model performance statistics at this point. Then at the bottom of the page, the authors could discuss PM model performance statistics (and again, specify that only one city does not meet the MFE for PM model performance). Right now, it is slightly confusing to discuss O3, then PM, then both in terms of the averages.

Response: A more centralized presentation of all gas/particle phase model performance statistics have been added in section 3 (results). See response to Reviewer 1 comments 6 and 8.

10. Page 10, line 231. While I recognize that the submitted Yu et al. manuscript describes the 'good agreement' for PM0.1 modeling assessment in California, I think it could be summarized more quantitatively here in only one or two sentences.

Response: Yu et al PM0.1 source contribution for gasoline, diesel engines, food cooking, wood burning, and "other sources FE and FB were within EPA criteria of +/- 0.5 and 0.75, respectively added to page 12 line 259-263.

12. Page 13, Figure 2. How does the model convert from OM to OC? Does the two product model used (Carlton et al.) predict OM or OC? I thought it was OM, but if I

am mistaken please correct me. If a conversion is done to estimate OC based on the simulated OM, it would be appropriate to include this in the caption to Figure 2.

Response: The primary carbon variable tracked in model calculations is organic matter (OM), and the SOA model also predicts OM directly. These values must be converted to organic carbon (OC) for comparisons to measured values. Primary organic matter was converted to OC by dividing by a factor of 1.1. SOA components were converted to OC by dividing by a factor of 1.5. These points have been clarified on line 245 of the revised manuscript.

13. Page 14, Figure 3; Page 15, Figure 4. Would it be possible to somehow show on these figures the relative contribution of primary PM0.1 versus secondary PM0.1? This would truly drive home (and quantify) the relative contributions of direct emission versus in situ formation (I realize it is predominantly primary, but doing this would show it).

Response: Unfortunately, it is not possible to show the relative contributions on Figures 3 and 4. The primary vs. secondary fraction of PM2.5 and PM0.1 at each city location has been summarized in tables S7-S16 of the supporting information. A sentence summarizing this information has been added on line 313-315 of the revised manuscript.

14. Page 18, line 343. This paragraph does not seem necessary to me, as it focuses on previous work that simulated PNC, which as the authors point out in the nucleation discussion (see comment above), is not equivalent to PM0.1 (the focus of this work).

Response: Even though the focus of the current work is PM0.1, many researchers still use particle number concentration to describe UFPs. The discussion of how previous studies handled natural gas combustion emissions also explains why this source was not identified in previous studies. We therefore respectfully request that the paragraph be retained, but will defer to the Editor's judgement if the length of the paper is too long.

15. Page 22, Figure 8. A suggestion for improved readability: break up this figure into four panels by geographic region of the nation (since the focus is determining how cities in the same region compare – as discussed as 'regional clusters' on page 21).

Response: Note that the Figure has been moved to SI in response to comments by Reviewer 3. The authors feel that keeping the figure as one panel shows that PM0.1 source contribution across cities (even regional ones) do not correlate highly with one another and therefore emission control strategies should be tailored to each specific city. The text on page 66 of SI has been revised to describe the Figure.

16. SI, Page 13, line 95. The MFE given in the caption (0.67) is for O3? The MFE for PM given in the text is 0.75. Please specify both in the caption here. Also, note that the bold lines reflect cities that do not meet one of those criteria.

Response: As requested, updates have been made to the SI regarding all model performance statistics (Tables S1-S6 and Figure S1)

17. SI, Tables S3 and S4. These do not appear to be called out anywhere (if they were, and I missed it, I apologize). I assume this is the data that were used to create the vectors for the dot products? If so, that discussion is an appropriate place for them to be called out.

Response: The vector analysis has been moved to the SI and text has been added reflecting the use of these tables in the vector analysis. Page 65 line 20-21 âĂČ Reviewer #3

1.) It is not entirely clear to me why the authors selected air pollution episodes lasting a few days as the sole basis for estimating PM0.1 mass concentrations in different cities? Would a few days be much too short time period to get reliable information on different sources, and would selection of photochemical pollution episodes bias the importance of some sources over the others?

Response: The simulation dates were selected to capture the peak photochemical

air pollution episode at each location in the year 2010 identified by the measured peak ozone concentrations during that year. Each domain was simulated for one week with 3 days spin up and 4 days analysis such that the peak photochemical episode occurred on the last day of simulation. All simulation dates are stated in Table 1. Simulation dates were selected in regional clusters to focus on photochemical episodes driven by regional stagnation leading to the concentration of emissions from routine sources rather than extreme events driven by factors such as wildfires. The simulation dates therefore overlap for many cities within the same region. A figure has been added to the manuscript to illustrate how the dates overlap (figure 1 page 6).

We agree that the current paper represents PM0.1 concentrations during a peak summer photochemical episode. Future studies will consider seasonal averages and annual averages, but this analysis is beyond the scope of the current manuscript.

2.) Since PM0.1 mass is the combined result of primary particle emissions (and nucleation) into this size range, and subsequent accumulatoin of secondary meterial by these particls, the authors should explain in more detail how they determined PM0.1 mass concentration (and the related source contribution) from their model simulations and what are the related uncertainties. There are several issues related to this. First, how many size bins the used model has in the sub-0.1 um size range and how close to the 0.1 is the border between the two nearest size bins? Now the authors only mention the number of size bin over the whole particle size range from 10 nm to 10 um (page 7). Second, what is the actual particle diameter used in model simulations? Mass measurements rely usually on aerodynamics diameters (impactors), while number size distribution measurements in the ultrafine size range rely usually on electric mobility diameters. These two diameters may differ substantially (up to a factor 2) for ambient aerosol particles, and the diameter used in a model can be either one of these two or something else. This is an important issue because PM mass size distributions often have a steep gradient at around 0.1 um, making the PM0.1 mass concentration very sensitive to the diameter chosen to represent the size 0.1 um. Third, the authors

state that they do not care about nucleation because it only affects the particle number concentration but not the PM mass concentration. This is not true. Think, for example, a situation where 2 sources dominate the ultrafine particle number concentration: nucleation and a combustion source that produces particles with a peak diameter slightly below 100 nm. When these particle age in the atmosphere for a while and accumulate secondary material from the gas phase, those originating from nucleation tend to remain in the sub-100 nm size range while a big part of combustion particle may grow past 100 nm. As a result, whether or not to include nucleation also affects PM0.1 um mass. This issue should, at the very least, mentioned in the manuscript.

Response: Nucleation using the ternary nucleation (TN) mechanisms involving H2SO4-H2O-ammonia (NH3) (Napari et al, 2002) has been added to the model configuration and the model simulations have been rerun. This mechanism has been applied in California with good agreement found between predicted and measured PM0.1 and N7 (Yu et al 2018). Nucleation did not significantly contribute to PM0.1 mass in the current study, and so the relative contributions from primary sources were unchanged due to the addition of nucleation.

Five (5) size bins equally spaced on a log diameter scale are used between 10 nm and 100 nm. The initial central diameters of each bin are: 12.6nm, 20nm, 32nm, 50nm, 79nm. Particle size bins "float" using the moving sectional approach. Condensation of secondary material causes particle growth while fresh emissions move the bin-averaged properties back towards the original emissions diameter. The model output therefore represents the competition between fresh emissions and atmospheric aging.

Number is tracked as an explicit variable for each moving size bin in the presence of all the major atmospheric processes (emissions, transport, deposition, gas-particle conversion, coagulation). The moving sectional approach naturally conserves particle number concentration since material is not transferred from one bin to another except through the relatively slow process of coagulation that mostly occurs between very

small particles and very large particles. The number concentration of the smaller bin involved in coagulation is reduced and the mass is transferred to the larger size bin. Number concentration is not the focus of the current manuscript, but additional details are provided by Yu et al. (2018).

3.) It is unclear to me how authors keep track on the different sources contributing to the PM0.1 mass concentration. I understand that keeping track particle numbers from different sources is possible, but how this is done for PM mass as a big fraction of it is formed secondarily in the atmosphere?

Response: Yu et al (2018), Ying et al., 2008b and Hu et al., 2017 provide a detailed description of how the model explicitly tracks mass in each particle size bin. A statement has been added to the main text of the current manuscript referencing those descriptions. In summary, the model explicitly tracks primary mass from different primary sources with an artificial tracer species. Tracer emissions are empirically set to be 1% of the total primary particle mass emitted from each source category. Tracers are carried through all major processes including transport, coagulation and deposition. The final tracer concentrations are directly proportional to the primary particle mass from the associated group.

Source contributions to PM0.1 SOA are tracked by tagging the emissions that feed into the chemical reaction mechanism. Reaction products inherit the tags from the parent compounds. Final semi-volatile reaction products that condense to the PM carry these same source tags allowing them to be quantified.

To be clear, 87% of the PM0.1 mass identified in the current study is primary, and so the tracer approach for primary emissions carries most of the source apportionment information.

4.) The authors use ozone and OC/BC concentrations in PM2.5 to evaluate their model. This is fine. However, it is clear that this sort of model evaluation does not guarantee that the model works well for PM0.1. While I do understand there are too few PM0.1

measurements around for a proper model evaluation in this respect, I still think that the authors should be more honest to state this explicitly in the manuscript (a good performance for ozone does, by no means, guarantee that also PM0.1 is simulated well).

Response: A recent study by Yu et al (2018) utilized the UCD-CIT CTM and compared predicted PM0.1 source contribution to CMB results using molecular markers (Xue et al 2018) in California. Source contributions to PM0.1 OC for gasoline (mobile), diesel (mobile), wood burning, meat cooking and "other" calculated using the UCD-CIT model and the molecular marker technique were in good agreement, which builds confidence in the accuracy of the UFP source predictions.

Good performance in California does not guarantee good performance across the entire US. The lack of data needed for model evaluation outside of California has been noted on Line 265 of the revised manuscript.

5.) I do not understand the last particle of the discussion before the conclusions, including equations 2 and 3 and figure 8. The obtained results misses a proper methodological description, and the actual results does not seem very helpful in the context of this paper. I would recommend removing this part of the analysis altogether from this paper.

Response: The current study identified that sources of PM2.5 and PM0.1 vary across major urban regions. The normalized dot product calculation allowed us to evaluate each city source contribution as a vector and quantitatively compare it to another city. The analysis found that few regional clusters were observed for PM0.1 source vectors, suggesting that emissions control programs may need to be tailored to each region.

The text describing Figure 8 has been clarified and the Figure has been moved to SI in the revised manuscript based on the Reviewer comment.

Minor and technical issues 1.) Section 2.3. Did the authors use real-meteorological

data when calculating biogenic emissions using the MEGAN model? This is important because biogenic emissions are very sensitive to ambient temperatures. Please explain in the text.

Response: MEGANv2.1 was configured with the same meteorology implemented into the UCD-CIT CTM. These met fields were determined using WRFv3.6. A statement has been added to the text to note this configurations (line 202)

2.) What is the "Actual Max" for in the caption of figure 3? The given numbers are extremely accurate (3-5 digits)

Response: Actual max is the predicted maximum PM2.5 and PM0.1 value (μg/m3).

Interactive comment on Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2018-833, 2018.