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Interactive comment on "Sources of Airborne Ultrafine Particle Number and Mass Concentrations in California" *by* Xin Yu et al.

Anonymous Referee #3

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The present paper attempts to quantify the sources of ultrafine particle number and mass in California using a chemical transport model. Determination of the sources of particle number is a challenging problem that has received a lot of attention for global scales, but relatively little for urban and regional scales. The work arrives at a rather controversial conclusion that contradicts most existing studies (including Hu et al. (2017) by the same team): natural gas combustion is the largest source of particle number and a significant source of ultrafine mass at most locations and periods at least in California. The paper is rather uneven: it does a good job dealing with the sources of ultrafine mass, but at the same time its treatment of the sources of particle number is problematic and potentially wrong. Its major weaknesses do not allow me to recommend its publication in its current state. However, the paper does offer important

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new insights and should be publishable if its weak parts are corrected or even deleted. The various issues that need to be addressed are described below.

(1) Major problems

(1.1) Sources of particle number. The authors calculate the particle number contributed by each source based on the corresponding mass (equation 1 in the paper). This is wrong for two reasons. First a significant fraction of the particle mass is secondary (sulfates, nitrates, secondary organic aerosol). When the secondary mass increases, the contribution of the corresponding source to particle number does not. Second, coagulation involves particles from different sources. It is not clear to which source the authors assign the particle resulting from the coagulation of two particles from different sources. Both of these problems are quite important for ultrafine particle number concentrations. The errors of this oversimplified approach should be estimated (at least for one period) with careful zero-out analysis (e.g., removing only the ultrafines and not the larger particles to avoid changes in the condensation and coagulation sinks). If the error is significant the corresponding part of the work should be redone or should be replaced with just a description of the contributions to emissions for different size ranges.

(1.2) Importance of non-residential natural gas combustion as a source of ultrafine particles. This is clearly the most important, but also the most controversial finding of the study. The evidence provided to support this potentially very important result is rather weak and the authors miss a lot of opportunities to strengthen their argument.

The first is the use of size distributions. The predicted size distributions from this source apparently peak in the 10-20 nm size range. There are a lot of available size distribution measurements in the area that can be directly compared with the model predictions. My understanding howeveris that the measured number size distributions (not immediately next to freeways) peak at the 35-40 nm range (see for example Sowlat et al., 2016). Some of these size distribution measurements are available for the periods that have been simulated so a comparison of size distributions (including sources) could be performed without much effort.

The second is the use of the spatial distribution of particle number. The predicted concentration maps are not shown, but one would expect much higher concentrations near the corresponding major source areas. Traffic should have quite a different spatial pattern. There have been also a lot of particle number distribution measurements in California during the last decade. An effort to test if the predicted patterns match the observed ones would help.

The third is the average diurnal variation. However, this study assumes that the non-residential natural gas emissions have a similar temporal pattern as traffic (Figure S2). So the observed rush-hour peak in particle number that all previous studies assign to traffic, here is explained by natural gas combustion. However, more careful spatio-temporal analysis could help strengthen (or weaken) the conclusion. For example, the predicted morning number peak in Roubidoux in summer does not exist in the measurements. The situation is even worse in midday during the winter suggesting that emissions from this source are clearly overestimated in this area. Is this helpful? Is this area dominated by these emissions or is the sampling site an exception? On the other hand, the model performs well in other areas so one could make the opposite argument site by site. However, without using all the information about predicted patterns in space and time it is difficult to reach a conclusion.

(1.3) Modeling of growth of ultrafine particles. The approach used to simulate condensation/evaporation of sulfuric acid, ammonium, nitric acid, secondary organics on the ultrafine particles in this study is not explained in any detail. There is a rather confusing statement in lines 129-137 that “dynamic condensation/evaporation is not considered”. Does this mean that the particles are assumed to be in equilibrium? If yes, how does the model deal with the effect of surface tension on the equilibrium vapor pressure especially in the 10-20 nm range? Do these particles evaporate because their equilibrium vapor pressure is higher than that of the bigger particles? This is a crucial process for the number concentration of the smaller particles and it is not clear that it is simulated properly.

(2) Other significant issues

(2.1) Definition of particle number concentration. The use of the term particle number concentration throughout this paper is often confusing and sometimes misleading. It is important to always define the lower threshold of the size range of the corresponding concentration. The total particle concentration can be easily a factor of 2 or 3 higher than the concentration of particles with diameter higher than 10 nm (N10).

(2.2) Growth of freshly nucleated particles to 10 nm. The authors state that they parameterize the growth process following the work of Kerminen and Kulmala (2002). However, this parameterization requires the growth rate (GR) of the particles. The calculation of this rate is non-trivial in a model with coarse aerosol size resolution such as the current one. Errors in the GR can lead to significant errors in the estimation of the contribution of nucleation as a source to particle number. The authors should evaluate the error of this parameterization for their aerosol model.

(2.3) There is little information provided about the frequency and spatial extent of nucleation in the simulations in the various seasons. This information is needed to understand the simulation results.

(2.4) Emissions from natural gas combustion. A map of the estimated N10 and PM0.1 emissions from this major source is needed (see also comment 1.2). Also the average diurnal profile of the emissions for the domain and the average size distribution should be shown.

(2.5) Temporal scale of evaluation. The authors present metrics of the model performance but they do not clarify if these are for hourly, daily, monthly, simulation averages or something else. The text and the corresponding tables do not include this information. Given the availability hourly measurements evaluation at this timescale should be

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also performed (if it has not been performed yet). The evaluation at a daily scale is also useful.

(2.6) The measurements of particle number refer to N6 and N7 while the predictions to N10. The authors suggest that the average error in the corresponding comparisons should be less than 10

(2.7) The use of qualitative terms (general agreement, agree reasonably well, good agreement) is not helpful and should be avoided.

(2.8) If my understanding of the paper is correct, the current model does not use the dynamic organic aerosol scheme used by Hu et al. (2017). If this is the case, the results regarding the contribution of SOA to PM0.1 in this work should be discussed and should be compared to that version of the model. If it is the same it should be clearly stated.

(2.9) Contribution of traffic particles. Ronkko et al. (PNAS, 114, 7549-7554, 2017) argued that traffic is an even more important source of particle number, because there are a lot of sub-10 nm particles emitted. Given that the current study does not include primary traffic particles smaller than 10 nm (which of course can grow to larger sizes), can it seriously underestimate the contribution of this source in urban environments?

(3) Minor issues

(3.1) The abstract says that simulations have been performed for 2012, 2015 and 2016. However, the presented results are only for 2012. This is important because there are available size distribution measurements for 2015-16 in the modeling domain that can be used for the evaluation of the model predictions (see comment 1.2).

(3.2) The predicted correlations between PM2.5 and particle number concentrations can be compared with the corresponding measured correlations as an indirect way to evaluate the model performance.

(3.3) Lines 66-67 “when nucleation algorithms were not standardized”. This statement

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is confusing.

(3.4) Are the sulfate and nitrate concentrations shown in Table S4 for PM2.5 or for another size range?

(3.5) Table 1 should probably also include the predicted and measured average number concentrations.

(3.6) The terms “measured” and “predicted” should be used everywhere in Section 3.2.1 and other parts of the paper in which predictions are compared to measurements.

(3.7) The number of samples and their duration corresponding to the results of Figs. 2-3 should be stated in the caption.

(3.8) Line 296. Figures 4-6 and 7-9 do not show the seasonal variation of the corresponding variables. They show data (are these daily averages or something else) for different days in different seasons. These figures could be improved if they were split in four parts for the different periods simulated. The discussion could also be improved if the actual seasonal averages were shown (may be in the SI) and discussed.

(3.9) Figure S2. What is A, B, and C? What is the average pattern in the domain?

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