## Dear Reviewer,

Thank you for the comments to help improve the quality of the paper. We have revised the manuscript to address your comments and a detailed response to each comment is provided in this file.

The comments are in regular font, the responses are in red, and the changes in the manuscript are in blue.

## **Anonymous Referee #2**

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The authors provided a long-term analysis for the spatial distribution of PM0.1, and its components including POA and SOA. By using the source apportionment method, the authors further discussed the contribution of different sources on PM0.1 and its components. The article is generally well-written, and has clearly expressed the conclusions clearly by showing convincing data analysis. I will suggest the paper published on ACP, after the authors address my following suggestions:

Pg 10: define the metric used for the evaluation: MFB and MFE. Can put the equation into the Supporting.

**Responses:** Accepted. We put the definitions in the Supplemental Materials and referred to the definition in the main text.

Pg 10: "in the first paper in the series": if the authors claimed this paper as "the fourth in the series" (Pg 5 line 70), then I suggest the authors change "the first paper" to "the third : : :." to avoid confusion, or do the other way.

Responses: We changed to "Part I paper" to avoid confusion.

Pg 11, line 189-190: I assume the authors were still talking about the winter when they say "Wood smoke is predicted to be : : :."?

**Responses:** To be more accurate, we changed to "Wood smoke is predicted to be the dominant OC source in winter....".

Pg 11, line 192-193: the authors implied that the overestimation of the PM2.5 in the San Jose site was due to the overestimated emission inventory. So how did the authors make that conclusion? Was the emission inventory data significant different from the other places, or more uncertain compared with others?

Responses: The PM2.5 OC was consistently over-predicted at San Jose in all winter seasons during the 9 year period (Figure 1b). While at other sites, wintertime PM2.5 OC was not always over-predicted in all winters. Considering that wood burning is the dominant source of winter PM2.5 OC in California, and similar meteorological performance among these sites, we attributed the consistent over-prediction of PM2.5 OC at San Jose more likely to overestimated wood burning emissions at this location.

Surveys of home heating methods conducted by the Bay Area Air Quality

Management District (BAAQMD) found that wood smoke emissions inventories were over-estimated in San Jose for the years 2012 and 2013. While these years are outside of the analysis window in the current manuscript, these findings support the hypothesis that wood smoke emissions in San Jose are over-estimated in the years 2000-2009.

This has been explained on lines 200-202 of the revised manuscript.

Pg 12: line 217-219: I suggest the authors move the brief introduction of the 6 Obs sites into Pg 10 to Pg 10 in front of Fig. 1. Also can the authors comment why they didn't use the EI Cajon site to evaluate the model's performance of simulating in PM2.5 in Fig. 1?

Responses: We moved the brief introduction of the observation sties to Pg10 in front of Fig.1. We kept six sites in Fig. 1 to make the figure clearer, but in the revised manuscript, we added El Cajon in Fig. 1.

Pg 15, line 287: change "PM2.5" to "PM2.5-SOA fraction" or "that in PM2.5". Also the authors concluded that the SOA fraction in PM0.1 lower than that in PM2.5, but in Figure 4, we can see the fractions are higher in PM0.1 than PM2.5 in rural areas. Can the authors explain why?

Responses: We changed to "that in PM2.5" and we added "in urban areas" to be more accurate.

The SOA/TOA fractions in PM0.1 are generally low at all locations where primary combustion emissions are significant. This includes all major urban areas or locations with major transportation corridors. The PM0.1 SOA/TOA fraction increases in regions with very low primary combustion emissions because few natural sources emit primary OA in this size range. Natural sources including windblown dust contribute more to the PM2.5 size fraction than the PM0.1 size fraction in these remote regions, which explains the different behavior illustrated in Figure 4 and Figure S1. All concentrations are very low in these remote regions, and so the points have minor importance for health effects analysis. We feel that the extended discussion would risk confusing the reader, and so we have made these points in the Figure caption for S1 rather than in the main text.

Pg 34, in Figure 7 and others, also in the supplementary, I am confused about the meaning of colorbar. I thought it stands for the fractions from each source category in the total PM0.1 POA, but it seems not. What is the "maximum concentration value", maximum of the monthly mean or maximum of the yearly mean? Also how the authors made the conclusion that the dominant regional sources are "wood smoke, meat cooking : : :"? Looking at the map, most of the data are in the range of "0-10" %, and you can't tell which regions are in the 1% and which regions are in the 9%. For sources with a Max value of 900 but fractions around 1% may not be larger than the

source with a Max value of 120 and fractions around 9%. Please quantify the fractions from each source before making conclusion. Also consider doing this for other similar plots.

Responses: The maximum concentration value is the maximum 9-year average concentrations. We made the changes in the figure captions to be clearer.

The dominant sources were determined based on the total contributions of the sources region wide. We added the fraction values in the discussion and we did the same changes for the discussion of Figure 8.

Pg 43 & 44: Switch the order S4 and S5 to follow when they are mentioned in the paper.

**Responses:** The order of figures in the supplemental materials was changed to follow when they are mentioned in the paper.