<u>**Anonymous referee #1**</u>

This manuscript described the spatial and temporal variability of sources of ambient PM$_{2.5}$ in California. It is a nice summary about PM source apportionment using a comprehensive dataset collected at multiple ambient monitoring stations across the state. The results will provide useful information for future epidemiological studies and help us improve the current understanding of the relationship between PM sources and health effects. The publication in Atmospheric Chemistry and Physics is recommended after the following comments are addressed.

We would like to thank the reviewer for his/her helpful feedback. The reviewer's comments are properly noted and his/her major concern about further clarification on the PMF analyses has been carefully addressed. Please find below detailed responses to each comment.

<u>*Major Comments:*</u>

Comment 1: *On page 20048, the authors state that previous studies are impacted by the limited number of sampling locations and short measurement periods. The readers may not know the background information about those studies. Could you please select 2 or 3 out of 10 papers cited here and briefly describe their limitations? How do they compare to your data (8 sites and 6 years)?*

Response: The reviewer's comment is noted. The following part is added to the revised manuscript to provide some succinct examples of the previous source apportionment studies conducted in California and describe their limitations in terms of spatial variability and/or studied time period.

"For instance, Kim et al. (2010) analyzed the PM$_{2.5}$ speciation data collected between 2003 and 2005 at 2 sampling sites in southern California (Los Angeles and Rubidoux) to identify and quantify major PM$_{2.5}$ sources, by application of a PMF model. Using similar source apportionment approach, Hwang and Hokpe (2006) evaluated the sources of ambient PM$_{2.5}$ at 2 sampling sites in San Jose during a large period of time between 2000 and 2005. In a more comprehensive study, Chen et al (2007) applied several receptor models (including PMF, CMB, and UNMIX) to the chemically speciated PM$_{2.5}$ measurements collected for one year (between 2000 and 2001) at 23 sites, all located in California's San Joaquin Valley, to estimate PM$_{2.5}$ source contributions."

Comment 2: Did you run the PMF model separately for 8 sites? Or you used all daily-averaged data in one input file for the PMF model? Please clarify.

Response: The PMF model was performed for each sampling site separately. To clarify this in the revised manuscript, the following sentence under the "source apportionment" section has been updated.

"In this study, the EPA positive matrix factorization (PMF) receptor model (version 3.0.2.2) was performed at each sampling site separately to identify the major sources of ambient $PM_{2.5}$ and quantify their relative contributions to total $PM_{2.5}$ mass.

Comment 3: On page 20050, the authors state that PMF analyses were conducted by a larger dataset between 2002 and 2013 for LA and Rubidoux, but average source contributions between 2002 and 2007 were calculated. Did you use any data between 2008 and 2013 for LA and Rubidoux in the input file when running the PMF model? If so, does that affect the PMF results as the measurement period is not consistent among different sites? Also, the PM sampling interval varies across 8 sites (every third day versus every sixth day). How did you prepare the concentration and uncertainty profiles in terms of data structure alignment? Did you use any values to replace those missing data points caused by different measurement period and sampling interval? Blank cells are not accepted by EPA PMF model 3.0.

Response:

1. The reviewer's comment is well taken. In the companion paper of this study (Hasheminassab et al., 2014), for LA and Rubidoux we used a combined chemical dataset from 2002 to 2013 as the input file when running the PMF model. For this paper, in order to be able to compare the results with those obtained for the rest of sampling sites, we calculated the average source contributions between 2002 and 2007, using the output of the same PMF runs, which were originally conducted using the 2002-2013 chemical dataset. Hasheminassab et al. (2014) performed a rigorous sensitivity analysis to evaluate whether the source contributions between 2002 and 2007 are biased when the PMF model is performed on the combined chemical dataset from 2002 to 2013. To that end, at each sampling site, the PMF model was first conducted using the entire chemical dataset (i.e. 2002-2013), and then the results were compared to the outputs of the PMF model performed separately on the pooled chemical datasets before and after 2007 (i.e. 2002-

2006 and 2008-2012, respectively). A similar number of sources with almost identical profiles were obtained from both approaches. In addition, the PMF outcome indicated comparable source contributions from the two approaches (less than 18% difference in average source contributions among all sources). The similarity between the source contributions obtained from the two approaches was also supported by Mann-Whitney rank sum tests, which showed that the differences between the daily-resolved source contributions are not statistically significant (p>0.05).

The below statements are added to the revised manuscript to address the reviewer's concern:

"Time-integrated 24 h $PM_{2.5}$ samples were collected between 2002 and 2007 at all sampling sites, except for LA and Rubidoux, at which a combined chemical dataset from 2002 to 2013 was used as the input file when running the PMF model (Hasheminassab et al., 2014). In the present study, in order to compare the results with those obtained for the rest of sampling sites, we calculated the average source contributions between 2002 and 2007 from the output of the same PMF runs which were originally conducted using the 2002-2013 chemical dataset. By performing a sensitivity analysis, Hasheminassab et al. (2014) showed that the results of the PMF model performed on the entire chemical dataset (i.e. 2002-2013) is comparable to the output of the PMF model conducted separately on 2002-2006 and 2008-2012 datasets, in terms of the sources identified (similar number of sources with almost identical compositions) and the absolute source contributions (less than 18% difference in average source contributions among all sources). The outcome of the sensitivity analysis thus indicated that the daily-resolved source contributions between 2002 and 2007 are not significantly biased when the chemical data between 2008 and 2013 are also included into the PMF input file."

2. As mentioned in response to the previous comment, PMF model was performed independently for each sampling site. Therefore, we had a separate input matrix of concentration and uncertainty for each site. Obviously, the dimension of the input matrices varies among all 8 sites, due to the differences in the sampling intervals and the number of species included in the model, following the screening procedure. We did not intend to align the structure and/or dimension of the input matrices among all 8 sites.

PMF model was performed separately for each sampling site and eventually the seasonal variability in the contributions of the identified sources was evaluated among all sites. In each individual input dataset, however, missing values of single species were treated following the method of Polissar et al. (1998), as mentioned in the manuscript.

Comment 4: On page 20051, could you please describe where PM filter weighing and chemical analysis were conducted? By EPA staff? On-site? Or samples were transported to other laboratories?

Response: Filter handling and custody procedures as well as protocols for the chemical analyses are thoroughly described in the U.S. EPA Quality Assurance Project Plan (QAPP) (EPA-454/R-01-001) for the field operations involved in the $PM_{2.5}$ Speciation Trends Network (STN). Briefly, the state and local air monitoring agencies are responsible for the operation of the STN sites. They are also responsible for training the personnel performing sampling, QA/QC, data handling, and other duties related to the $PM_{2.5}$ chemical speciation network. For STN sampling, filters are tested, equilibrated, and weighted in the U.S. EPA contract laboratories, and then they are shipped to the field. After sampling, filters bearing $PM_{2.5}$ deposits are promptly shipped back to the laboratories for weight determination and other chemical analyses.

The following statements are added to the revised manuscript to further clarify this:

"Filter weighing and chemical analyses were performed according to the U.S. EPA Quality Assurance Project Plan (QAPP) (EPA-454/R-01-001) adopted for the STN field sampling. According to the QAPP, filters are tested, equilibrated, and weighted in the U.S. EPA contract laboratories, and then they are shipped to the field. After sampling, filters bearing $PM_{2.5}$ deposits are promptly shipped back to the laboratories for weight determination and other chemical analyses."

**Minor Comments:**

In addition to the comments mentioned above, the following changes in the manuscript are suggested.

Comment 1: Page 20046 Line 2: dp –> subscript p

Response: The correction was made in the revised manuscript.

Comment 2: Page 20048 Line 29: replace CAL EPA with Cal EPA (to be consistent with the abbreviation used in acknowledgements)

Response: "CAL EPA" was replaced with "Cal EPA" in the revised manuscript.

Comment 3: Page 20050 Line 7: remove (SoCAB) as it is not used in the following sections

Response: In accordance with the reviewer's comment, "(SoCAB)" was removed from the revised manuscript.

Comment 4: Page 20050 Line 8: LA should be defined when Los Angeles appears for the first time.

Response: The reviewer's comment is noted. In the revised manuscript we defined LA when Los Angeles appeared for the first time.

Comment 5: Page 20051 Line 13: replace positive matrix factorization (PMF) with PMF (it has been defined previously in the introduction section).

Response: In the revised manuscript we replaced "positive matrix factorization (PMF)" with "PMF".

Comment 6: Page 20051 Line 26 and 20052 Line 2: BDL is defined twice and differently (w/ and w/o the).

Response: The reviewer's comment is noted. In the revised manuscript we first defined "below detection limit (BDL)" (without the), and then used the acronym (BDL).

Comment 7: Page 20053 Line 12, 16, 18: replace relative humidity with RH as it has been defined previously.

Response: "relative humidity" was replaced with RH in the revised manuscript.

Comment 8: Page 20058 Line 3: remove (CMB) as it is not used in the following sections.

Response: "(CMB)" was removed from the revised manuscript.

Comment 9: Page 20058 Line 18: replace relative humidity with RH.

Response: "relative humidity" was replaced with RH in the revised manuscript.

Comment 10: Page 20060 Line 15, 19: replace relative humidity with RH.

Response: "relative humidity" was replaced with RH in the revised manuscript.

References

Chen, L.W.A., Watson, J.G., Chow, J.C., Magliano, K.L., 2007. Quantifying PM2.5 Source Contributions for the San Joaquin Valley with Multivariate Receptor Models. Environmental Science & Technology 41, 2818-2826.

Hasheminassab, S., Daher, N., Ostro, B.D., Sioutas, C., 2014. Long-term source apportionment of ambient fine particulate matter (PM2.5) in the Los Angeles Basin: A focus on emissions reduction from vehicular sources. Environmental Pollution 193, 54-64.

Hwang, I., Hopke, P.K., 2006. Comparison of source apportionments of fine particulate matter at two San Jose speciation trends network sites. Journal of the Air & Waste Management Association (1995) 56, 1287-1300.

Kim, E., Turkiewicz, K., Zulawnick, S.A., Magliano, K.L., 2010. Sources of fine particles in the South Coast area, California. Atmospheric Environment 44, 3095-3100.

Polissar, A.V., Hopke, P.K., Paatero, P., Malm, W.C., Sisler, J.F., 1998. Atmospheric aerosol over Alaska: 2. Elemental composition and sources. Journal of Geophysical Research: Atmospheres 103, 19045-19057.