

**Reviewer #2:**

**The study is focused on the characterization of the major sources of PM number concentrations and on the quantification of their contributions using the PMF receptor model applied to PM number size distributions combined with several auxiliary variables in central Los Angeles. The topic is interesting, the data set is large and reliable, the paper is well organized and data interpretation seems to be sound. While there are many articles regarding aerosol source identification by PMF, there are few regarding the analysis of particle number concentrations by PMF. This work gives a very good quantitative identification of the sources which contribute to the particle number as a function of particle size in Los Angeles. I have only few minor remarks:**

**Authors:** We thank the reviewer for his/her valuable comments on the paper that has improved the quality of the work. Please find below our detailed response and the modification made to the manuscript according to each comment.

**1) p 14 l.32-34: why the same explanation is not valid for traffic 1?**

**Response:** The reviewer's comment is properly noted and the response is provided in details below.

As mentioned in the manuscript, because of its larger mode diameter, "Traffic 2" factor was attributed to more aged particles that could also have come from more distant sources, compared to the "Traffic 1" factor that is attributed to freshly emitted particles from nearby traffic sources. Therefore, one possible source that could affect Traffic 2, but not Traffic 1, is the increased traffic volume in Downtown Los Angeles (located 4 km northeast of our sampling site), mainly due to the increased nighttime activities in this part of the city, especially during weekend nights. To further support this hypothesis, we analyzed the wind direction data during our sampling campaign and realized that in weekends, the prevailing wind was from NE direction during night, as can be seen in the table below. This observation further corroborates our hypothesis that Traffic 2 particles

could have come from Downtown LA during weekend nights, when the nighttime activities, and therefore traffic volume, peak in this area of the city. These sources are sufficiently far not to affect the Traffic 1 factor (i.e. freshly emitted particles), because by the time these particles reach our sampling site, their size would have grown to larger ranges not captured in Traffic 1 factor.

Average wind speed and wind direction during the sampling campaign in central Los Angeles

Hour	Wind speed		
	Avg	STD	WD
0	3.43	1.74	NE
1	3.51	1.83	NE
2	3.65	2.10	NE
3	3.76	1.89	NE
4	3.63	1.99	NE
5	3.78	1.93	NE
6	4.00	2.23	NE
7	3.93	2.29	NE
8	3.60	2.14	NE
9	3.10	1.88	E
10	3.36	1.81	S
11	3.89	1.94	SW
12	5.16	2.47	SW
13	6.31	2.60	SW
14	7.34	2.49	SW
15	7.54	2.17	W
16	7.23	2.11	W
17	6.14	2.19	W
18	4.63	2.15	W
19	3.91	1.84	W
20	3.45	1.70	W
21	3.21	1.59	N
22	3.08	1.77	NE
23	3.25	1.75	NE

**2) Factor 5: is there any explanation why this factor gives such a small contribution to particle number? This factor is attributed to secondary nitrates and organics (quite reasonable). Is not there any contribution from secondary sulfates in Los Angeles? If yes, in which of the identified factors is?**

**Response:** The reviewer's comment is properly taken and detailed response is provided below.

Regarding the first part of the comment (i.e., relatively low contribution of this factor to particle number), it should be noted that it is mostly because of the size range in which this factor lies, making this source factor a relatively small contributor to particle number concentrations; as the particles grow in size, their contribution in mass concentrations increases drastically, while their contribution to number concentrations decreases (Figure 4). The “secondary aerosol” source factor identified in this study ranged between 400-500  $\mu\text{m}$ , which, based on the results presented in Figure 2 (i.e. number and volume size distribution) contributes greatly to particle volume/mass concentration, but does not have a large contribution to the particle number concentration. This observation is also consistent with the results presented in many previous studies. For instance, in a study conducted by (Ogulei et al., 2006), the contribution of secondary sources to the total particle number concentrations in Baltimore was found to be in the order of 100 particles/cm<sup>3</sup>, while the contribution from traffic and other important sources (such as power plant) were found to be in the order of thousands of particles/cm<sup>3</sup>. (Beddows et al., 2015) also reported that, in London, the contribution of the identified “secondary aerosol” sources was around 400 particles/cm<sup>3</sup>, while that of traffic sources was as high as 3500 particles/cm<sup>3</sup>. Additionally, in a study performed by (Friend et al., 2013), a source that was identified as “secondary aerosol” had a contribution to particle number concentration of around 5% in the sampling site that was mostly affected by traffic sources, while its contribution was 10% in the sampling site that was less affected by traffic sources (mainly because the particle number concentrations in the first traffic site were mostly affected by traffic sources, making the relative contribution from other sources less important). This was also the case in the present study, in which traffic sources together

were found to contribute more than 60% to the total particle number concentrations, overwhelming the relative contribution from other sources.

Regarding the second part of the comment (i.e., the possible contribution from secondary sulfate), we do acknowledge the existence of secondary sulfate in Los Angeles, given that this source has been identified in several previous source apportionment studies, using PMF, in this area (Hasheminassab et al., 2014; Kim and Hopke, 2007). However, it should be noted that these studies were performed on particle mass concentrations using chemically-speciated data. Using a chemically-speciated dataset, one can easily distinguish secondary nitrate from secondary sulfate, because of the existence of chemical markers of such sources, i.e.  $\text{NO}_3^-$  and  $\text{SO}_4^{2-}$ . However, using solely particle number size distribution data, discerning these two factors (along with secondary organic aerosols) is rather impossible, because they lie within the same size range, which is the most important criterion for PMF to differentiate source factors.

It should be noted that the only PMF source apportionment studies on particle number concentrations that have been able to separately identify secondary sulfate and secondary nitrate are those that also included chemically-speciated data along with the particle number size distributions.

Lastly, although the PMF model in this study was not able to identify separate source factors for secondary sulfate and nitrate, the contribution of secondary sulfate is expected to coexist in the “secondary aerosol” factor resolved by the PMF solution. However, we were not able to observe any fingerprints (distinct patterns of diurnal or seasonal variation) pertinent to secondary sulfate. This is mainly because the seasonal and diurnal variations of secondary sulfate and nitrate are actually reverse, the former peaking in summer and in mid-day, while the latter peaking in the cold season and at night. Moreover, previous studies in this area (Hasheminassab et al., 2014) found that the concentrations of secondary nitrate aerosols far exceed those of secondary sulfate (2-3 times higher); therefore, it can be concluded that the seasonal as well as the diurnal variation of the “secondary aerosol” resolved in this study are probably governed by the contributions of secondary nitrate aerosols, as these particles are believed to be the major component of the secondary aerosols, at least in the Los Angeles area.

**3) p 18 l.9: the factor was called “soil/road dust” but the time trend of this source does not justify road dust as a source**

**Response:** The reviewer's comment is properly noted and a detailed response is provided below.

Based on the previous studies conducted in Los Angeles at the same sampling location (Cheung et al., 2012; Shirmohammadi et al., 2015), and also given the close proximity of our sampling site to major traffic sources (surface streets and freeways), we are confident that the "road dust" exists as a source of PM in the study area. However, as mentioned above, since we only used particle number size distribution data for the source apportionment analysis without any chemically-specified data or unique source tracers, the PMF model could not separately identify two distinct "road dust" and "soil" factors. Nonetheless, given the typical size range of road dust particles, which mainly exist in the coarse mode (Cheung et al., 2012), we believe that road dust would be partitioned in the identified "soil" factor as well.

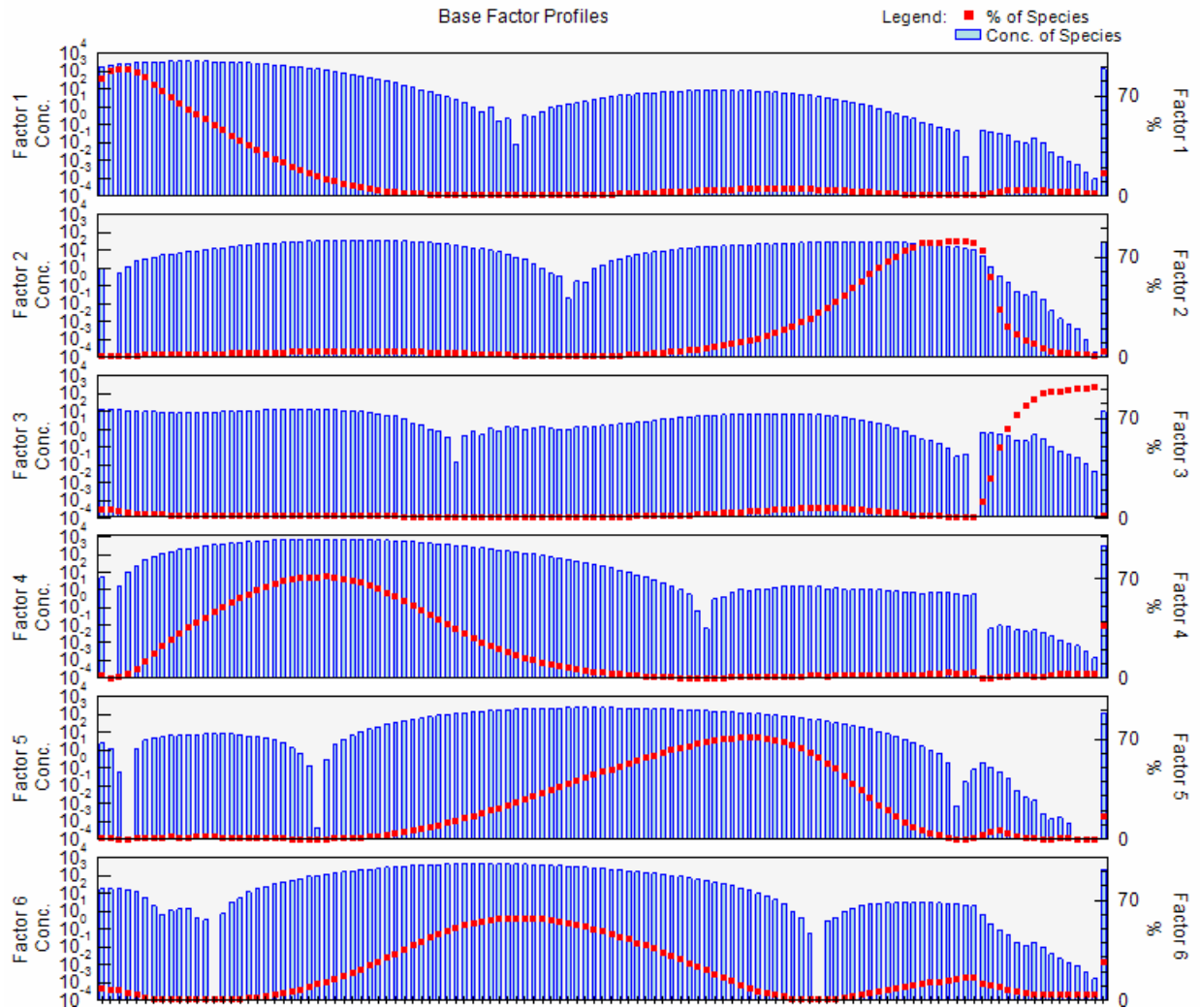
Additionally, based on the study of (Cheung et al., 2012), the contribution of "road dust" is expected to be higher in summer, which is consistent with the seasonal trend observed for the "soil/road dust" factor in this study, with much higher contribution in the warm phase (Figure 6).. Moreover, prior studies in the same area indicated that the contribution of soil is larger compared to road dust (Cheung et al. 2012) Therefore, the overall diurnal trend for this factor, i.e. soil/road dust, may have been dominated by the soil particles, not enabling us to see a distinct diurnal variability between the two sources.

**4) Fig 3: the normalized concentrations of PM<sub>2.5-10</sub> are higher respect to PM<sub>2.5</sub> for both traffic 2 and urban background; is there any possible explanation?**

**Response:** The reviewer's comment is properly taken and the author's response is provided in details below.

We concur with the reviewer that the size range of particles identified in the “Traffic 2” and “Urban Background” are larger than those of the “Traffic 1” factor (as shown by the number size distributions). This is mainly because that they are more aged and more likely to originate from more distant sources (especially the urban background aerosol) compared to the freshly emitted particles that come from the “Traffic 1” factor. However, this cannot solely justify the relatively high loadings of PM<sub>10-2.5</sub> in these two factors. This is due to the fact that PM<sub>10-2.5</sub> particles are different in sources and chemical composition compared to PM<sub>2.5</sub>, and the only factor, among all of the identified source factors, that can be attributed to this size fraction is “soil/road dust” (Figure 2). Therefore, we believe the loading of PM<sub>10-2.5</sub> observed in these two factors (around 20%) may reflect a PMF artifact, which may occasionally fail to distinguish the profiles of two or more sources, and on several instances residuals from other factors may be observed in the factor of interest, as also noted by reviewer #1.

Additionally, it should be noted that our main scope in this study was particle number apportionment, rather than particle mass apportionment. Mass concentrations, along with some other parameters (e.g. gaseous pollutants, traffic data, etc.) were included as auxiliary data only to help the interpretation of the resolved factors. A sensitivity analysis was run to identify the impact of these auxiliary variables on the PMF results. As can be seen below, results of the analysis indicated that the results of the PMF model are quite robust even after excluding the data pertaining to the auxiliary variables (i.e., PM mass, gaseous pollutants, EC/OC, meteorological parameters, and traffic count data).



5) Fig. 8: why on weekend there is a night peak only for traffic 2?

**Response:** We have fully addressed this comment in response to the first comment raised by the respected reviewer. So you are kindly referred to the response to the first comment.

## References

- Beddows, D. C. S., Harrison, R. M., Green, D. C., and Fuller, G. W.: Receptor modelling of both particle composition and size distribution from a background site in London, UK, *Atmospheric Chemistry and Physics Discussions*, 15, 10123-10162, 2015.
- Cheung, K., Shafer, M. M., Schauer, J. J., and Sioutas, C.: Diurnal trends in oxidative potential of coarse particulate matter in the Los Angeles Basin and their relation to sources and chemical composition, *Environmental science & technology*, 46, 3779-3787, 2012.
- Friend, A. J., Ayoko, G. A., Jager, D., Wust, M., Jayaratne, E. R., Jamriska, M., and Morawska, L.: Sources of ultrafine particles and chemical species along a traffic corridor: comparison of the results from two receptor models, *Environmental Chemistry*, 10, 54-63, 2013.
- Hasheminassab, S., Daher, N., Saffari, A., Wang, D., Ostro, B. D., and Sioutas, C.: Spatial and temporal variability of sources of ambient fine particulate matter (PM<sub>2.5</sub>) in California, *Atmos. Chem. Phys.*, 14, 12085-12097, 10.5194/acp-14-12085-2014, 2014.
- Kim, E., and Hopke, P. K.: Source characterization of ambient fine particles in the Los Angeles basin, *Journal of Environmental Engineering and Science*, 6, 343-353, 2007.
- Ogulei, D., Hopke, P. K., Zhou, L., Pancras, J. P., Nair, N., and Ondov, J. M.: Source apportionment of Baltimore aerosol from combined size distribution and chemical composition data, *Atmospheric Environment*, 40, 396-410, 2006.
- Shirmohammadi, F., Hasheminassab, S., Wang, D., Saffari, A., Schauer, J. J., Shafer, M. M., Delfino, R. J., and Sioutas, C.: Oxidative potential of coarse particulate matter (PM<sub>10-2.5</sub>) and its relation to water solubility and sources of trace elements and metals in the Los Angeles Basin, *Environmental Science: Processes & Impacts*, 17, 2110-2121, 2015.