Reviewer #1:

SOURCE APPORTIONMENT OF PARTICLE NUMBERS IN LOS ANGELES
There is very widespread interest currently in particle number concentrations in the atmosphere, and the determinants of those concentrations. Much of the recent research has focused on the global atmosphere, with particular attention being paid to nucleation processes. While such research is important in the context of global climate, there is also a very important research area associated with particulate matter in the urban atmosphere. While a huge number of source apportionment studies of particle mass have been carried out in urban environments around the world, there has been a relative scarcity of source apportionment of particle number concentrations. The latter requires data sets which are not available from many locations, and the rather challenging application of source apportionment programs. Consequently, there are few high quality studies in the literature and this paper presents a valuable addition for a major city with a well characterized air pollution climate. It can be very challenging to extract meaningful factors from PMF, but the authors appear to have succeeded in doing so, and have provided convincing associations with the sources to which they attribute the factors. In summary, the data interpretation appears to be sound, and consequently the paper offers excellent quantitative insights into the sources contributing to the particle number count as a function of particle size within Los Angeles. There are some relatively minor points which need to be addressed, relating to both the methodology and the data interpretation.

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 modifications made to the manuscript according to each comment.

(A) At no point is it clearly stated how the particle size distribution data are sub-divided for uploading to the PMF. Were the size bins from the SMPS used without subsequent combination into larger size fractions and what size ranges were used from the OPC? There is also a question as to how the errors in the SMPS and OPC

data were quantified in order to populate the error matrix of the PMF. These are points of interest which should be included in the paper.

Response: The reviewer's comment is properly taken and carefully addressed as discussed in detail below.

Regarding the first part of the reviewer's question (i.e. the size bins used from SMPS and OPS), it should be mentioned that the size bins from the SMPS were used without subsequent combination into larger size fractions. Additionally, the SMPS size bins from 13.6-514 nm were merged with the OPS size channels covering 0.522-9.01 μm as the input data to the PMF model. This information is now added in the revised manuscript (Page 6, Lines 3-5):

"Therefore, size bins covering the range of 13.6-514 nm from SMPS (without subsequent combination into larger size fractions) were merged with the OPS channels from 0.522 to 9.01 μm as the input data to the PMF model. "

Regarding the second part of the comment (i.e. the estimation of errors from the SMPS and OPS), we had already provided detailed information as to how these errors were estimated and used to calculate the uncertainties associated with each single data point (Page 9, Lines 7-24):

"In the present work, since no measurement uncertainties were available for the input parameters, we applied the method suggested by Ogulei et al. (2006a;2006b) and Zhou et al. (2014) to calculate the uncertainties for individual data points inserted into the model. For this purpose, measurement errors were first estimated for each data point using the following equation:

$$\sigma_{ij} = C_1(N_{ij} + \overline{N}_j) \qquad (6)$$

where, σ_{ij} is the estimated measurement error for the *i*th sample and *j*th size bin (or concentration of auxiliary variables); C_1 is an empirical constant usually between 0.01 and 0.05; N_{ij} is the observed number concentration for the *i*th sample and *j*th size bin (or concentration of auxiliary variables); and $\overline{N}_j$ is the arithmetic mean of the PM number concentrations for the *j*th size bin (or concentration of auxiliary variables).

The value of the measurement method obtained from the above equation is then used to calculate the measurement uncertainty, according to the following equation:

$$S_{ij} = \sigma_{ij} + C_2 \max(|x_{ij}|, |y_{ij}|) \qquad (7)$$

where, S_{ij} is the calculated uncertainty associated with the ith sample and jth size bin; C_2 is an empirical constant usually between 0.1 and 0.5; and Y_{ij} is the value calculated by the model for x_{ij}. In the present work, C_1 and C_2 values of 0.05 and 0.1 were chosen to obtain the most physically interpretable solution using a trial and error approach."

It should be noted that this method has been successfully used in a large number of studies on PM source apportionment using particle number size distribution, including but not limited to (Beddows et al., 2015;Friend et al., 2012;Harrison et al., 2011;Krecl et al., 2008;Ogulei et al., 2007;Ogulei et al., 2006a;Ogulei et al., 2006b).

(B1) The PMF does not appear to have separated the factors wholly cleanly (which is not unusual). Thus, the nucleation factor (Factor 1) appears to show some influence of morning rush hour traffic seen in Figures 7 and 8. A comment on the magnitude of this overlap would be useful.
(B2) The nucleation factor has significant magnitude, even in the depths of the night, which require some explanation. Is this a reflection that nucleation continues through the night, or is it an artefact of the PMF?

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

(B1) We do not believe that the minor peak observed in the morning rush hours in the nucleation factor is due to PMF artifact. Although the maximum amount of nucleation is expected to occur in early afternoon, when temperature and solar radiation are highest leading to maximum photochemical activity in the atmosphere, we expect to observe some influence of nucleation during morning rush hours due to the cooling, following dilution, of vehicular emissions and the partitioning of semi-volatile species into the

particle phase (Harrison et al., 2011). A Similar trend was observed by Harrison et al. (2011) in number size distributions on data collected near a major road way in central London. The "nucleation" factor resolved by Harrison et al. (2011) exhibited even a larger peak in the morning rush hours than that observed in the early afternoon, mainly because their measurements were carried out near a major roadway, while in the present study measurements were performed in an urban background location that is affected by vehicular emissions. Similar to what was observed in our study, the authors argued that this nucleation peak was due to the dilution of diesel exhaust emissions in the low temperatures observed during that time of day, and has been supported by the results from other studies, including (Charron and Harrison, 2003;Janhäll et al., 2004;Ntziachristos et al., 2007). In addition, our results are also consistent with those from the study of Brines et al. (2016), in which the authors observed very similar diurnal variation for nucleation, with a minor peak in the early morning and a major peak in early afternoon at the same sampling location in Los Angeles, using the k-means clustering approach for PM number apportionment.

Therefore, to fully address the reviewer's comment we have added the following text to the revised manuscript:

Page 13, Lines 12-14: "A minor peak was also observed during morning rush hours (6-8 am), which suggests the partial influence from traffic sources, as also observed by loadings of HDV and LDV in this factor (Figure 3)."

Page 13, Lines 20-23: " The minor peak in the early morning can also be explained by the cooling, following dilution, of vehicular exhaust emissions, which leads to the partitioning of semi-volatile exhaust gases into the particle phase; this process is further enhanced by to the lower temperatures during that time of day (Harrison et al. 2011; Charron and Harrison, 2003; Janhall et al. 2004; Ntziachristos et al. 2007)."

Page 13, Lines 26-28: "They observed very similar diurnal variation for nucleation, with a minor peak in the early morning and a major peak in early afternoon at the same sampling location in Los Angeles."

(B2) In regards to the second part of the comment, we believe that it may have been due to both the PMF artifacts and occurrence of low levels of nucleation during the night. As the respected reviewer has mentioned, it is not unusual for the PMF model to fail to

cleanly resolve different factors, and it is likely that the trends do not exactly follow the characteristics that are expected from the resolved factor; thus, they are assigned to PMF artifacts. This can also be the case here, as can be true for all other PMF source apportionment studies. On the other hand, looking at the results of previous studies, it is unlikely that the contribution of a factor such as nucleation would go down to zero after peaking at specific times of the day (i.e., early morning and early afternoon). For example, Harrison et al. (2011) presented a diurnal variation chart for the nucleation factor, peaking in early morning and early afternoon hours, but not decreasing down to very low levels following these peaks or during nighttime. As can be seen in Figure 7, the contributions of nucleation factor are reduced substantially to lower levels at night, reaching background levels, but are still detectable. Moreover, the diurnal trend of nucleation factor in this study is in very good agreement with the findings of Brines et al. (2016), in which the authors reported nucleation as one of the major sources of UFPs in Los Angeles using the data obtained from the same sampling location.

(C) The Traffic 2 factor shows a rather odd diurnal variation, which differs substantially between the cold phase and the warm phase and between weekday and weekend data. The diurnal profiles seem to suggest a substantial contribution of semi-volatile materials, especially during the cold phase, as reflected in the substantial increase at nighttime.

Response: The reviewer's comment is properly taken and the following sentence has been added to the manuscript text:

Page 15, Lines 10-12 "This diurnal profile suggests a major contribution from semi-volatile compounds in the atmosphere, particularly in the cold phase, as reflected in the substantial increase at nighttime."

In addition, more detailed discussion on the characteristics of Traffic 2 and its diurnal variability, particularly during weekday vs. weekend, have been presented in response to comment 1 of the second reviewer.

(D) The huge difference between the cold phase and the warm phase seen in Figure 7 for the soil/road dust factor is rather surprising. Are the weather conditions really that different between the cold phase and the warm phase, such that they can explain such a huge seasonality?

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

Based on Figure 1, it can be observed that the meteorological conditions are indeed different in the cold and warm phases; there is an average 7 °C difference in temperature between cold and warm phases; a 10% difference in RH; wind speed is twice as high in the warm phase compared to the cold phase; and solar radiation is approximately twice as high in the warm phase than in the cold phase. The appreciable differences in all of the aforementioned parameters significantly influence atmospheric stability and mixing height in different phases, favoring the wind-induced resuspension of road dust and crustal material (i.e. soil), thereby enhancing the contribution of soil/road dust in the warm phase compared to the cold phase. Hasheminassab et al. (2014) also found in a PMF source apportionment study across the state of California that the "soil" factor had a maximum contribution in summer (due to the increased temperature and wind speed, as well as atmospheric instability), particularly in areas with lower levels of RH and precipitation, which is consistent with the results observed in the present study.

(E) Some recent papers have attributed a significant proportion of particulate matter mass in California to emissions from cooking. Such attribution usually comes from AMS data, largely on the basis of diurnal profiles. Is there any indication of such a source within this data set? One might expect it to be more prominent at the weekend than on weekdays, especially if related to outdoor barbequing.

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

Although we acknowledge the existence of "cooking" source in the study area, we did not expect that the PMF would be able to resolve such a factor. This is mainly because our sampling site is located in the vicinity of a major freeway (about 150 m downwind), in the middle of a parking center, and is quite far from (at least obvious) sources of food cooking emissions (e.g. restaurants, residential areas). So we can conclude that compared to traffic sources, the contribution of such a minor source, such as cooking, would be very small, making it difficult to be detected/resolved by the PMF model. Moreover, previous studies conducted at the same sampling location were not able to identify/quantify "cooking" sources even using specific tracers of cooking, such as cholesterol, in the chemical mass balance (CMB) source apportionment model (Arhami et al., 2010;Hasheminassab et al., 2013).

Nonetheless, the contribution from "cooking" sources, although very minor, is expected to be within the resolved ""urban background aerosol" factor. However, since many sources are involved in this factor to form the background concentrations, detecting a distinct diurnal trend from each of the participating sources is quite unlikely.

(F) Page 3, line 6. This implies that regulations on PM number emissions from road vehicles in Europe have been set on the basis of health studies of UFPs. This is not the case. The PMP number limits, which have been introduced as part of recent European vehicle emission standards, arose because of the difficulty encountered in achieving repeatable measurements of very low mass concentrations of PM in diluted engine exhaust, whereas particle number could be measured very repeatedly after removable of the semi-volatile fraction. Hence a test procedure, based upon solid particle number, was considered a more practicable option than determination of mass.

Response: We thank the reviewer for his/her insightful comment. To address the reviewer's comment we changed the sentence in the introduction in a way to dispel the implication that the European particle number concentration standard was set because of the body of evidence linking health effects to particle number concentrations. The revised paragraph can be found below.

Page 3, Lines 6-7: "Regulations on PM number concentrations have already been implemented on motor vehicle emissions in a few countries."

G. Page 5, line 34. Last word should read 'factor' rather than 'actor'.

Response: The reviewer's comment was noted and the change was made in the revised manuscript (Page 5, Line 32).

H. Page 6, line 30. 'Ration' should read 'ratio'.

Response: The reviewer's comment was noted and the change was made in the revised manuscript (Page 6, Line 31).

I. It would be good to quote the OC/EC gradient determined through equation 1, as there is still considerable interest in applications of the EC Tracer Method to source apportionment of organic carbon.

Response: The reviewer's comment is properly taken and the following information was added to the revised manuscript accordingly.

Page 7, Lines 1-3: "Using equation (1), the slope and the intercept of the regression line were found to be 1.55 (±0.07) and 0.45 (±0.24), respectively. More detailed information on the results obtained using the EC tracer method can found elsewhere (Saffari et al., 2016)"

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