

March 31th, 2016

Dear Editor,

We would like to thank you and the reviewers for their helpful comments on our manuscript entitled: "**Source apportionment of ambient particle number concentrations in central Los Angeles using positive matrix factorization (PMF) (Ms. Ref. No.: acp-2015-959)**".

We believe the constructive comments improved the manuscript for the readership of *Atmospheric Chemistry and Physics*.

Attached is a document in which we provide detailed responses to the comments. I am submitting this file on behalf of Dr. Constantinos Sioutas (sioutas@usc.edu), the corresponding author of this paper.

We hope that the revised manuscript is now in an acceptable form for publications in *Atmospheric Chemistry and Physics*.

Sincerely,

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The authors would like to thank the reviewers and editor for their helpful and constructive comments on the manuscript. Attached are detailed replies to each reviewer's comment and the appropriate changes made to address each comment.

Response to reviewers' comments

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:

$$\sigma_{ij} = C_1(N_{ij} + \bar{N}_j) \quad (6)$$

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 \bar{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:

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

where, S_{ij} is the calculated uncertainty associated with the i th sample and j th 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 barbecuing.

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 removal 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 be found elsewhere (Saffari et al., 2016)."

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		WD
	Avg	STD	
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 μm , 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., 2006b), the contribution of secondary sources to the total particle number concentrations in Baltimore was found to be in the order of 100 particles/cm³, while the contribution from traffic and other important sources (such as power plant) were found to be in the order of thousands of particles/cm³. (Beddows et al., 2015) also reported that, in London, the contribution of the identified “secondary aerosol” sources was around 400 particles/cm³, while that of traffic sources was as high as 3500 particles/cm³. 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. NO_3^- and 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 1.9: 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.

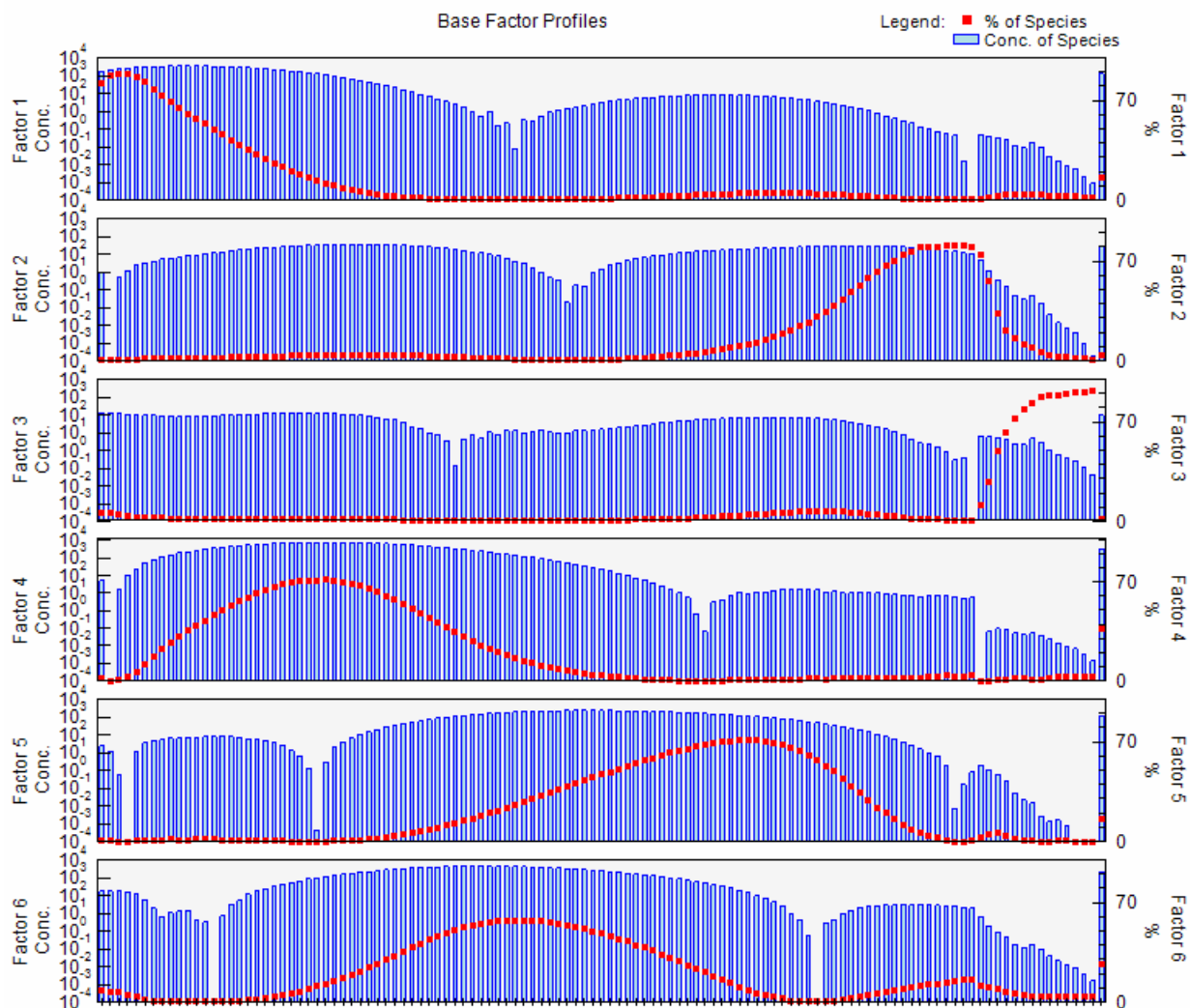
4) Fig 3: the normalized concentrations of PM_{2.5-10} are higher respect to PM_{2.5} for both traffic 2 and urban background; is there any possible explanation?

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_{10-2.5} in these two factors. This is due to the

fact that PM_{10-2.5} particles are different in sources and chemical composition compared to PM_{2.5}, 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_{10-2.5} 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.

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1 **Source apportionment of ambient particle number concentrations in central Los**
2 **Angeles using positive matrix factorization (PMF)**

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16 **Abstract**

17 In this study, the Positive Matrix Factorization (PMF) receptor model (version 5.0)
18 was used to identify and quantify major sources contributing to particulate matter (PM)
19 number concentrations, using PM number size distributions in the range of 13 nm to 10 μ m
20 combined with several auxiliary variables, including black carbon (BC), elemental and
21 organic carbon (EC/OC), PM mass concentrations, gaseous pollutants, meteorological, and
22 traffic counts data, collected for about 9 months between August 2014 and 2015 in central
23 Los Angeles, CA. Several parameters, including particle number and volume size distribution
24 profiles, profiles of auxiliary variables, contributions of different factors in different seasons
25 to the total number concentrations, diurnal variations of each of the resolved factors in the
26 cold and warm phases, weekday/weekend analysis for each of the resolved factors, and
27 correlation between auxiliary variables and the relative contribution of each of the resolved
28 factors, were used to identify PM sources. A six-factor solution was identified as the
29 optimum for the aforementioned input data. The resolved factors comprised nucleation,
30 traffic 1, traffic 2 (having a larger mode diameter than traffic 1 factor), urban background
31 aerosol, secondary aerosol, and soil/road dust. Traffic sources (1 and 2) were the major
32 contributor to PM number concentrations, collectively making up to above 60% (60.8-68.4%)
33 of the total number concentrations during the study period. Their contribution was also
34 significantly higher in the cold phase compared to the warm phase. Nucleation was another
35 major factor significantly contributing to the total number concentrations (an overall

1 contribution of 17%, ranging from 11.7% to 24%), having a larger contribution during the
2 warm phase than in the cold phase. The other identified factors were urban background
3 aerosol, secondary aerosol, and soil/road dust, with relative contributions of approximately
4 12% (7.4-17.1), 2.1% (1.5-2.5%), and 1.1% (0.2-6.3%), respectively, overall accounting for
5 about 15% (15.2-19.8%) of PM number concentrations. As expected, PM number
6 concentrations were dominated by factors with smaller mode diameters, such as traffic and
7 nucleation. On the other hand, PM volume and mass concentrations in the study area were
8 mostly affected by sources having larger mode diameters, including secondary aerosols and
9 soil/road dust. Results from the present study can be used as input parameters in future
10 epidemiological studies to link PM sources to adverse health effects as well as by policy
11 makers to set targeted and more protective emission standards for PM.

12

13 **1. Introduction**

14 Numerous epidemiological studies have provided compelling evidence linking exposure
15 to ambient particulate matter (PM) with increased risk of respiratory and cardiovascular
16 diseases, hospitalization, and premature mortality (Brunekreef and Forsberg, 2005; Dockery
17 and Stone, 2007; Miller et al., 2007; Pope et al., 2004; Pope Iii et al., 2002; Gauderman et al.,
18 2015). According to the most recent global burden of disease study, over 3 million premature
19 deaths annually occur all around the globe due to exposure to ambient PM (Lim et al., 2013).
20 It should, however, be noted that most of these epidemiological studies have related the
21 aforementioned health outcomes with solely the mass concentrations of PM and, therefore, do
22 not adequately represent submicron particles (Ogulei et al., 2007), mainly because this PM
23 fraction contributes negligibly to total ambient PM mass (Delfino et al., 2005; Vu et al.,
24 2015). More recently, studies have associated human health effects with particles
25 characteristics other than mass concentration, including size, number concentration, chemical
26 composition, and even surface area (Brook et al., 2010; Kasumba et al., 2009; Lighty et al.,
27 2000; Chen et al., 1991; Dreher et al., 1997; Oberdörster et al., 1994; Peters et al., 1997; Delfino
28 et al., 2010; Davis et al., 2013). Even though our knowledge of which particle characteristic
29 (mass, size, surface area, etc.) can be considered as the best predictor for human health
30 outcomes is limited, there is growing evidence highlighting the critical role of particle size
31 and number concentrations from a human health effect perspective (Vu et al., 2015). For
32 example, studies have indicated that ultrafine particles (UFP, i.e. particles with an
33 aerodynamic diameter of <100 nm) have higher toxicity per unit mass (Donaldson et al.,
34 1998; Li et al., 2003; Nel et al., 2006; Oberdörster et al., 2002), have higher deposition

1 efficiencies in the lung (Venkataraman, 1999), and penetrate deeper into the alveolar regions
2 of lungs (Sioutas et al., 2005). Additionally, several studies have also found that PM number
3 concentrations (mostly UFPs) can be associated with adverse effects on human health,
4 particularly for cardiovascular diseases (Delfino et al., 2005;Peters et al., 1997;Wichmann et
5 al., 2000).

6 **Regulations on PM number concentrations have already been implemented on motor**
7 **vehicle emissions in a few countries.** For instance, the Euro 5b and 6 have set a limit to
8 particle number emission factors, in addition to particle mass emission limits, for heavy-duty
9 and gasoline vehicles (<http://www.dieselnet.com/standards/eu/ld.php>, accessed 20 October
10 2015). It is also expected that this approach be gradually adopted in other parts of the world
11 (Friend et al., 2013), based mainly on the critical health implications of the PM number
12 concentrations, especially in smaller fractions like UFP. This emphasizes the necessity of
13 identification and quantification of PM sources based on number as well as mass (Friend et
14 al., 2013). This allows for source-specific assessment of health effects of exposure to PM to
15 provide us with the knowledge required to develop efficient control strategies for PM
16 emissions from major sources to minimize those health effects (Yue et al., 2008).

17 Positive Matrix Factorization (PMF) is one of the most widely used receptor models
18 that have been successfully applied to identify and quantify sources of atmospheric particles.
19 The vast majority of previous efforts has been devoted to the identification of sources that
20 contribute to the mass of particles using PMF on chemically-speciated PM mass data in
21 different parts of the world (Sowlat et al., 2013;Sowlat et al., 2012;Dutton et al., 2010;Lim et
22 al., 2010;Alleman et al., 2010;Sofowote et al., 2015). Recently, attempts have been made to
23 characterize sources that contribute to particle number, rather than mass, using PMF applied
24 to particle number size distribution data. These studies have adopted different approaches in
25 their source apportionment, including: (1) using particle number size distribution together
26 with gaseous pollutants, chemical composition, meteorological, or traffic data in the PMF
27 analysis (Beddows et al., 2015;Harrison et al., 2011;Kasumba et al., 2009;Ogulei et al.,
28 2007;Ogulei et al., 2006b;Thimmaiah et al., 2009;Zhou et al., 2005), (2) using particle
29 number size distribution and chemical composition data in separate and/or combined PMF
30 runs (Beddows et al., 2015;Gu et al., 2011), (3) comparing PMF results with actual events
31 during the study period (Ogulei et al., 2007), and (4) simply correlating the PMF results with
32 gaseous pollutants data (Friend et al., 2013;Friend et al., 2012;Kim et al., 2004). It is
33 noteworthy that the major factors resolved by these studies have been nucleation, traffic,
34 secondary aerosol, urban background, and wood burning.

1 Numerous studies have been performed in Los Angeles evaluating PM number
2 concentrations as well as size distributions, with a focus on vehicular emissions as a major
3 source of particle number in urban areas (Singh et al., 2006;Zhang et al., 2005). Source
4 apportionment of atmospheric particles has also been extensively studied in Los Angeles, but
5 almost all of the studies have focused on the contribution of different sources to PM mass
6 rather than PM number concentration (Ham and Kleeman, 2011;Hasheminassab et al.,
7 2013;Hwang and Hopke, 2006;Kim and Hopke, 2007;Kim et al., 2010;Schauer and Cass,
8 2000). To the best of our knowledge, no source apportionment studies have ever been
9 performed in Los Angeles on particle number size distributions using PMF. The only study
10 providing a source apportionment of particle number concentrations in Los Angeles is that of
11 Brines et al. (2015), in which major sources contributing to particle number concentrations
12 were identified in five high-insolation cities around the world (Barcelona, Madrid, Roma, Los
13 Angeles, and Brisbane) using the k-means clustering method. It should, however, be noted
14 that, in case of Los Angeles, the Brines et al. (2015) study used particle number size
15 distribution data for a rather limited time period (i.e., 3 months); moreover, the studied size
16 distributions ranged from 13 nm to 400 nm, thus excluding potentially important PM sources
17 contributing to the larger size fractions of PM_{2.5} and/or PM₁₀.

18 In the present work, we collected high-resolution (5-min measurements), wide-spectrum
19 particle number size distribution data (i.e., 13 nm to 10 μ m, covering the nucleation, Aitken,
20 accumulation, and coarse PM modes) over a long period of time (i.e., 9 months, covering
21 both warm and cold seasons) in a location near downtown of Los Angeles, California, to
22 identify and quantify sources contributing to particle number concentrations using the most
23 recent version of the PMF model (version 5.0). We also included gaseous pollutants (i.e., CO,
24 NO, NO₂, O₃), particle mass (PM_{10-2.5} and PM_{2.5}), meteorological (temperature, relative
25 humidity (RH), and wind speed), black carbon (BC), elemental carbon (EC) as well as
26 primary (POC) and secondary organic carbon (SOC), and traffic (counts of light-duty
27 vehicles (LDVs) and heavy-duty vehicles (HDVs)) data as inputs to help identify the factors
28 resolved by the model. Results from the present study can be used as a platform for future
29 health effect studies to estimate the source-specific impact of exposure to PM from a number
30 concentration perspective, which is critical for development and establishment of abatement
31 strategies and standards in order to minimize the most relevant health outcomes.

32

33 **2. Methodology**

34 *2.1. Sampling site*

1 Continuous measurements were carried out at the particle instrumentation unit (PIU)
2 located on the University of Southern California's (USC) park campus, approximately 3 km
3 south of downtown Los Angeles, CA. The PIU is located within approximately 150 m
4 downwind of a routinely congested interstate freeway, i.e. I-110, and is also in close
5 proximity to parking and construction facilities. Previous studies conducted by this research
6 group have indicated that the PIU is a mixed urban site that is also heavily impacted by
7 vehicular emissions (Geller et al., 2004; Moore et al., 2007; Hasheminassab et al., 2014b).

9 *2.2. Sampling time, method, and instrumentation*

10 Continuous measurements were conducted at the PIU from August 2014 through March
11 2015 as well as in August 2015. To obtain number size distribution of atmospheric particles
12 in the size range of 14 -760 nm (mobility diameter), a Scanning Mobility Particle Sizer
13 (SMPSTM, TSI Model 3081) was used, which was connected to a Condensation Particle
14 Counter (CPC, model 3020, TSI Inc., USA). Particles in the size range of 0.3-10 μm (optical
15 diameter) were measured using an Optical Particle Sizer (OPSTM, Model 3330, TSI Inc.,
16 USA). The time resolution for these two instruments was 5 min. The OPS instrument was
17 calibrated by the manufacturer using Polystyrene Latex (PSL) particles, which have a
18 dynamic shape factor of 1 (i.e., spherical particles) and a refractive index of 1.59. It should
19 also be noted that the measurements provided by the OPS instrument depend primarily on the
20 refractive index and the dynamic shape factor (Hasheminassab et al., 2014b). Numerous
21 studies have indicated that for spherical particles, the size selection offered by optical particle
22 counters, such as the OPS instrument, is quite similar to the actual physical diameter of the
23 particle being measured (Chen et al., 2011; Hasheminassab et al., 2014b; Hering and
24 McMurry, 1991; Reid et al., 1994). That said, there is compelling evidence in the literature
25 supporting the fact that the refractive index and the dynamic shape factor for ambient
26 aerosols in urban areas (such as Los Angeles) are quite similar to those of PSL
27 particles (Covert et al., 1990; Ebert et al., 2004; Hänel, 1968; Kent et al., 1983; Stolzenburg et
28 al., 1998; Strawa et al., 2006; Watson et al., 2002). To further evaluate this assumption, we
29 used the Multi-Instrument Manager (MIMTM) software, developed by TSI Inc., USA, which
30 estimates the refractive index and dynamic shape factor of aerosols from parallel SMPS and
31 OPS scans. The output from this software indicated that the average real part of the refractive
32 index for the aerosols collected in this study was 1.59 ± 0.01 and their dynamic shape factor
33 was 0.99 ± 0.02 . This finding is also in concert with the results of Hasheminassab et al.
34 (2014b), which reported an average shape factor of near unity at the same sampling site,

1 using the apparent and material density of aerosols. Hence, further adjustment of OPS sizing
2 was deemed unnecessary and the OPS size distribution, with the original size selection, was
3 merged with the SMPS size spectra. **Therefore, size bins covering the range of 13.6-514 nm**
4 **from SMPS (without subsequent combination into larger size fractions) were merged with the**
5 **OPS channels from 0.522 to 9.01 μm as the input data to the PMF model.** More detailed
6 information on the sensitivity of the OPS sizing to the refractive index and the dynamic shape
7 factor of aerosols can be found in (Hasheminassab et al., 2014b).

8
9 Black carbon (BC) concentrations, with a time resolution of 15 min, were measured
10 using a portable Aethalometer (Magee scientific, model AE-42). Hourly concentrations of
11 elemental carbon (EC) and organic carbon (OC) were measured using a semi-continuous
12 EC/OC carbon aerosol analyzer (Model 4, Sunset Laboratory Inc., USA), using the
13 thermal/optical transmittance measurement protocol of the National Institute of Occupational
14 Safety and Health (NIOSH 5040). By applying the “EC tracer method”, Saffari et al. (2016)
15 estimated the primary organic carbon (POC) and secondary organic carbon (SOC)
16 concentrations from total OC at the same location. These two parameters (i.e., POC and
17 SOC) were also used as input parameters in the PMF model, as they can provide valuable
18 input regarding the detection of primary and secondary sources of PM. The EC tracer method
19 has been discussed in detail elsewhere (Day et al., 2015;Saffari et al., 2016). Briefly, the main
20 assumption in this method is that EC and POC are released from similar sources; therefore,
21 this approach is most applicable where combustion is the main source of ambient POC (Day
22 et al., 2015). It is noteworthy that, in the present study, the sampling site was located in close
23 proximity to a major freeway, thereby making the EC tracer suitable for the data collected in
24 this location, as it has also been used in previous studies in the same sampling site (Polidori et
25 al., 2007;Saffari et al. 2016) as well as similar locations in the Los Angeles basin (Na et al.,
26 2004; Strader et al., 1999). In this method, the following equations can be used after
27 determining the ratio of POC to EC to estimate the concentration of SOC:

$$\text{POC} = [\text{OC/EC}]_p \times \text{EC} + b \quad (1)$$

$$\text{SOC} = \text{OC} - \text{POC} \quad (2)$$

30 where, $[\text{OC/EC}]_p$ is the POC to EC **ratio**; b is the intercept of the linear regression between
31 POC and EC, which is considered to be the portion of POC associated with non-combustion
32 emissions. **Using equation (1), the slope and the intercept of the regression line were found to**

1 be 1.55 (± 0.07) and 0.45 (± 0.24), respectively. More detailed information on the results
2 obtained using the EC tracer method can be found elsewhere (Saffari et al., 2016). It is also
3 noteworthy that we used the “high EC edge method” to determine observations with a high
4 probability of dominant POC contribution, which is believed to be a more accurate method
5 for the identification of the $[OC/EC]_p$ ratio compared to the traditional approach, as discussed
6 by Day et al. (2015), and has also been successfully applied in a number of previous studies
7 (Harrison and Yin, 2008; Lim and Turpin, 2002; Na et al. 2004).

8 9 *2.3. Auxiliary variables*

10 To help better identify the factors resolved by the PMF model, additional parameters,
11 including gaseous pollutants (i.e., CO, NO, NO₂, and O₃) and particulate matter mass
12 concentrations in two size fractions (i.e., PM_{10-2.5} and PM_{2.5}), meteorological parameters (i.e.,
13 temperature, relative humidity, and wind speed), and traffic flow data (counts of LDVs and
14 HDVs), were also included in the model as auxiliary variables. Hourly concentrations of
15 particulate mass and gaseous pollutants together with hourly measurements of meteorological
16 parameters were acquired from the online data base of California Air Resources Board
17 (CARB), for the sampling site located in Downtown Los Angeles (North Main St.),
18 approximately 3 km to the northeast of the PIU. The hourly traffic flow data were acquired
19 from the nearest vehicle detection station (VDS) to our sampling site on the Freeway I-110,
20 operated by the freeway performance measurement system (PeMS), under the
21 California Department of Transportation (CalTrans). Table 1 provides a summary statistics of
22 the input parameters to the PMF model in this study. To achieve the same time resolution
23 across all variables, we calculated hourly-averaged data points for all variables.

24 25 *2.4. Meteorology in central Los Angeles*

26 To evaluate the impact of meteorological conditions on factor contributions as well as
27 to better identify the resolved factors based on their expected seasonal trends, the study
28 period was partitioned into two phases, i.e., colder phase (from November to February) and
29 warmer phase (from August to October as well as March), and the model outputs, except for
30 factor profiles, are presented for each phase accordingly. Figure 1 illustrates the diurnal
31 variation of important meteorological parameters, namely, temperature, RH, wind speed, and
32 solar radiation, in the cold and warm phases. As can be seen from the figure, on average,
33 temperature was 5-7 °C higher in the warm phase than in the cold phase, although the trends

1 were similar in both phases. Minimum temperatures were observed in the early morning
 2 (coinciding with morning rush hours), while maximum temperatures were seen at around
 3 noon. Conversely, RH peaked at night and exhibited a minimum in the early afternoon. RH
 4 was also slightly higher in the warm phase than in the cold phase. As expected, wind speed
 5 peaked in the early afternoon during the warm phase and slightly shifted to the evening in the
 6 cold phase, while the slowest winds were blown during nighttime. The wind speeds were also
 7 higher in the warm phase compared to the cold phase. Solar radiation had a consistent trend
 8 in both phases, peaking at noon, with the levels being higher in the warm phase than in the
 9 cold phase, as one would expect. Similar trends and levels were also observed by
 10 Hasheminassab et al. (2014b) in central LA, indicating the occurrence of stable atmospheric
 11 conditions during nighttime until morning rush hours, especially in colder months of the year.

12

13 2.5. PMF model

14 PMF, first developed by Paatero and Tapper (1993), is a multivariate statistical model
 15 used for identifying and quantifying the contribution of different sources to a set of samples
 16 using the fingerprints of those sources. This multivariate factor analysis tool decomposes a
 17 matrix of speciated data into two sub-matrices, i.e., factor profiles and source contributions,
 18 as shown below (Krecl et al., 2008):

$$19 \quad X = G \cdot F + E \quad (3)$$

20 where, X is the matrix of samples (here, particle number size distribution together with
 21 auxiliary variables data); G is the matrix containing source contributions; F is the matrix
 22 containing factor profiles; and E is the residual matrix.

23 The above equation can also be expressed mathematically, as the following (Norris et
 24 al., 2014):

$$25 \quad x_{ij} = \sum_{k=1}^p g_{ik} f_{kj} + e_{ij} \quad (4)$$

26 where, x_{ij} is the PM number concentration (or concentration of another auxiliary
 27 species) for the i th sample and the j th size bin (or species); p is the number of factors that
 28 contribute to the PM number concentrations; g_{ik} is the relative contribution of k th factor to i th
 29 sample; f_{kj} is the PM number concentration of j th size bin in the k th factor; and e_{ij} is the
 30 residual (observed–estimated) value for the i th sample and j th size bin.

31 With the constraint that no sample can have a significantly negative contribution and
 32 using a least-square method, the PMF then resolves factor profiles and contributions by

1 attempting to minimize the Q value, as shown below (Paatero, 1997; Paatero and Tapper,
2 1994):

$$3 \quad Q = \sum_{i=1}^n \sum_{j=1}^m \left[\frac{x_{ij} - \sum_{k=1}^p g_{ik} \cdot f_{kj}}{u_{ij}} \right]^2 \quad (5)$$

4 where, u_{ij} is the uncertainty associated with the sample x_{ij} .

5 One of the advantages of the PMF model is weighting every single value in the input
6 data matrix using user-provided uncertainties, enabling the model to allow for measurement
7 confidence in resolving the factor profiles and contributions (Paatero et al., 2014). In the
8 present work, since no measurement uncertainties were available for the input parameters, we
9 applied the method suggested by Ogulei et al. (2006a; 2006b) and Zhou et al. (2014) to
10 calculate the uncertainties for individual data points inserted into the model. For this purpose,
11 measurement errors were first estimated for each data point using the following equation:

$$12 \quad \sigma_{ij} = C_1(N_{ij} + \bar{N}_j) \quad (6)$$

13 where, σ_{ij} is the estimated measurement error for the i th sample and j th size bin (or
14 concentration of auxiliary variables); C_1 is an empirical constant usually between 0.01 and
15 0.05; N_{ij} is the observed number concentration for the i th sample and j th size bin (or
16 concentration of auxiliary variables); and \bar{N}_j is the arithmetic mean of the PM number
17 concentrations for the j th size bin (or concentration of auxiliary variables).

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

$$20 \quad S_{ij} = \sigma_{ij} + C_2 \max(|x_{ij}|, |y_{ij}|) \quad (7)$$

21 where, S_{ij} is the calculated uncertainty associated with the i th sample and j th size bin; C_2
22 is an empirical constant usually between 0.1 and 0.5; and Y_{ij} is the value calculated by the
23 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
24 most physically interpretable solution using a trial and error approach.

25 In the present study, the most recent version of the PMF model, version 5.0, newly
26 released by the United States Environmental Protection Agency (PMF guide), was used.
27 Uncertainties associated with the resolved factor profiles were estimated using three error
28 estimation methods, namely, Displacement (DISP) analysis, Bootstraps (BS) method, and a
29 combination of DISP and BS methods (BS-DISP). For the DISP analysis, a solution was
30 considered valid if the observed drop in the Q value was below 0.1% and there was no factor

1 swaps for the smallest dQ_{\max} (i.e., 4). For the BS method, 100 runs were selected and a
2 solution was considered valid if all of the factors had a mapping of above 90%. For the BS-
3 DISP analysis, a solution was considered valid if the observed drop in the Q value was below
4 0.5% (Brown et al., 2015; Paatero et al., 2014).

5 The PMF model was run in the robust mode, which down-weights the effect of values
6 with high uncertainties (i.e., values set as “weak” in the model) on the final solution resolved
7 by the model (Brown et al., 2015). Missing values were replaced by interpolating the
8 previous and the next data points in the matrix; however, to decrease the effect of these
9 replaced values on the final solution, their uncertainty was set as three times the mean
10 uncertainty for that species (that is practically what the model does to set a species as
11 “weak”). Based on the recommendations presented by Brown et al. (2015), genuine zero
12 values were included in the input matrix. Particle number concentration (PNC) was selected
13 as the “total variable”, and the PMF model automatically turned it into a weak species by
14 increasing its uncertainty by a factor of 3. An extra model uncertainty of 5% was also set to
15 account for errors that are not covered in the input uncertainty values (Reff et al., 2007), since
16 the uncertainty matrix only includes the effect of random as well as experimental errors.

17 18 *2.6. Input matrices*

19 The model was run in two different scenarios, one with EC/OC data, which included
20 1053 samples of 131 species, and one without EC/OC data, which included 2976 samples of
21 129 species. This was due mainly to the fact that the EC/OC data were being collected in
22 parallel for a different study that coincided with the current work in a span of time shorter
23 than the entire study period. Therefore, in order to keep the large number of samples from the
24 main study (i.e., 2976) as well as to use the critical advantage of having EC/OC data in the
25 factor identification process, it was decided to run the PMF model in two different scenarios,
26 one including EC/OC data and one without these data. It should also be noted that although
27 the latter matrix contained BC data, this variable was excluded from the former matrix to
28 avoid double counting, as EC was already included in the dataset. The results of the PMF run
29 including the EC/OC data are provided in the supplementary information (Figure S1).

1 **3. Results and discussion**

2 *3.1. Overview of the data*

3 Table 2 presents the statistical characteristics of the species included in the PMF model.
4 In this table, signal-to-noise (S/N) ratio is a parameter that indicates if the variability in the
5 measurements is real or within the data noise. In the current version of the model, i.e., PMF
6 5.0, the method used for calculating the S/N ratio has been updated compared to the previous
7 versions, resolving the disadvantages associated with the previous method of S/N calculation
8 (for a more detailed discussion on the S/N calculation methods, see SI). In the current
9 method, if the resulting S/N ratio is above 1, it can be concluded that the species has a
10 reliable signal. As reported in Table 2, all the species in the input matrix had S/N ratios well
11 above 1, indicating very strong signals for all the variables. Figure S2 also illustrates the
12 correlation between the measured and PMF-predicted total number concentrations for the
13 entire sampling period. As can be seen in the figure, the high correlation between measured
14 and predicted values ($R^2=0.99$) and very close to 1 slope of the regression line indicate that
15 the PMF model has been successful in modeling the input data and apportioning the total PM
16 number concentrations to the resolved factors.

17 Figure 2 depicts the average number and volume size distributions of all the input data
18 to the PMF model by phase, which were collected during the entire study period. As shown in
19 the figure, the vast majority of the particles were smaller than 100 nm, and the number
20 concentration had a mode diameter at around 40 nm. Additionally, a significantly higher
21 number concentration was observed in the cold phase compared to the warm phase, which is
22 consistent with the results from the previous studies conducted in Los Angeles (Hudda et al.,
23 2010;Singh et al., 2006). Regarding volume concentrations, we observed one minor volume
24 mode at the size range of 300-500 nm and a major mode at around 4-6 μm . In this case, the
25 volume concentration was higher in the cold phase than in the warm phase for the minor
26 mode diameter (at 300-500 nm), while a sharper peak was observed for the major mode
27 diameter (at around 4-6 μm) in the warm phase compared to the cold phase. This PM volume
28 size distribution is typical of urban areas (Vu et al., 2015), and is also consistent with the
29 findings of a previous study conducted recently by this research group at the same sampling
30 location (Hasheminassab et al., 2014b).

31

32 *3.2. Number of Factors*

33 In the present work, the PMF model was run several times using different number of
34 factors, input uncertainty matrices (as noted in the methods section), and extra modeling

1 uncertainties to obtain the best and most physically applicable solution. Additionally, we used
2 several criteria to determine the best solution resolved by the model, including: 1) particle
3 number size distribution profiles for different factors; 2) volume size distribution profiles for
4 the resolved factors; 3) profiles of auxiliary variables for different factors; 4) contribution of
5 each factor in different seasons to the total number concentrations; 5) diurnal variations of
6 each of the resolved factors in the cold and warm phases; 6) diurnal variations of each of the
7 resolved factors in weekdays versus weekends; and 7) correlation between auxiliary variables
8 and the relative contribution of each of the resolved factors. The six-factor solution was
9 found to present the most physically explainable one, and was, therefore, chosen as the final
10 solution. When the model was run with one less factor (i.e., 5-factor solution), the model
11 could not distinguish the two traffic factors, and Traffic 1 and Traffic 2 factors were merged
12 together. On the other hand, when the model was run with one more factor (i.e., 7-factor
13 solution), a new factor was resolved by the model, having a mode diameter between that of
14 “urban background aerosol” and “secondary aerosol”, but without having any distinct diurnal,
15 seasonal, or weekday/weekend trends or auxiliary variables profile. Therefore, this factor
16 could not be meaningfully interpreted and identified, prompting us to choose the 6-factor as
17 the optimal solution.

18 Figure 3 illustrates the number size distributions as well as the auxiliary variables
19 profiles for each of the factors resolved by the PMF. Figure 4 indicates volume size
20 distribution of each factor. In Figures 3, 4, and S1, the black solid lines represent absolute
21 concentrations (number or volume) of each size bin and should be read from the left Y axis,
22 while the grey triangles represent the explained variation of each size bin and should be read
23 from the right Y axis. The relative contributions (overall, and by cold or warm phases) of
24 each factor to the total number concentrations are shown in Figure 5. Figure 6 illustrates the
25 contribution (particles/cm³) of each of the PMF-resolved factors to the total number
26 concentrations in the cold and warm phases within box and whisker plot. The diurnal
27 variations and the weekday/weekend trends (geometric means) for each of the factors are
28 illustrated in Figures 7 and 8, respectively. The spearman correlation coefficient matrix
29 indicating the association between the auxiliary variables and the factors resolved by the
30 PMF model is also presented in Table 3.

31

32 3.3. Factor identification

33 *Factor 1:* Factor 1 has a number mode at <20 nm, a volume mode at <20 nm, and
34 contributes 17.3% (11.7-24%) to the total number concentrations (Figures 3, 4, and 5). This

1 factor has strong positive (except for RH, with which this factor has negative correlation)
2 associations with temperature, wind speed, SOC, and O₃ (Table 3), which are also
3 statistically significant (p<0.05). These associations are also apparent from high loadings of
4 temperature, RH, wind speed, and O₃ in the auxiliary variables profile (Figures 3 and S1).
5 The contribution of this factor to the total number concentration was also higher in the warm
6 phase than in the cold phase, when higher temperatures, wind speeds, and solar radiation are
7 observed (Figure 1); this was the case both in terms of percent contribution (24% in the warm
8 phase vs. 11.7% in the cold phase) and number concentration (589±25 particles/cm³ in the
9 cold phase vs. 1153±28 particles/cm³ in the warm phase) (Figures 5 and 6 and Table S1). The
10 diurnal variations for this factor also revealed a sharp peak in the afternoon (2-6 PM) (Figure
11 7), which coincides with very high temperatures, wind speeds, and solar radiation as well as
12 with minimum RH (Figure 1). **A minor peak was also observed during morning rush hours**
13 **(6-8 am), which suggests the partial influence from traffic sources, as also observed by**
14 **loadings of HDV and LDV in this factor (Figure 3).** However, there was no significant
15 distinction in the diurnal variation patterns of this factor in weekdays compared to weekends
16 (Figure 8).

17 The above characteristics are all typical of a "nucleation" factor, during which new
18 particles are formed via photochemical events under high temperatures, high wind speeds,
19 and low RH (Beddows et al., 2015; Brines et al., 2015; Dall'Osto et al., 2012; Vu et al., 2015).
20 **The minor peak in the early morning can also be explained by the cooling, following dilution,**
21 **of vehicular exhaust emissions, which leads to the partitioning of semi-volatile exhaust gases**
22 **into the particle phase; this process is further enhanced by to the lower temperatures during**
23 **that time of day (Harrison et al. 2011; Ntziachristos et al. 2007; Janhall et al. 2004; Charron**
24 **and Harrison, 2003).** Our findings are most specifically consistent with those of the study of
25 Brines et al. (2015), in which the authors had reported nucleation as one of the major sources
26 of UFPs in five high-insolation cities, including Los Angeles using the data obtained from the
27 same sampling location. **They observed very similar diurnal variation for nucleation, with a**
28 **minor peak in the early morning and a major peak in early afternoon at the same sampling**
29 **location in Los Angeles.**

30

31 *Factor 2:* Factor 2 is mostly represented by particles at 20-40 nm and contributes about
32 40% (33.2-43.4%) to the total number concentration (Figures 3 and 5). It also has a volume
33 concentration peak at around 30-40 nm (Figure 4). Judging by the loadings presented in the
34 auxiliary variables profile (Figures 3) and correlation coefficients presented in Table 3, this

1 factor has clear associations with gaseous pollutants (e.g., CO, NO, and NO₂), BC, EC, and
2 POC (from the scenario containing EC/OC data (Figure S1), which themselves are indicators
3 of vehicular emissions (Gu et al., 2011;Ogulei et al., 2006b). In addition, high species
4 loadings (Figure 3) and correlation coefficients (Table 3) of LDV and HDV counts can also
5 be observed for this factor, indicating the influence of nearby passing traffic on this factor.
6 The contribution of this factor to the total number concentration was also much higher in the
7 cold phase than in the warm phase, when lower temperatures, wind speeds, and solar
8 radiation (Figure 1) lead to increased atmospheric stability and lower mixing height
9 (Hasheminassab et al., 2014a); this was the case both in terms of percent contribution (43.4%
10 in the cold phase vs. 33.2% in the warm phase) and number concentration (3166±66
11 particles/cm³ in the cold phase vs. 1201±61 particles/cm³ in the warm phase) (Figures 5 and 6
12 and Table S1). The diurnal variations also revealed a distinctive pattern peaking in the
13 morning rush hours (around 7-8 AM) (Fig. 7). The weekday/weekend analysis also indicated
14 that this factor had higher contributions during the weekdays compared to the weekends
15 (Figure 8). Therefore, this factor can be attributed to traffic tailpipe emissions. Previous
16 source apportionment studies on number size distributions have also associated such
17 characteristics with fresh vehicular emissions (Beddows et al., 2015;Dall'Osto et al., 2012;Vu
18 et al., 2015). This factor is denoted as “traffic 1”, given that another factor attributed to traffic
19 emissions was resolved, which will be discussed in the following section. The characteristics
20 of this traffic factor are in agreement with what Brines et al. (2015) reported for five high-
21 insolation cities, including Los Angeles.

22
23 *Factor 3:* This factor has a major peak in the Aitken mode (60-100 nm) and contributes
24 27.5% (25-27.6%) to the total number concentration (Figures 3 and 5). It also exhibited a
25 volume concentration peak at around 100 nm (Figure 4). Judging by the loadings presented in
26 the auxiliary variables profile (Figures 3 and S1) and correlation coefficients presented in
27 Table 3, significant associations can be observed between this factor and gaseous pollutants
28 (e.g., CO, NO, and NO₂), as well as with BC (and EC from the scenario containing EC/OC
29 data (Fig. S1)). Although weaker than those of Factor 2, there are significant positive
30 associations between this factor and LDV and HDV counts (Figure 3 and Table 3),
31 suggesting the likely influence of nearby passing traffic. This factor also had a significantly
32 higher contribution to the total number concentrations in the cold phase than in the warm
33 phase (an average of 1755±56 particles/cm³ in the cold phase vs. 1059±43 particles/cm³ in
34 the warm phase (Figure 6)), in spite of the fact that its percent contribution to the total PM

1 number concentrations was comparable in both phases, and slightly higher in the warm phase
2 (25% in the cold phase vs. 27.6% in the warm phase (Figure 5)). This is due mainly to the
3 fact that the contribution of the "traffic 1" factor is so large in the cold phase that has
4 significantly obscured the percent contribution of other factors in this phase, even though
5 their absolute contributions in terms of total number concentrations were higher in the cold
6 phase.

7 The diurnal variations for this factor also indicated clear peaks during the morning rush
8 hours (6-8 am) in both phases, along with another peak at late night during the cold phase,
9 most likely due to the stagnant atmospheric conditions during this time of the year, which
10 traps the emissions in lower altitudes (Figure 7). **This diurnal profile suggests a major
11 contribution from semi-volatile compounds in the atmosphere, particularly in the cold phase,
12 as reflected in the substantial increase at nighttime.** The weekday/weekend analysis for this
13 factor revealed larger contributions in weekdays than in weekends, especially during daytime
14 hours. The slightly higher nighttime contribution of this factor in the weekends compared to
15 the weekdays can be attributed to the larger number of within-city travels being made on
16 holiday nights. These levels and trends are, overall, suggestive of emissions from vehicular
17 sources. However, the larger size range of this factor compared to factor 2, combined with the
18 involvement of EC and SOC (as observed from the scenario containing EC/OC data (Fig.
19 S1)) as well as BC, suggest that although this factor also originates from "traffic", the
20 particles are "older" (i.e., more aged) than those observed in factor 2 and are mostly in the
21 Aitken and Accumulation modes; therefore, it was labeled as "traffic 2". This finding is also
22 consistent with those of the previous studies (e.g., Brines et al. (2015)), in which the authors
23 detected distinct traffic factors (with a collective relative contribution of approximately 60%
24 in Los Angeles at the same sampling site) using a different source apportionment method,
25 named k-means cluster analysis. It should be noted that it is quite common in source
26 apportionment studies performed on size-segregated PM number concentrations to detect
27 more than one traffic factors, due primarily to the fact that particle sizes may change, as
28 particles undergo processes including agglomeration as well as evaporation or condensation
29 of semi-volatile species from- or onto their surface following their release in the atmosphere
30 (Harrison et al., 2016;Kim et al., 2004;Zhou et al., 2005).

31 It is also noteworthy that the traffic 2 factor has a slightly higher HDV loading than
32 traffic 1 factor (Figures 3 and S1). It also has a somewhat stronger positive correlation with
33 HDV ($R=0.43$) than with LDV ($R=0.41$), while the traffic 1 factor has a stronger correlation
34 with LDV ($R=0.69$) than with HDV ($R=0.52$). Additionally, the stronger correlation of traffic

1 2 factor with EC and BC compared to traffic 1 leads us to the hypothesis that HDV might be
2 contributing more to this factor than LDV is. Vu et al. (2015) have also suggested that
3 observing a number concentration mode at the size range of 60-100 nm can be a result of
4 incomplete combustion of diesel fuel, consisting of pyrolytic EC and OC. Other studies have
5 also found two particle modes, or factors, for traffic-related emissions. Although the
6 emissions in both of these two modes are believed to come from the same fleet of vehicles,
7 they have different formation mechanisms and chemistry, with particles associated with the
8 second mode (i.e., soot mode) assumed to have an elemental carbon core. This is consistent
9 with the findings of the present study, judging by the mode diameter and high loading of BC,
10 EC, and OC in the traffic 2 factor (Figures 3 and S1) and the strong correlation of this factor
11 with BC, EC, and OC (Table 3). Additionally, studies have indicated that a fraction of diesel
12 PM emissions, which is generally in the range of 50-200 nm, comprises particles that have an
13 elemental core, with low-vapor-pressure hydrocarbons and sulfur compounds being adsorbed
14 on their surface (Burtcher, 2005). Therefore, it might be likely that this factor is representing
15 a higher contribution of HDV emissions, although stronger evidence is required to confirm
16 this hypothesis.

17

18 *Factor 4:* Factor 4, which contributes 12.2% (7.4-17.1%) to the total number
19 concentration, is represented by a number mode at around 220 nm and a volume mode at
20 around 250 nm (Figures 3, 4, and 5). The profile for the auxiliary variables also indicates
21 high loadings for gaseous pollutants (e.g., CO, NO, and NO₂) and BC (Figure 3) as well as
22 for EC and SOC (when the PMF model was run with the EC/OC data (Fig. S1)). The large
23 correlation coefficients of this factor with the aforementioned species also confirm its strong
24 association with these parameters (Table 3). The lower-than-unity NO/NO₂ ratio for this
25 factor also suggests that these particles are aged compared to the newly formed particles (Liu
26 et al., 2014). This is also supported by the stronger positive correlation of this factor with
27 SOC than with POC, suggesting the fact that this factor is not coming from direct emissions
28 and has most likely undergone processes and reactions in the atmosphere. As can be inferred
29 from Figures 5 and 6, the contribution of this factor is significantly higher in the cold phase
30 than in the warm phase, both in terms of percent contribution (17.1% in the cold phase vs.
31 7.4% in the warm phase) and the absolute contribution to the total number concentration
32 (1200±41 particles/cm³ in the cold phase vs. 284±23 particles/cm³ in the warm phase). As
33 seen in Figure 7, the diurnal variations for this factor also exhibit a clear peak during morning
34 hours, which indicates higher concentrations when atmosphere is more stable and wind

1 speeds are low, especially in the cold phase when these conditions are even more intense
2 (Figure 1). The weekday/weekend analysis also revealed a slightly elevated contribution of
3 this factor to the total number concentrations during morning rush hours, especially during
4 the weekdays, suggesting the small influence of traffic emissions on this factor. Previous
5 studies have indicated that these are characteristics of the “urban background aerosol”, as
6 suggested by (Beddows et al., 2015;Dall'Osto et al., 2012).

7
8 *Factor 5:* Factor 5 has a number and volume mode at around 500 nm and a minor
9 number mode at 50 nm (looking at the black dots, representing the explained variations)
10 (Figures 3 and 4). This factor contributes 2.1% (1.5-2.5%) to the total number concentration
11 (Figure 5). It is also associated with high loadings of PM_{2.5} mass concentration (i.e., major
12 contributor to PM_{2.5} mass), NO, NO₂, Temperature, RH (Figure 3), and SOC (as observed
13 from the scenario containing EC/OC data (Figure S1)). This is also supported by the results
14 of the correlation analysis presented in Table 3, indicating that this factor has strong positive
15 correlations with PM_{2.5}, NO, NO₂, Temperature, RH, and SOC. The overall small
16 contribution of this factor to the total number concentration was slightly higher in the cold
17 phase than in the warm phase; this was the case both in terms of percent contribution (2.5%
18 in the cold phase vs. 1.5% in the warm phase) and number concentration (111±11
19 particles/cm³ in the cold phase vs. 100±5 particles/cm³ in the warm phase) (Figures 5 and 6
20 and Table S1). The diurnal variation for this factor also reveals a significant increase during
21 nighttime, especially during the cold phase (Figure 7). However, the weekday/weekend
22 analysis did not reveal any distinctive trend pertaining to the day of the week for this factor
23 (Figure 8). These pieces of evidence point to “secondary aerosols” as the most appropriate
24 title for this factor, which is consistent with the results of previous PMF studies both on
25 number size distributions and chemical speciation data (Beddows et al., 2015;Hasheminassab
26 et al., 2014a). Table 3 indicates a much higher correlation of this factor with SOC than POC
27 (R values of 0.5 and 0.2, respectively). The association of this factor with RH and
28 temperature, along with its higher contribution to particle number during the cold phase,
29 particularly at night, support the hypothesis that this factor likely represents the fraction of
30 aerosols produced by secondary reactions on a regional scale, including ammonium nitrate
31 (whose partitioning in the PM phase increases with decreasing temperature and increased
32 RH), but also secondary organic aerosols from nighttime and/or aqueous phase reactions, as
33 indicated in earlier studies in this area (Hersey et al., 2011;Venkatachari et al., 2005). In a
34 previous source apportionment study on PM_{2.5} chemical speciation data in downtown Los

1 Angeles, we also found a similar factor profile, representing a mixture of secondary
2 components (dominated by secondary nitrate and SOC) with higher contribution during the
3 cold season (Hasheminassab et al., 2014a). Moreover, previous studies have shown that
4 secondary organic aerosol formed at nighttime together with ammonium nitrate are major
5 contributors to the mass concentrations of PM_{2.5}, which was also observed in the present
6 work from the high loading of PM_{2.5} mass concentration in this profile (Figure 3)
7 (Hasheminassab et al., 2014a;Arhami et al., 2010;Saffari et al., 2016).

8
9 *Factor 6:* Factor 6 is dominated by particles at around 1 μm and above (Figure 3). This
10 factor also had a volume mode at > 1 μm (Figure 4). Although this factor contributes only
11 1.1% (0.2-6.3%) to the total number concentration (Figure 5), it is associated with high
12 loadings of coarse PM and PM_{2.5} (great contributor to mass) (Figure 3). In addition, high
13 loadings of temperature and wind speed were observed for this factor (Figure 3). Table 3 also
14 indicates strong correlation of this factor with coarse PM, PM_{2.5}, temperature, and wind
15 speed. The contribution of this factor to the total number concentration was also higher in the
16 warm phase than in the cold phase, both in terms of percent contribution (0.2% in the cold
17 phase vs. 6.3% in the warm phase) and number concentration (14±1 particles/cm³ in the cold
18 phase vs. 243±3 particles/cm³ in the warm phase) (Figures 5 and 6 and Table S1). The diurnal
19 variations for this factor exhibited significantly higher contributions during daytime,
20 especially in the warm phase (Figure 7), when atmosphere is unstable, wind speed is high,
21 and the mixing height is at its maximum (Figure 1). However, the weekday/weekend analysis
22 did not reveal any distinctive trend pertaining to the day of the week for this factor (Figure 8).
23 Based on all of the abovementioned characteristics, this factor was named “soil/road dust”
24 (Gietl et al., 2010;Harrison and Booker, 2001;Harrison et al., 2012). This is also quite
25 consistent with the findings of the study of Hasheminassab et al. (2014a), in which the
26 authors apportioned the sources of ambient fine particulate matter across the state of
27 California. In that study, the authors observed a lower contribution of the soil factor to
28 particle mass concentrations in the northern regions of the state of California, mainly because
29 of higher RH and increased precipitation that inhibit the re-suspension of soil due to strong
30 winds (Harrison and Booker, 2001). In the present study, similarly, the contribution of this
31 factor was higher in the warm phase, when higher temperatures and wind speeds facilitate the
32 re-suspension of soil and dust (Figure 1).

1 **4. Summary and conclusions**

2 The present study was the first attempt to characterize major sources of PM number
3 concentrations and quantify their contributions using the PMF receptor model applied on PM
4 number size distributions in the range of 13 nm to 10 μm combined with several auxiliary
5 variables, including BC, EC/OC, PM mass, gaseous pollutants, meteorological, and traffic
6 flow data, in central Los Angeles. The six-factor solution was found to be the most physically
7 applicable solution for the input data: nucleation, traffic 1, traffic 2, urban background
8 aerosol, secondary aerosol, and soil. Traffic sources (1 and 2) were the major contributor to
9 PM number concentrations, making up to above 60% of the total number concentrations
10 combined, with larger contributions in the cold phase compared to the warm phase, when
11 lower temperatures, wind speeds, and solar radiation lead to increased atmospheric stability
12 and lower mixing height. The contribution of traffic factors was largest during morning and
13 afternoon rush hours; it was also higher in the weekdays compared to the weekends, as
14 expected. In agreement with the findings of previous studies in Los Angeles, nucleation was
15 another major factor contributing to the total number concentrations (17%), having a larger
16 contribution in the warm phase than in the cold phase. The diurnal variations for this factor
17 also revealed a sharp peak in the afternoon (2-6 PM), which coincides with high
18 temperatures, wind speeds, and solar radiation as well as with minimum RH, providing ideal
19 conditions for the occurrence of photochemical nucleation processes, especially during
20 warmer seasons. Urban background aerosol, secondary aerosol, and soil, with relative
21 contributions of approximately 12%, 2.1%, and 1.1%, respectively, overall accounted for
22 approximately 15% of PM number concentrations. However, these factors dominated the PM
23 volume and mass concentrations, due mainly to their larger mode diameters.

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28 29 **References**

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1 Table 1. Summary of the input parameters to the PMF model in this study.

Parameter	Source of data	Time resolution in original data set
EC, OC	Sunset monitor	1 hr
Size Distribution (14-760 nm)	SMPS	5 min
Size Distribution (0.3-10 μm)	OPS	5 min
BC	Aethalometer	15 min
PM mass concentration data ($\text{PM}_{10-2.5}$, $\text{PM}_{2.5}$)	CARB	1 hr
Gaseous Pollutants (NO , NO_2 , CO , O_3 , SO_2)	CARB	1 hr
Meteorological data (T, RH, WS)	CARB	1 hr
Traffic data (counts of LDV and HDV)	PeMS	1 hr

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Table 2. Summary statistics for the parameters included in the PMF model.

Species	Geometric Mean	Standard Error	Min	Max	S/N ratio
Total number concentration (#/cm ⁻³)	6860.00	94.10	524.00	32400.00	7.00
PM _{10-2.5} (µg/m ³)	15.90	0.19	2.00	77.00	7.00
PM _{2.5} (µg/m ³)	14.50	0.23	1.00	101.00	6.90
CO (ppm)	0.58	0.01	0.10	2.19	7.10
NO (ppb)	8.46	0.57	1.00	212.00	5.80
NO ₂ (ppb)	22.50	0.23	1.90	75.00	7.10
O ₃ (ppb)	17.40	0.33	2.00	105.00	6.80
BC (µg/m ³)	1.14	0.02	0.124	9.13	6.90
POC*(µg/m ³)	2.20	0.08	0.10	19.20	6.80
SOC* (µg/m ³)	2.13	0.05	0.04	16.30	7.10
EC* (µg/m ³)	1.01	0.03	0.01	7.34	8.80
RH (%)	50.40	0.40	6.00	99.00	7.10
Temperature (°C)	18.80	0.13	3.89	38.33	7.30
Wind speed (m/s)	4.03	0.04	1.00	14.00	6.80
LDV (#/h)	3790	34	691	7620	7.10
HDV (#/h)	153	3	5	920	6.80

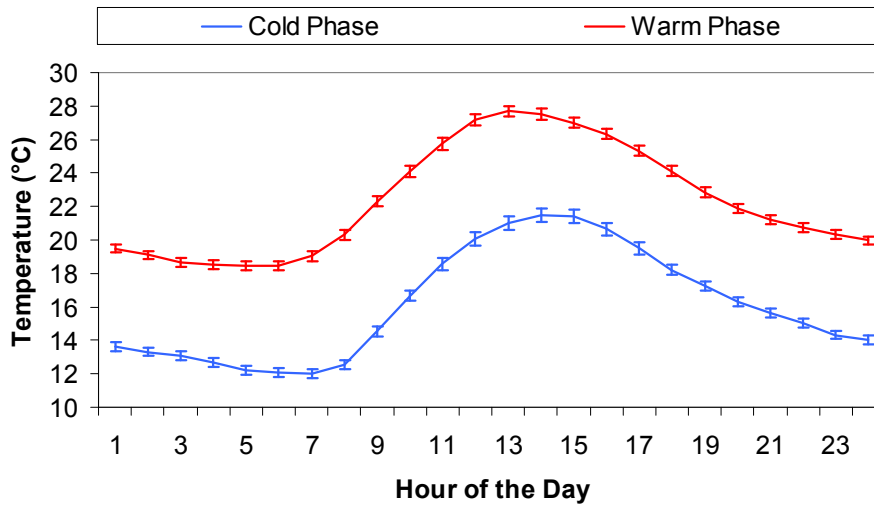
*Values are pertaining to the runs including EC/OC data.

1 Table 3. Spearman correlation coefficient matrix indicating the association between the
 2 auxiliary variables and the factors resolved by the PMF model. R values above 0.5 are
 3 bolded.
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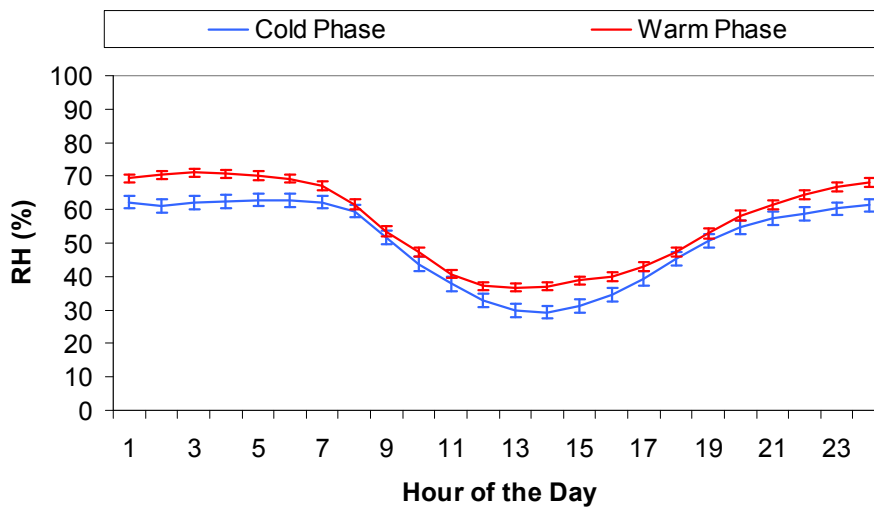
Species	Nucleation	Traffic 1	Traffic 2	Urban Background Aerosol	Secondary Aerosol	Soil/Road Dust
PM _{10-2.5}	0.17*	0.21*	0.35*	0.27*	0.09*	0.39*
PM _{2.5}	-0.24*	-0.09*	0.05*	0.33*	0.69*	0.23*
CO	0.04	0.41*	0.58*	0.64*	0.17*	0.28*
NO	-0.01	0.48*	0.59*	0.52*	0.27*	0.24*
NO ₂	0.08*	0.50*	0.60*	0.57*	0.33*	0.14*
O ₃	0.57*	0.34*	0.40*	-0.35*	0.46*	0.19*
BC	0.01	0.53*	0.70*	0.71*	0.13*	0.22*
POC	0.09*	0.62*	0.28*	0.30*	0.24*	0.29*
SOC	0.46*	0.12*	0.43*	0.58*	0.46*	0.20*
EC	0.17*	0.47*	0.56*	0.60*	0.20*	0.17*
RH	-0.26*	-0.32*	-0.30*	-0.05*	0.43*	0.33*
Temp	0.52*	-0.23*	-0.18*	-0.39*	0.34*	0.47*
WS	0.57*	0.00	0.07*	-0.04*	-0.25*	0.62*
LDV	0.22*	0.70*	0.42*	0.05*	0.01	0.02
HDV	0.23*	0.52	0.43*	-0.08*	-0.12*	-0.01

5 * Indicates R values that are statistically significant (P<0.05).
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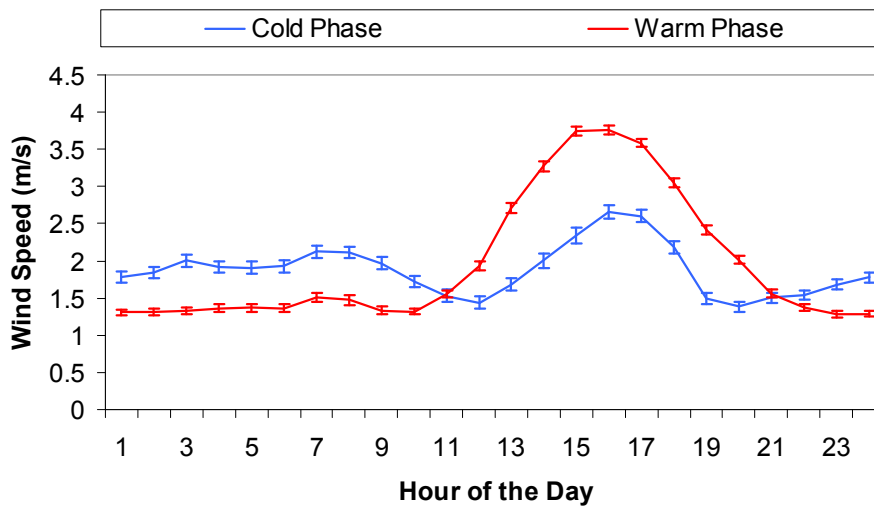
1 Figure 1. Diurnal variations of important meteorological parameters in the cold and warm phases. Error bars correspond to one standard error.
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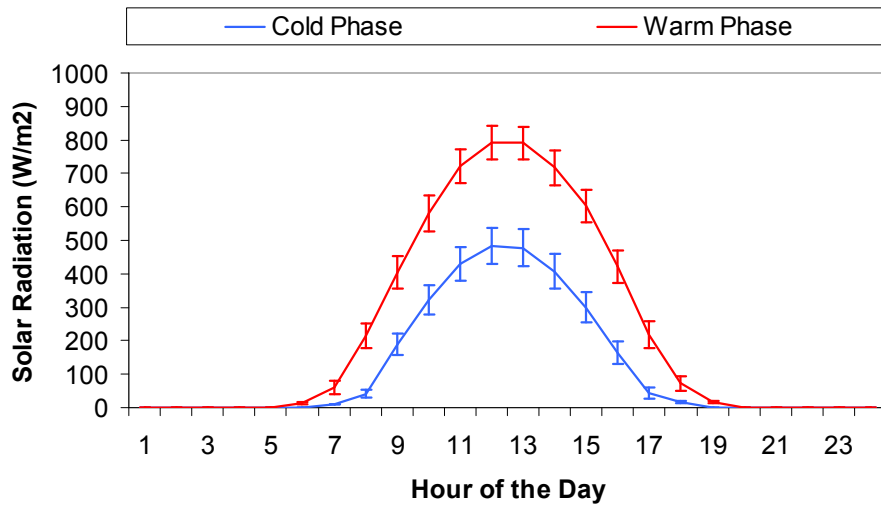
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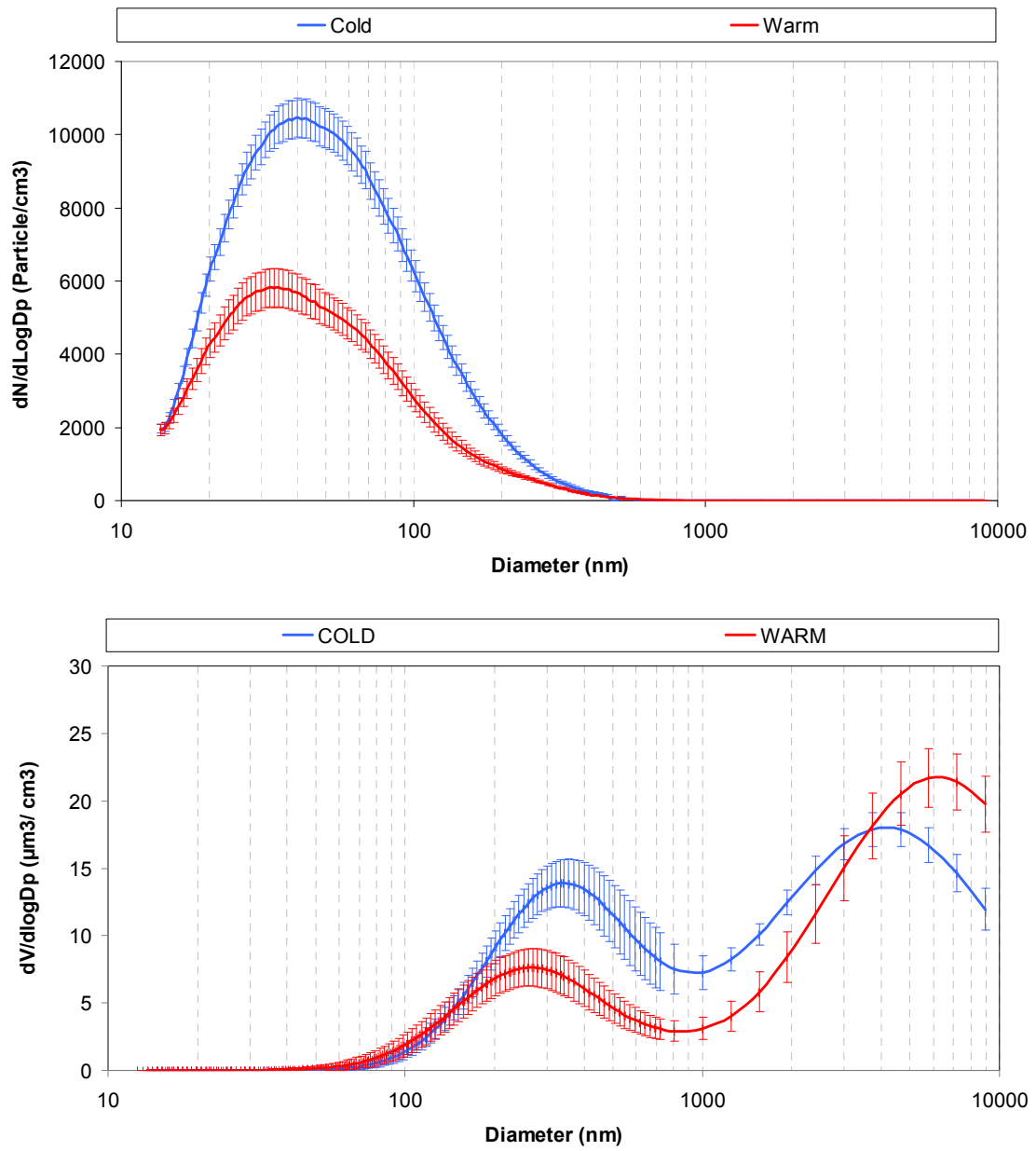


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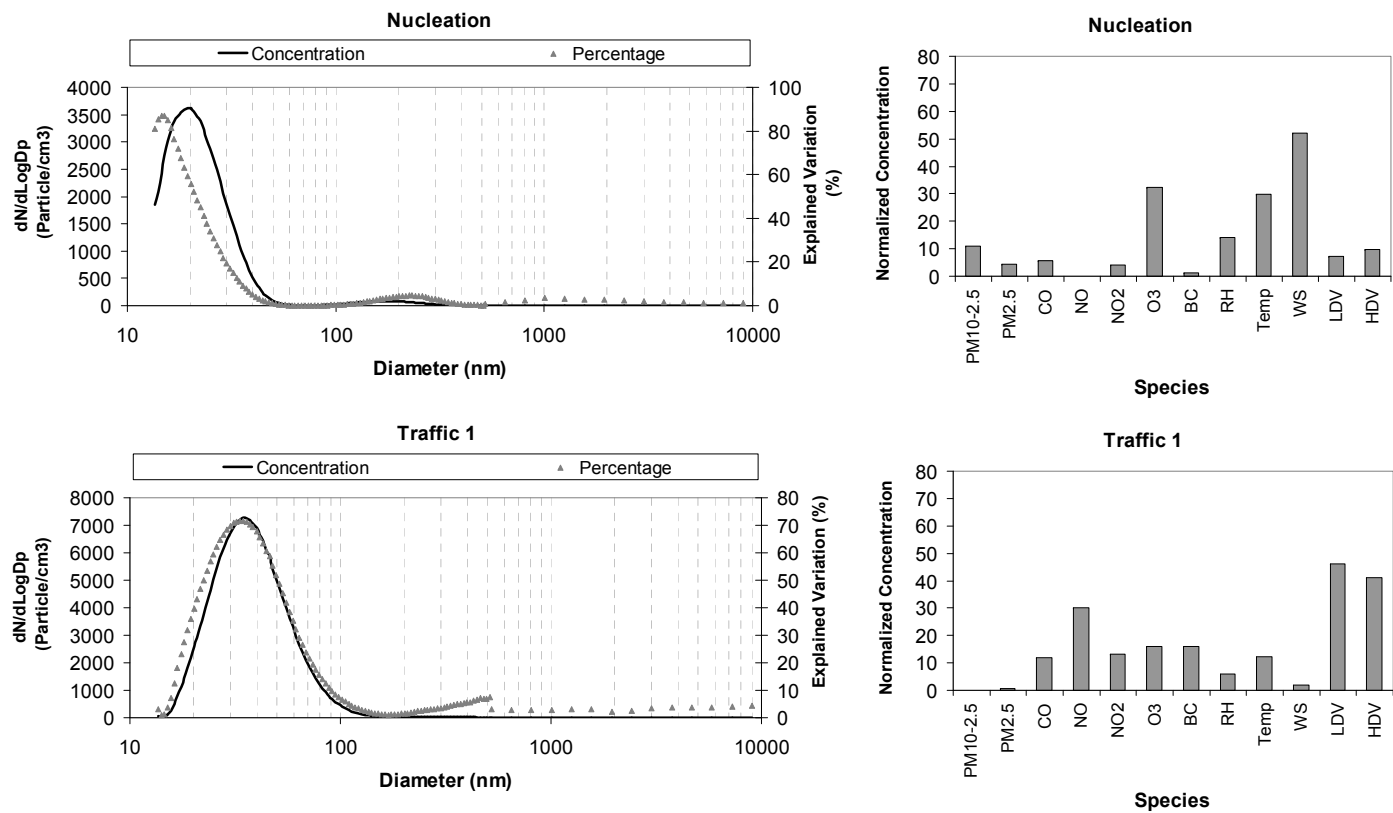
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1 Figure 2. Average number and volume size distributions of all the input samples to the PMF
2 model in the cold and warm phases (the graphs represent geometric means \pm SE).



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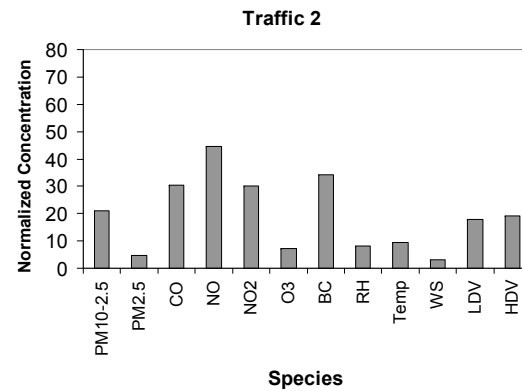
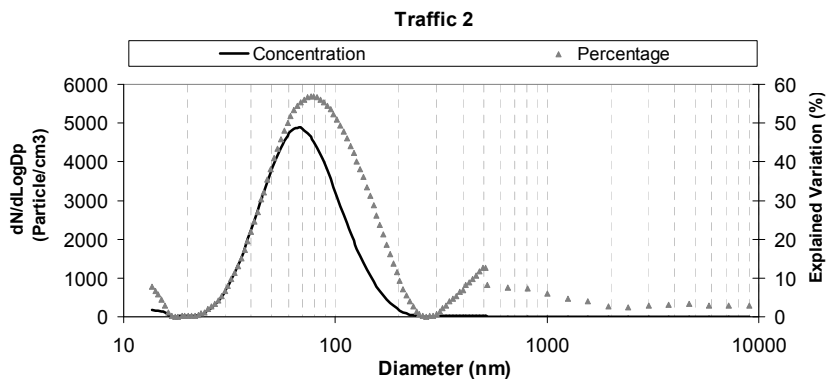
1 Figure 3. The number size distributions as well as the auxiliary variables profiles for each of the factors resolved by the PMF model.



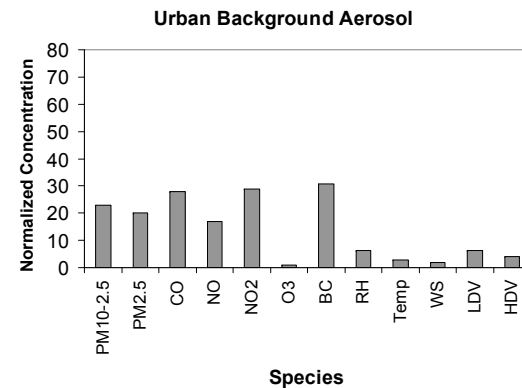
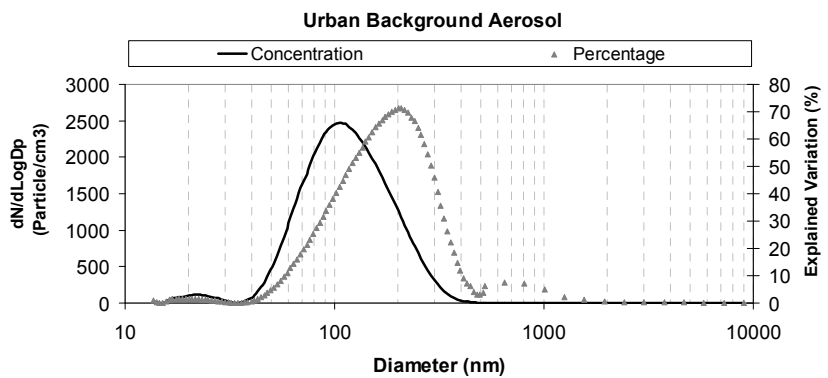
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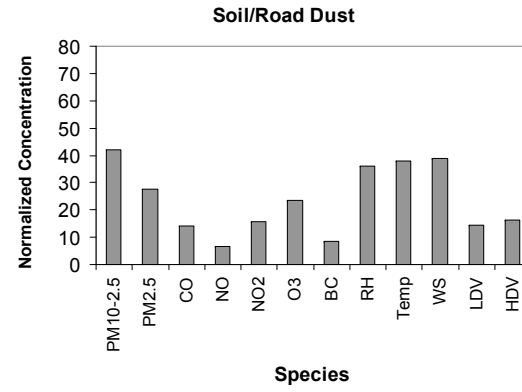
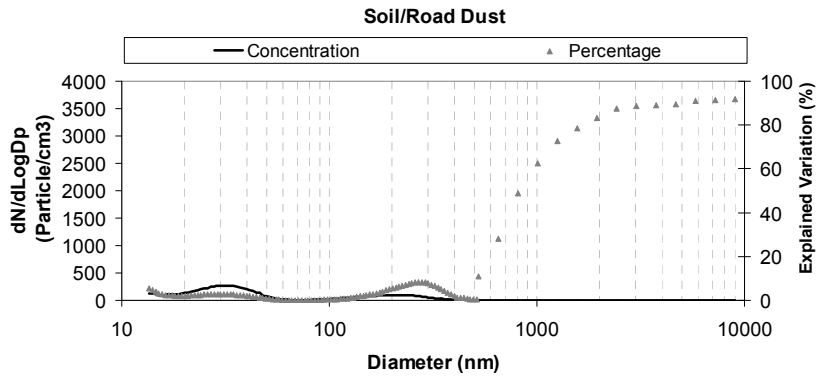
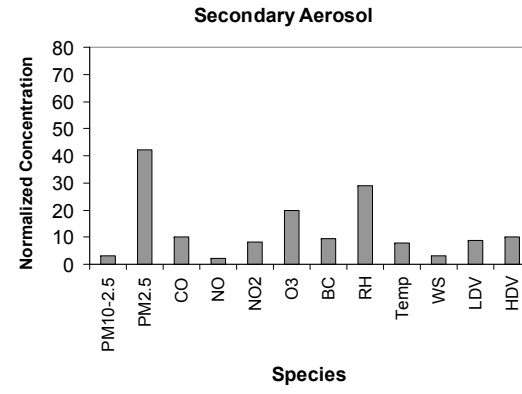
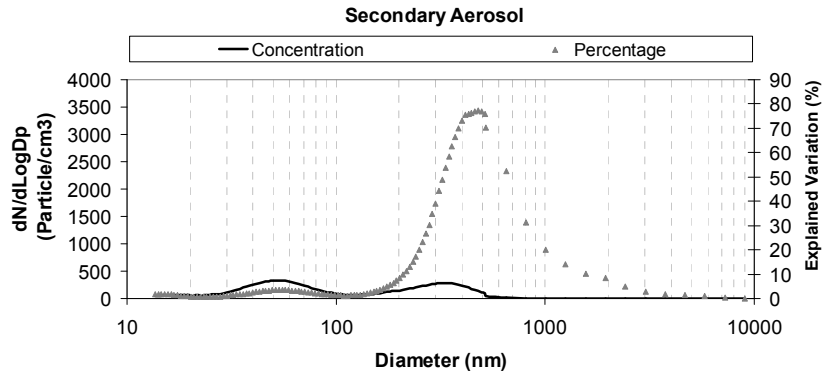
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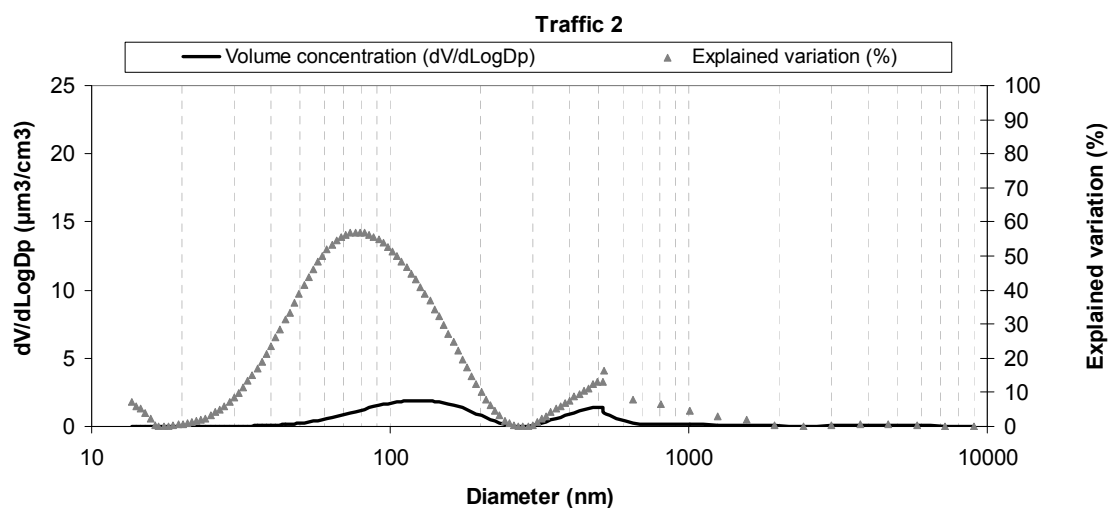
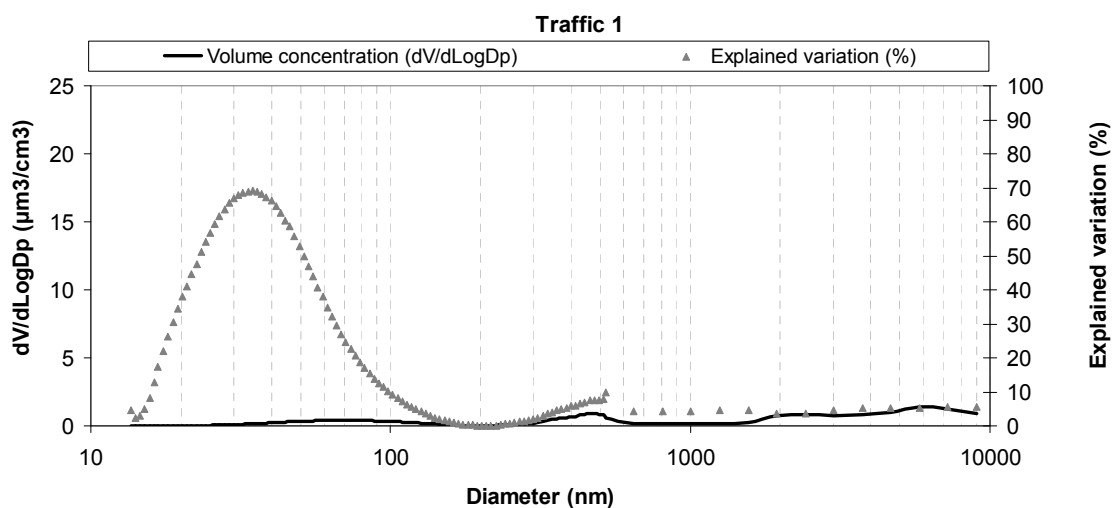
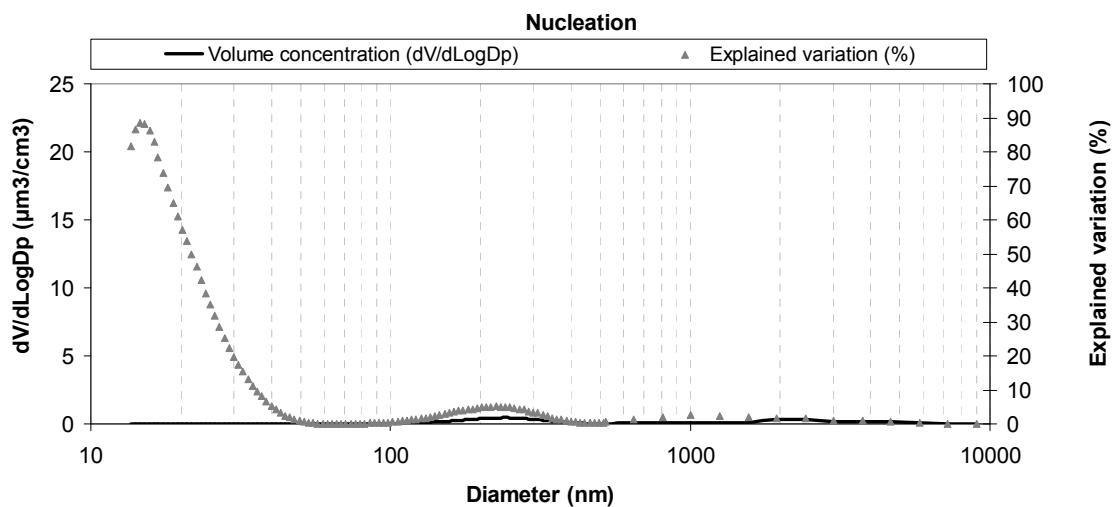
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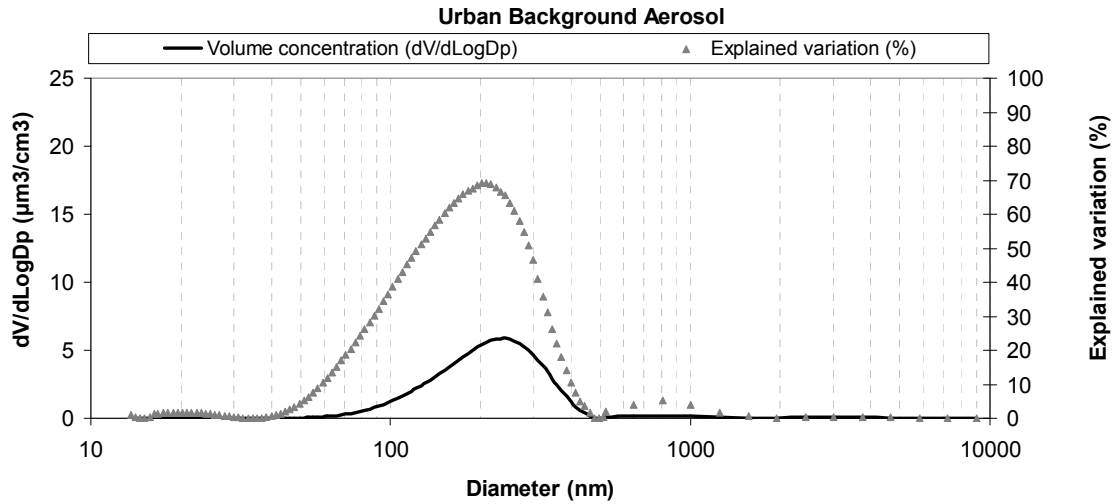
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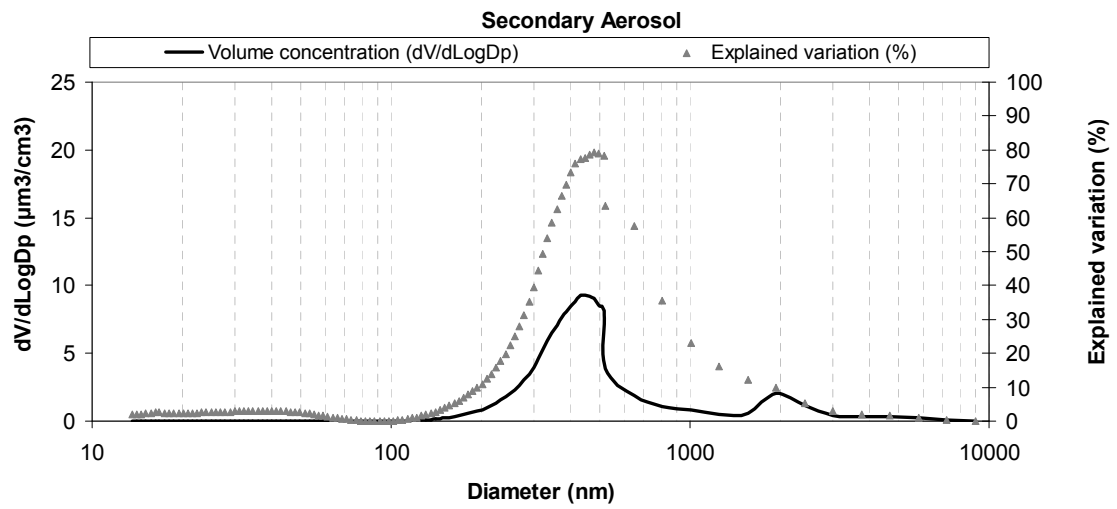
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1 Figure 4. Volume size distributions along with the explained variation (%) of each factor
2 profile resolved by the PMF model.

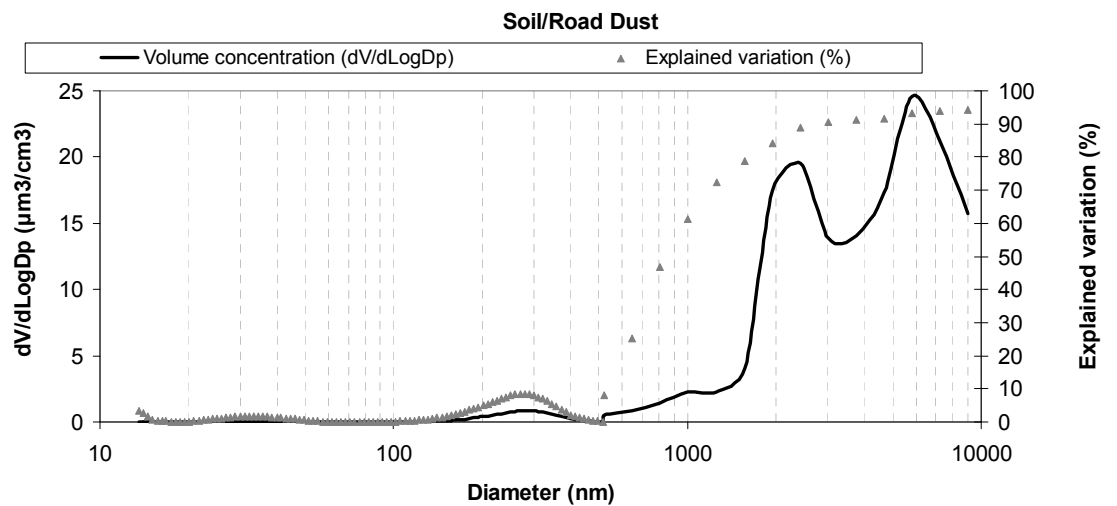




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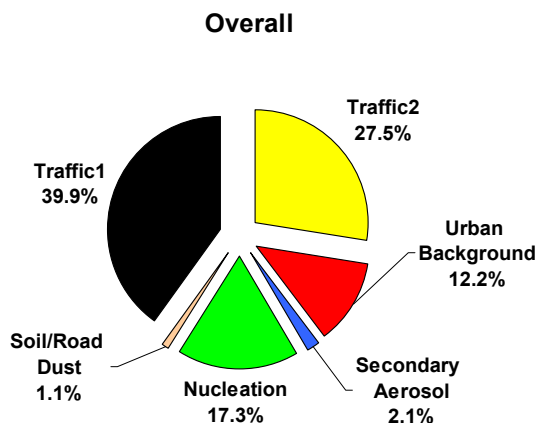
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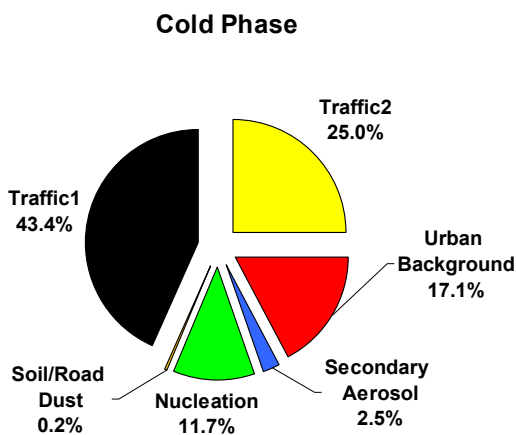
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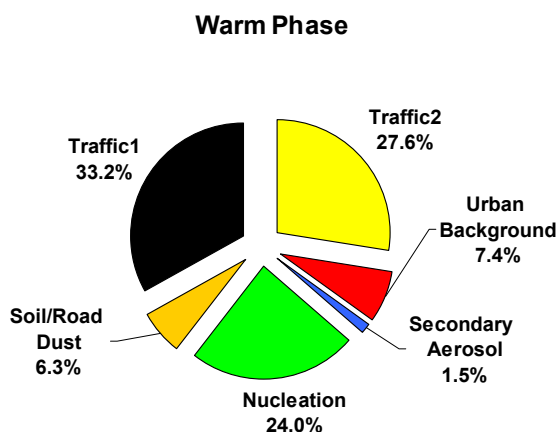
1 Figure 5. Relative contribution of each factor to the total number concentrations: a) overall
2 phases; b) cold phase; and c) warm phase.



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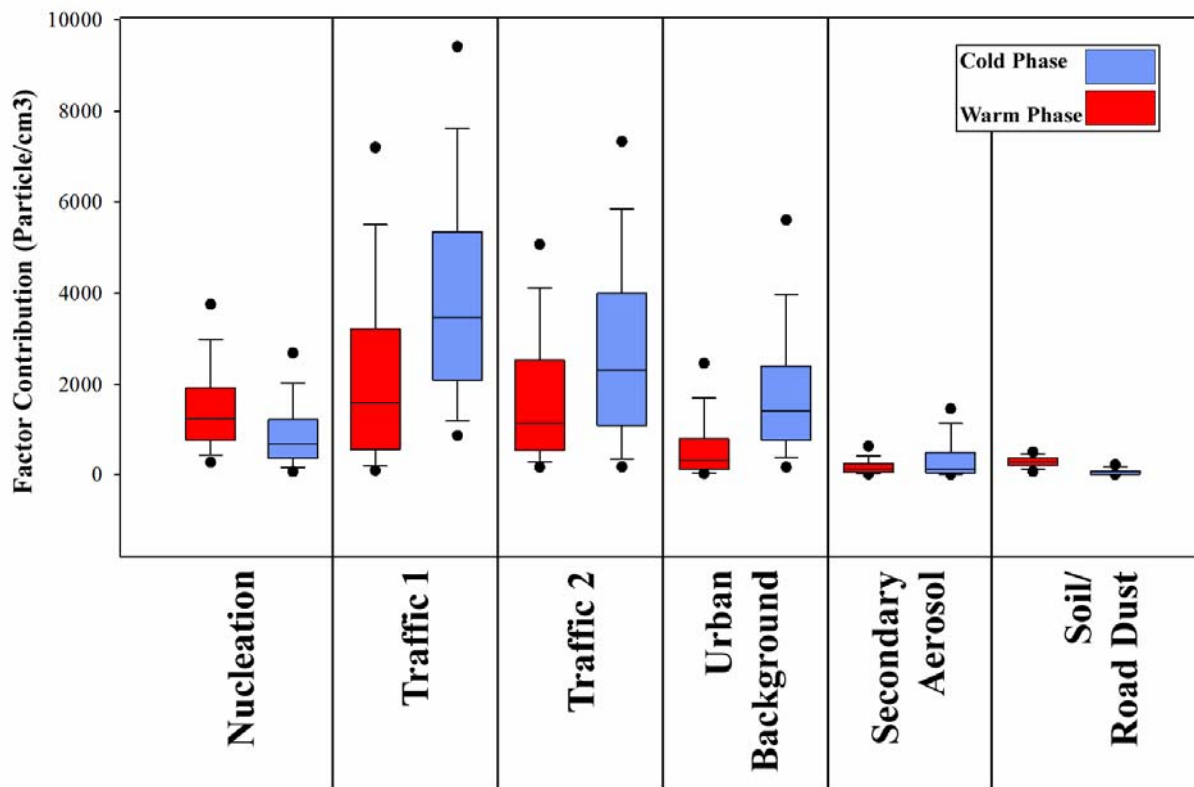


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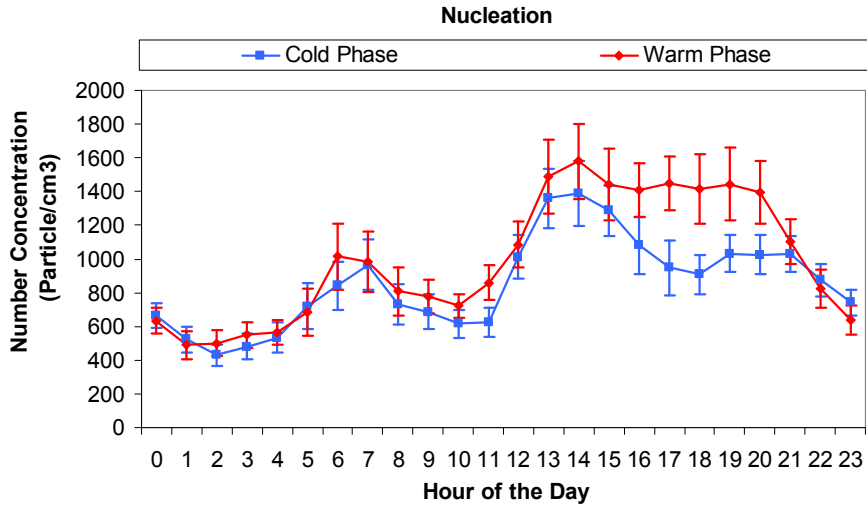
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1 Figure 6. Contribution (particles/cm³) of each of the PMF-resolved factors to the total number
 2 concentrations in the cold and warm phases.

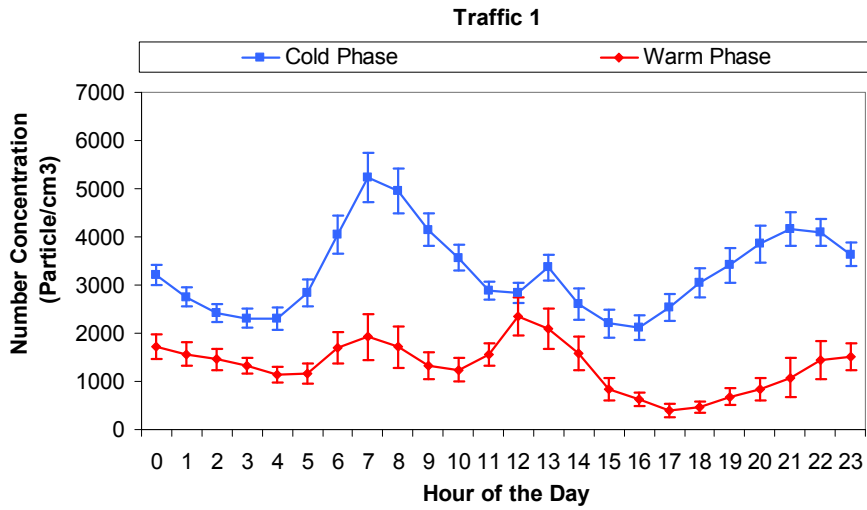


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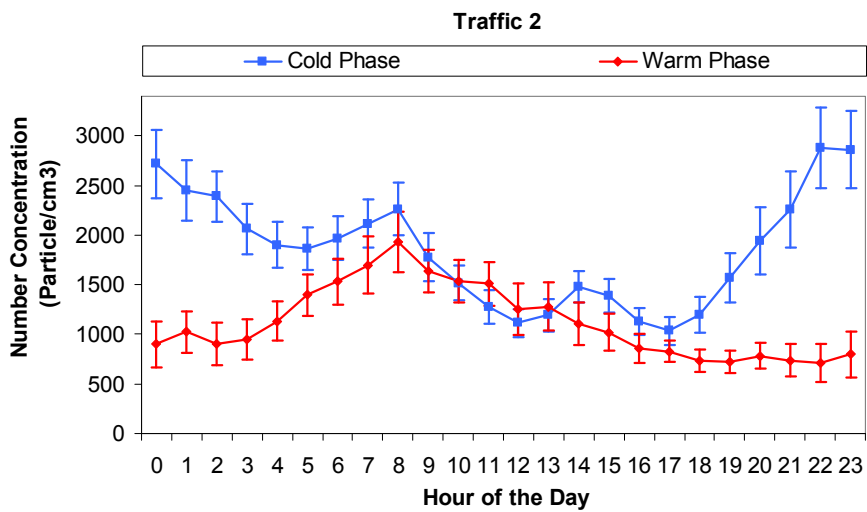
1 Figure 7. Diurnal variations (geometric means) of number concentrations (particles /cm³)
 2 from each factor resolved by the PMF model in the cold and warm phases. Error bars
 3 correspond to one standard error.



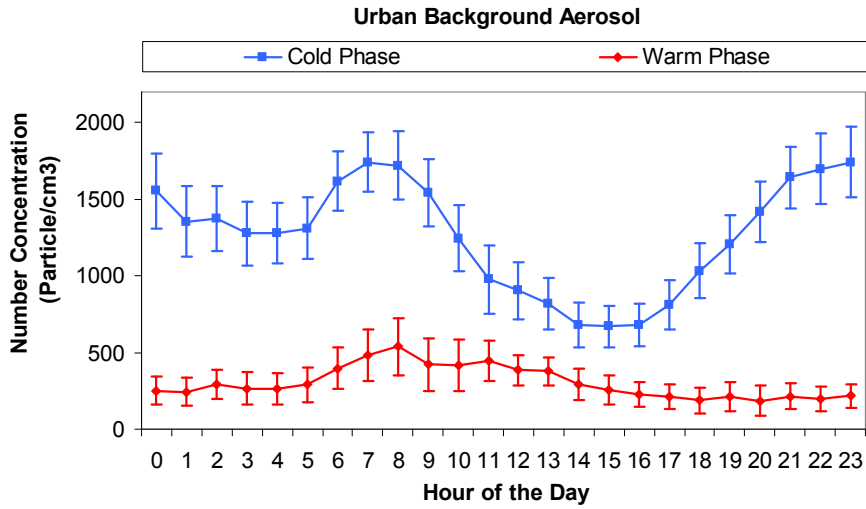
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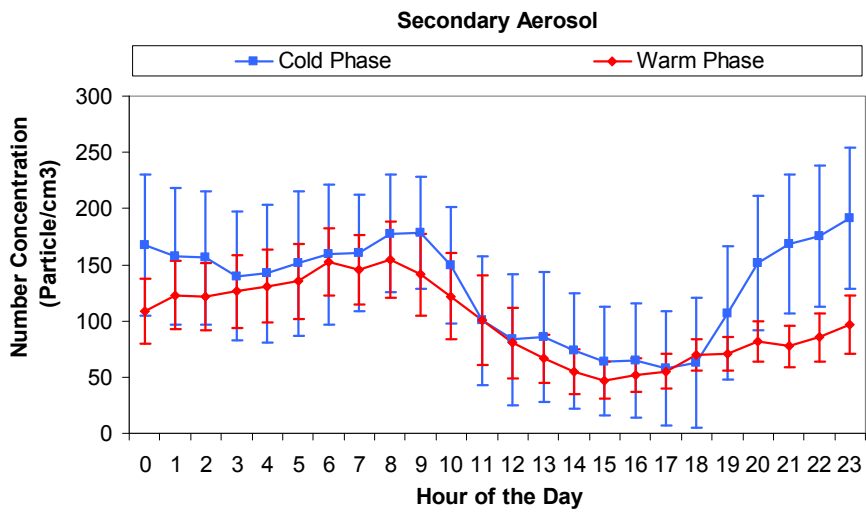
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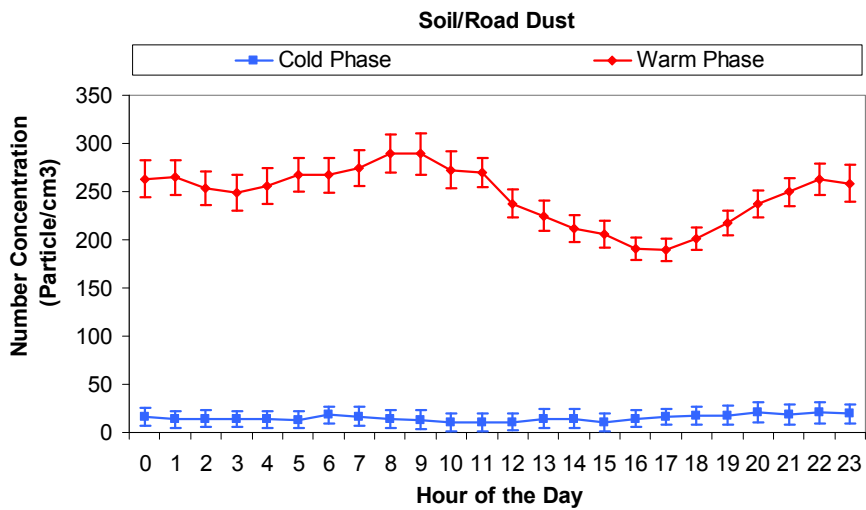
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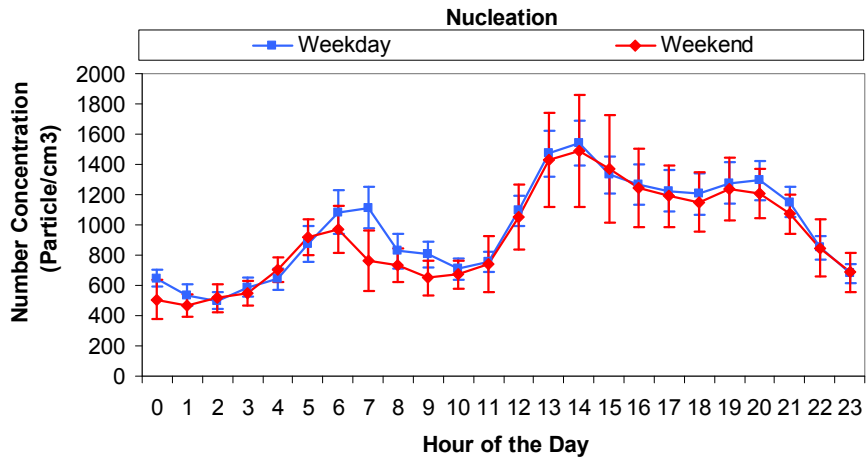
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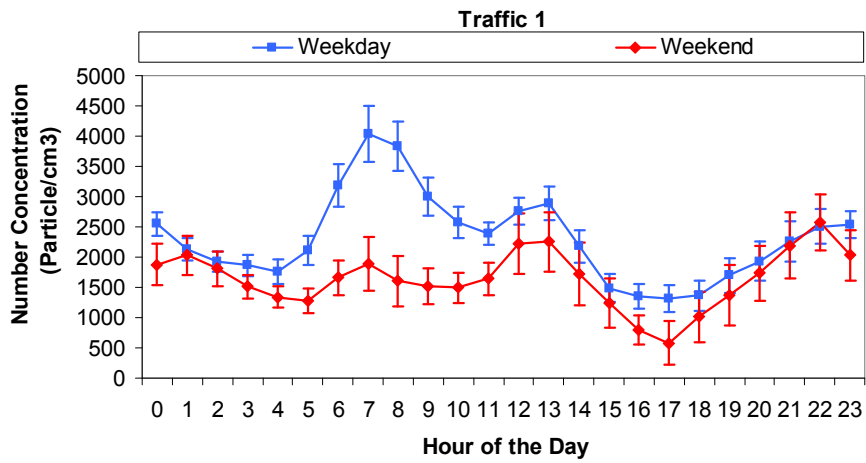
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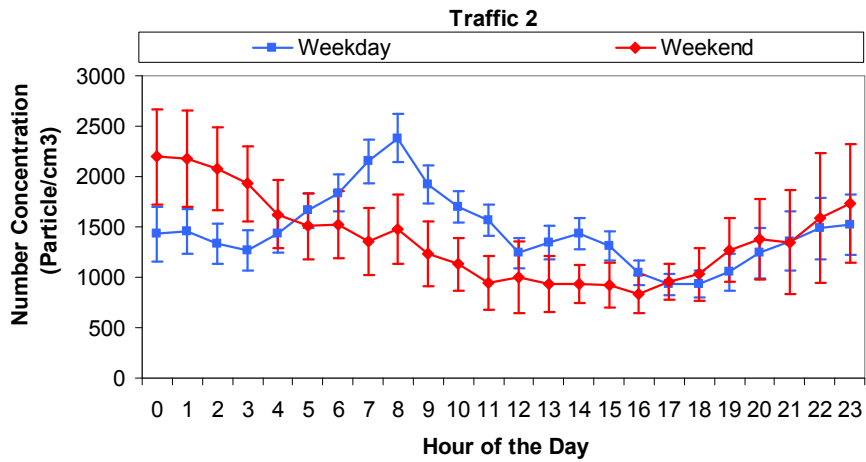
1 Figure 8. Weekday/weekend analysis of each of the factors resolved by the PMF model
2 (values are geometric means). Error bars correspond to one standard error.



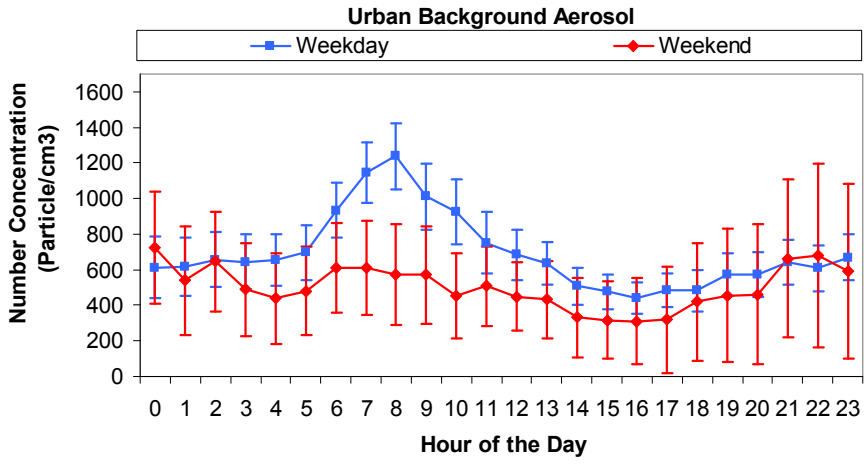
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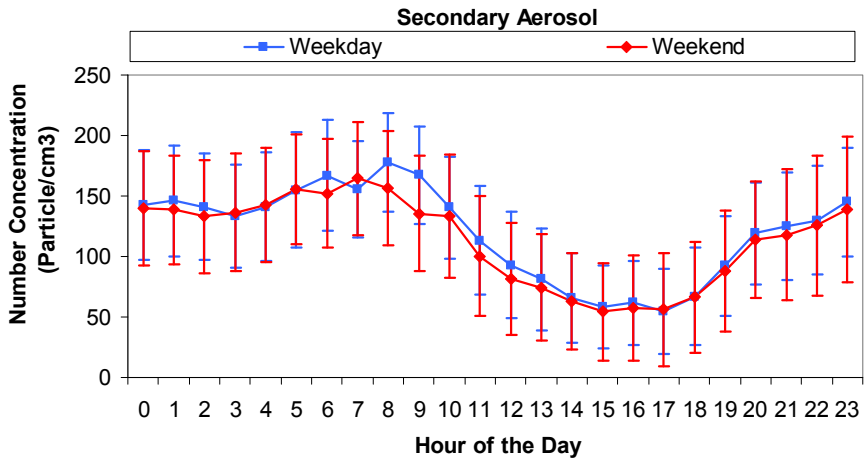
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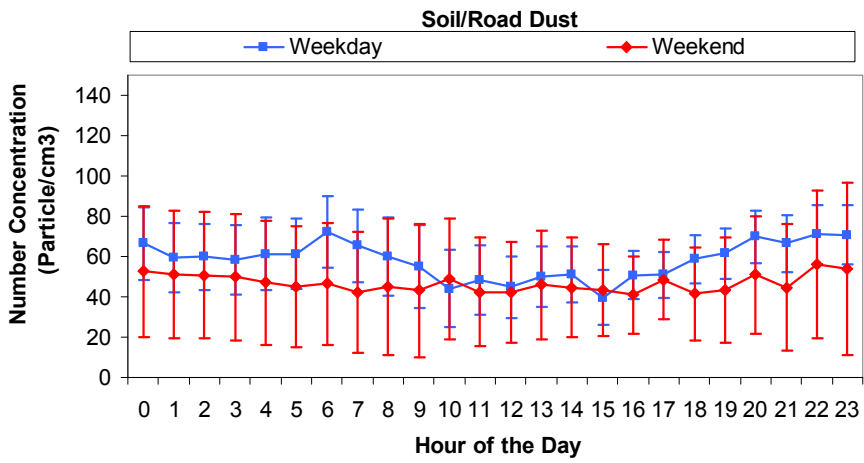
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