

# Environmental Science Processes & Impacts

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## Environmental Impact

Little is currently known about the sources of airborne nanoparticles during summertime hot, arid and dusty climatic conditions in the Middle East region. This is a first *source apportionment* study for the airborne nanoparticles in this region that has quantified the contribution to particle number concentrations from numerous major sources, along with determining particle number distribution profiles of individual sources. Besides policy makers and environmental authorities findings of this work study could be important for modelling community to treat major nanoparticle sources in dispersion modelling and health impact assessments in the region.



1 emissions), 9% (industrial emissions), 9% (regional background), 6% (miscellaneous  
2 sources) and 3% (Arabian dust transport) of total PNCs. The sources of nanoparticles and  
3 their particle number distribution profiles identified could serve as a reference data to design  
4 more detailed field studies in future and treat these sources in dispersion modelling and health  
5 impact assessment studies.

6 **Keywords:** *Positive matrix factorization; Particle number size distribution; Source*  
7 *identification; Summertime nanoparticles; Middle East region*

## 8 **1. Introduction**

9 Exposure to particulate matter (PM) is known to adversely affect human health.<sup>1</sup>  
10 Ambient concentrations of PM are currently regulated through mass-based standards of PM<sub>10</sub>  
11 and PM<sub>2.5</sub>, i.e., aerodynamic diameters less than 10 and 2.5  $\mu\text{m}$ , respectively.<sup>2</sup> Because of  
12 possessing negligible mass compared to the regulated PM,<sup>3,4</sup> these standards do not control  
13 airborne nanoparticles that are referred to as those below 300 nm in diameter and represent  
14 the majority (~99%) of total particle number concentrations, PNCs.<sup>5</sup> Nanoparticles are  
15 characterised by their vast numbers and high surface area.<sup>6,7</sup> As a result, they can adsorb  
16 large concentrations of toxic hazardous chemicals on their surfaces, translocate and deposit in  
17 different parts of the human body and thereby causing adverse health effects.<sup>8,9</sup> Evidences  
18 from a large number of studies link the exposure of nanoparticles to the occurrence of  
19 cardiovascular diseases.<sup>8</sup> This effect is attributed to the translocation of the redox-active  
20 components of the nanoparticles in the human body, which promotes the progression of  
21 atherosclerosis.<sup>10</sup> Furthermore, preliminary estimates of excess mortality related to  
22 nanoparticle exposure have been reported to be notable at 11,252 deaths in 2010 in Delhi<sup>11</sup>  
23 and ~310,000 deaths per year in Asian megacities.<sup>9</sup> However, such estimates are currently

1 unavailable for the Middle East region, clearly showing a need for field studies that can  
2 provide an in-depth insight into the sources of nanoparticles and associated health impacts.

3 Pollutants measured at a receptor site are a combination of various local and regional sources  
4 situated at varying distances from a site. Nanoparticles are dynamic in nature with a potential  
5 to change in the atmosphere through transformation processes such as dilution, nucleation,  
6 coagulation, condensation/evaporation and deposition during their transport from the source  
7 to the receptor site.<sup>12, 13</sup> However, majority of the transformation occurs close to the source  
8 and the particle number distributions (PNDs) may not change considerably at large distances  
9 from their original emission source such as road traffic and petroleum refineries.<sup>14, 15</sup> Hence,  
10 the application of source apportionment models on the data collected at a receptor site could  
11 allow the extraction of the latent factors contributing to the total PND data and potentially  
12 reveal the nearby or faraway sources, along with their individual PND profiles. Our previous  
13 study<sup>16</sup> showed different PND profiles during variable wind directions at different times of  
14 the day, thereby representing the contribution of different sources to the measured size-  
15 resolved PND data. What remains unknown is the contribution of these different sources to  
16 the PNCs and the PND data collected during hot and arid weather conditions. These  
17 unstudied aspects are taken up for a detailed investigation in this study.

18 Source apportionment models are important to identify various unknown sources and  
19 quantify their contributions towards the total measured concentrations. Such information is  
20 important to design efficient abatement strategies to control emissions. One of the most  
21 common receptor-based source apportionment models is positive matrix factorization (PMF),  
22 which can overcome the drawbacks of principle component analysis, PCA.<sup>17, 18</sup> The output of  
23 PMF is more physically realistic than that of PCA because the former allows the  
24 implementation of non-negative constraints and production of explainable positive elements

1 among all factors. Other models such as the chemical mass balance (CMB) and Unmix are  
2 comparable to PMF, to some extent. However, PMF does not require prior knowledge of the  
3 sources and their profiles, as required in the case of CMB, thereby making it an easier and  
4 more cost-effective solution. Furthermore, PMF allows for the weighting of each data point  
5 individually<sup>18</sup> – a feature that is not available in the Unmix model.

6 Prior to the incorporation of PND data in the PMF, this source apportionment technique has  
7 been applied for the identification of particles sources in many previous studies.<sup>19-22</sup>

8 However, these studies have mainly focused on PM mass concentrations and compositional  
9 data. PMF analysis, based on the PM chemical composition data, is often time-consuming  
10 and expensive, and does not segregate PNDs according to their sources. The knowledge of  
11 source-specific PNDs is of great relevance to epidemiological studies because of the size-  
12 dependency of respiratory tract deposition pattern in the human body on particle diameter.<sup>23</sup>

13 Several studies worldwide have successfully deployed total PNC data in the application of  
14 PMF to identify sources and their contributions over the past decade (see summary of  
15 relevant studies in Table 1). Few of these studies have used only PND data for the PMF  
16 analysis,<sup>24</sup> while others have included PM chemical composition data,<sup>25</sup> gaseous pollutant  
17 data,<sup>26</sup> and chemical composition and gaseous pollutant data<sup>15</sup> in their PMF analysis. In fact,  
18 none of the studies till date have applied PMF to the distinct PND characteristics found in the  
19 Middle East region, and therefore, the contributions of the different sources of PNC are  
20 currently unknown.

21 In order to fill the above-noted research gaps, we have applied PMF to our PND data set,  
22 ranging from 5–1000 nm, collected continuously over a 31-day period during summertime  
23 conditions at a roadside location in Fahaheel, Kuwait, by using a fast response differential  
24 mobility spectrometer (Cambustion DMS500). In addition, PM<sub>10</sub>, gaseous pollutants (NO<sub>x</sub>,

1 O<sub>3</sub>, CO and SO<sub>2</sub>) and meteorological data were used to assist the interpretation of the PMF  
2 results by using conditional probability function (CPF).

3 The following are the unique features of our work. Firstly, the use of DMS500 is  
4 advantageous because it can provide real-time measurements of nanoparticles at a sampling  
5 rate of 10 Hz, allowing for the rapid capture of the fast transformation processes. The  
6 DMS500 is currently one of the commercially available fastest response particle sizers,  
7 requiring only ~100 ms to complete one full spectrum of PND. This enabled us to capture the  
8 peaks of PNCs that occur within a few seconds in urban environments.<sup>27</sup> Furthermore, the  
9 sampling height of the DMS500 inlet was ~1.60 m above the ground, representing the typical  
10 breathing height of the people, which can be easily used in epidemiological studies in  
11 calculating deposition doses. Secondly, the application of PMF was applied at a high  
12 temporal resolution (5-min based measurements), which is higher than that in most of the  
13 previous work (see Table 1), and on a continuous measured data of all studied parameters as  
14 opposed to the intermittent data used by some of previous studies (Table 1). Thirdly, most of  
15 the published work has only used wind direction in their CPF application (Table 1), but our  
16 study used both wind direction and speed, providing a better understanding of the  
17 directionality and position of the potential sources. Finally, to the best of our knowledge, this  
18 is a first instance when a source apportionment technique is used on high-resolution PND  
19 data in Kuwait, and the Middle East in general, which was collected during severe  
20 summertime conditions (maximum temperature ~48 °C and minimum relative humidity  
21 ~0.20%) with frequent dust events (Section 2.1).

22 In the light of the existing research gaps, the aims of this study are: (i) to identify the possible  
23 sources of nanoparticles in the studied area which represents a typical roadside environment

1 of the Middle East region, (ii) to quantify the sources contribution to total PNCs and (iii) to  
2 determine the individual PND spectrum of various sources in a Middle Eastern city, Kuwait.

## 3 **2. Experimental methods**

### 4 **2.1 Site description**

5 This study was conducted at a near-road location in the urban area of Fahaheel,  
6 Kuwait (Figure 1). The geographic coordinates of the sampling site are 29°4'52.70" N and  
7 48°6'52.08" E. The sampling instruments were placed inside an air-conditioned cabin, located  
8 at a distance of ~15 m east of the kerbside of Fahaheel highway. This highway runs in the  
9 north-south direction, linking The State of Kuwait with the Kingdom of Saudi Arabia. This  
10 six-lane highway is one of the busiest highways in Kuwait, consisting of three lanes (~3.70 m  
11 wide) in each direction. These lanes are separated by a paved median strip, and there are two  
12 additional lanes in each direction reserved for emergency. The areas to the immediate east  
13 and west of the sampling site are intra-city activities and open flat desert, respectively. The  
14 intra-city activities in Fahaheel area consist of vehicular movement, gas stations and small  
15 businesses. Additionally, the sampling site is influenced from south-east direction by a vast  
16 range of petroleum, petrochemical, cement, caustic and small industries, located at a distance  
17 of 1200 m from the edge of these petroleum activities.<sup>36</sup>

18 Measurements were made during summertime in the month of May and June 2013 when  
19 ambient temperature reached to ~48 °C, the relative humidity decreased to a minimum of  
20 0.20% and the dust events (i.e., when  $PM_{10} > 200 \mu\text{g m}^{-3}$ ) were observed for ~49% of the  
21 total measurement time. The average temperature, relative humidity and wind speed were  
22 found to be  $37 \pm 4.5$  °C,  $13.6 \pm 10.0\%$  and  $6.3 \pm 3.0 \text{ m s}^{-1}$ , respectively. The prevailing wind  
23 direction was north-west (~311°N). Wind speed and ambient temperature affected the PNCs



1 notably. For example, ambient temperature was found to linearly decrease the PNCs due to  
2 partial evaporation<sup>16</sup>; see details in Supplementary Information, SI, Section S1.

3 The sampling site and the Fahaheel area are ideal for this study because of the following  
4 reasons. Firstly, Fahaheel is a typical urban area in Kuwait surrounded by heavy petroleum  
5 industries, reflecting typical characteristics of the oil-rich State of Kuwait and the intra-city  
6 activities, as well as is a good representative of the Middle East region (especially the  
7 Arabian Peninsula region), in terms of topography and climatic conditions. Secondly, no  
8 other major highways directly influence the sampling site, except the studied Fahaheel  
9 highway, allowing a clear identification of the highway impact on the measured PND data.  
10 Thirdly, the sampling site is characterised by the absence of obstacles for at least ~300 m  
11 radius, eliminating the downwash effects. Finally, the surrounding potential sources of the  
12 sampling site are well-distributed at different directions and distances, allowing the  
13 development of CPF plots using local wind data to aid in the source identification by PMF.  
14 Further details on the sampling site characteristics, including traffic and meteorology, can be  
15 seen in Al-Dabbous and Kumar<sup>16</sup>.

## 16 **2.2 Data acquisition**

17 A total of 8675 valid 5-minute PND observations, each in 36 size classes, covering 5–  
18 1000 nm size range, were continuously measured from 27 May to 26 June 2013 by using a  
19 DMS500. These measurements were collected at 0.10-second time resolutions and then  
20 averaged to 5-min interval means to synchronise them with the pollutants and meteorological  
21 data. DMS500 is a parent version of DMS50 (i.e., portable instrument with similar features)  
22 that has been successfully used in a variety of our studies, related to roadside and kerbside  
23 measurements,<sup>16, 37</sup> vehicle-wake,<sup>38</sup> vehicle in-cabin<sup>39-41</sup> and indoor construction  
24 environments.<sup>42, 43</sup> The DMS500 detects particles based on their electrical mobility.<sup>3</sup>  
25 Additionally, a suite of pollutants (PM<sub>10</sub>, O<sub>3</sub>, NO<sub>x</sub>, SO<sub>2</sub> and CO) and meteorological

1 parameters (temperature, relative humidity, wind speed and direction) were obtained from the  
2 adjacent (~300 m away from site) Environmental Protection Agency (EPA) monitoring  
3 station. These continuous data are well-maintained and quality-controlled by the Kuwait  
4 EPA. Further details on the experimental setup, instrumentation and working principle of  
5 various instruments can be seen elsewhere.<sup>16</sup>

### 6 **2.3 Statistical analysis**

7 PMF analysis was applied using the US EPA's PMF program (version 5.0) on the  
8 dataset composed of 36 variables. These variables included PNDs in 36 size classes covering  
9 a size range of 5–1000 nm, following the methodology described in Paatero<sup>18</sup>. PMF is a  
10 multivariate factor analysis model used to identify the contribution and profile by exposing  
11 the dataset to a multi-linear engine algorithm and a gradient algorithm approach in order to  
12 find the best-fit solution.<sup>44, 45</sup> This method is featured by the non-negative constraints and the  
13 use of uncertainties to scale individual data points. The uncertainty data file supplied by the  
14 instrument manufacturer (Cambustion Ltd., Cambridge), consisting of size-specific  
15 minimum detection limits and error fractions, was also included in the PMF. An extra  
16 modelling uncertainty of 5% was added to the model to account for any additional  
17 measurement errors that were not covered by the uncertainty data file.<sup>46</sup> The missing  
18 sampling values due to instrument failure were modest (i.e., <3% of the entire sampling  
19 period) and simply excluded from the analysis. In addition, CPF plots were prepared using  
20 the threshold of the upper 25<sup>th</sup> percentile of the fractional contribution of each factor/source.  
21 These plots complemented the PMF analysis by depicting the trend in the factors score with  
22 wind direction and speed so that factors could be tentatively assigned to the potential sources  
23 in the area.<sup>47</sup> Furthermore, CPF plots were also drawn for the routinely measured pollutants  
24 (PM<sub>10</sub>, O<sub>3</sub>, NO<sub>x</sub>, SO<sub>2</sub> and CO) by using the same criterion. Open Air (R package), which is an

1 open-source statistical tool,<sup>48</sup> was used to derive the CPF plots that assisted in the  
2 interpretation of the measured air pollution data.

### 3 **3. Results and discussion**

4 Using the PMF approach described in Section 2.3, six different factors were identified  
5 that were then tentatively assigned to the potential sources based on the following  
6 information: (i) factor-specific PNDs (Figure 2 and Figure 3g-l, middle vertical panel), (ii)  
7 diurnal variation of the factors (Figure 3m-r, right vertical panel), (iii) contribution of each  
8 factor to the total PNC (Figure 4), (iv) hourly Pearson product–moment correlations, along  
9 with the significance level (p-value), between each factor contribution and measured gaseous  
10 ( $O_3$ ,  $NO_x$ ,  $SO_2$  and  $CO$ ) and  $PM_{10}$  pollutants (Table 2), and (v) the CPF plots for each factor  
11 contribution (Figure 3a-f, left vertical panel) and measured gaseous ( $O_3$ ,  $NO_x$ ,  $SO_2$  and  $CO$ )  
12 and  $PM_{10}$  pollutants (Figure 5).

#### 13 **3.1 Factor 1: Miscellaneous sources**

14 This factor showed multiple PND modes, with the major peaks at about 365 nm and  
15 1000 nm, and a positive correlation with  $PM_{10}$  ( $r = 0.39$ ; p-value  $<0.01$ ; Table 2). Factor 1  
16 also showed a minor peak at  $\sim 5$  nm, which could represent fresh traffic emissions but to  
17 lesser extent than that observed for factors 4 and 5. Furthermore, the wind directionality and  
18 the relatively high wind speed (up to  $10 \text{ m s}^{-1}$ ) of this factor (Figure 3a) and  $PM_{10}$  (Figure 5a)  
19 indicated that the particle emissions had travelled from a remote location and grown to larger  
20 sizes through coagulation. Al-Dabbous and Kumar<sup>16</sup> previously reported a dominating role of  
21  $PM_{10}$  in suppressing PNCs due to coagulation process. For instance, PNCs were found to be  
22 reduced by  $\sim 23\%$  when  $PM_{10}$  concentration increased by  $\sim 500\%$ , compared to the values  
23 prior to the arrival of the dust event (i.e., when  $PM_{10} < 200 \mu\text{g m}^{-3}$ ). A similar observation on  
24 coagulation scavenging has been reported by Jayaratne *et al.*<sup>49</sup> with respect to the influence

1 of the Australian dust storm on the PNCs. This factor made the second lowest contribution  
2 (6%) to the total PNCs. The directionality of the CPF plots and the association with PM<sub>10</sub>  
3 clearly corresponds to the west Shuaiba industrial area and the dust blown by high wind  
4 speed from the desert during the south-westerly winds.

5 Furthermore, particles emitted from the industrial area appears to be aged particles that have  
6 spent time in the atmospheric environment and grown to larger sizes during their travel from  
7 their far sources (for example, west Shuaiba industrial area during the south westerly winds,  
8 in this case). These particles could be attributed to the vehicle movements within the  
9 industrial area such as those found in Factor 5 (Section 3.5), but neither the factor  
10 contribution did show any nocturnal variation (Figure 3m) nor the PND profile (Figure 3g)  
11 and the poor correlations with the NO<sub>x</sub> and CO (Table 2) support any direct association with  
12 the traffic emissions. For example, the diurnal behaviour of factor 1 (Figure 3m) showed a  
13 slight drop in factor contribution during the afternoon hours; otherwise this remains fairly  
14 constant during the rest of the period. The reason for this slight drop could be attributed to the  
15 unstable atmospheric conditions, induced by the intensive solar radiation ( $800\pm 548 \text{ W m}^{-2}$   
16 during the afternoon hours compared with an average value of  $323\pm 373 \text{ W m}^{-2}$  during the  
17 entire period), leading to relatively larger mixing of these particles.<sup>16</sup> Although this factor  
18 was tentatively assigned to shared sources, information available from the correlations  
19 between factor contribution and gaseous pollutants (Table 2), and diurnal profile of factor  
20 contribution (Figure 3m), was insufficient to assign a separate weighting to each of these two  
21 different sources.

### 22 **3.2 Factor 2: Arabian dust transport**

23 This factor showed a bimodal PND (Figure 3i) with a major peak at 560 nm, and a  
24 minor peak at 60 nm, along with a distinctively high correlation with PM<sub>10</sub> ( $r = 0.71$ ; p-value  
25  $< 0.01$ ; Table 2). The wind directionality and the associated high speed levels (more than 15

1 m s<sup>-1</sup>) noted in CFP plots of this factor (Figure 3b), as well as PM<sub>10</sub> (Figure 5a), indicate the  
2 influence of the dust from the long-range transport that is associated with the typical Arabian  
3 dust events. This factor showed behaviour similar to that of factor 1, but to a greater extent in  
4 terms of higher PM<sub>10</sub>, wind speed levels and the typical directionality (i.e., north-westerly  
5 direction) associated with the frequent dust events in the region. In an extended analysis of  
6 the same dataset,<sup>16</sup> Arabian dust events were found to suppress PNCs due to the influence of  
7 coagulation process, which explains the minimum contribution (3%) of this factor to the total  
8 PNC (Figure 4). It is worth pointing out that both the factors 1 (6%) and 2 (3%) made the  
9 lowest contributions (Figure 4), among the six resolved factors, but showed the highest  
10 correlations with PM<sub>10</sub>; these characteristics support the possible effects of the coagulation  
11 process during high concentrations of PM<sub>10</sub> approaching the site from the westerly wind  
12 direction (i.e., open desert; Figure 1). Furthermore, the diurnal profile of this factor showed  
13 an increased contributions during the afternoon (12:00 to 14:00 h; Figure 3n) due to relatively  
14 higher wind speeds and associated saltation process.<sup>50</sup> In an extended analysis on the same  
15 dataset (SI Figure S1), but excluding the major dust event periods (i.e., when PM<sub>10</sub> >1000 µg  
16 m<sup>-3</sup>), we observed almost similar contribution to the total PNCs (Figure S2) to those observed  
17 in Figure 4 for all the six sources. Pearson product-moment correlations between each factor  
18 and measured gaseous (O<sub>3</sub>, NO<sub>x</sub>, SO<sub>2</sub> and CO) and PM<sub>10</sub> pollutants also exhibited similar  
19 correlations (Table S1) to those observed in Table 2. This similarity confirms that the input  
20 dataset were not highly affected by the Arabian dust events, mainly because the major dust  
21 event periods were only 5.7% of the total measurements period.

### 22 3.3 Factor 3: Industrial emissions

23 This factor showed a monomodal distribution with a peak at ~42 nm (Figure 3i), and  
24 made a 9% contribution to the total PNCs (Figure 4). The diameter of this peak was in  
25 accordance with those recorded for industrial emissions in previously published studies. For

1 instance, Ogulei *et al.*<sup>31</sup> reported a peak at 44 nm during their one-year long measurements  
2 (2004–2005) at an urban background location in New York (USA) that was significantly  
3 influenced by the industrial activities. We have several reasons to believe that factor 3  
4 represents industrial emissions. For example, the CPF plots of this factor are strongly  
5 associated with south easterly winds (Figure 3c), which is consistent with the wind  
6 directionality of SO<sub>2</sub> (Figure 5b). The directionality of these plots clearly correspond to the  
7 Shuaiba industrial area, which hosts a range of oil refineries (i.e., Mina Al-Ahmadi, Shuaiba  
8 and Mina Abdullah refinery), petrochemical industries (e.g., ammonia, urea, polyethylene  
9 and polypropylene plant) and two power desalination plants.<sup>51, 52</sup> Furthermore, this factor had  
10 the highest correlation ( $r = 0.31$ ;  $p$ -value  $< 0.01$ ) with SO<sub>2</sub> among all the factors (Table 2),  
11 which supports the fact that industrial emissions are clearly associated with this factor.  
12 Moreover, NO<sub>x</sub> ( $r = 0.37$ ;  $p$ -value  $< 0.01$ ) and CO ( $r = 0.23$ ;  $p$ -value  $< 0.01$ ) also showed a  
13 positive correlation with this factor, indicating an association with the combustion activities  
14 within the vicinity of the industrial area. Past studies have also linked industrial emissions  
15 with the combustion related pollutants, mainly SO<sub>2</sub>.<sup>29, 31, 33, 35</sup> The association with SO<sub>2</sub> may  
16 indicate the influence of secondary particle formation in the form of photo-chemically  
17 induced sulphuric-acid nucleation.<sup>53, 54</sup> The diurnal profile of this factor displayed a typical  
18 diurnal variation, linked with the meteorological conditions and the associated boundary layer  
19 <sup>55</sup>. For example, a decreased factor contribution was observed during the afternoon, which  
20 was caused by the expanded depth of the boundary layer and the associated dilution with the  
21 background air. Based on the above concluding evidences, we attributed this factor to the  
22 industrial emissions.

### 23 **3.4 Factor 4: Fresh traffic emissions**

24 This factor showed a major PND peak between 5–12 nm and another minor peak at ~60  
25 nm (Figure 3j) and explained nearly half (46%; Figure 4) of the total PNC contribution.

1 Looking at the PND and the peaks, this contribution was believed to be from the local traffic.  
2 For example, this bimodal profiles of PNDs are consistent with those observed by Fujitani *et*  
3 *al.*<sup>56</sup> at 10 nm and 40–60 nm during their near road measurements in Kanagawa Prefecture,  
4 Japan. Furthermore, similar PND peaks related to local traffic were observed by numerous  
5 studies performed in cities worldwide, such as at 20 nm (major peak) and 100 nm (minor  
6 peak) in Beijing, China,<sup>15</sup> 20 nm in Brisbane, Australia,<sup>28</sup> 9–40 nm in Augsburg, Germany,<sup>25</sup>  
7 10–100 nm in Erfurt, Germany,<sup>24</sup> 20 nm in London, UK,<sup>26</sup> 13.3 nm in Cambridge, UK,<sup>5</sup> 10  
8 nm in New York, USA,<sup>31</sup> 15 nm in Pittsburgh, USA.<sup>35</sup> The wind directionality (Figure 3d)  
9 corresponded to the highway located at 15 m west of the measurement location, and the wind  
10 speed was observed to be relatively low ( $<5 \text{ m s}^{-1}$ ) compared with much higher levels noted  
11 during the major dust events. This low level of wind speed indicates an association with  
12 close-range source (i.e., local traffic). The directionality of the factor contribution is also  
13 consistent with those for  $\text{NO}_x$  (Figure 5c) and CO (Figure 5d), especially from the westerly  
14 wind direction, indicating the same emission source. Furthermore, this factor contribution  
15 correlated positively with the  $\text{NO}_x$  ( $r = 0.30$ ;  $p\text{-value} < 0.01$ ), which is a primary traffic-  
16 generated pollutant.<sup>57, 58</sup> The diurnal profile of this factor contribution (Figure 3p) was in  
17 agreement with the diurnal pattern of the traffic volume, except during the noon hours when  
18 the high traffic volume corresponded to low factor contribution. The reason for this odd  
19 behaviour was previously studied in an extended analysis by Al-Dabbous and Kumar<sup>16</sup> and  
20 explained by the extreme temperature (reaching up to  $\sim 50 \text{ }^\circ\text{C}$ ) that resulted in partial  
21 evaporation and increased rate of coagulation with larger particles.<sup>59</sup> Most of the above-  
22 discussed studies also observed higher PND magnitude in the morning rush hours compared  
23 with those during evening rush hours; this is consistent with the findings of our current study.  
24 Based on the above observations, we attributed this factor to local traffic emissions, seen  
25 through the newly formed particles (i.e., fresh traffic emissions) in nucleation mode.

### 1 3.5 Factor 5: Aged traffic emissions

2 This factor showed a major peak at 24 nm, followed by a minor peak at 130 nm (Figure  
3 3k). The former peak is presumably attributed to the nearby highway emissions, and the  
4 latter, to aged particles transported from the industrial area. These bimodal profiles of PNDs  
5 are similar to those observed by Gu *et al.*<sup>25</sup> at 20 and 100 nm during their measurements in  
6 Augsburg, Germany, and attributed them to aged traffic emissions. This factor showed the  
7 second highest contribution to the total PNC (27%; Figure 4). This factor was positively  
8 correlated with NO<sub>x</sub> (r = 0.54; p-value <0.01) and CO (r = 0.23; p-value <0.01) and showed  
9 no correlation with SO<sub>2</sub>. Moreover, the CPF shown in Figure 3e clearly pointed out the wind  
10 direction from the Shuaiba industrial area (i.e., south-easterly direction) and the traffic  
11 emission from Fahaheel highway (i.e., westerly direction). This wind directionality is  
12 identical to those obtained for NO<sub>x</sub> (Figure 5c) and CO (figure 5d). Therefore, the  
13 correlations with the NO<sub>x</sub> and CO as well as the CPF suggest that there is a contribution from  
14 *primary* (solid carbonaceous) particles from diesel vehicles from the nearby industrial area  
15 and the Fahaheel highway. However, absence of such correlations with the SO<sub>2</sub> suggests a  
16 negligible contribution of *secondary* particle formation through photo-chemically induced  
17 sulphuric-acid nucleation like what is noticed in case of factor 3. Furthermore, the diurnal  
18 profile of this factor contribution (Figure 3q) was similar to the profile of factor 4, with a  
19 slight increase in the evening hours, indicating the influence of nocturnal commercial traffic  
20 (e.g., heavy duty trucks) operating on the Fahaheel highway and within the industrial area. In  
21 total, both the fresh (factor 4) and aged (factor 5) traffic emissions accounted for about 73%  
22 of the total PNCs, which is comparable to roadside studies in London, UK (~72%)<sup>26</sup> and  
23 Brisbane, Australia (~74%)<sup>28</sup>.



### 1 3.6 Factor 6: Regional background

2 This factor showed multiple PND peaks with a major peak at 150 nm, followed by a  
3 minor peak at 750 nm (Figure 3l), and contributed to 9% of the total PNCs (Figure 4).  
4 Particles in the size range (diameter >100 nm) could possibly be originated: (i) either locally,  
5 through direct emissions from local sources such as exhaust emissions or brake dust, or  
6 coagulation of smaller particles with each other and with their larger counterparts,<sup>3</sup> or (ii)  
7 regionally that are transported to the receptor site.<sup>29</sup> However, the wind directionality shown  
8 in Figure 3f indicate that the PNC emissions were approaching to the site from all the wind  
9 directions and the association with the high wind speed indicated a contribution from the far-  
10 range sources. Particles larger than 100 nm contain low volatility and solid cores.<sup>23</sup>  
11 Therefore, these can travel relatively larger distances compared with highly volatile  
12 nucleation mode particles<sup>12, 60</sup>. This factor also showed the highest correlation with PM<sub>10</sub> (r =  
13 0.31; p-value <0.01) compared with other pollutants (Table 2), agreeing with those reported  
14 by Ogulei *et al.*<sup>31</sup> where they found a high correlation with regionally transported PM<sub>2.5</sub>.  
15 Both the factors 6 and 1 showed identical correlations with the PM<sub>10</sub>, but information  
16 available from the wind directionality and PNDs profile assist in attributing the factor 6 to  
17 regional background. Furthermore, the lack of obvious diurnal variation in factor contribution  
18 (Figure 3r) also suggests that this is a regional background source.

## 19 4. Summary and conclusions

20 Particle number and size distributions in the size range of 5–1000 nm were  
21 continuously measured for a period of one month, starting from 27 May to 26 June 2013, at a  
22 roadside location in Kuwait. The aims of the study were to identify the sources size-resolved  
23 particles under summertime climatic conditions, as well as with quantifying their  
24 contributions, and understanding their influencing parameters (PM<sub>10</sub>, gaseous pollutants and  
25 meteorological parameters).

1 The application of PMF helped in identifying six probable sources: miscellaneous sources,  
2 Arabian dust transport, industrial emissions, fresh as well as aged traffic emissions, and  
3 regional background. Traffic emissions made the highest (73%) contributions to the total  
4 PNC, followed by industrial emissions (9%), regional background (9%), miscellaneous  
5 sources (6%) and Arabian dust transport (3%). The high correlations between  $PM_{10}$  and the  
6 factor contribution of the last three sources indicated the possible influence of coagulation of  
7 PNCs with their larger counterparts and thus resulting in the suppression of total PNCs. The  
8 diurnal profile of the factor contribution of the traffic sources (i.e., *factor 4* and *factor 5*) were  
9 categorised by a bimodal distribution, coinciding with the morning and evening rush hours,  
10 whereas Arabian dust transport (i.e., *factor 2*) was characterised by an increased factor  
11 contribution in the noon hours, where high wind speed approached the sampling site loaded  
12 with high levels of  $PM_{10}$ . Miscellaneous sources (*factor 1*) and regional background (*factor*  
13 *6*) displayed no diurnal variation in their factor contribution, except during noon hours where  
14 high dilution was expected due to the expanded boundary layer and the associated high wind  
15 speed. Traffic sources (i.e., *factors 4* and *5*) showed a typical bimodal PND, while all the  
16 long-range transport sources (i.e., *factors 1*, *2*, and *6*) consisted mostly of particles greater  
17 than 100 nm in diameter, resulting from their growth in size during transport from sources far  
18 away. Industrial emissions (i.e., *factor 3*) displayed a unique monomodal PND, peaking at  
19 about 42 nm. The similarities in the wind directionality of the factors contribution and the  
20 pollutants, using CPF at 75<sup>th</sup> percentile threshold criterion, assisted in sources allocation.

21 This study covers a hitherto overlooked topic in the Middle East region. The findings of this  
22 work make contributions towards the understanding of potential sources of nanoparticles in  
23 the area and their probable contribution to the PNCs. Furthermore, PND profiles associated  
24 with individual sources present an important reference data for future studies in the Middle  
25 East region. Long-term measurement studies, involving more pollutants (e.g., trace metals

1 and organic compounds), are recommended to elucidate further on specific source  
2 characteristics and their emission strengths.

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9 Network and Kuwait Environmental Protection Agency for their cooperation in providing us  
10 the meteorological and gaseous pollutants data.

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- 1 **List of figure captions**
- 2 **Figure 1.** Location of the sampling site in the Fahaheel area, showing the major sources  
3 surrounding the site. Satellite image includes material ©NSPO 2014 distribution Spot Image  
4 S.A.; courtesy of Airbus Defence and Space, all rights reserved. Note: SIA = Shuaiba  
5 Industrial Area; WSIA = West Shuaiba Industrial Area.
- 6 **Figure 2.** Percent contribution of each source identified towards the PNCs in different size  
7 ranges.
- 8 **Figure 3.** Directionality of the factor contribution using CPF plots at 75<sup>th</sup> percentile level,  
9 considering both local wind direction and speed (Figure 3a-f). The colours in Figure 3a-f  
10 represent the probability of factor contribution with respect to wind direction and speed.  
11 Figures 3g-l represent the factor-specific PND profiles while Figures 3m-r show the diurnal  
12 variation of the normalised factor contribution.
- 13 **Figure 4.** Sources contribution (%) to the total PNC data in the urban area of Fahaheel,  
14 Kuwait.
- 15 **Figure 5.** Directionality of (a) PM<sub>10</sub>, (b) SO<sub>2</sub>, (c) NO<sub>x</sub>, (d) CO and (e) O<sub>3</sub> using CPF plots at  
16 75<sup>th</sup> percentile level, considering both local wind direction and speed. The colours in these  
17 figures represent the probability of the aforementioned pollutants with respect to wind  
18 direction and speed.

## 1 List of Tables

2 **Table 1.** Summary of recent PMF studies focusing on PND data set, together with other  
 3 auxiliary parameters (e.g., gaseous pollutants, particulate matter, chemical composition and  
 4 traffic).

| Author (year)                         | Location (type)                           | Size range (nm) | Instruments        | Additional data   | Sources identified (contribution of each source to the total apportioned PNC, %)   |
|---------------------------------------|---|-----------------|--------------------|---|--|
| This study                            | Fahaheel, Kuwait (roadside)               | 5–1000          | DMS500             | PM <sub>10</sub> and Gaseous (O <sub>3</sub> , NO, NO <sub>x</sub> , SO <sub>2</sub> and CO)  | Local traffic (46% of the total apportioned PNC), mixture of local traffic and industrial emissions (27%), industrial emissions (9%), regional background (9%), miscellaneous sources (6%), Arabian dust transport (3%)  |
| Liu <i>et al.</i> <sup>15</sup>       | Beijing, China (urban background)         | 14.5–2514       | SMPS               | Gaseous (O <sub>3</sub> , NO, NO <sub>2</sub> , CO and SO <sub>2</sub> ), and chemical composition (organic matter, sulphate, nitrate, ammonium and chlorine) | Local sources: cooking (22.8%), solid-mode exhaust (18.8%), nucleation-mode exhaust (18.7%), secondary nitrate (8.9%), secondary sulphate (7.9%), coal-fired power plant (6.8%) and road dust (2.3%)<br>Regional sources: accumulation mode (13.8%)  |
| Friend <i>et al.</i> <sup>28</sup>    | Brisbane, Australia (roadside)            | 14–715          | SMPS               | PM <sub>10</sub> , gaseous (CO, NO and NO <sub>2</sub> )  | Petrol vehicles (30.8%), diesel traffic (28.1%), local traffic (14.9%), biomass burning (20.1%) and two unidentified sources (6%)  |
| Gu <i>et al.</i> <sup>25</sup>        | Augsburg, Germany (urban background)      | 3–10000         | UDMA, UCPC and APS | Metals, water-soluble ions, elemental carbon (EC) and organic carbon (OC)   | Aged traffic (40.3%), re-suspended dust (32.6%), stationary combustion (26.1%), fresh traffic (24.9%), nucleation particles (3.7%), secondary aerosols (1.2%), and long-range transported dust (1.1%)  |
| Harrison <i>et al.</i> <sup>26</sup>  | London, UK (curbside)                     | 15–10000        | SMPS and APS       | Gaseous (O <sub>3</sub> , NO, NO <sub>2</sub> and CO) and traffic flow  | Road emissions: solid-mode exhaust (18.8%), brake dust (13.7%), re-suspended dust (4.4%) and nucleation-mode exhaust (3.6%)<br>Urban background: well-aged regional (26.8%), accumulation mode (12.8%), solid fuel/nitrate (8.4%), cooking (6.7%), regional (2.5%) and suburban traffic (2.3%) |
| Kasumba <i>et al.</i> <sup>29</sup>   | New York, USA (urban background)          | 100–470         | SMPS               | PM <sub>2.5</sub> and gaseous (CO, SO <sub>2</sub> and O <sub>3</sub> )   | Local traffic or gasoline traffic (21.7%), mixture of nucleation and traffic (20.1%), industrial emissions (17.2%), distant traffic or diesel traffic (15.2%), nucleation (17.6%), secondary sulphate (6.4%), ozone-rich secondary aerosol (0.9%), and regionally transported aerosol (1.1%)   |
| Thimmaiah <i>et al.</i> <sup>30</sup> | Prague, Czech Republic (urban background) | 18.8–723.5      | SMPS               | Gaseous (CO, SO <sub>2</sub> , NO <sub>x</sub> , O <sub>3</sub> , CH <sub>4</sub> , Non Methane Hydrocarbons)   | NO <sub>x</sub> -rich (influenced by diesel emissions, 37.8%), gasoline traffic (34.2%), heating (24.6%) and ozone-rich (mainly influenced by meteorology, 3.5%)   |



|                                    |                                    |          |              | and Total Hydrocarbons   |   |
|------------------------------------|------------------------------------|----------|--------------|--|---|
| Yue <i>et al.</i> <sup>24</sup>    | Erfurt, Germany (roadside)         | 10–3000  | MAS          | Gaseous (O <sub>3</sub> , NO, NO <sub>2</sub> , CO and SO <sub>2</sub> ) and Chemical composition (sulphate, EC and OC)                          | Ultrafine particles from local traffic (79%), secondary aerosols from multiple sources (6%), particles from remote traffic sources (5%) and airborne soil (1%)  |
| Ogulei <i>et al.</i> <sup>31</sup> | New York, USA (urban background)   | 10–470   | SMPS         | PM <sub>2.5</sub> , gaseous (CO, SO <sub>2</sub> and O <sub>3</sub> )  | Diesel/distant traffic (23.3%), mixture of gasoline/local traffic and nucleation (22%), industrial emissions (21.4%), nucleation (15.7%), secondary sulphate (10.9%), ozone-rich secondary aerosol (4.7%) and regionally transported aerosol (1.9%)                     |
| Ogulei <i>et al.</i> <sup>32</sup> | New York, USA (on-road, mobile)    | 6–500    | EEPS         | --   | Background urban emissions (39.5%), local/street diesel traffic (21.2%), aged/evolved diesel particles (15.5), fresh tail-pipe diesel exhaust (15.4%), spark-ignition gasoline emissions (4.3%) and secondary/transported material (4%)                                 |
| Ogulei <i>et al.</i> <sup>33</sup> | Baltimore, USA (roadside)          | 9.6–2458 | SMPS and APS | PM <sub>2.5</sub> , Gaseous (O <sub>3</sub> , NO <sub>x</sub> and CO), Metals and Chemical composition (sulphate, nitrate, EC and OC)            | Oil-fired power plant emissions, two secondary nitrates, local gasoline traffic, coal-fired power plant, secondary sulphate, diesel emissions/bus maintenance, Quebec wildfire episode, nucleation, incinerator, airborne soil/road-way dust, and steel plant emissions |
| Zhou <i>et al.</i> <sup>34</sup>   | Pittsburgh, USA (urban background) | 3–2500   | SMPS and APS | PM <sub>2.5</sub> , Gaseous (O <sub>3</sub> , NO <sub>x</sub> , NO, SO <sub>2</sub> and CO), Metals and Chemical composition (sulphate, nitrate) | Two secondary nitrates, remote traffic, secondary sulphate, lead, diesel traffic, coal-fired power plant, steel mill, nucleation, local traffic, and coke plant.  |
| Kim <i>et al.</i> <sup>14</sup>    | Seattle, USA (urban background)    | 20–400   | DMPS         | Gaseous (NO <sub>x</sub> and CO),  | Wood burning (48%*), secondary aerosol (21%*), diesel emissions (20%*) and motor vehicle emissions (11%*)   |
| Zhou <i>et al.</i> <sup>35</sup>   | Pittsburgh, USA (urban background) | 3–2500   | SMPS and APS | PM <sub>2.5</sub> , Gaseous (O <sub>3</sub> , NO, NO <sub>x</sub> , SO <sub>2</sub> and CO) and Chemical composition (sulphate, OC and EC).      | Sparse nucleation (28.2%), local traffic (21.7%), stationary combustion (21.1%), grown particles and remote traffic (20%) and secondary aerosol (9%)  |

- 1 Note: \*Contributions to particle volume concentration. SMPS = Scanning mobility
- 2 spectrometer; UDMA = Ultrafine differential mobility analyser; UCPC = Ultrafine
- 3 condensation particle counter; APS = Aerodynamic particle sizer; OPC = Optical particle
- 4 counter; MAS = Mobile aerosol spectrometer (comprising a combination of differential
- 5 mobility spectrometer, DMPS, and an optical laser aerosol spectrometer); EEPS = Engine
- 6 exhaust particle spectrometer.

1

2 Table 2: Hourly Pearson product-moment correlations, along with the significance level (p-

3 value), between each factor contribution and measured pollutants (PM<sub>10</sub>, O<sub>3</sub>, NO<sub>x</sub>, SO<sub>2</sub> and

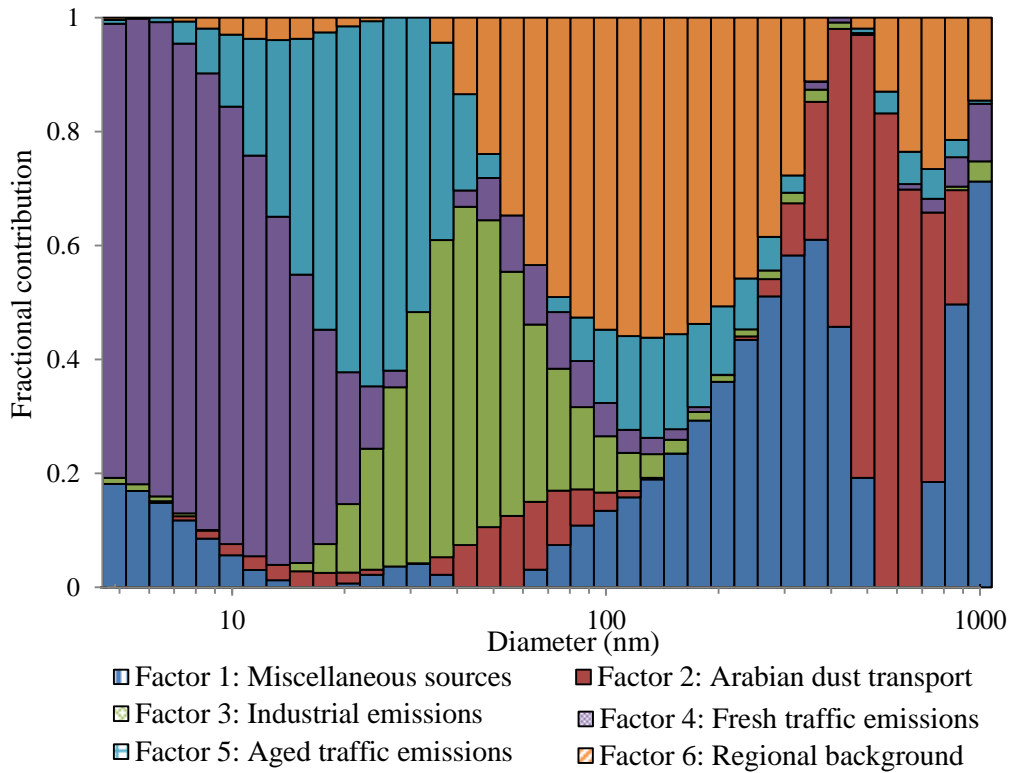
4 CO).

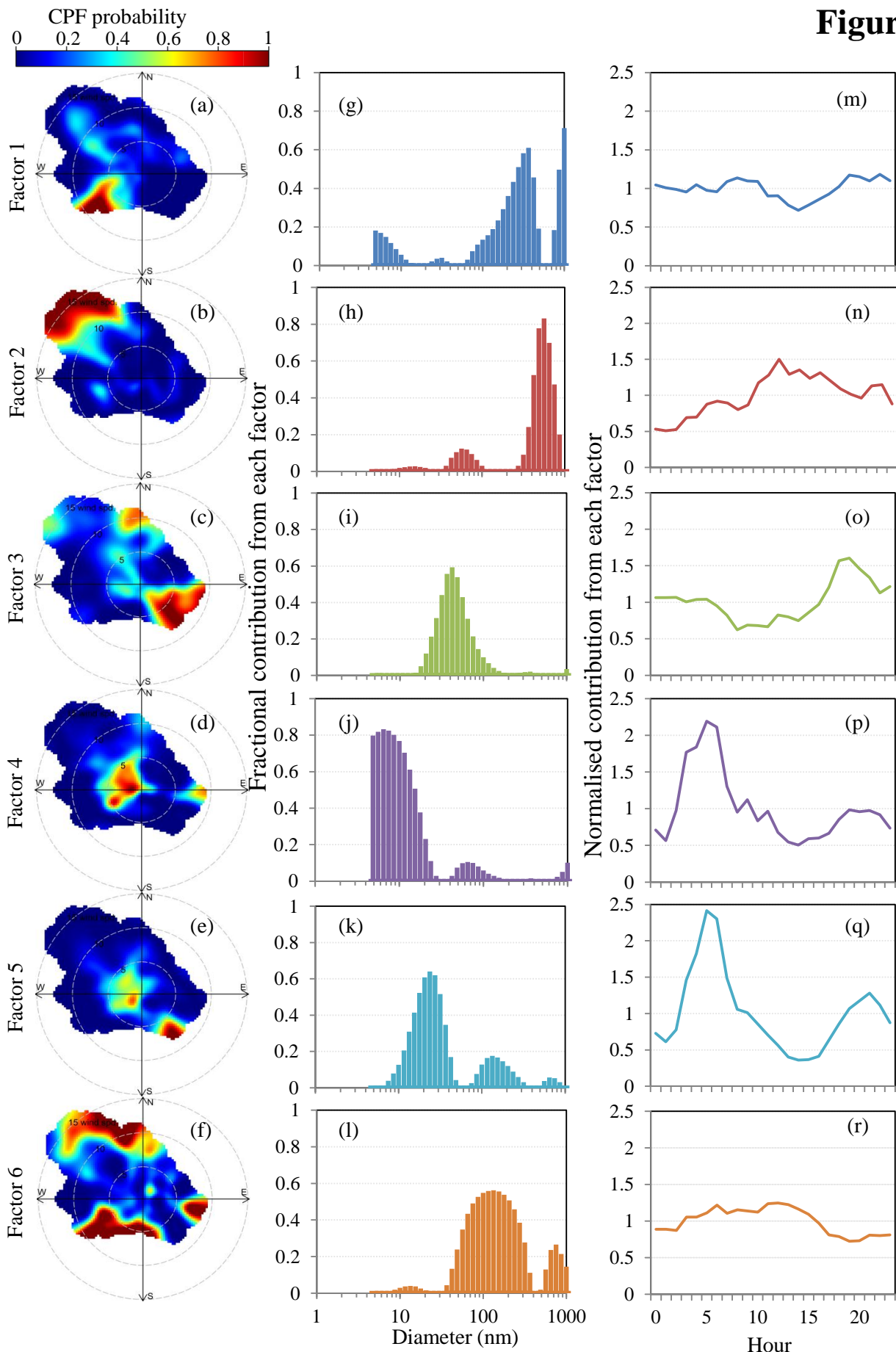
|                  | Factor 1           | Factor 2           | Factor 3           | Factor 4           | Factor 5           | Factor 6          |
|------------------|--------------------|--------------------|--------------------|--------------------|--------------------|-------------------|
| PM <sub>10</sub> | 0.39 <sup>a</sup>  | 0.71 <sup>a</sup>  | -0.10 <sup>a</sup> | -0.15 <sup>a</sup> | -0.16 <sup>a</sup> | 0.31 <sup>a</sup> |
| O <sub>3</sub>   | -0.14 <sup>a</sup> | 0.03 <sup>b</sup>  | -0.15 <sup>a</sup> | -0.39 <sup>a</sup> | -0.49 <sup>a</sup> | 0.14 <sup>a</sup> |
| NO <sub>x</sub>  | -0.04 <sup>a</sup> | -0.11 <sup>a</sup> | 0.37 <sup>a</sup>  | 0.30 <sup>a</sup>  | 0.54 <sup>a</sup>  | 0.01              |
| SO <sub>2</sub>  | -0.04 <sup>a</sup> | -0.04 <sup>a</sup> | 0.31 <sup>a</sup>  | -0.05 <sup>a</sup> | 0.05 <sup>a</sup>  | 0.19 <sup>a</sup> |
| CO               | -0.13 <sup>a</sup> | 0.01               | 0.23 <sup>a</sup>  | 0.07 <sup>a</sup>  | 0.23 <sup>a</sup>  | 0.02              |

5 <sup>a</sup>Correlation is significant at the 0.01 level. <sup>b</sup>Correlation is significant at the 0.05 level.

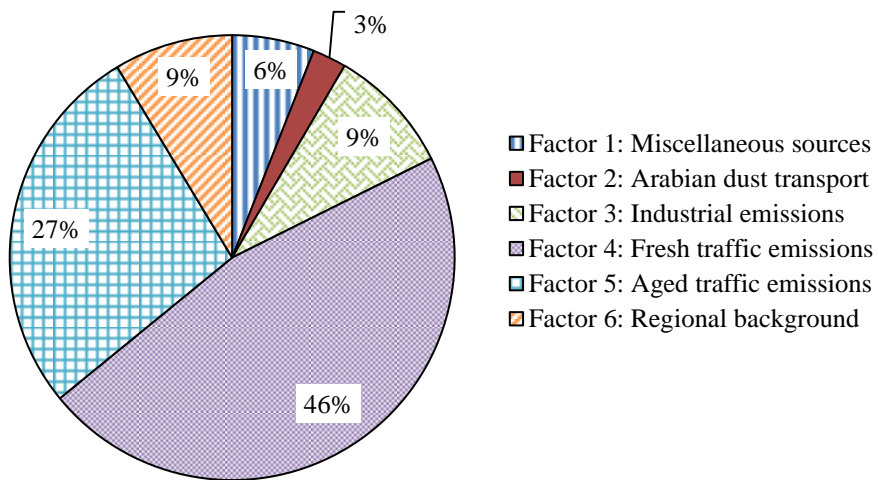


**Figure 1**

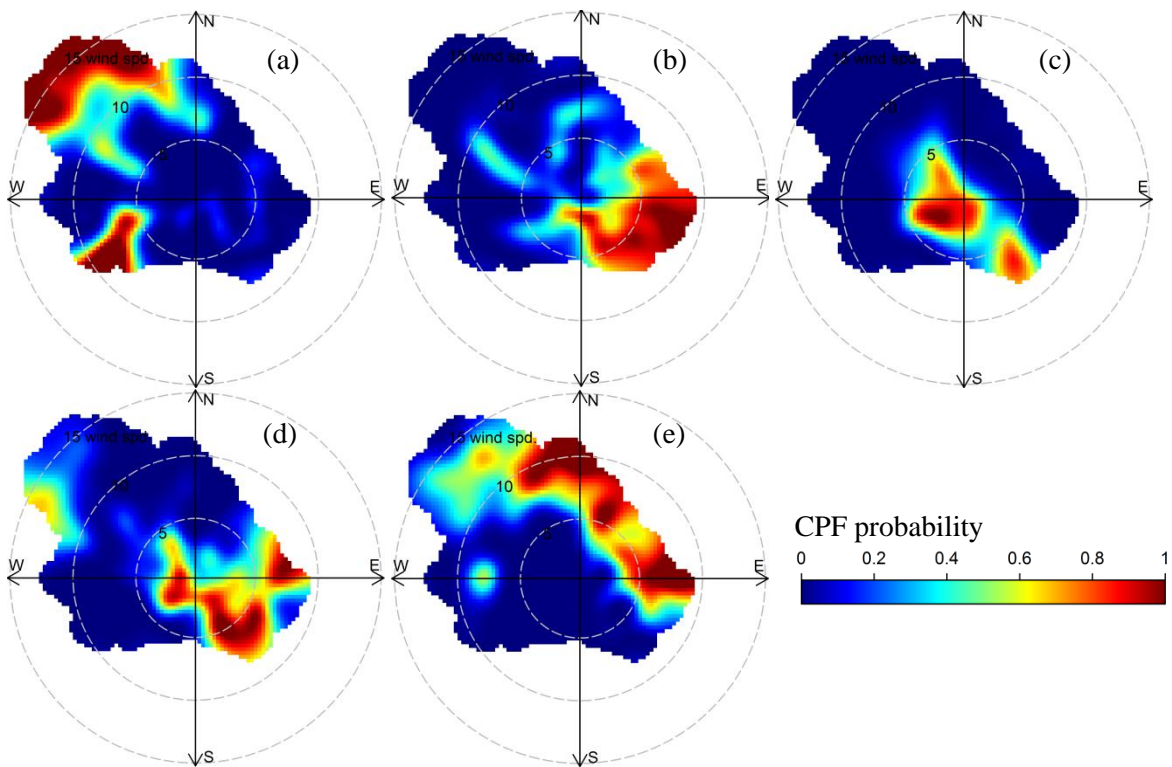
**Figure 2**







**Figure 4**

**Figure 5**