Source apportionment of airborne nanoparticles in a Middle Eastern city using positive matrix factorization

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ABSTRACT

Airborne nanoparticles have been studied worldwide, but little is known about their sources in the Middle East region, where hot, arid and dusty climatic conditions generally prevail. For the first time in Kuwait, we carried out size-resolved measurements of particle number distributions (PNDs) and concentrations (PNCs) in the 5–1000 nm size range. Measurements were made continuously for 31 days during the summer months of May and June 2013 using a fast–response differential mobility spectrometer (Cambustion DMS500) at a sampling rate of 10 Hz. Sources and their contributions were identified using the positive matrix factorization (PMF) approach that was applied to the PND data. Simultaneous measurements of gaseous pollutants (i.e., \(O_3\), NO, NO\(_x\), SO\(_2\) and CO), PM\(_{10}\), wind speed and direction were also carried out to aid the interpretation of the PMF results through the conditional probability function plots and Pearson product-moment correlations. Six major sources of PNCs were identified, contributing \(\sim\)46\% (fresh traffic emissions), 27\% (aged traffic

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

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

1. **Introduction**

Exposure to particulate matter (PM) is known to adversely affect human health. Ambient concentrations of PM are currently regulated through mass–based standards of PM_{10} and PM_{2.5}, i.e., aerodynamic diameters less than 10 and 2.5 \( \mu \text{m} \), respectively. Because of possessing negligible mass compared to the regulated PM, these standards do not control airborne nanoparticles that are referred to as those below 300 nm in diameter and represent the majority (~99%) of total particle number concentrations, PNCs. Nanoparticles are characterised by their vast numbers and high surface area. As a result, they can adsorb large concentrations of toxic hazardous chemicals on their surfaces, translocate and deposit in different parts of the human body and thereby causing adverse health effects. Evidences from a large number of studies link the exposure of nanoparticles to the occurrence of cardiovascular diseases. This effect is attributed to the translocation of the redox-active components of the nanoparticles in the human body, which promotes the progression of atherosclerosis. Furthermore, preliminary estimates of excess mortality related to nanoparticle exposure have been reported to be notable at 11,252 deaths in 2010 in Delhi and ~310,000 deaths per year in Asian megacities. However, such estimates are currently
unavailable for the Middle East region, clearly showing a need for field studies that can provide an in-depth insight into the sources of nanoparticles and associated health impacts.

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

Source apportionment models are important to identify various unknown sources and quantify their contributions towards the total measured concentrations. Such information is important to design efficient abatement strategies to control emissions. One of the most common receptor–based source apportionment models is positive matrix factorization (PMF), which can overcome the drawbacks of principle component analysis, PCA.\textsuperscript{17, 18} The output of PMF is more physically realistic than that of PCA because the former allows the implementation of non–negative constraints and production of explainable positive elements
among all factors. Other models such as the chemical mass balance (CMB) and Unmix are comparable to PMF, to some extent. However, PMF does not require prior knowledge of the sources and their profiles, as required in the case of CMB, thereby making it an easier and more cost-effective solution. Furthermore, PMF allows for the weighting of each data point individually\textsuperscript{18} – a feature that is not available in the Unmix model.

Prior to the incorporation of PND data in the PMF, this source apportionment technique has been applied for the identification of particles sources in many previous studies.\textsuperscript{19-22} However, these studies have mainly focused on PM mass concentrations and compositional data. PMF analysis, based on the PM chemical composition data, is often time-consuming and expensive, and does not segregate PNDs according to their sources. The knowledge of source-specific PNDs is of great relevance to epidemiological studies because of the size-dependency of respiratory tract deposition pattern in the human body on particle diameter.\textsuperscript{23} Several studies worldwide have successfully deployed total PNC data in the application of PMF to identify sources and their contributions over the past decade (see summary of relevant studies in Table 1). Few of these studies have used only PND data for the PMF analysis,\textsuperscript{24} while others have included PM chemical composition data,\textsuperscript{25} gaseous pollutant data,\textsuperscript{26} and chemical composition and gaseous pollutant data\textsuperscript{15} in their PMF analysis. In fact, none of the studies till date have applied PMF to the distinct PND characteristics found in the Middle East region, and therefore, the contributions of the different sources of PNC are currently unknown.

In order to fill the above–noted research gaps, we have applied PMF to our PND data set, ranging from 5–1000 nm, collected continuously over a 31-day period during summertime conditions at a roadside location in Fahaheel, Kuwait, by using a fast response differential mobility spectrometer (Cambustion DMS500). In addition, PM\textsubscript{10}, gaseous pollutants (NO\textsubscript{x},
O₃, CO and SO₂) and meteorological data were used to assist the interpretation of the PMF results by using conditional probability function (CPF).

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

In the light of the existing research gaps, the aims of this study are: (i) to identify the possible sources of nanoparticles in the studied area which represents a typical roadside environment
of the Middle East region, (ii) to quantify the sources contribution to total PNCs and (iii) to determine the individual PND spectrum of various sources in a Middle Eastern city, Kuwait.

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

<table>
<thead>
<tr>
<th>Author (year)</th>
<th>Location (type)</th>
<th>Size range (nm)</th>
<th>Instruments</th>
<th>Additional data</th>
<th>Sources identified (contribution of each source to the total apportioned PNC, %)</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>This study</strong></td>
<td>Fahaheel, Kuwait (roadside)</td>
<td>5–1000</td>
<td>DMS500</td>
<td>PM$_{10}$ and gaseous (O$_3$, NO, NO$_x$, SO$_2$ and CO)</td>
<td>Fresh traffic emissions (46% of the total apportioned PNC), aged traffic emissions (27%), industrial emissions (9%), regional background (9%), miscellaneous sources (6%), Arabian dust transport (3%)</td>
</tr>
<tr>
<td>Liu <em>et al.</em></td>
<td>Beijing, China (urban background)</td>
<td>14.5–2514</td>
<td>SMPS</td>
<td>Gaseous (O$_3$, NO, NO$_x$, CO and SO$_2$), and chemical composition (organic matter, sulphate, nitrate, ammonium and chlorine)</td>
<td>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%). Regional sources: accumulation mode (13.8%)</td>
</tr>
<tr>
<td><strong>Friend <em>et al.</em></strong></td>
<td>Brisbane, Australia (roadside)</td>
<td>14–715</td>
<td>SMPS</td>
<td>PM$_{10}$, gaseous (CO, NO and NO$_2$)</td>
<td>Petrol vehicles (30.8%), diesel traffic (28.1%), local traffic (14.9%), biomass burning (20.1%) and two unidentified sources (6%)</td>
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<tr>
<td><strong>Gu <em>et al.</em></strong></td>
<td>Augsburg, Germany (urban background)</td>
<td>3–10000</td>
<td>UDMA, UCPC and APS</td>
<td>Metals, water-soluble ions, elemental carbon (EC) and organic carbon (OC)</td>
<td>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%)</td>
</tr>
<tr>
<td>Harrison <em>et al.</em></td>
<td>London, UK (curbside)</td>
<td>15–10000</td>
<td>SMPS and APS</td>
<td>Gaseous (O$_3$, NO, NO$_x$ and CO) and traffic flow</td>
<td>Road emissions: solid-mode exhaust (18.8%), brake dust (13.7%), re-suspended dust (4.4%) and nucleation-mode exhaust (3.6%). 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%)</td>
</tr>
<tr>
<td>Kasumba <em>et al.</em></td>
<td>New York, USA (urban background)</td>
<td>100–470</td>
<td>SMPS</td>
<td>PM$_{2.5}$ and gaseous (CO, SO$_2$ and O$_3$)</td>
<td>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%)</td>
</tr>
<tr>
<td>Thimmaiah <em>et al.</em></td>
<td>Prague, Czech</td>
<td>18.8–723.5</td>
<td>SMPS</td>
<td>Gaseous (CO, SO$_2$, NO$_x$, O$_3$)</td>
<td>NO$_x$-rich (influenced by diesel emissions, 37.8%), gasoline traffic (34.2%), heating (24.6%) and...</td>
</tr>
<tr>
<td>Authors</td>
<td>Location</td>
<td>Methodology</td>
<td>PM$_{2.5}$ Composition</td>
<td>Source Apportionment</td>
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<tr>
<td>Al-Dabbous, A.N., Kumar, P., 2015</td>
<td>Republic (urban background)</td>
<td>DMPS</td>
<td>CH$_4$, Non Methane Hydrocarbons and Total Hydrocarbons</td>
<td>oozne-rich (mainly influenced by meteorology, 3.5%)</td>
<td></td>
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<tr>
<td>Yue et al., 2004</td>
<td>Erfurt, Germany (roadside)</td>
<td>MAS</td>
<td>Gaseous (O$_3$, NO, NO$_2$, CO and SO$_2$) and Chemical composition (sulphate, EC and OC)</td>
<td>Ultrafine particles from local traffic (79%), secondary aerosols from multiple sources (6%), particles from remote traffic sources (5%) and airborne soil (1%)</td>
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<tr>
<td>Ogulei et al., 2014</td>
<td>New York, USA (urban background)</td>
<td>SMPS</td>
<td>PM$_{2.5}$, gaseous (CO, SO$_2$ and O$_3$)</td>
<td>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%)</td>
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<tr>
<td>Ogulei et al., 2014</td>
<td>New York, USA (on-road, mobile)</td>
<td>EEPS</td>
<td>--</td>
<td>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%)</td>
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<tr>
<td>Ogulei et al., 2014</td>
<td>Baltimore, USA (roadside)</td>
<td>SMPS and APS</td>
<td>PM$_{2.5}$, Gaseous (O$_3$, NO$_x$, CO), Metals and Chemical composition (sulphate, nitrate, EC and OC)</td>
<td>Oil-fired power plant emissions, second 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</td>
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<tr>
<td>Zhou et al., 2014</td>
<td>Pittsburgh, USA (urban background)</td>
<td>SMPS and APS</td>
<td>PM$_{2.5}$, Gaseous (O$_3$, NO$_x$, NO, SO$_2$ and CO), Metals and Chemical composition (sulphate, nitrate)</td>
<td>Two secondary nitrates, remote traffic, secondary sulphate, lead, diesel traffic, coal-fired power plant, steel mill, nucleation, local traffic, and coke plant.</td>
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<tr>
<td>Kim et al., 2014</td>
<td>Seattle, USA (urban background)</td>
<td>DMPS</td>
<td>Gaseous (NO$_x$ and CO),</td>
<td>Wood burning (48%<em>), secondary aerosol (21%</em>), diesel emissions (20%<em>) and motor vehicle emissions (11%</em>)</td>
<td></td>
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<tr>
<td>Zhou et al., 2014</td>
<td>Pittsburgh, USA (urban background)</td>
<td>SMPS and APS</td>
<td>PM$_{2.5}$, Gaseous (O$_3$, NO, NO$_x$, SO$_2$ and CO) and Chemical composition (sulphate, OC and EC)</td>
<td>Sparse nucleation (28.2%), local traffic (21.7%), stationary combustion (21.1%), grown particles and remote traffic (20%) and secondary aerosol (9%)</td>
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</tr>
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</table>

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

2. Experimental methods

2.1 Site description

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

Measurements were made during summertime in the month of May and June 2013 when ambient temperature reached to ~48 °C, the relative humidity decreased to a minimum of 0.20% and the dust events (i.e., when PM$_{10}$ >200 µg m$^{-3}$) were observed for ~49% of the total measurement time. The average temperature, relative humidity and wind speed were found to be 37±4.5 °C, 13.6±10.0% and 6.3±3.0 m s$^{-1}$, respectively. The prevailing wind direction was north-west (~311°N). Wind speed and ambient temperature affected the PNCs
notably. For example, ambient temperature was found to linearly decrease the PNCs due to partial evaporation\textsuperscript{16}; see details in Supplementary Information, SI, Section S1.

\textbf{Figure 1.} Location of the sampling site in the Fahaheel area, showing the major sources surrounding the site. Satellite image includes material ©NSPO 2014 distribution Spot Image S.A.; courtesy of Airbus Defence and Space, all rights reserved. Note: SIA = Shuaiba Industrial Area; WSIA = West Shuaiba Industrial Area.

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

2.2 Data acquisition

A total of 8675 valid 5–minute PND observations, each in 36 size classes, covering 5–1000 nm size range, were continuously measured from 27 May to 26 June 2013 by using a DMS500. These measurements were collected at 0.10–second time resolutions and then averaged to 5–min interval means to synchronise them with the pollutants and meteorological data. DMS500 is a parent version of DMS50 (i.e., portable instrument with similar features) that has been successfully used in a variety of our studies, related to roadside and kerbside measurements, vehicle-wake, vehicle in-cabin and indoor construction environments. The DMS500 detects particles based on their electrical mobility. Additionally, a suite of pollutants (PM$_{10}$, O$_3$, NO$_x$, SO$_2$ and CO) and meteorological parameters (temperature, relative humidity, wind speed and direction) were obtained from the adjacent (~300 m away from site) Environmental Protection Agency (EPA) monitoring station. These continuous data are well-maintained and quality-controlled by the Kuwait EPA. Further details on the experimental setup, instrumentation and working principle of various instruments can be seen elsewhere.

2.3 Statistical analysis

PMF analysis was applied using the US EPA’s PMF program (version 5.0) on the dataset composed of 36 variables. These variables included PNDs in 36 size classes covering
a size range of 5–1000 nm, following the methodology described in Paatero\textsuperscript{18}. PMF is a multivariate factor analysis model used to identify the contribution and profile by exposing the dataset to a multi-linear engine algorithm and a gradient algorithm approach in order to find the best-fit solution.\textsuperscript{44,45} This method is featured by the non–negative constraints and the use of uncertainties to scale individual data points. The uncertainty data file supplied by the instrument manufacturer (Cambustion Ltd., Cambridge), consisting of size–specific minimum detection limits and error fractions, was also included in the PMF. An extra modelling uncertainty of 5\% was added to the model to account for any additional measurement errors that were not covered by the uncertainty data file.\textsuperscript{46} The missing sampling values due to instrument failure were modest (i.e., <3\% of the entire sampling period) and simply excluded from the analysis. In addition, CPF plots were prepared using the threshold of the upper 25\textsuperscript{th} percentile of the fractional contribution of each factor/source. These plots complemented the PMF analysis by depicting the trend in the factors score with wind direction and speed so that factors could be tentatively assigned to the potential sources in the area.\textsuperscript{47} Furthermore, CPF plots were also drawn for the routinely measured pollutants (PM\textsubscript{10}, O\textsubscript{3}, NO\textsubscript{x}, SO\textsubscript{2} and CO) by using the same criterion. Open Air (R package), which is an open-source statistical tool,\textsuperscript{48} was used to derive the CPF plots that assisted in the interpretation of the measured air pollution data.

3. Results and discussion

Using the PMF approach described in Section 2.3, six different factors were identified that were then tentatively assigned to the potential sources based on the following information: (i) factor-specific PNDs (Figure 2 and Figure 3g-l, middle vertical panel), (ii) diurnal variation of the factors (Figure 3m-r, right vertical panel), (iii) contribution of each factor to the total PNC (Figure 4), (iv) hourly Pearson product–moment correlations, along
with the significance level (p-value), between each factor contribution and measured gaseous
(O₃, NOₓ, SO₂ and CO) and PM₁₀ pollutants (Table 2), and (v) the CPF plots for each factor
contribution (Figure 3a-f, left vertical panel) and measured gaseous (O₃, NOₓ, SO₂ and CO)
and PM₁₀ pollutants (Figure 5).

Figure 2. Percent contribution of each source identified towards the PNCs in different size
ranges.

3.1 Factor 1: Miscellaneous sources

This factor showed multiple PND modes, with the major peaks at about 365 nm and
1000 nm, and a positive correlation with PM₁₀ (r = 0.39; p-value < 0.01; Table 2). Factor 1
also showed a minor peak at ~5 nm, which could represent fresh traffic emissions but to
lesser extent than that observed for factors 4 and 5. Furthermore, the wind directionality and
the relatively high wind speed (up to 10 m s⁻¹) of this factor (Figure 3a) and PM₁₀ (Figure 5a)
indicated that the particle emissions had travelled from a remote location and grown to larger
sizes through coagulation. Al-Dabbous and Kumar¹⁶ previously reported a dominating role of
PM$_{10}$ in suppressing PNCs due to coagulation process. For instance, PNCs were found to be reduced by ~23% when PM$_{10}$ concentration increased by ~500%, compared to the values prior to the arrival of the dust event (i.e., when PM$_{10} < 200$ $\mu$g m$^{-3}$). A similar observation on coagulation scavenging has been reported by Jayaratne et al.$^{49}$ with respect to the influence of the Australian dust storm on the PNCs. This factor made the second lowest contribution (6%) to the total PNCs. The directionality of the CPF plots and the association with PM$_{10}$ clearly corresponds to the west Shuaiba industrial area and the dust blown by high wind speed from the desert during the south-westerly winds.

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

3.2 Factor 2: Arabian dust transport

This factor showed a bimodal PND (Figure 3i) with a major peak at 560 nm, and a minor peak at 60 nm, along with a distinctively high correlation with PM$_{10}$ ($r = 0.71$; p-value <0.01; Table 2). The wind directionality and the associated high speed levels (more than 15 m s$^{-1}$) noted in CFP plots of this factor (Figure 3b), as well as PM$_{10}$ (Figure 5a), indicate the influence of the dust from the long–range transport that is associated with the typical Arabian dust events. This factor showed behaviour similar to that of factor 1, but to a greater extent in terms of higher PM$_{10}$, wind speed levels and the typical directionality (i.e., north–westerly direction) associated with the frequent dust events in the region. In an extended analysis of the same dataset, Arabian dust events were found to suppress PNCs due to the influence of coagulation process, which explains the minimum contribution (3%) of this factor to the total PNC (Figure 4). It is worth pointing out that both the factors 1 (6%) and 2 (3%) made the lowest contributions (Figure 4), among the six resolved factors, but showed the highest correlations with PM$_{10}$; these characteristics support the possible effects of the coagulation process during high concentrations of PM$_{10}$ approaching the site from the westerly wind direction (i.e., open desert; Figure 1). Furthermore, the diurnal profile of this factor showed an increased contributions during the afternoon (12:00 to 14:00 h; Figure 3n) due to relatively higher wind speeds and associated saltation process. In an extended analysis on the same dataset (SI Figure S1), but excluding the major dust event periods (i.e., when PM$_{10}$ >1000 µg m$^{-3}$), we observed almost similar contribution to the total PNCs (Figure S2) to those observed in Figure 4 for all the six sources. Pearson product–moment correlations between each factor and measured gaseous (O$_3$, NO$_x$, SO$_2$ and CO) and PM$_{10}$ pollutants also exhibited similar
correlations (Table S1) to those observed in Table 2. This similarity confirms that the input dataset were not highly affected by the Arabian dust events, mainly because the major dust event periods were only 5.7% of the total measurements period.

Figure 3. Directionality of the factor contribution using CPF plots at 75th percentile level, considering both local wind direction and speed (Figure 3a-f). The colours in Figure 3a-f represent the probability of factor contribution with respect to wind direction and speed.
Figures 3g-l represent the factor-specific PND profiles while Figures 3m-r show the diurnal variation of the normalised factor contribution.

### 3.3 Factor 3: Industrial emissions

This factor showed a monomodal distribution with a peak at ~42 nm (Figure 3i), and made a 9% contribution to the total PNCs (Figure 4). The diameter of this peak was in accordance with those recorded for industrial emissions in previously published studies. For instance, Ogulei et al.\(^3\) reported a peak at 44 nm during their one–year long measurements (2004–2005) at an urban background location in New York (USA) that was significantly influenced by the industrial activities. We have several reasons to believe that factor 3 represents industrial emissions. For example, the CPF plots of this factor are strongly associated with south easterly winds (Figure 3c), which is consistent with the wind directionality of SO\(_2\) (Figure 5b). The directionality of these plots clearly correspond to the Shuaiba industrial area, which hosts a range of oil refineries (i.e., Mina Al-Ahmadi, Shuaiba and Mina Abdullah refinery), petrochemical industries (e.g., ammonia, urea, polyethylene and polypropylene plant) and two power desalination plants.\(^5\)\(^1\), \(^5\)\(^2\) Furthermore, this factor had the highest correlation (\(r = 0.31; \text{p-value} < 0.01\)) with SO\(_2\) among all the factors (Table 2), which supports the fact that industrial emissions are clearly associated with this factor. Moreover, NO\(_x\) (\(r = 0.37; \text{p-value} < 0.01\)) and CO (\(r = 0.23; \text{p-value} < 0.01\)) also showed a positive correlation with this factor, indicating an association with the combustion activities within the vicinity of the industrial area. Past studies have also linked industrial emissions with the combustion related pollutants, mainly SO\(_2\).\(^2\)\(^9\), \(^3\)\(^1\), \(^3\)\(^3\), \(^3\)\(^5\) The association with SO\(_2\) may indicate the influence of secondary particle formation in the form of photo-chemically induced sulphuric-acid nucleation.\(^5\)\(^3\), \(^5\)\(^4\) The diurnal profile of this factor displayed a typical diurnal variation, linked with the meteorological conditions and the associated boundary layer.\(^5\)\(^5\) For example, a decreased factor contribution was observed during the afternoon, which
was caused by the expanded depth of the boundary layer and the associated dilution with the background air. Based on the above concluding evidences, we attributed this factor to the industrial emissions.

![Factor 1: Miscellaneous sources
Factor 2: Arabian dust transport
Factor 3: Industrial emissions
Factor 4: Fresh traffic emissions
Factor 5: Aged traffic emissions
Factor 6: Regional background]

**Figure 4.** Sources contribution (%) to the total PNC data in the urban area of Fahaheel, Kuwait.

Table 2: Hourly Pearson product-moment correlations, along with the significance level (p-value), between each factor contribution and measured pollutants (PM$_{10}$, O$_3$, NO$_x$, SO$_2$ and CO).

<table>
<thead>
<tr>
<th></th>
<th>Factor 1</th>
<th>Factor 2</th>
<th>Factor 3</th>
<th>Factor 4</th>
<th>Factor 5</th>
<th>Factor 6</th>
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</thead>
<tbody>
<tr>
<td>PM$_{10}$</td>
<td>0.39$^a$</td>
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<td>-0.16$^a$</td>
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<td>0.03$^b$</td>
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<td>-0.39$^a$</td>
<td>-0.49$^a$</td>
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<tr>
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<td>0.37$^a$</td>
<td>0.30$^a$</td>
<td>0.54$^a$</td>
<td>0.01</td>
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<td>-0.04$^a$</td>
<td>0.31$^a$</td>
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<td>0.05$^a$</td>
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<td>CO</td>
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<td>0.23$^a$</td>
<td>0.07$^a$</td>
<td>0.23$^a$</td>
<td>0.02</td>
</tr>
</tbody>
</table>

$^a$Correlation is significant at the 0.01 level. $^b$Correlation is significant at the 0.05 level.

### 3.4 Factor 4: Fresh traffic emissions

This factor showed a major PND peak between 5–12 nm and another minor peak at ~60 nm (Figure 3j) and explained nearly half (46%; Figure 4) of the total PNC contribution. Looking at the PND and the peaks, this contribution was believed to be from the local traffic.
For example, this bimodal profiles of PNDs are consistent with those observed by Fujitani et al.\(^{56}\) at 10 nm and 40–60 nm during their near road measurements in Kanagawa Prefecture, Japan. Furthermore, similar PND peaks related to local traffic were observed by numerous studies performed in cities worldwide, such as at 20 nm (major peak) and 100 nm (minor peak) in Beijing, China,\(^{15}\) 20 nm in Brisbane, Australia,\(^{28}\) 9–40 nm in Augsburg, Germany,\(^{25}\) 10–100 nm in Erfurt, Germany,\(^{24}\) 20 nm in London, UK,\(^{26}\) 13.3 nm in Cambridge, UK,\(^{5}\) 10 nm in New York, USA,\(^{31}\) 15 nm in Pittsburgh, USA.\(^{35}\) The wind directionality (Figure 3d) corresponded to the highway located at 15 m west of the measurement location, and the wind speed was observed to be relatively low (<5 m \(s^{-1}\)) compared with much higher levels noted during the major dust events. This low level of wind speed indicates an association with close-range source (i.e., local traffic). The directionality of the factor contribution is also consistent with those for NO\(_x\) (Figure 5c) and CO (Figure 5d), especially from the westerly wind direction, indicating the same emission source. Furthermore, this factor contribution correlated positively with the NO\(_x\) (\(r = 0.30;\) p-value <0.01), which is a primary traffic–generated pollutant.\(^{57, 58}\) The diurnal profile of this factor contribution (Figure 3p) was in agreement with the diurnal pattern of the traffic volume, except during the noon hours when the high traffic volume corresponded to low factor contribution. The reason for this odd behaviour was previously studied in an extended analysis by Al-Dabbous and Kumar\(^{16}\) and explained by the extreme temperature (reaching up to \(\sim 50\) °C) that resulted in partial evaporation and increased rate of coagulation with larger particles.\(^{59}\) Most of the above-discussed studies also observed higher PND magnitude in the morning rush hours compared with those during evening rush hours; this is consistent with the findings of our current study. Based on the above observations, we attributed this factor to local traffic emissions, seen through the newly formed particles (i.e., fresh traffic emissions) in nucleation mode.
3.5 Factor 5: Aged traffic emissions

This factor showed a major peak at 24 nm, followed by a minor peak at 130 nm (Figure 3k). The former peak is presumably attributed to the nearby highway emissions, and the latter, to aged particles transported from the industrial area. These bimodal profiles of PNDs are similar to those observed by Gu et al.\textsuperscript{25} at 20 and 100 nm during their measurements in Augsburg, Germany, and attributed them to aged traffic emissions. This factor showed the second highest contribution to the total PNC (27%; Figure 4). This factor was positively correlated with NO\textsubscript{x} ($r = 0.54$; p-value <0.01) and CO ($r = 0.23$; p-value <0.01) and showed no correlation with SO\textsubscript{2}. Moreover, the CPF shown in Figure 3e clearly pointed out the wind direction from the Shuaiba industrial area (i.e., south-easterly direction) and the traffic emission from Fahaheel highway (i.e., westerly direction). This wind directionality is
identical to those obtained for NO\(_x\) (Figure 5c) and CO (figure 5d). Therefore, the correlations with the NO\(_x\) and CO as well as the CPF suggest that there is a contribution from primary (solid carbonaceous) particles from diesel vehicles from the nearby industrial area and the Fahaheel highway. However, absence of such correlations with the SO\(_2\) suggests a negligible contribution of secondary particle formation through photo–chemically induced sulphuric–acid nucleation like what is noticed in case of factor 3. Furthermore, the diurnal profile of this factor contribution (Figure 3q) was similar to the profile of factor 4, with a slight increase in the evening hours, indicating the influence of nocturnal commercial traffic (e.g., heavy duty trucks) operating on the Fahaheel highway and within the industrial area. In total, both the fresh (factor 4) and aged (factor 5) traffic emissions accounted for about 73% of the total PNCs, which is comparable to roadside studies in London, UK (~72%)\(^{26}\) and Brisbane, Australia (~74%)\(^{28}\).

### 3.6 Factor 6: Regional background

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

4. **Summary and conclusions**

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

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

This study covers a hitherto overlooked topic in the Middle East region. The findings of this work make contributions towards the understanding of potential sources of nanoparticles in the area and their probable contribution to the PNCs. Furthermore, PND profiles associated with individual sources present an important reference data for future studies in the Middle East region. Long-term measurement studies, involving more pollutants (e.g., trace metals and organic compounds), are recommended to elucidate further on specific source characteristics and their emission strengths.

5. Acknowledgements

The authors are grateful to the Kuwait Institute for Scientific Research (KISR) for experimental and funding support for Abdullah’s PhD research. We also thank Professors Alan Robins (University of Surrey) and Min Hu (Peking University), and Drs Jianfei Peng (Peking University) and Jonathan Symonds (Cambustion Ltd) for their valuable contributions and discussion to develop this article. We also thank the Kuwait National Meteorological Network and Kuwait Environmental Protection Agency for their cooperation in providing us the meteorological and gaseous pollutants data.
6. References


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Supporting Information (SI)

for

Source apportionment of airborne nanoparticles in a Middle Eastern city using Positive Matrix Factorization

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The SI includes:

Section S1

Figures S1-S3

Tables S1

References used
S1. Effect of ambient temperature and wind speed on PNCs

The studied area is featured by its high temperature (up to ~48 °C). Ambient temperature and wind speed were key parameters affecting the PNCs in various size ranges. The variations in ambient temperature and wind speed varied in the 28-48 °C and 0.26-15.25 m s⁻¹ ranges, respectively. Sufficient variation in ambient temperature allows assessing the influence of temperature on PNCs. We normalised the total PNCs by the traffic volume to remove the dependency of PNCs on the traffic volume. The influence of wind direction on PNCs were also removed by only selecting one wind direction (i.e., data from the north-westerly winds was used that represented the majority of the data¹). Then, the influence of temperature on PNCs were analysed under three wind speed ranges that is low (2-3 m s⁻¹; representing 10% of the selected data), medium (5-7 m s⁻¹; representing 25% of the selected data), and high (9.5-10.5 m s⁻¹; representing 10% of the selected data) wind speeds ranges. A decrease in PNCs was observed with the increase in ambient temperature for all studied wind speed ranges, when fitted to a linear trend line, and this decrease was optimum for the low wind speed range (see Figure S3). The reason for this decrease was attributed to the partial evaporation,² caused by the increase temperature, and hence an increase in coagulation rate.² Detailed information about the influence of meteorological parameters on PNCs are thoroughly discussed in our previous work.¹
Figure S1: Directionality of the factor contribution using CPF plots at 75th percentile level for the non-dusty period, considering both local wind direction and speed (left vertical panels). The colours in CPF plots represent the probability of factor contribution with respect to wind direction and speed. Figures in the middle vertical panels represent the factor-specific PND profiles for the non-dusty period. Figures in the right vertical panels represent the diurnal variation of the normalised factor contribution for the non-dusty period.
Figure S2: Sources contribution (%) to the total PNC data in the urban area of Fahaheel, Kuwait, for the non-dusty period.

Figure S3: The relationship between normalised PNCs and ambient temperature under low (2-3 m s⁻¹; representing 10% of the selected data) medium (5-7 m s⁻¹; representing 25% of the selected data), and high (9.5-10.5 m s⁻¹; representing 10% of the selected data) wind speed ranges.

Table S1: Hourly Pearson product-moment correlations, along with the significance level (p-value), between each factor contribution and measured pollutants (PM$_{10}$, O$_3$, NO$_x$, SO$_2$ and CO) for the non-dusty period.

<table>
<thead>
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<th>Factor 4</th>
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<td>PM$_{10}$</td>
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<td>0.15</td>
<td>0.31</td>
<td>0.13</td>
</tr>
</tbody>
</table>

*Correlation is significant at the 0.01 level.

References