

We thank the reviewer for the suggestions to improve the manuscript. Our responses to the two comments are provided as follows.

Two minor revisions are suggested:

1) The authors say that they are unable to separate local vehicular emissions from the mixed source, but they also find that EC emissions are lower during the weekend. I would argue that is a good indicator of local traffic emissions, even if not all of it (some people still drive on the weekends.) Gas flares and cement industries probably are active all week long, thus the weekday/weekend difference should indicate local sources.

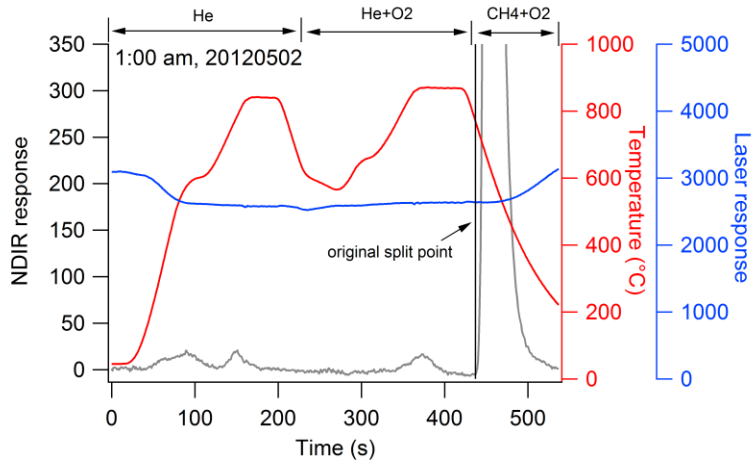
Response:

We modified line 290 to 295 as follows: "This reduction may be ascribed to the decrease in the local vehicular activities and industrial activities during the weekend. Therefore, local EC can be roughly estimated to be $0.51 \mu\text{g m}^{-3}$, about 22% of total EC, by the difference in average EC concentrations between weekday and weekend. As there were still some local traffic and industrial activities during the weekend, this estimate is likely a lower bound of the local contribution to EC concentrations in the study. " We also added one sentence near line 474 of the conclusion, "at least 22% of EC was ascribed to local sources".

2) The authors should check if the low EC in May samples is due to premature EC evolution due to increased refractory matter on the filter substrate. If not, they could add a statement to that effect.

Response:

We checked the thermograms of May samples and most of the samples are similar to Fig. A. 2 (b), shown in the following figure. Unlike Fig. A. 2 (c), where the incorrect split point can be seen clearly in He phase, it is not possible to state that premature EC evolution did or did not occur. In general, however, the sum of OC+EC was low in May.



Sources of PM_{2.5} carbonaceous aerosol in Riyadh, Saudi Arabia

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Abstract

Knowledge of the sources of carbonaceous aerosol affecting air quality in Riyadh, Saudi Arabia is limited, but needed for the development of pollution control strategies. We conducted sampling of PM_{2.5} from April to September, 2012 at various sites in the city, and used a thermo-optical semi-continuous method to quantify the organic carbon (OC) and elemental carbon (EC) concentrations. The average OC and EC concentrations were 4.7 ± 4.4 and 2.1 ± 2.5 $\mu\text{g m}^{-3}$, respectively, during this period. Both OC and EC concentrations had strong diurnal variations, with peaks at 6-8 am and 20-22 pm, attributed to the combined effect of increased vehicle emissions during rush hour and the shallow boundary layer in the early morning and at night. This finding suggested a significant influence of local vehicular emissions on OC and EC. The OC/EC ratio in primary emissions was estimated to be 1.01, close to documented values for diesel emissions. Estimated primary (POC) and secondary (SOC) organic carbon concentrations were comparable, with average concentrations of 2.0 ± 2.4 and 2.8 ± 3.4 $\mu\text{g m}^{-3}$, respectively.

We also collected 24 hour samples of PM₁₀ onto quartz microfiber filters and analyzed these for an array of metals by ICP-OES. Total OC was correlated with Ca (R^2 of 0.63), suggesting that OC precursors and Ca may have similar sources, and the possibility that they underwent similar atmospheric processing. In addition to a ubiquitous dust source, Ca is emitted during desalting processes in the numerous refineries in the region and from cement kilns, suggesting these sources may also contribute to observed OC

34 concentrations in Riyadh. Concentration weighted trajectory (CWT) analysis showed that
35 high OC and EC concentrations were associated with air masses arriving from the
36 Persian Gulf and the region around Baghdad, locations with high densities of oil fields
37 and refineries as well as a large Saudi Arabian cement plant. We further applied positive
38 matrix factorization to the aligned data set of EC, OC and metal concentrations (Al, Ca,
39 Cu, Fe, K, Mg, Mn, Na, Ni, Pb and V). Three factors were derived, and were proposed to
40 be associated with oil combustion, industrial emissions (Pb-based), and a combined
41 source from oil fields, cement production, and local vehicular emissions. The dominant
42 OC and EC source was the combined source, contributing $3.9 \mu\text{g m}^{-3}$ (80%) to observed
43 OC and $1.9 \mu\text{g m}^{-3}$ (92%) to observed EC.

44 **1. Introduction**

45 Organic carbon (OC) and elemental carbon (EC) (or black carbon, BC, operationally
46 identified based on detection method) are key components of the atmospheric aerosol
47 (Jacobson et al., 2000). The contribution of carbonaceous components to total particulate
48 matter (PM) concentrations varies with site and season, comprising from 20 to 90% of the
49 total mass (Kanakidou, et al., 2005). EC is emitted from a variety of combustion processes
50 (Bond et al., 2013), is classified as a short-lived climate forcer that contributes to
51 atmospheric warming (Ramanathan and Carmichael, 2008), and is also associated with
52 human morbidity and mortality (Weinhold, 2012). OC includes both direct emissions
53 (primary organic carbon, POC) and secondary OC (SOC) formed in the atmosphere via
54 oxidation (Robinson et al., 2007). Common sources of atmospheric POC and of SOC
55 precursors are vehicular exhaust, industrial emissions, biogenic emissions, and biomass
56 burning (Millet et al., 2005; Saarikoski et al., 2008; Genberg et al., 2011; Hu et al., 2012;
57 Vodička et al., 2013; Heal and Hammonds, 2014; Huang et al., 2014a, b). Except near
58 strong emission sources, secondary organic aerosol is the main contributor to the total
59 organic aerosol mass concentration, frequently accounting for $72 \pm 21\%$ (Zhang et al. 2007;
60 Jimenez et al., 2009).

61 Trace metals account for only a small fraction of PM mass concentrations, but they can
62 adversely impact human health (e.g., Lippmann et al., 2006; Hong et al., 2010). As some
63 emission sources release specific trace elements, these elements can serve as useful
64 source markers in PM source apportionment studies (Lee et al., 2011; Peltier and
65 Lippmann, 2010; Han et al., 2005; Harrison et al., 2012; Karanasiou et al., 2009; Ondov
66 et al., 2006; Querol et al., 2007; Viana et al., 2008; Yu et al., 2013). Elemental
67 enrichments can also be used to roughly differentiate natural and anthropogenic sources
68 (Khodeir et al., 2012; Rushdi, et al., 2013). Relative abundances of crustal elements can
69 help identify the sources of suspended dust, as these abundances are known to be
70 different for different dust source regions (Engelbrecht et al., 2009).

72 In this study, we report measurements of ambient particulate matter in Riyadh, the capital
of Saudi Arabia. In prior studies conducted in the Middle East, dust was identified as the
74 major source of PM₁₀ (Givehchi, et al., 2013); however, contributions from anthropogenic
sources to PM mass concentrations were found to be significant (>82% of total PM₁₀ mass,
76 Al-Dabbous and Kumar, 2015; >50% of PM₁₀ Tsiouri et al., 2015). Tsiouri et al. (2015)
summarized the major sources of PM₁₀ in ambient air in the Middle East as oil combustion,
78 re-suspended soil, road traffic, crustal dust, and marine aerosol; significant sources of
PM_{2.5} were oil combustion in power plants, re-suspended soil, sand dust, and road traffic.
80 Carbonaceous particles were estimated to account for 50-60% of PM_{2.5} in cities in
Palestine, Jordan, and Israel (Abdeen et al., 2014). Not surprisingly, since oil production
and processing was widespread across the Middle East, heavy oil combustion was
82 estimated to contribute 69% to PM_{2.5} mass and 18% to PM₁₀ in in Jeddah, Saudi Arabia
(Khodeir et al., 2012). Air quality in Riyadh reflects not only the impact of local and
84 regional dust and regional oil extraction and refining, but also significant local sources
that include a heavy traffic load and multiple industries. We focus here on identifying the
86 major sources of PM_{2.5} carbonaceous aerosol in Riyadh to provide a basis for formulating
air pollutant mitigation strategies.

88 **2. Methodology**

2.1 Sampling sites and data collection

90 Riyadh and its environs were divided into 16 12 km × 12 km sampling cells as shown in
Fig. 1. Sampling locations within each cell were carefully chosen to best represent the
92 mix of land use and other characteristics of the cell. From April through September, 2012,
an in-situ semi-continuous OC/EC analyzer (Sunset Laboratory Inc., Model-4), installed
94 in a mobile laboratory, moved from cell to cell and measured hourly EC and OC, with
some interruptions due to instrument maintenance or holidays. The sampling strategy is
96 documented in Table S1. In this instrument, volatile gases were removed from the
samples by carbon denuders prior to collection. Airborne particles smaller than 2.5 μm
98 were then collected on quartz fiber filters at a flow rate of 8 l min⁻¹. Upon completion of a
preset sampling duration, all carbon that had been accumulated on the filter was removed
100 by heating the sample in multiple increasing temperature steps, first in a completely
oxygen-free helium environment and then in a He/O₂ environment. The vaporized
102 compounds flowed through an oxidizer oven, were oxidized to carbon dioxide, and were
detected via an infrared analyzer. An external methane (CH₄) standard was injected at
104 the end of every analysis and used to normalize the analytical result. Since in theory the
quartz filter has had all of the collected carbonaceous aerosol removed during each
106 analysis cycle, the filter was re-used for multiple samples and changed only periodically.

108 A detailed description of the PM₁₀ sample collection and elemental analysis
methodologies can be found in Alharbi et al. (2015). In brief, sampling was conducted

110 from the same mobile platform and concurrent with the OC/EC sampling. A PM₁₀ inlet
112 was used to sample ambient aerosol onto quartz microfiber filters over a 24 h period.
114 These samples were collected every three days and elemental analyses for Al, As, B, Ca,
116 Cd, Co, Cr, Cu, Fe, K, Li, Mg, Mn, Mo, Na, Ni, Pb, Te, V, and Zn were performed by ICP-
OES. NO and NO₂ (NO_x) were measured by chemiluminescence and O₃ was measured
by UV photometer simultaneously using Signal Ambirak air quality monitoring system
(Signal Ambitech Ltd, UK).

2.2 EC and OC re-split method

118 The Sunset semi-continuous EC/OC analyzer adopts the same thermal-optical analysis
120 method for determination of OC and EC that is commonly applied to the offline analysis
122 of filter samples. The OC and EC mass concentrations (as mass of C) are quantified by
124 a calibrated non-dispersive infrared sensor (NDIR) signal that detects the evolved CO₂.
126 Ideally, OC is defined as the carbon evolved under increasing temperature ramps
128 conducted in an inert atmosphere (100% He) and EC is defined to be the subsequent
130 carbon evolution in an oxidizing atmosphere (He/10% O₂ mixture). In the inert
132 atmosphere, rather than simply volatilizing, a fraction of OC may be pyrolyzed due to
134 insufficient oxygen, and this pyrolyzed OC may be evolved in the subsequent oxidizing
136 atmosphere, appearing as EC. This fraction of OC is usually called pyrolyzed organic
138 carbon (PyOC). To subtract PyOC from EC, laser transmittance or reflectance is deployed
140 to monitor the variations in filter darkness; the transmittance or reflectance responds to
142 the presence of EC throughout the analysis, but then drops when PyOC is formed and
144 rises again as PyOC is evolved. The fraction of total assigned EC evolved in the oxidizing
146 atmosphere before the laser signal returns to its initial value is believed to be due to PyOC,
148 so in post-analysis the final EC is reported as the difference between the total carbon
evolved in the oxidizing atmosphere and the PyOC. This methodology has been
automated in the Sunset instrument. However, unusual EC and OC splits for a large
number of samples were observed during the study period: (a) split points jumped to the
end of the analysis because the laser response did not rebound to its initial value before
the CH₄ calibration phase; or (b) split-points were located in the pre-oxygen position.
These split point deviations were ascribed to refractory residue on the filters: the laser
correction factor supplied in the standard manufacturer software may not be applicable
to the dusty environment of Riyadh (Polidori et al., 2006; Jung et al., 2011; Wang et al.,
2012). Therefore, observed relationships between laser response and temperature in the
CH₄ + O₂ injection calibration phase were used to develop a corrected split point. The
correction methodology assumed that only refractory material was present on the filter in
this phase, so that effects of this refractory material on the laser response to temperature
variations could be isolated, corrected, and these corrections applied during the other
analysis phases. A full description of the methodology is found in the Appendix. We noted,
however, that measurement artifacts from carbonates in dust may have been present in

150 this study, which would result in a high bias in the OC measurements. As noted in
152 Karanasiou et al. (2011) and in the standard operating procedure (SOP) document
published by the Research Triangle Institute (RTI) (<https://www3.epa.gov/ttnamti1/files/ambient/pm25/spec/RTIIMPROVEACarbonAnalysisSSLSOP.pdf>), the evolution of carbonates from filter samples during thermal analysis can
154 occur over several carbon peaks. While it is preferred to use acid decomposition of
carbonates (on separate sample punches) to obtain the best quantification, Karanasiou
156 et al. (2011) demonstrated that the protocol used in this study completely evolves
carbonates in the OC fraction, and that manual integration to isolate the carbonate
158 concentration was possible but carried large uncertainty. Hence, we did not attempt to
separately quantify carbonate in this work.

160 **2.3 SOC estimation by minimum R squared (MRS) method**

The EC tracer method is widely used to estimate secondary organic carbon mass
162 concentrations, applying the following equations, which assume that EC has only
combustion sources:

$$164 \quad POC = \left(\frac{OC}{EC}\right)_{pri} \times EC \quad \text{Eqn 1}$$

$$SOC = OC_{total} - \left(\frac{OC}{EC}\right)_{pri} \times EC - b \quad \text{Eqn 2}$$

166 where $(OC/EC)_{pri}$ is the OC/EC ratio in fresh combustion emissions, b denotes non-
combustion-derived POC, and OC_{total} and EC are ambient measurements. The key to
168 successful application of this method is to begin with an appropriate estimate of the
 $(OC/EC)_{pri}$ ratio. Several approaches have been documented to determine $(OC/EC)_{pri}$.
170 Gray et al. (1986) directly adopted the ratios from emission inventories. Turpin and
Huntzicker (1995) used the measured OC/EC ratio when local emissions were dominant
172 in a certain location or over a specified period. Based on the expectation that co-emitted
POC and EC are well correlated, Lim and Turpin (2002) took the slope of OC against EC
174 using OC/EC ratio data for the lowest 5-10% values of that ratio. Millet et al. (2005)
proposed that a critical point where SOC was independent of EC should represent the
176 primary OC/EC ratio; the critical point was found by a minimum R-squared (MRS) method.
Assuming that non-combustion sources (i.e., the b term in Eqn 2) are negligible, this
178 method can derive the most accurate primary OC/EC ratio, compared with previously-
proposed approaches (Wu and Yu, 2016). However, this method may underestimate the
180 SOC concentration if some SOC is associated with EC: co-emitted semi-volatile POC
could rapidly oxidize to low-volatility SOC and partition on the surface of EC. However,
182 given that accurate emission inventories were not available for Riyadh, we employed this
method in the absence of a priori knowledge of $(OC/EC)_{pri}$ to provide a conservative
184 estimate of the SOC concentration during our observational period.

186 The methodology for and applications of the MRS method were documented in Millet et
 188 al., (2005), Hu et al., (2012), and Wu and Yu (2016). The non-combustion source (*b* term)
 190 was assumed to be zero. A series of coefficients of determination (R^2) between EC and
 192 SOC calculated by Eqns 1 and 2, varying $(OC/EC)_{pri}$ from 0 to 10 using steps of 0.01 in
 the ratio, was generated. At low $(OC/EC)_{pri}$ ratio, a significant portion of the estimated
 SOC still belonged to POC. At high $(OC/EC)_{pri}$ ratio, the term $(OC/EC)_{pri} \times EC$ largely
 exceeded OC_{total} and became dominant. At the correct ratio, all the POC was removed
 and R^2 of SOC and EC reached a minimum. This ratio was then used to estimate SOC in
 all samples.

194 2.4 Back trajectory analysis

To develop an understanding of potential regional influences on observed PM, we
 196 calculated 24-hr back trajectories (BTs) every 3 hours during each sampling period using
 the National Oceanic and Atmospheric Administration (NOAA) Hybrid Single-Particle
 198 Lagrangian Integrated Trajectory (HYSPLIT; Stein, et al., 2015; Rolph, 2016). Trajectories
 were initiated for a starting height of 500 m above ground level (AGL). Residence time
 200 analysis (RTA), describing the probability of air mass origins, was also performed
 (Ashbaugh et al., 1985). The probability (P_{ij}), representing the residence time of a
 202 randomly selected air mass in the ij^{th} cell during the observational period, was calculated
 as follows:

$$204 \quad P_{ij} \cong \frac{n_{ij}}{N} \quad \text{Eqn 3}$$

where n_{ij} is the number of trajectory segment endpoints that fell in the ij^{th} cell and N is the
 206 total number of endpoints.

Concentration weighted trajectory analysis (CWT) is another effective tool that we
 208 combined with back trajectory data and pollutant concentration to trace the source origin
 for certain species. The calculation formula is as follows:

$$210 \quad C_{ij} = \frac{1}{\sum_{i=1}^M \tau_{ijl}} \sum_{i=1}^M C_i \tau_{ijl} \quad \text{Eqn 4}$$

where C_{ij} is the average weighted concentration in the grid cell (i, j), C_i is the measured
 212 species concentration, τ_{ij} is the number of trajectory endpoints in the grid cell (i, j) and M
 is the number of samples that have trajectory endpoints in the grid cell (i, j).

214 2.5 Positive matrix factorization (PMF) analysis

Positive matrix factorization (PMF) has been successfully applied to aerosol composition
 216 data to suggest sources impacting the sampling site (Reff et al., 2007 and Viana et al.,
 2008). We aligned daily-average OC and EC with concurrent averaged measurements of

218 metal concentrations in the PM₁₀ fraction (Al, Ca, Cu, Fe, K, Mg, Mn, Na, Ni, Pb and V)
and prepared a matrix of size 35 ×13 for input to the USEPA PMF, version 5.0
220 ([https://www.epa.gov/air-research/positive-matrix-factorization-model-environmental-](https://www.epa.gov/air-research/positive-matrix-factorization-model-environmental-data-analyses)
data-analyses). Data points with “ND” were replaced by 1/2 of the detection limit and the
222 corresponding uncertainties were assigned as 5/6 of the detection limit. The uncertainties
for all other data were calculated as $s_{ij} + DL_{ij}/3$, where s_{ij} represents the analytical
224 uncertainty for species i in the sample j and DL_{ij} represents the detection limit (Polissar
et al., 1998; Reff et al., 2007). In this study, the analytical uncertainty was assumed to be
226 5% of the corresponding concentration for metal species. Uncertainties for the EC and
OC data were not reported. Norris et al. (2014) suggested that, for such cases, the initial
228 uncertainties be set to a proportion of the concentration. The uncertainties for OC and EC
were therefore calculated as 10% of the corresponding concentrations for this study.

230 **3. Results and discussion**

3.1 Overview of EC and OC concentrations

232 Fig. S1.a shows the time series of OC and EC concentrations during the study period and
denotes the corresponding sampling cells in which the measurements were obtained.
234 Average OC and EC concentrations during the observational period were 4.8 ± 4.4 and
 $2.1 \pm 2.5 \mu\text{g C m}^{-3}$, respectively (we will use $\mu\text{g m}^{-3}$ for OC and EC hereafter when referring
236 to $\mu\text{g C m}^{-3}$). Table 1 presents some comparative values of measured EC and OC
concentrations in PM_{2.5} in urban areas world-wide, since urban areas are expected to
238 share some similar anthropogenic source types (e.g. vehicular and industrial emissions)
with Riyadh. The average concentrations in this work for both EC and OC were
240 remarkably consistent with those reported by von Schneidemesser et al. (2010) and
Abdeen et al. (2014) for 11 Middle Eastern sampling sites, including Tel Aviv, a major city
242 in Israel (OC: 4.8 and EC: 1.6 $\mu\text{g m}^{-3}$). The average OC concentrations were also
comparable to those reported for suburban Hong Kong (4.7 $\mu\text{g m}^{-3}$, Huang et al., 2014b),
244 higher than Cleveland and Detroit, US (3.10 and 3.54 $\mu\text{g m}^{-3}$, Snyder et al., 2010), but
lower than those reported for Gwangju, Korea (5.0 $\mu\text{g m}^{-3}$, Batmunkh et al., 2016), Veneto,
246 Italy (5.5 $\mu\text{g m}^{-3}$, Khan et al., 2016), Athens, Greece (6.8 $\mu\text{g m}^{-3}$, Grivas et al., 2012),
urban Hong Kong (10.1 $\mu\text{g m}^{-3}$, Ho et al., 2006), Delhi, Indian ($16.5 \pm 6.6 \mu\text{g m}^{-3}$, Satsangi
248 et al., 2012), and Beijing, China ($18.2 \pm 13.8 \mu\text{g m}^{-3}$, Zhao et al., 2013), reflective of the
different mix of sources and different photochemical environments. EC concentrations
250 also vary widely among urban regions, depending on the characteristics of local sources.

The Riyadh sampling site characteristics and the corresponding average OC and EC
252 concentrations in each grid cell are summarized in Table S1. Results of a one-sided t-test
($p < 0.001$) on OC and EC data from industrial and residential sites suggested a significant
254 difference in carbonaceous aerosol concentrations between the two site types: OC mass
concentrations in industrial sites were 1.4 times those in the residential sites, and EC

256 mass concentrations were 1.7 times higher (Fig. 2). The mean OC/EC ratio was lower in
the industrial sites (3.1) than in residential sites (6.0), suggesting the importance of POC
258 emissions in industrial regions and a larger SOC contribution in residential areas. We also
divided Riyadh into four quadrants to investigate the spatial variation of OC and EC across
260 the city. Fig. 3 shows that OC and EC concentrations were higher in the eastern quadrants.

Fig. 4 shows the results of the RTA, demonstrating that air masses arriving in Riyadh
262 were mainly from within Saudi Arabia and from the south / southwest of the city in April
and May, and from the north / northeast from June to September, extending to the west
264 coast of the Persian Gulf. These two dominant wind directions were used to stratify data
in Fig. S1b, which shows that the average OC concentration increased from 3.8 to 5.3 μg
266 m^{-3} and EC from 1.1 to 2.7 $\mu\text{g m}^{-3}$ when the air mass source region shifted from
south/southeast to north/northeast, respectively.

268 **3.2 Diurnal variation of OC and EC**

Fig. 5 shows the diurnal variations in OC and EC mass concentrations. OC and EC
270 concentrations peaked from 6-9 am and were elevated during nighttime (after 1600 pm).
 NO_x also showed a similar diurnal pattern (Fig. S2). The morning peak coincided with
272 traffic rush hours. The diurnal variations of OC and EC on weekdays and weekends
exhibited similar trends (Fig, S3), but EC was higher during weekdays. The elevation of
274 OC, EC and NO_x at night after 1600 pm may be attributed to the accumulation of
pollutants in the shallower nocturnal boundary layer. Average OC/EC ratios showed no
276 obvious trends; however, the median OC/EC ratio decreased slightly over the time period
when OC and EC concentrations built up, probably due to the increased contributions
278 from primary emissions. The average OC/EC ratio had a peak around 14:00 pm,
corresponding with peak concentrations of O_3 , suggestive of secondary aerosol formation.

280 **3.3 Weekend effect in OC and EC concentrations**

A “weekend effect” in concentrations of traffic-derived PM has been noted in previous
282 studies (e.g. Grivas et al., 2012; Bae et al., 2004; Moteballi et al., 2003; Lim and Turpin,
2002; Jeong et al., 2004; Lough et al., 2006). To investigate whether a weekend effect
284 could be discerned in the Riyadh dataset, two-sample t-tests assuming unequal variances
were performed for hourly EC and OC samples, grouped according to whether they were
286 obtained on weekdays (Saturday to Wednesday) or on weekends (Thursday and Friday).
The test indicated a statistically significant difference (29% lower on weekends) in EC
288 concentrations between weekday and weekend, but no significant difference in OC ($p <$
0.001 with a 95% confidence level), as shown in Fig. 6. NO_x concentrations were also
290 reduced during weekends, by 48% compared to weekdays (Fig. S4). This reduction may
be ascribed to the decrease in the local vehicular activities and industrial activities during
292 the weekend. Therefore, local EC can be roughly estimated to be 0.51 $\mu\text{g m}^{-3}$, about 22%

294 of total EC, by the difference in average EC concentrations between weekday and
296 weekend. As there were still some local traffic and industrial activities during the weekend,
298 this estimation is likely a lower bound of the local contribution to EC concentration in this
study. OC concentrations had no significant weekday-weekend variation. The decrease
of EC was the main driver of the increasing OC/EC ratio during the weekends, indicating
the reduced primary emission and effective SOC formation / transport during the
weekends.

300 3.4 SOC estimation

Fig. 7 shows the determination of $(OC/EC)_{pri}$ using the minimum R squared method
302 (MRS). The value of this ratio derived in this study was 1.01, which occurred at the 14th
percentile in the observed OC/EC ratios. In the compilation of PM_{2.5} OC and EC emission
304 profiles presented by Chow et al. (2011), the $(OC/EC)_{pri}$ for oil combustion was
documented to range from 0.2 to 2.5 with an average of 1.0 ± 0.2 , 0.9 to 8.1 with an
306 average of 3.4 ± 2.2 for gasoline emissions, and 0.2 to 2.7 with an average of 1.0 ± 0.8 for
diesel emissions. Our estimate was within with these ranges and was closer to the
308 averages for oil combustion and diesel emissions, consistent with expected important
contributions from these sources to PM_{2.5} carbonaceous aerosol in Riyadh. Using our
310 MRS-derived $(OC/EC)_{pri}$ in equations (1) and (2), we computed average POC and SOC
concentrations of 2.0 ± 2.4 and $2.8 \pm 3.4 \mu\text{g m}^{-3}$, respectively, suggesting that POC and
312 SOC contributions to PM_{2.5} were comparable during our study. The average POC and
SOC concentrations were 1.0 ± 1.0 and $2.7 \pm 4.0 \mu\text{g m}^{-3}$, respectively, when transport was
314 from the south/southwest. POC increased to $2.5 \pm 2.7 \mu\text{g m}^{-3}$ and SOC was almost
unchanged when the direction of transport was from the north / northeast. Variability in
316 OC was thus mainly due to variability in POC. The sampling locations were in cells
classified as being in the outskirts of the city when south/southwesterly transport was
318 prevalent, but included both outskirts and in-city grids when north/northeasterly transport
was prevalent. The increase in POC during northerly transport regimes could not
320 therefore be attributed solely to the influence of local primary emissions, since transport
of POC from outside Riyadh was also possible.

322 The diurnal variation of SOC (Fig. S3) showed a small peak of SOC concentration in the
morning from 7-9 am, lagging behind the POC and EC morning peaks by about two hours;
324 this result was not unexpected since photochemical production of SOC will require time
for reactions to proceed once precursors have accumulated in the atmosphere. A second
326 small peak in SOC concentration occurred at 14:00 pm, concurrent with ozone formation
(Fig. S2) and consistent with the variation in OC/EC ratios discussed in Section 3.3. The
328 diurnal variations of POC and SOC were similar on weekdays and weekends, but the
weekday-to-weekend changes in POC and SOC had opposite trends. The estimated
330 POC was $2.2 \pm 2.5 \mu\text{g m}^{-3}$ on weekdays and decreased to $1.5 \pm 1.9 \mu\text{g m}^{-3}$ on weekends.
The estimated SOC was $2.6 \pm 2.9 \mu\text{g m}^{-3}$ on weekdays and increased by 23% to 3.2 ± 4.5

332 $\mu\text{g m}^{-3}$ on weekends. The elevated SOC during weekends was likely due to regional
334 production and transport. With regards to spatial variation, POC and SOC were 3.5 ± 2.7
335 and $3.2\pm 2.9 \mu\text{g m}^{-3}$ in the industrial sites, 2.1 ± 2.6 and $2.6\pm 3.0 \mu\text{g m}^{-3}$ in the residential
336 sites, and 1.1 ± 1.1 and $2.8\pm 4.1 \mu\text{g m}^{-3}$ in the outskirts sites, respectively. SOC
337 concentrations were 2.5 times those of POC in the outskirts sites, an expected result
338 since these latter sites are farther removed from the sources of primary emissions within
339 the city proper. The results were consistent with the study of von Schneidmesser et al.
340 (2010) that SOC (i.e., OC that was left unapportioned by a chemical mass balance model)
341 was estimated to be 30-74% of the total OC in 11 sites in the Middle East, having
342 climatological conditions similar to those in Riyadh.

342 **3.5 Possible sources of PM_{2.5} carbonaceous aerosols**

343 **3.5.1 Correlation between OC, EC and other elemental species**

344 As a first step in seeking signatures of sources of carbonaceous aerosol in Riyadh, we
345 conducted an analysis of the correlations between OC or EC and measured elemental
346 species. We note that OC and EC were measured in the PM_{2.5} fraction, while elemental
347 species concentrations were obtained for the PM₁₀ fraction, which also included the PM_{2.5}.
348 OC and EC were poorly correlated with K, which we interpreted as indicating a negligible
349 influence of biomass burning on PM. Al, Fe, Mg, Mn, and Ca are found in crustal soils
350 and in PM samples of windblown dust. EC did not correlate well with these species ($R^2 <$
351 0.35 ; not shown). However, OC had a relatively strong correlation with Ca (R^2 of 0.63)
352 (Fig. 8 and Fig. S7) but, similar to EC, a poor correlation with other dust species (not
353 shown). These findings indicated that OC may have shared a source with Ca, but this
354 source was not likely to be associated with windblown dust. The correlation between SOC
355 and Ca was stronger than that between POC and Ca (Fig. S6). The thermo-optical
356 method may have measured CaCO_3 as OC, and the subsequent estimates of SOC
357 separated two sources, one associated with combustion and EC ("primary"), and another
358 associated with CaCO_3 (and mis-labeled "secondary"). Concentrations of Al and of other
359 metals (Fe, K, Mg and Mn) were strongly correlated ($R^2 > 0.9$), supporting their common
360 dust origin (Fig. 8). The correlation between Ca and other dust metal species (Al, Fe, K,
361 Fe and Mg), however, showed two divergent regimes, suggestive of an additional Ca-
362 containing source besides dust, that may have shared the same sources as OC.
363 Therefore, understanding the sources of Ca became a prerequisite in understanding the
364 sources of OC.

365 The enrichment factor (EF) is a practical and convenient tool to differentiate natural and
366 anthropogenic sources of metal species (Khodeir et al., 2012; Rushdi, et al., 2013). The
367 EF can be calculated using the following equation (Taylor, 1964):

368
$$EF = \frac{(X / C_{ref})_{air}}{(X / C_{ref})_{source}}$$
 Eqn 5

370 where X is the measured metal concentration, and C_{ref} is the concentration of the
 371 reference metal species. The equation compares the ambient elemental abundance of
 372 two species with their source abundance. An EF less than 10 suggests that the sample
 373 may come from a natural crustal source and an EF value > 10 indicates possible
 374 anthropogenic influence (Biegalski et al., 1998). Al, Fe, and K were all used as reference
 375 species to test for robustness of the findings. Fig. S6 shows that, for all three reference
 376 species, the EFs for Al, Fe, K, Mn, Mg, Na and V were calculated to be less than 10,
 377 suggesting a dominant crustal type origin. The EFs of Ni, Zn, Cr, Co, Pb, Li, B, As, Mo,
 378 Cd, and Te were calculated to be larger than 10, suggestive of the influence of
 379 anthropogenic emissions, e.g. traffic emissions, fossil fuel combustion and non-ferrous
 380 metal industries. The EF for Ca was calculated to be ~ 10 , consistent with the idea that it
 may have two sources in Riyadh, one natural and one anthropogenic.

382 Cement kilns have been documented to be important sources of elemental Ca in the
 383 atmospheric aerosol (Zhang et al., 2014). Chow et al. (2004) noted an important
 384 contribution of $PM_{2.5}$ POC from cement factories. Hence, contributions from cement
 385 production sources may have led to the good correlation between OC and Ca at the
 386 receptor sites. In the Middle East, another possible anthropogenic source for Ca is from
 387 the desalting and demetalization of crude oil in refineries (Wu et al., 2014); refineries are
 388 certainly contributing to the observed OC in Riyadh. A third possibility was that the Ca is
 389 crustal in origin, but from a different source region than most of the other sampled dust.
 390 Ca enrichment in dusts may vary across the Middle East region (Coz et al., 2010), and
 391 thus the correlation between Ca and other crustal species could diverge depending upon
 392 the dust source region. Regardless of dust source region, during transport to Riyadh, as
 393 ambient SOC precursors were oxidized, the products may have partitioned to particle
 394 surfaces, resulting in simultaneous transport of Ca and OC. Finally, we note that a
 395 correlation between Ca and OC may have occurred if calcium carbonate was being
 396 sampled and the carbonate detected as OC in the thermal analysis protocol, as
 397 mentioned in the Methods section above. While it was not possible to definitively
 398 distinguish between these various possibilities based only on EF, the large dust loadings
 that were present in nearly all samples suggest that this latter explanation could play a
 significant role in producing the observed Ca-OC correlations.

400 3.5.2 CWT analysis for Ca/Al ratio, Pb, OC and EC

401 We used CWT analysis to identify possible source origins for the observed highest values
 402 of Ca/Al ratio, Pb, OC and EC (Fig. 9). The CWT plot for the Ca/Al ratio showed that,
 when this ratio was high in Riyadh PM samples, air masses were most likely to have

404 passed over regions along the western shoreline of the Persian Gulf, and in particular,
406 the highest ratio was found for air masses passing over the site of a large Saudi Arabian
408 cement plant (Fig. S8). This transport pathway was thus consistent with the idea that
410 refineries and cement plants may represent anthropogenic sources of Ca. CWT analysis
412 of Pb showed that high observed concentrations in Riyadh aerosol were associated with
414 transport from Iraq, consistent with the continued usage of leaded fuel in that country
416 (Shaik et al., 2014). PM_{10} Pb concentrations were $0.035 \pm 0.088 \mu\text{g m}^{-3}$ in this study, lower
418 than measurements reported for eastern China (0.05 to $0.5 \mu\text{g m}^{-3}$, Li et al., 2010) and
420 the greater Cairo area ($0.3 \mu\text{g m}^{-3}$, Safar and Labib, 2010), both locations for which leaded
422 fuel has been phased out of usage, and lower than the U.S ambient concentration
standard for lead ($0.15 \mu\text{g m}^{-3}$ on a 3-month rolling basis; U.S. EPA, 2006). The
comparison showed that although Pb may have multiple potential sources in Riyadh, the
concentration levels were still below those of concern for human health. Industrial
emissions along the Saudi Arabian coast may also contribute some Pb to the measured
aerosol. While high OC concentrations were associated with transport from a similar
region of the Persian Gulf as was high Pb, the high-concentration source region extended
further north, encompassing areas with oil fields and refineries and the Baghdad urban
region (Fig. S9). Finally, the CWT plots for OC and EC were similar, suggesting their
highest concentrations may be attributed to similar sources, i.e., refineries, cement
factories and urban pollution.

424 **3.6 PMF analysis**

Three- to five-factor solutions were tested in the PMF model; the three-factor solution was
426 found to have the best solution characteristics (Fig.10). Most of the OC (77%) and EC
(90%) together with fractions of the crustal elements appeared in the first factor. We note
428 that 54% of Ca was loaded in this factor, as expected since OC was found to be correlated
with Ca. No significant crude oil tracers (Ni and V) appeared in the factor, indicating that
430 this factor was not related to oil combustion (Ganor et al., 1988). The CWT analysis
suggested that high OC and EC coming from the shoreline of the Persian Gulf may be
432 associated with industrial emissions, including refineries, gas flares in oil fields, and
cement production. However, we could not rule out potential contributions to this factor
434 from local vehicular emissions. Therefore, this factor was identified as a mixed source:
cement industries / gas flares / local vehicles.

436 A key signature in the second factor was the significant loading of Pb (98%); it also
included some dust species. While leaded fuels have been phased out in Saudi Arabia,
438 as mentioned above, they were still in use in Iraq; further, deposition of lead to soils and
resuspension is a documented exposure pathway (Laidlaw and Filippelli, 2008). CWT
440 analysis also supported a source origin of Pb from Iraq (Fig. 9). Hence Pb may have
served as a regional transport tracer in this PMF analysis. However, Pb could also be
442 contained in other industrial emissions, including cement manufacturing in the city. The

444 second factor was thus identified as leaded fuel combustion from long range transport /
industrial emissions.

446 The third factor contained almost all of the V and a large fraction of Ni (>60%), as well as
some crustal elements and OC. V and Ni and their ratios have been suggested as
448 markers of emissions from oil fired power plants (Ganor et al., 1988). Barwise (1990)
found that the highest V/Ni ratios (>1) among oil samples that they characterized were
450 associated with Abu Dhabi and Suez oils, as contrasted with samples from the North Sea,
China, Indonesia, and Australia, reflecting geological differences. The ratio of V/Ni in
452 factor 3 is 3.5, consistent with the Arabian Gulf source of oil in this region. Dust species
and some OC and EC were also associated with this factor, which we therefore identified
as oil combustion.

454 Fig. 11 shows the source contribution to OC and EC from these three factors in individual
samples. On average, the OC concentration was dominated by the mixed source (factor
456 1) ($3.8 \mu\text{g m}^{-3}$, 77%), followed by leaded fuel from long range transport ($0.8 \mu\text{g m}^{-3}$, 27%)
and oil combustion ($0.3 \mu\text{g m}^{-3}$, 6%). The contribution of the mixed source ranged from
458 37% in May ($0.7 \mu\text{g m}^{-3}$) to 95% in September ($6.7 \mu\text{g m}^{-3}$). The EC concentration was
also mainly attributed to the mixed source ($1.9 \mu\text{g m}^{-3}$, 92%). In some May samples, the
460 mixed source contribution was negligible, as the source tracer, EC, was only 0.1-0.4 $\mu\text{g m}^{-3}$,
about one order of magnitude lower than that in other periods. The tracer analysis
462 suggested that long-range transport was dominant for those samples.

4. Conclusions

464 To our knowledge, this study represents the first reported long-term and spatially resolved
hourly measurements of ambient OC and EC concentrations for Riyadh, Saudi Arabia,
466 along with supporting measurements that enabled a source apportionment of these
important aerosol species. We found that OC and EC average concentrations were
468 comparable to other reported measurements in Middle Eastern cities, and diurnal and
weekly variations indicated a clear influence from local emissions. However, OC and EC
470 concentrations varied with air mass source origin, indicative of not only variations across
Riyadh and its outskirts, but also of the influence of regional sources on carbonaceous
472 aerosol concentrations. About half of the measured OC was attributed to secondary
formation, at least 22% of EC was ascribed to local sources, and positive matrix
474 factorization suggested that EC and OC were mainly attributed to a mixed source
category comprising cement industries, gas flaring activities, and local vehicles.

476 Measurement of OC and EC via the online thermo-optical technique was found to be
challenging in the dusty environment encountered year-round in Riyadh. Our dataset
478 required correction via a hand analysis, as reported in the supplementary materials, as
the automated split method implemented by the manufacturer frequently failed for our

480 samples. The lack of a separate independent carbonate analysis, however, meant that
482 our reported OC concentrations may be biased high, as also suggested by the strong
484 correlation between OC and Ca. However, the correlation between OC and Ca may also
486 suggest co-emission of OC and its precursors with metal Ca from desalting and
488 demetalization processes in refineries; co-emission of Ca and OC from cement plants; or
490 condensation of OC on Ca-rich dust during long-range transport. In future studies of
ambient aerosol OC concentrations in dusty environments via online thermo-optical
techniques, additional observations or different measurement protocols are needed to
separate the contributions of carbonates to the measured OC and EC concentrations.
With such added information, the implied sources of Ca and OC can be further
investigated and their potential contributions to observed OC quantified.

492 **Appendix A: Correction method for OC/EC splits in data from the Sunset semi- continuous analyzer**

494 Laser response and temperature for individual blanks were well correlated,
496 suggesting that the influence of temperature on laser response may indirectly affect the
498 EC/OC split points (Figure A.1). This phenomenon has been pointed out previously, and
500 Versions RT-Calc 114 and newer of the Sunset instrument analysis software introduced
502 a laser correction factor to counteract the influence of temperature on the laser signal.
504 This correction factor is calculated in each cycle from the variation in the laser signal when
the analysis enters the methane calculation stage (Jung et al., 2011). However, it was
obvious that this correction approach did not work well for the Riyadh samples, since
many returned EC/TC=0, the case when the initial reflectance is not recovered in the
analysis. A revised method of finding the point of return to the original laser signal, and
thus determining the POC and EC contributions, was therefore proposed for this study
and used to correct the dataset.

506 The relationship for the Riyadh samples between laser response and temperature
508 during the calibration phase of the CH₄ + O₂ injection was used to develop a corrected
510 split point, assuming that only refractory material is present in this phase, and the effects
512 of this refractory material on the laser response to temperature variations could be
514 isolated and then applied during the other analysis phases. A correlation between laser
response and temperature in the calibration phase was derived using linear and quadratic
functions. The derived parameters from the two functions were applied in the following
equations to recompute a corrected laser signal for each analysis, instead of the laser
correction factor automatically generated by the Sunset program:

$$516 \text{Signal}_{new} = \text{Signal}_{original} + a (\text{Temp}^2_{initial} - \text{Temp}^2_{original})$$

$$+ b (Temp_{initial} - Temp_{original}) \quad (A.1)$$

518

$$Signal_{new} = Signal_{original} + c (Temp_{initial} - Temp_{original}) \quad (A.2)$$

520

522 where $Signal_{original}$ represented the original laser signal, $Signal_{new}$ represented the signal
524 after correction to the initial temperature, $Temp_{initial}$ represented the temperature at the
526 initial condition when each analysis began, and $Temp_{original}$ represented the original
528 temperature for each analysis; a and b in Eq. (A.1) were derived from the quadratic
530 equation for each analysis, and c in Eq. (A.2) was derived from a linear fit.

532 Since refractory residues accumulated on the filter during the measurement period,
534 the derived correlation between laser response and temperature varied sample by sample.
536 The equations to derive the corrected laser signal were, therefore, applied individually to
538 each sample. In the blank sample, the quadratic-function-generated laser signal was
540 smoother than the linear-function-generated one, especially during the calibration phase
542 of the $CH_4 + O_2$ injection (Figure A.2a). The relationship between temperature and laser
544 signal for the newly replaced filter tended to be closer to linear, while the signal for the
546 aged filter with residue accumulation showed a better fit using a quadratic equation. A
548 quadratic equation was therefore selected to correct the laser signal for the entire dataset.
The new split points were then set to where the corrected laser signal rebounded to its
value just before OC pyrolyzed and the laser signal decreased due to pyrolyzed organic
carbon formation. The method worked for both incorrect split-point cases, bringing the
split point back to the He + O_2 phase as expected and leading to more reasonable EC/OC
split points, i.e., neither at the end of the analysis nor in the pre-oxygen analysis phase.
It is noted that although the quadratic equation correction produced a better laser signal
for purposes of the carbon analyses, this correction did not work perfectly in the low
temperature He phase, where the corrected laser signal exhibited unexpected increases.
However, this shortcoming did not substantially influence the accuracy of the correction
during subsequent carbon evolution. We note that premature evolution of EC, leading to
an increasing laser signal in the inert environment due to the existence of refractory metal
oxides, was observed in the studies of Wang et al. (2010) and Bladt et al. (2012). The
increases in the corrected laser signal during the He stage in this study may be partially
due to the same cause, as Riyadh samples contained abundant metal oxides.

550

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Table 1 Comparison of OC and EC concentrations ($\mu\text{g m}^{-3}$) measured in urban areas world-wide.

City	Duration	EC	OC	EC	OC	References
		Conc. ($\mu\text{g m}^{-3}$)		S.D. ($\mu\text{g m}^{-3}$)		
Athens, Greece	Jan to Aug, 2003	2.2	6.8			Grivas et al., 2012
Gwangju, Korea	Winter of 2011	1.7	5.0	0.9	2.5	Batmunkh et al., 2016
Cleveland, US	Jul, 2007 and Jan, 2008	0.33	3.1	0.01	0.78	Snyder et al., 2010
Detroit, US		0.35	3.54	0.01	0.86	
Beijing, China	Selective days in four seasons from 2009 to 2010	6.3	18.2	2.9	13.8	Zhao et al., 2013
Urban, Hong Kong	Nov, 2000 to Feb, 2001 and Jun, 2001 to Aug, 2001	5.71	10.12	0.89	1.92	Ho et al., 2006
Suburban, Hong Kong	Mar, 2011 to Feb, 2012	0.86	4.7	0.53	2.87	Huang et al., 2014b
Veneto, Italy	Apr 2012 to Feb 2013	1.3	5.5			Khan et al., 2016
Delhi, India	Dec 20, 2012 to Feb 26, 2013	12.04	16.46	4.43	6.61	Panda et al., 2016
Middle East (11 sampling sites in Palestine, Jordan and Israel)	Jan to Dec, 2007	2.1	5.3	2.2	4	Von Schneidmesser, et al., 2010 Abdeen, et al., 2014
Riyadh, Saudi Arabia	Apr to Sep, 2012	2.13	4.76	2.52	4.4	this study

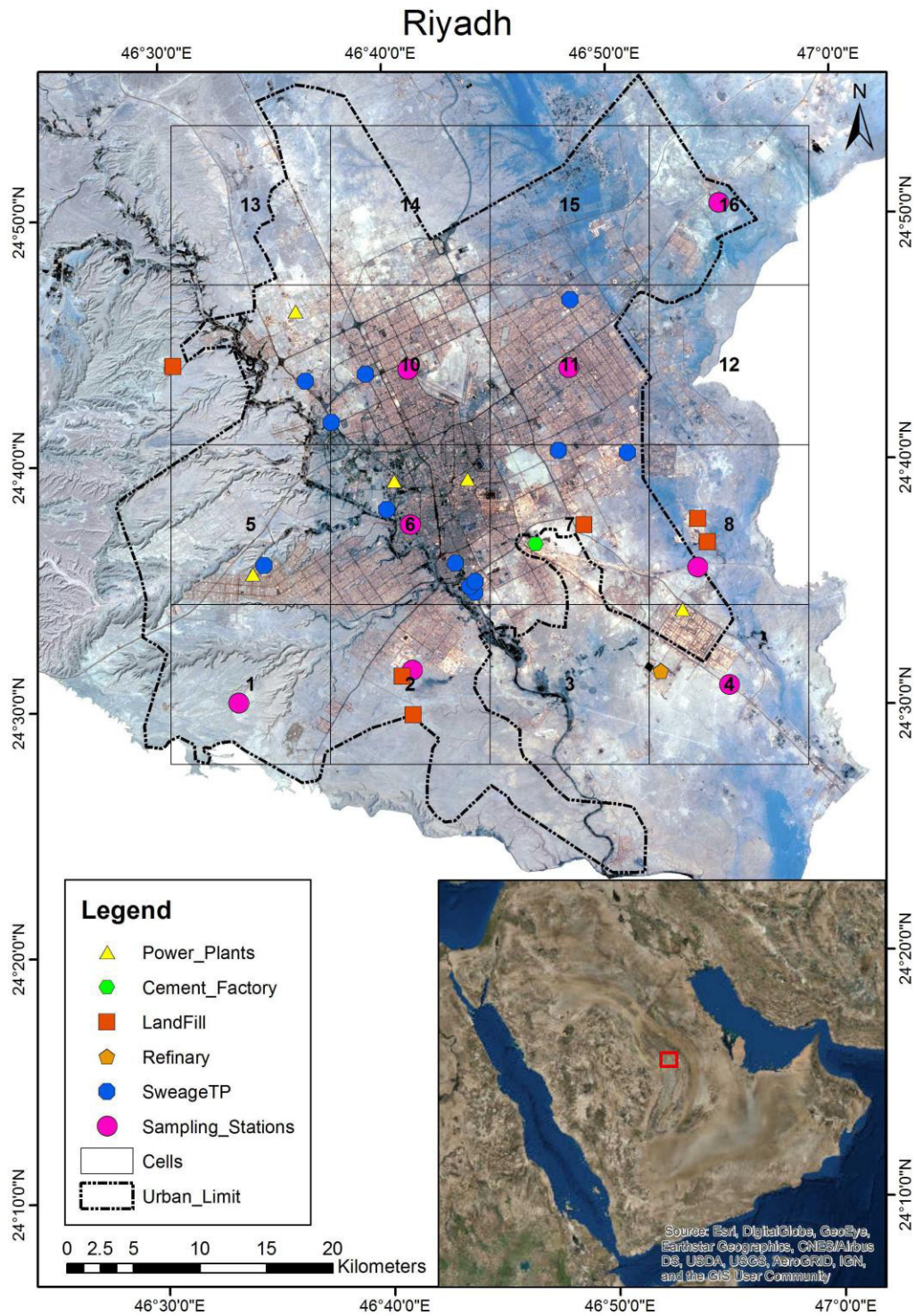


Figure 1 Image of Riyadh and immediate surroundings. Potential emission sources and 16 sampling locations are indicated. The characteristics of the sampling locations are listed in Table 1.

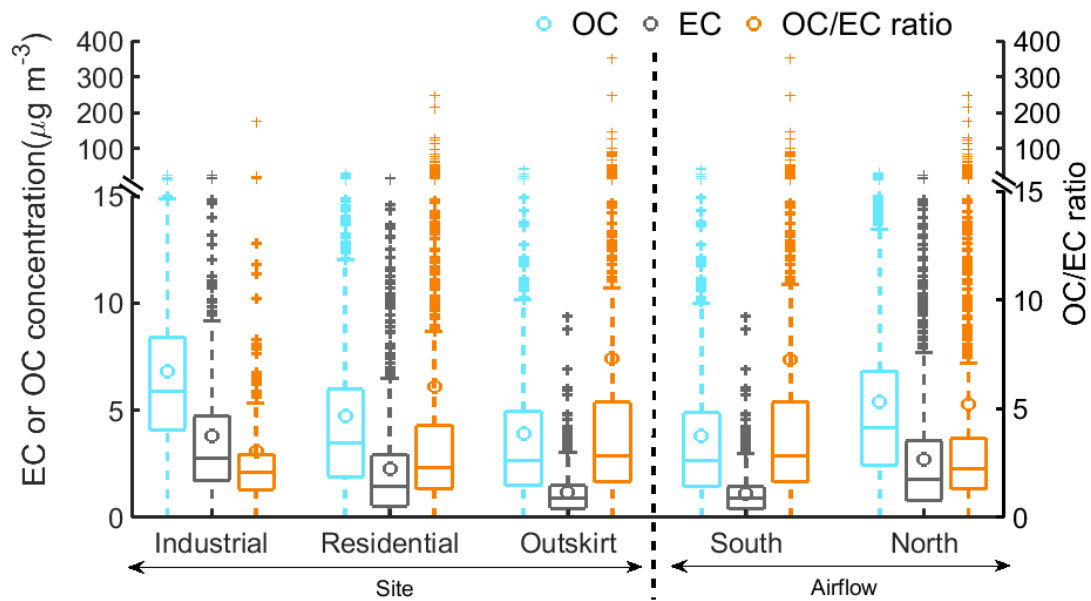


Figure 2 Observed OC and EC concentrations ($\mu\text{g m}^{-3}$) separated by site types and air mass source region according to Table 1 and Figure 1b. Box and whisker plots show median and quartile values; averages are shown as circles and outliers as crosses.

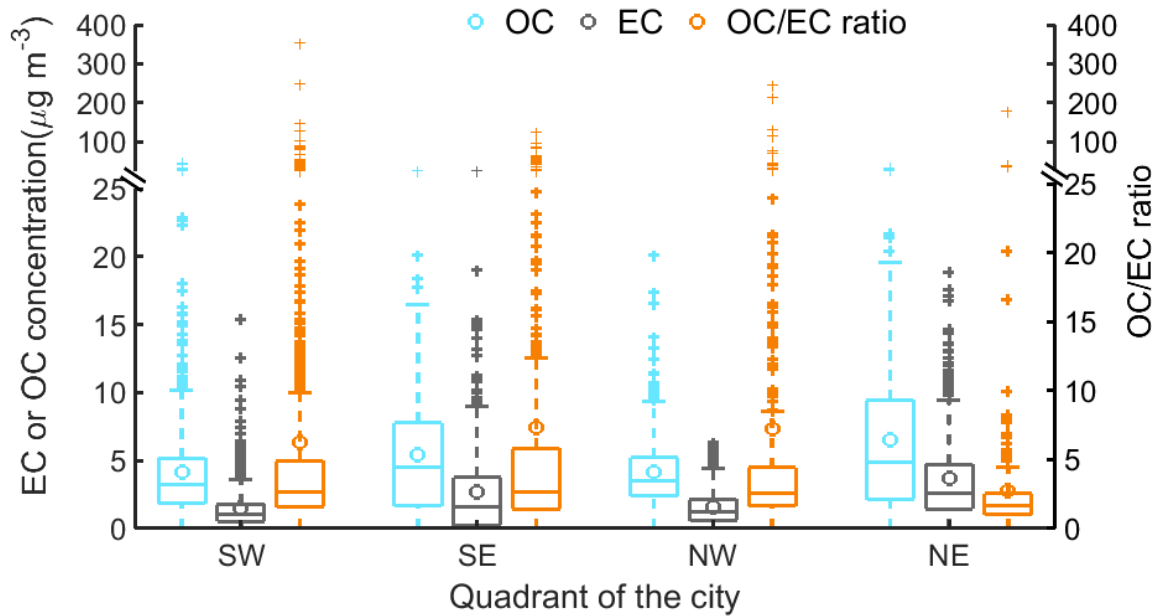


Figure 3 Spatial variation of OC and EC concentrations ($\mu\text{g m}^{-3}$) and OC/EC ratios in each quadrant of Riyadh. SW represents southwest Riyadh and includes the sampling cells 1, 2, 5 and 6; SE represents

southeast Riyadh and includes the cells 3, 4, 7, and 8; NW represents northwest Riyadh and includes the cells 9, 10, 13, and 14; and NE represents northeast Riyadh, and includes cells 11, 12, 15, and 16.

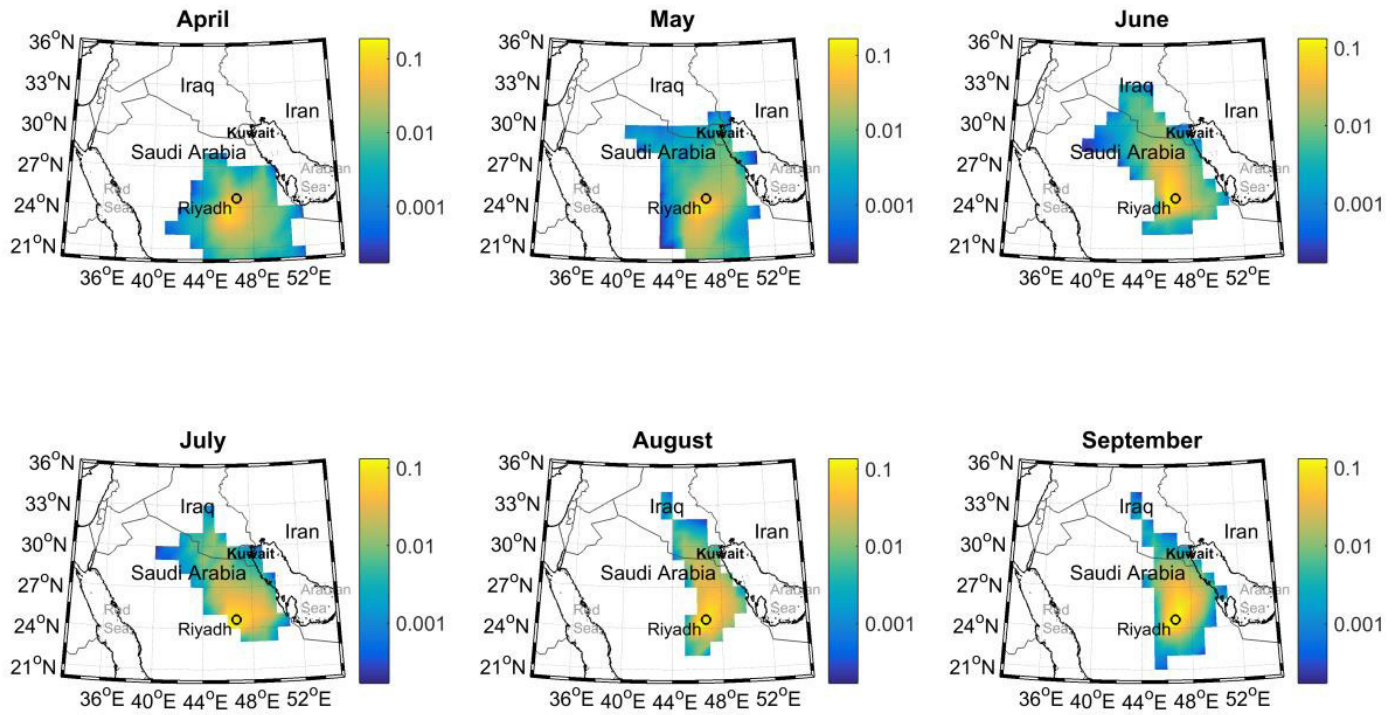
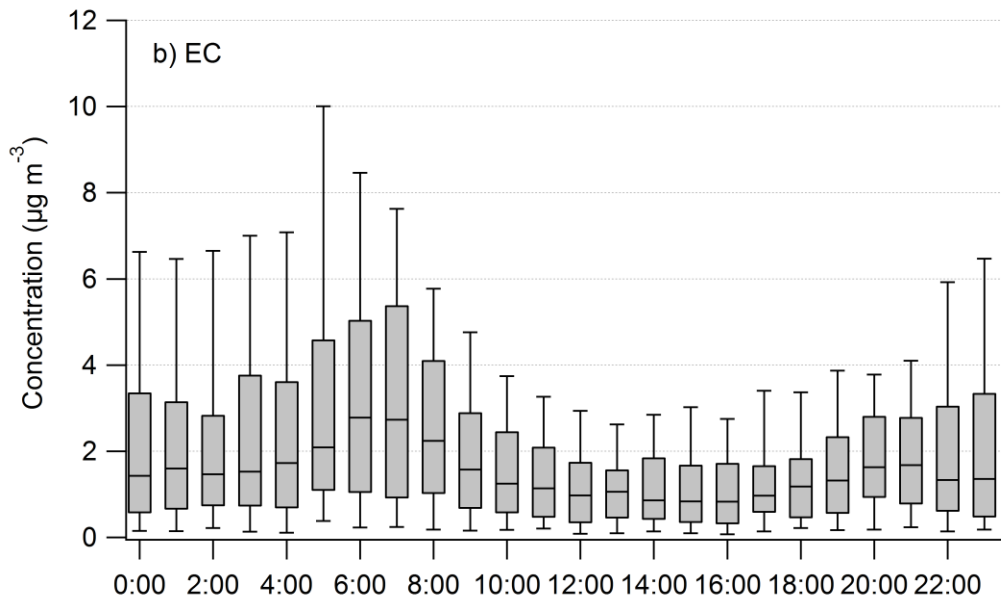
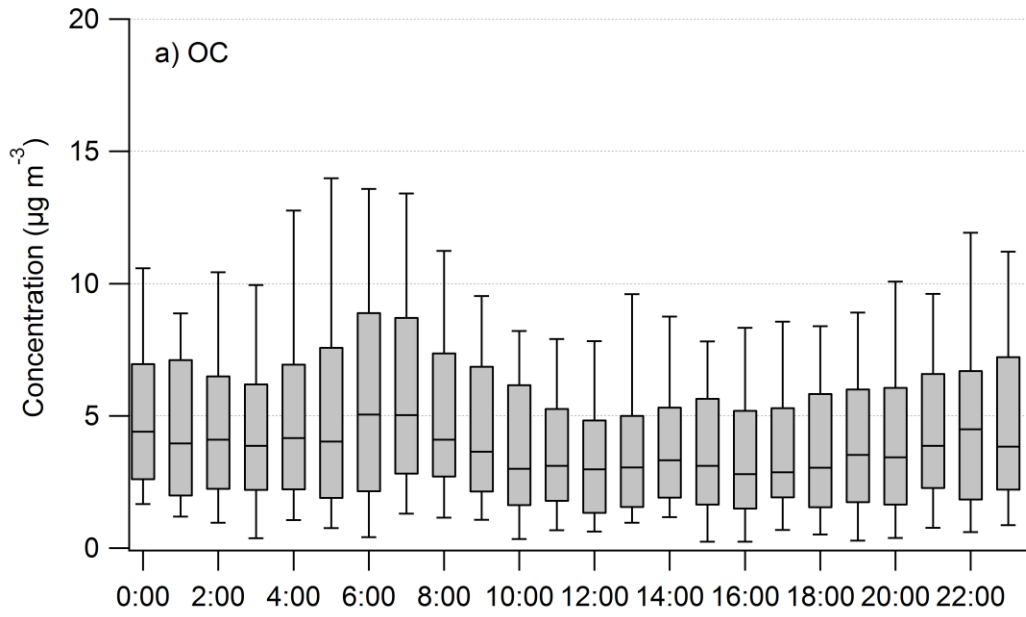


Figure 4 Back trajectory (24 h) residence time analysis of air masses arriving at Riyadh from April to September, 2011. Back trajectories were initiated from a starting height of 500 m above ground level. The color bar represents the normalized number count of the end points.



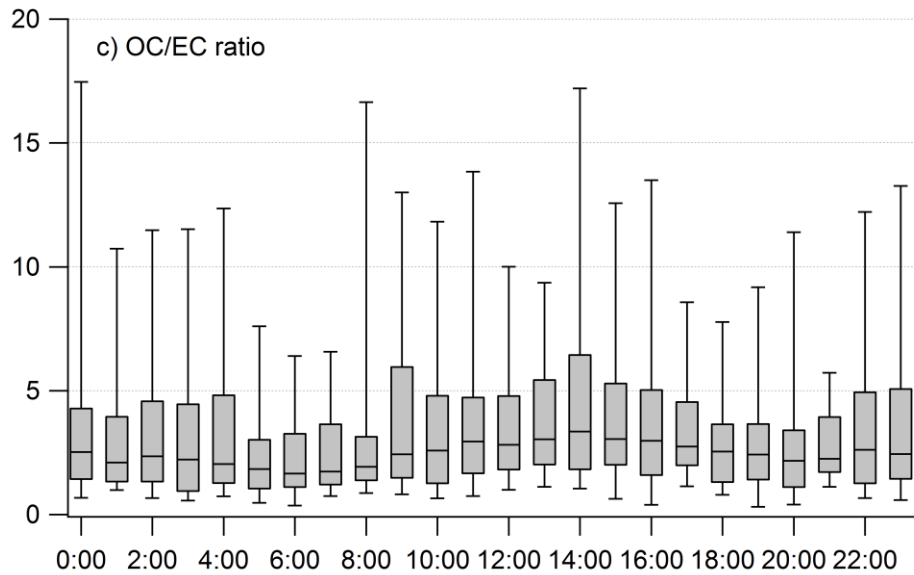


Figure 5 Diurnal variation of a) OC, b) EC and c) OC/EC ratio. Box represent the interquartile range and the upper and lower whisker represent 90% and 10%, respectively.

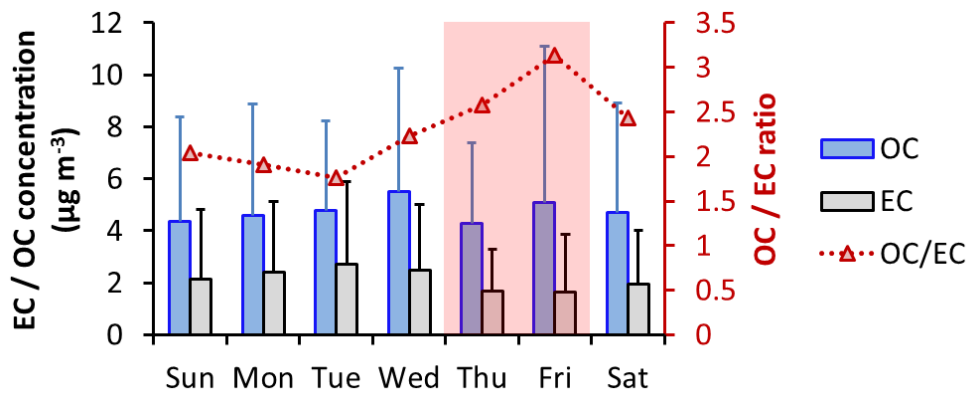


Figure 6 Day-of-week variation in OC ($\mu\text{g m}^{-3}$), EC ($\mu\text{g m}^{-3}$) and OC/EC ratio during the observational period. The shading days (Thu and Fri) were the weekends in Saudi Arabia in 2012.

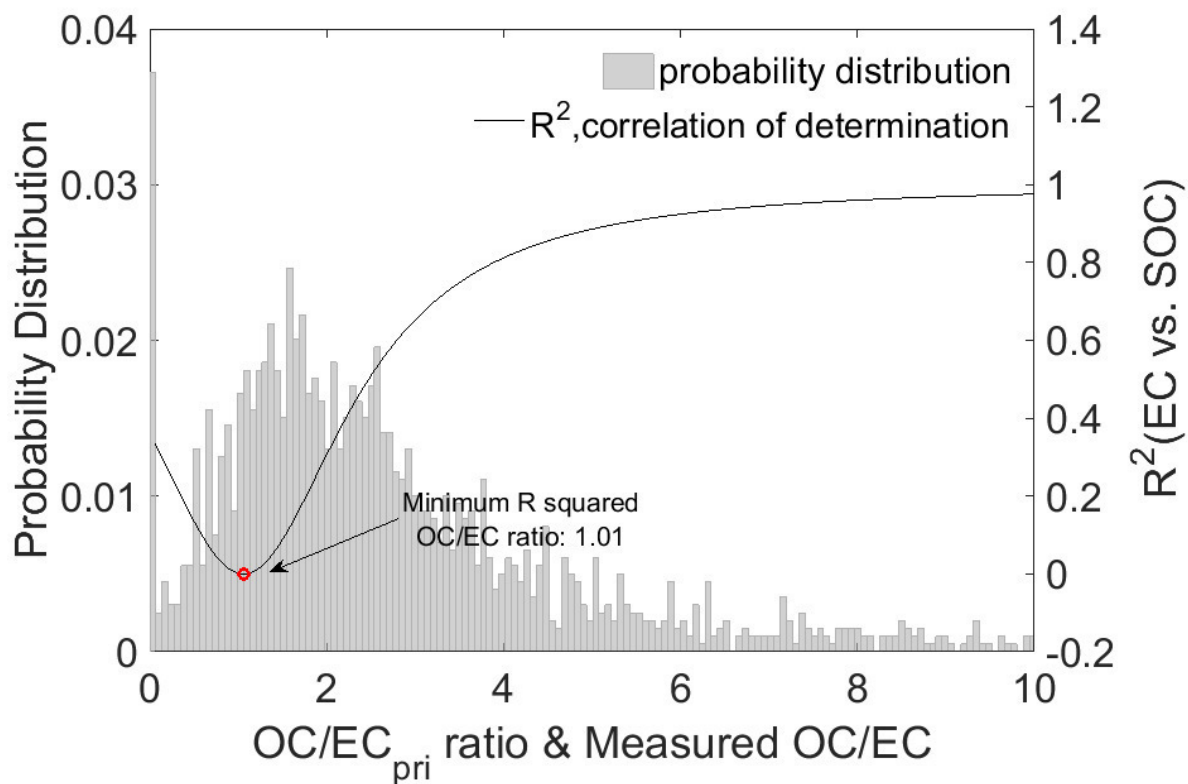


Figure 7 Determination of $(OC/EC)_{pri}$ using the minimum R squared method (MRS). The black curve is the coefficient of determination (R^2) between SOC and EC as a function of the assumed primary OC/EC ratio. The grey shaded area represents the probability distribution of the measured OC / EC ratios. The turning point (red circle) in the curve gives the best-fit primary emission ratio $(OC/EC)_{pri}$.

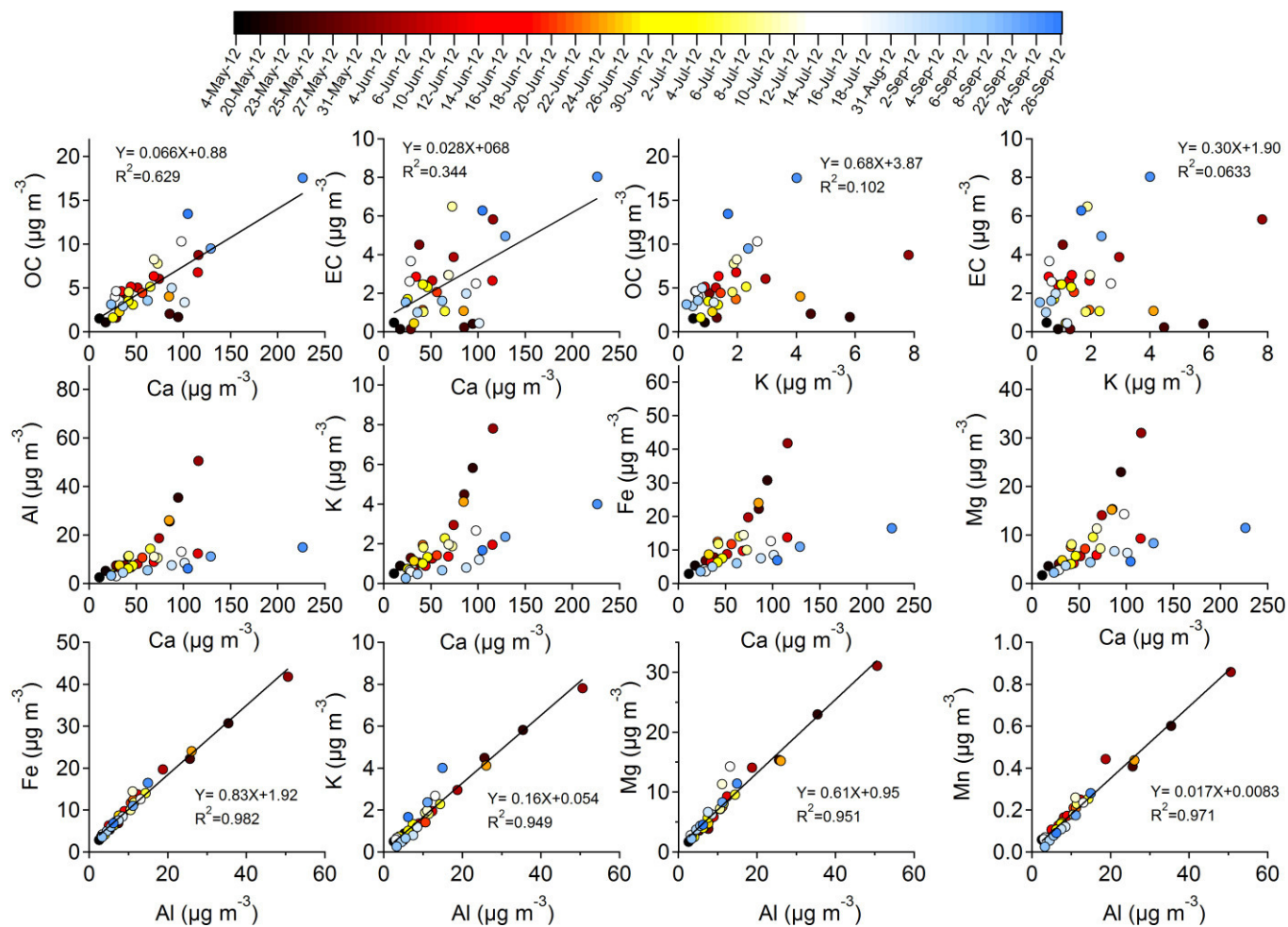


Figure 8 Correlation between dust species (Al, Fe, K, Mg, Mn and Ca), organic carbon (OC) and elemental carbon (EC) concentrations ($\mu\text{g m}^{-3}$). Color bar represents the corresponding sampling date.

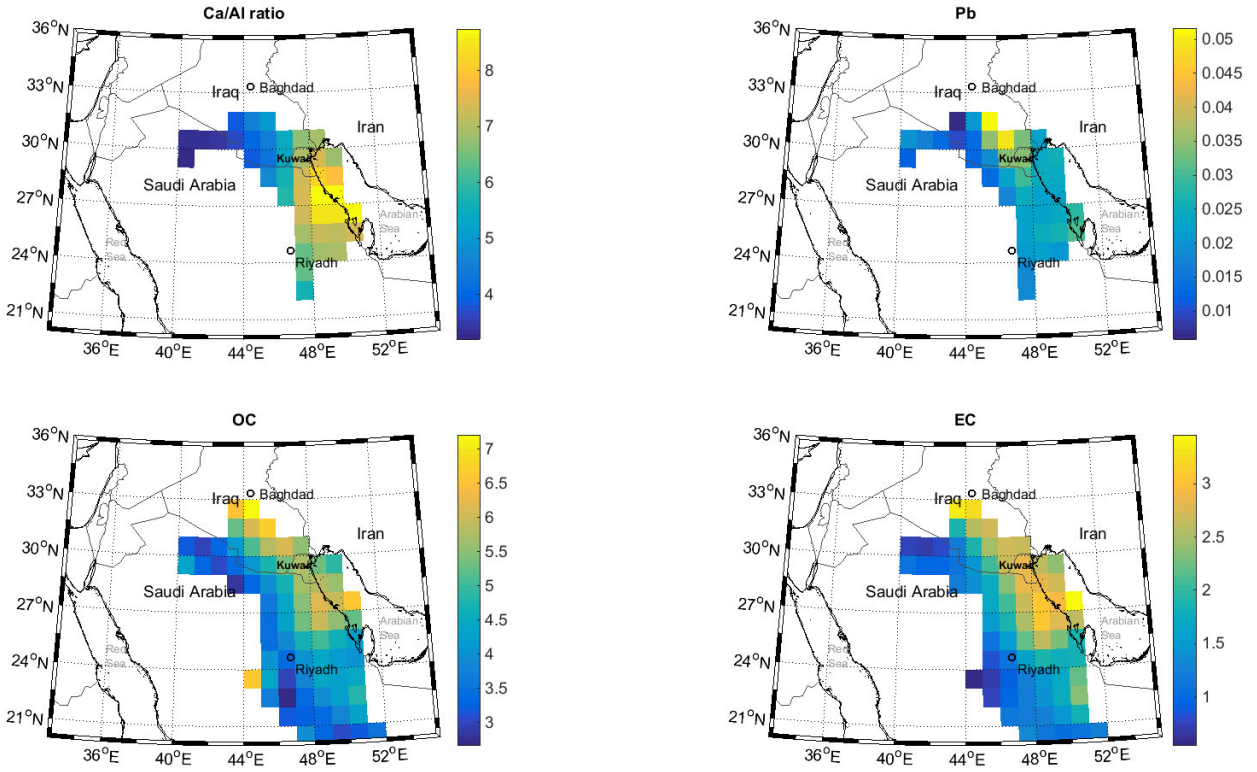


Figure 9 Concentration weighted trajectory analysis for indicated species, for 24-hr back trajectories with a starting height of 500 m. Color bars represent Ca/Al ratio, Pb concentrations (ng m^{-3}), OC concentrations ($\mu\text{g m}^{-3}$), and EC concentrations ($\mu\text{g m}^{-3}$).

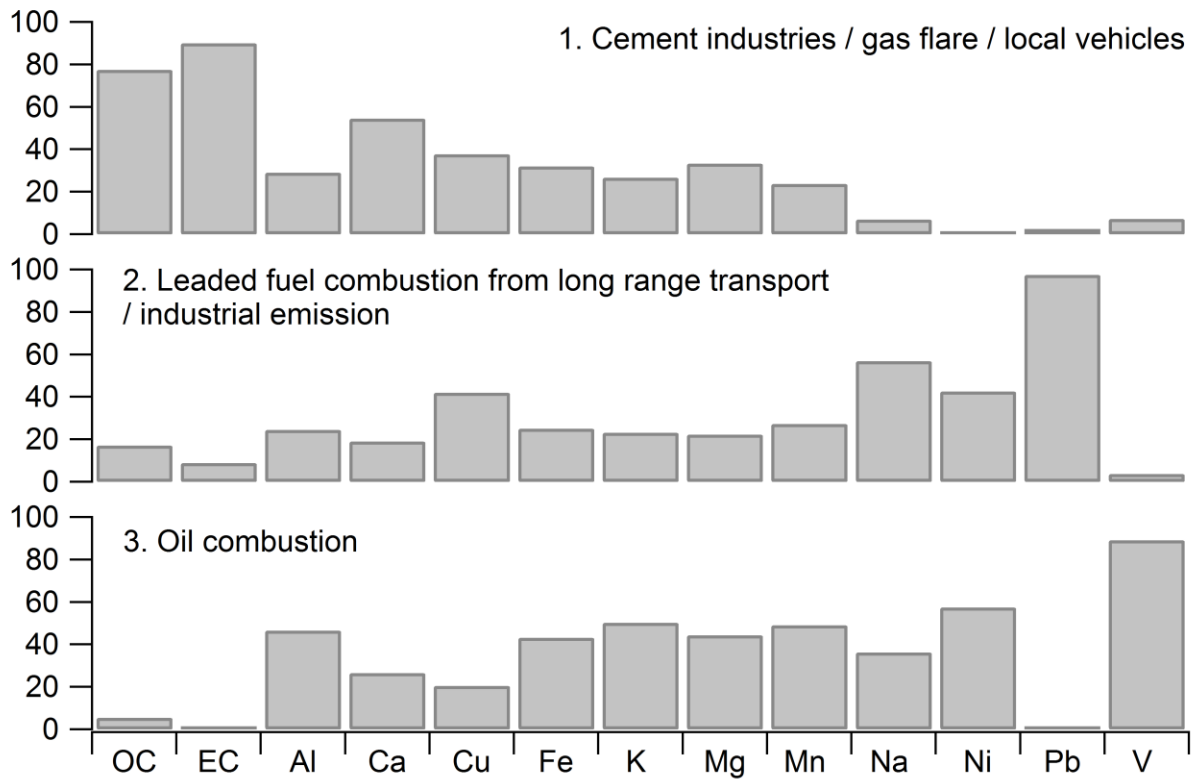


Figure 10 Source profile of PMF analysis of combined PM_{2.5} OC and EC and PM₁₀ metals concentrations. The sum of the species for all the factors was normalized to unity.

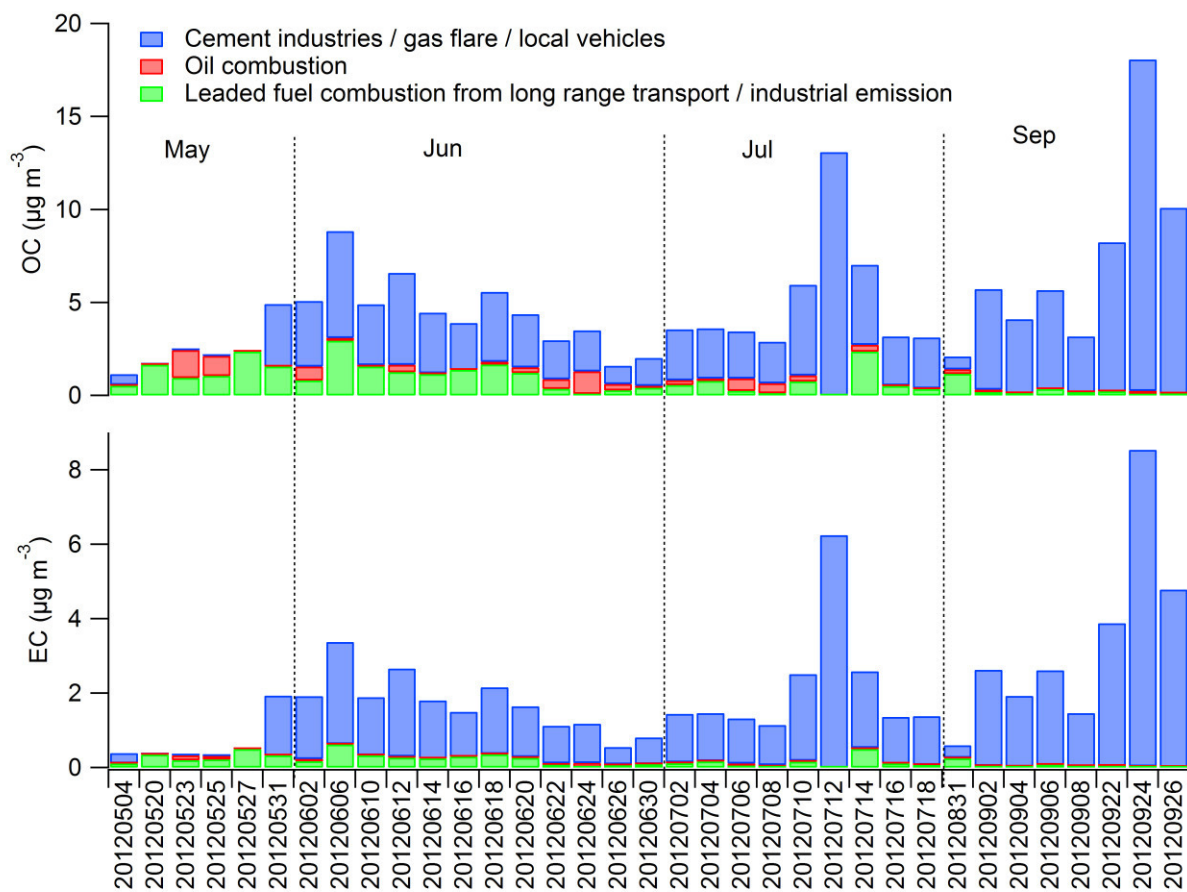


Figure 11 Source contributions to (a) OC and (b) EC ($\mu\text{g m}^{-3}$) from three sources, for each sample.

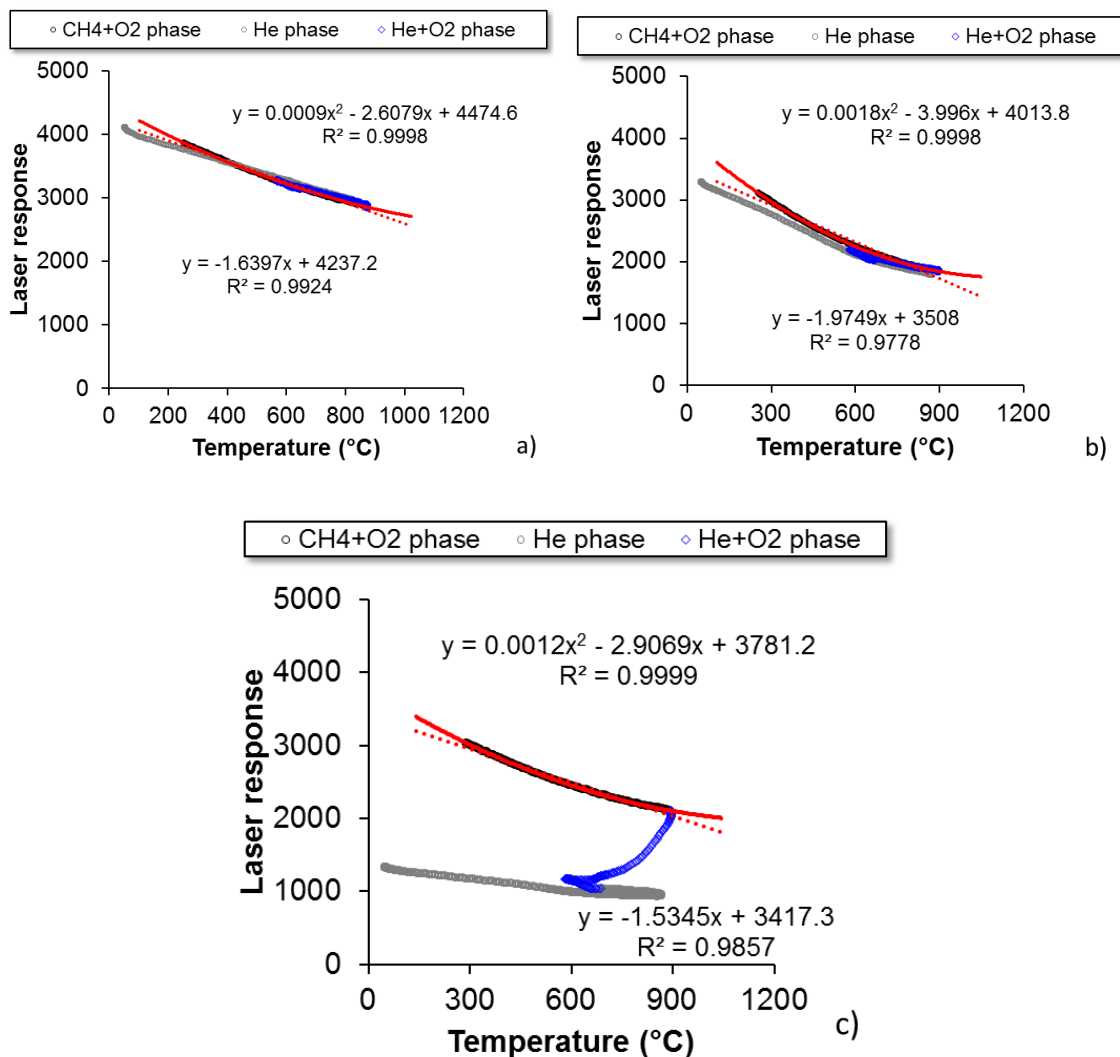


Figure A.1: Correlation between laser response and temperature (°C) for the three samples whose thermograms are shown in Figure A.2. (a) blank at 00:15 am, 20120706; (b) ambient sample at 20:00 pm, 20120706 (c) ambient sample at 6:00 am, 20120709. The gray lines indicate points during the oxygen-free (He only) phase of the analysis, the blue line is for points during the oxidizing stage (He+O₂) of the analysis and the black line is for the points during the calibration stage (CH₄+O₂). The red line is a best-fit polynomial through the CH₄+O₂ points, while the dashed red lines are linear fits.

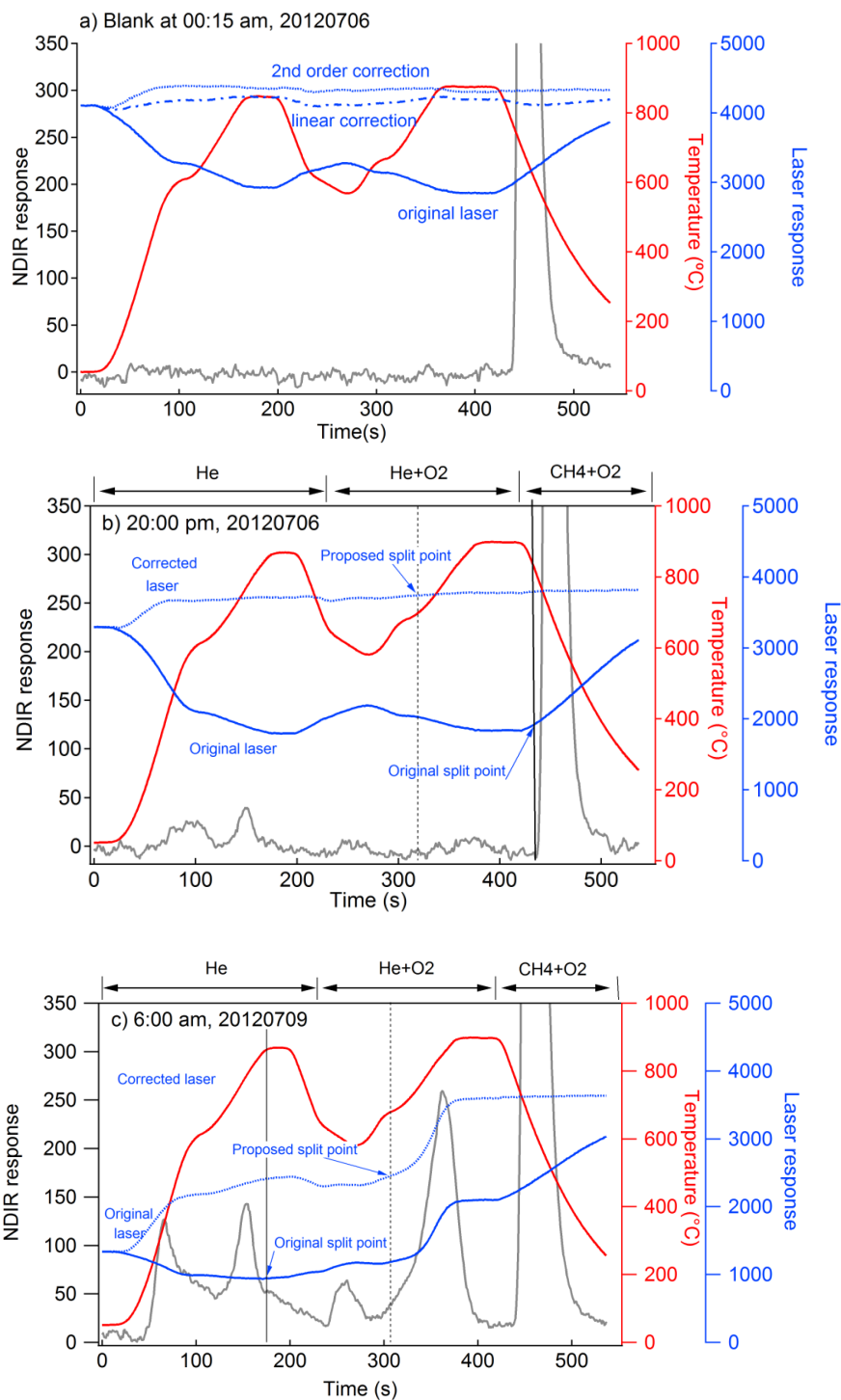


Figure A.2: Thermograms of selected Riyadh samples: (a) blank at 00:15 am, 20120706 (YYYYMMDD); (b) ambient sample at 20:00 pm, 20120706 with relatively low EC loading; (c) ambient sample at 6:00 am, 20120709 with relatively high EC loading.