



## Source apportionment and elemental composition of PM<sub>2.5</sub> and PM<sub>10</sub> in Jeddah City, Saudi Arabia

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### ABSTRACT

This paper presents the first comprehensive investigation of particulate matter with an aerodynamic diameter less than or equal to 2.5 and 10 microns (PM<sub>2.5</sub> and PM<sub>10</sub>) composition and sources in Saudi Arabia. We conducted a multi-week multiple sites sampling campaign in Jeddah between June and September, 2011, and analyzed samples by X-ray fluorescence (XRF). The overall mean mass concentration was 28.4±25.4 µg m<sup>-3</sup> for PM<sub>2.5</sub> and 87.3±47.3 µg m<sup>-3</sup> for PM<sub>10</sub>, with significant temporal and spatial variability. The average ratio of PM<sub>2.5</sub>/PM<sub>10</sub> was 0.33. Chemical composition data were modeled using factor analysis with Varimax orthogonal rotation to determine five and four particle source categories contributing significant amount of for PM<sub>2.5</sub> and PM<sub>10</sub> mass, respectively. In both PM<sub>2.5</sub> and PM<sub>10</sub> sources were (1) heavy oil combustion characterized by high Ni and V; (2) re-suspended soil characterized by high concentrations of Ca, Fe, Al, and Si; and (3) a mixed industrial source. The two other sources in PM<sub>2.5</sub> were (4) traffic source identified by presence of Pb, Br, and Se; (5) other industrial source mixture; while in PM<sub>10</sub> it was marine aerosol. To estimate the mass contributions of each individual source category, the PM mass concentration was regressed against the factor scores. Cumulatively, re-suspended soil and oil combustion contributed 77 and 82% mass of PM<sub>2.5</sub> and PM<sub>10</sub>, respectively.

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### 1. Introduction

Atmospheric pollution in Saudi Arabia, as a serious consequence of the rapid economic and social growth associated with fuel over-consumption, has recently become of considerable interest. Also, just as they are worldwide, chronic illnesses such as cancer, cardiovascular and respiratory diseases constitute a serious public health problem in Saudi Arabia (National Cancer Registry, 2009). It is now established that exposure to PM<sub>2.5</sub> and PM<sub>10</sub> are associated with damaging effects on the human respiratory and cardiovascular systems (e.g., Pope and Dockery, 2006; Peng et al., 2008). In general, the studies of the causes of air pollution on chronic diseases are not conclusive due to the difficulty of assembling large cohorts, following subjects through a long enough period of time, and the difficulty in measuring personal exposures to ambient air pollution (Chen and Goldberg, 2009). However, most recent reviews agree that the human health effects should not be attributed simply to the total mass concentration, and call to establish the toxicity of particulate matter (PM) components (Fanning et al., 2009; Lippmann, 2010), specifically metallic elements (Chen and Lippmann, 2009), inorganic compounds (Schlesinger, 2007) and carbonaceous PM components (Mauderly and Chow, 2008). Identifying the sources of ambient air pollution will support further research of specific mechanistic pathways of pollution and diseases.

Jeddah is the second largest city and the most significant commercial center in the Kingdom of Saudi Arabia. The growth of

the city over the last thirty years has been rapid and diverse, and continues to date. Unfortunately, due to lack of awareness and proper regulations, these development activities were accompanied by environmental degradation, and over the years the air quality progressively deteriorated. Similarly to almost everywhere else in the world, Jeddah's environment and its citizens' health are affected by both stationary and mobile sources. Here, the stationary sources include an oil refinery, a desalination plant, a power generation plant and several industries (Figure 1), as well as desert storms that are common to this area. The city has a major airport in the northern part, and various industrial activities in southern Jeddah. Many of the industrial facilities, such as the oil refinery, were originally built in nonresidential areas, but with urban development that ensued, are now in the middle of highly populated areas, and some of Jeddah's residential areas are particularly affected by several concrete factories. Mobile sources of pollution (all forms of transportation) also became Jeddah's most significant source of air pollution with 1.4 million cars currently registered in Jeddah. Vehicle fuels used in Jeddah are mainly unleaded gasoline and diesel although some Pb is still permissible.

While the earlier study by Nasralla (1983) concluded that concentrations of airborne particulates and other pollutants in Jeddah exceeded air quality standards, there is no published systematic research of sources of atmospheric PM in Jeddah. Several studies have been conducted to assess concentrations of total suspended particulates (TSP) and/or PM<sub>10</sub>, as well as their

lead content (Nasrallah, 1984; Abulfaraj et al., 1990). Most recent study by El-Assouli et al. (2007) reported that  $PM_{10}$  in the city of Jeddah routinely exceeds the average hourly standard for  $PM_{10}$  (established by the Presidency of Meteorology and Environment, (PME, 2001)) of  $80 \mu\text{g m}^{-3}$ . Although most of that PM was contributed by sand storms, the  $PM_{10}$  extractable organic matter collected at 11 sites was found to be genotoxic. The  $PM_{2.5}$  studies are either limited to a specific component such as lead in Saudi Arabia after leaded gasoline phase-out (Aburas et al., 2011), or specific source, such as a study on roadside soil pollution (Kadi, 2009).

We were able for the first time to clearly derive the comprehensive elemental composition, concentrations, and sources of  $PM_{2.5}$  and  $PM_{10}$  in Jeddah, Saudi Arabia. Our study enhances an understanding of chemical and physical properties of atmospheric pollution in the area and apportions the chemical constituents of PM to their emitting sources. The results of this exploratory study established the need for further sampling that will lay groundwork to evaluate the association between particulate air pollution and epigenetic modifications, and the onset of environmentally-related diseases in this area.

## 2. Methods

### 2.1. Geographical location and climate

Jeddah, population 3.4 million, is located on the Red Sea coast in the western part of Saudi Arabia. The city is surrounded by mountains in the north-east, east and south-east. The general climate of Jeddah is warm and moderate in winter; however, in summer it is characterized by high temperature and humidity. Rainfall is generally sparse. Meteorological data was obtained from PME. During the entire sampling campaign the prevailing winds were from west (32%) and north-west (40%). The average temperature was  $33^\circ\text{C}$ , and there was no precipitation.

### 2.2. Particulate sample collection and analysis

The  $PM_{2.5}$  and  $PM_{10}$  samplers [Automated Cartridge Collector Unit (ACCU) Sampler] were installed during June – September, 2011, at seven sites throughout Jeddah. The sites and the location of the study area in relation to the Middle East are shown on Figure 1. Sampling sites in Jeddah were selected according to traffic densities and human activities – three in urban (Site 4–University Campus, Site 5– Al-Nuzlah Al Yamaneyyah and Site 6 – Pitrumin), one in suburban (Site 3 – Al-Rughama), two in residential (Site 1 – Al-Muhammadiyah and Site 2 – Al-Rehab) and one in new residential (Site 7 – Al-Alfiyyah) areas. The urban and suburban areas in Jeddah are characterized by high traffic density and commercial activities. The residential areas are characterized by relatively high traffic density. In contrast, the new residential area is characterized by construction activities and low traffic density. Sampling inlets were installed at a height of 5 to 8 m above ground, as recommended by WMO/GAW (2003), to minimize local influences of surrounding obstructive vegetation, topography or buildings. Daily 24-hr samples were collected on Teflon filters (GelmanTeflo, 37 mm,  $0.2 \mu\text{m}$  pore-size) from midnight to midnight every other day for two weeks at each site, but not concurrently at all sites, except for the University Campus where samples were collected for three months. The specific time periods at each site are shown in Table 1.

The description for gravimetric and elemental analysis via energy dispersive X-ray fluorescence, ED-XRF, of all PM samples is provided in Maciejczyk et al. (2005). Briefly, filter masses were measured on a microbalance (model MT5, Mettler-Toledo Inc., Highstown, NJ). Samples were then analyzed for 33 elements by nondestructive XRF (model EX-6600-AF, Jordan Valley) using five secondary fluorescers (Si, Ti, Fe, Ge, and Mo), and spectral software XRF2000v3.1 (U.S. EPA and ManTech Environmental Technology, Inc.). Concentrations of elements above detection limit, defined as 3 times of the uncertainty of the measurements ( $3\sigma$ ), are reported in Table 3.

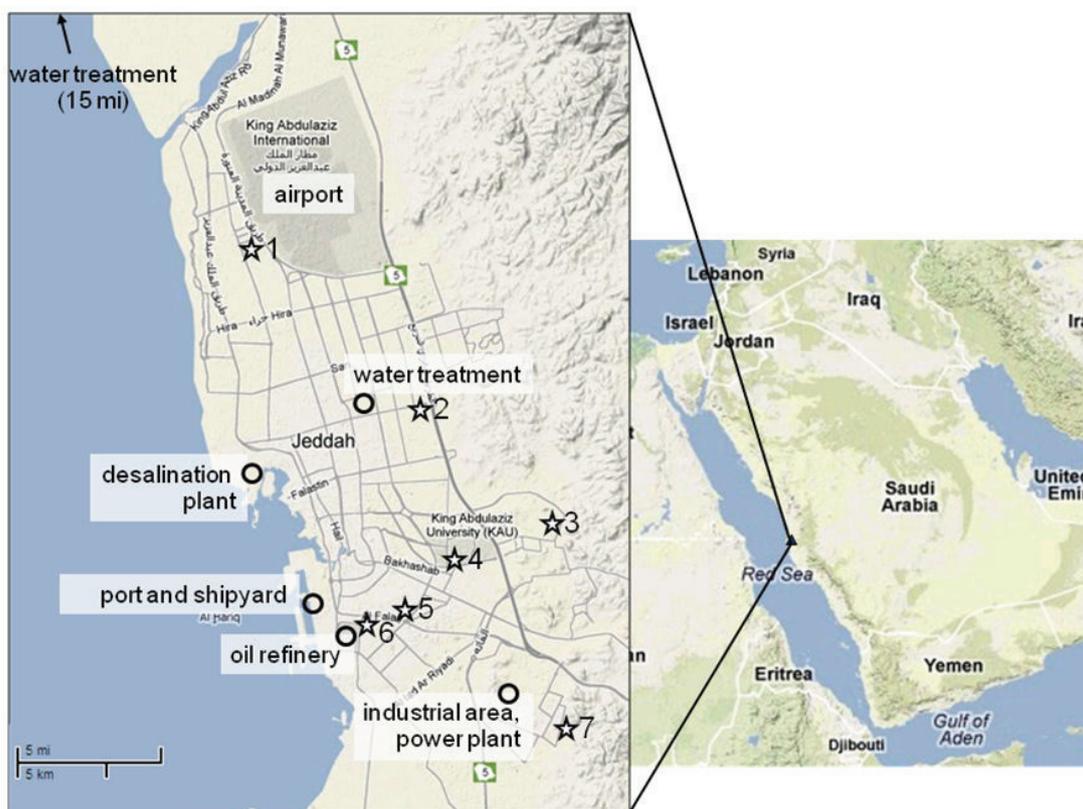


Figure 1. Location of sampling sites (stars) and major industrial sources (circles) in Jeddah, Saudi Arabia.

**Table 1.** Sampling sites in Jeddah and PM concentrations ( $\mu\text{g m}^{-3}$ ) summary

Site Name	Location type <sup>a</sup>	Sampling period in 2011	Number of samples (PM <sub>2.5</sub> , PM <sub>10</sub> )	PM <sub>2.5</sub> (mean±S.D.)	PM <sub>10</sub> (mean±S.D.)	PM <sub>2.5</sub> /PM <sub>10</sub>
1. Al-Muhammadiyah	residential	Sept 10 – 22	7, 7	15.8±3.1	47.0±6.5	0.34
2. Al-Rehab	residential	Aug 13 – 25	7, 7	18.0±4.0	68.9±20.5	0.26
3. Al-Rughama	suburban	July 30 – Aug 11	7, 6	73.2±65.1	141±124	0.52
4. University Campus	urban	June 18 – Sept 24	43, 40	23.8±11.7	84.9±33.1	0.27
5. Al-Nuzlah Al Yamaneyyah	urban	June 20 – 30	6, 6	29.1±14.1	73.5±10.1	0.40
6. Pitrumin	urban	July 2 – 14	7, 7	31.1±5.8	107±28.7	0.29
7. Al-Alfiyyah	residential	July 16 – 28	7, 7	24.5±11.8	99.4±43.9	0.25

<sup>a</sup>Urban= residential + commercial area

Suburban = urban area located at the city border

### 2.3. Source apportionment

To identify major particle source categories in each size fraction, we applied the SPlus Factor Analysis model with Varimax rotations. Since our study is the first source apportionment in this area, the information for some contributing sources is lacking. Thus, multivariate statistical analysis is a reasonable choice for an exploratory investigation that requires no a priori knowledge of the number of sources or source profiles. This and similar multivariate methods are well developed in source apportionment of air pollution for the purpose of epidemiology (see most recent critical review by Heal et al., 2012). Instead, we interpreted the likely physical sources with aid of particular species in each factor. Ambient concentrations of Na, Mg, Al, Si, S, Cl, K, Ca, V, Mn, Fe, Ni, Cu, Zn, Se, Br, and Pb were used as independent variable for both PM<sub>2.5</sub> and PM<sub>10</sub> modeling. The PM<sub>2.5</sub> source apportionment modeling also included Ti and Ba. The selection of species was based on existing knowledge of elemental tracers (e.g., Gordon, 1988; Thurston et al., 2005) as well as XRF detectability. Several major constituents of PM, such as nitrates, elemental and organic carbon, were not measured. Due to the limited number of samples at individual sites, the samples for each size fraction were pooled. Although pooling samples decreased our ability to identify sources unique for each site, it increased the statistical power of the analysis, and allowed to identify sources common to all sites. The number of factors in the final solution was determined by experimenting with different number of factors that yield high eigenvalue ( $\geq 1$ ). The final choice was based on the evaluation of the interpretability of the resulting source profiles and their explanation of total variance of the data set. To obtain the daily mass contribution from each source, the "absolute" scores were regressed onto the gravimetric mass concentrations (Thurston and Spangler 1985; Maciejczyk and Chen, 2005).

## 3. Results and Discussion

### 3.1. PM mass and elemental concentrations

In total, 80 PM<sub>10</sub> and 84 PM<sub>2.5</sub> samples with 17 and 20 variables, respectively, were used for modeling purposes. The mean concentrations of elements with standard deviations, and total mass are shown in Tables 1 and 3. The mean mass concentration was  $28.4 \pm 25.4 \mu\text{g m}^{-3}$  for PM<sub>2.5</sub>, and  $87.3 \pm 47.3 \mu\text{g m}^{-3}$  for PM<sub>10</sub>. It is apparent from Table 1 that PM concentrations also had significant spatial and temporal variability, and these elevated averages with corresponding high standard deviations are mainly due to comparatively large values from Site 3. All sites average concentrations exceeded World Health Organization for both PM<sub>2.5</sub> and PM<sub>10</sub> annual averages of 10 and  $20 \mu\text{g m}^{-3}$ , respectively. The concentration of PM<sub>10</sub> exceeded the European Union Air Quality Standard for daily average of  $50 \mu\text{g m}^{-3}$

in 87.5% of samples, and mean PM<sub>2.5</sub> concentration exceeded the allowable EU annual PM<sub>2.5</sub> average of  $25 \mu\text{g m}^{-3}$ . However, in comparison with U.S. 24-hr average standards, only 10% of PM<sub>10</sub> samples exceeded the standard of  $150 \mu\text{g m}^{-3}$ , and 18% of PM<sub>2.5</sub> samples exceeded the standard of  $35 \mu\text{g m}^{-3}$ . These results clearly indicated that the airborne particulate pollution has been high in Jeddah.

Jeddah's PM levels and ratio of PM<sub>2.5</sub>/PM<sub>10</sub> were compared with those in different urban and coastal urban locations across Europe and Asia (Table 2). The Jeddah's average ratio of PM<sub>2.5</sub>/PM<sub>10</sub> was 0.33 which was significantly lower than in most other urban locations. Only suburban Site 3 had a ratio of 0.52 comparable with the European urban ratios of 0.60 – 0.70. Excluding Site 3, other Jeddah's sites clearly had a significant contribution from coarse PM resulting in an average PM<sub>2.5</sub>/PM<sub>10</sub> ratio of 0.30. These were similar to the ones in Lebanon and India where PM<sub>10</sub> concentrations were also high, most likely due to windblown dust, unpaved roads, and difference in urban landscaping.

As shown in Table 3, the largest elemental contributors to the PM<sub>2.5</sub> mass were S and Si, followed by Al, Fe, and Ca; while the largest contributors to the PM<sub>10</sub> mass were Si and Ca, followed by S, Al, and Fe. The sum of the elements that were measured accounted for 36.5% of PM<sub>2.5</sub> mass and 36.4% of PM<sub>10</sub> mass. Reconstructed mass was computed from the elemental data for comparison with the measured mass concentrations for the same days. This was accomplished by using the following equations that account for the sum of typical PM components observed in the ambient atmosphere: ammonium sulfate, soil, sea salt (Malm et al., 1994; IMPROVE, 2000):

$$\text{Ammonium sulfate} = 4.125 S \quad (1)$$

$$\text{Soil} = 2.20 Al + 2.49 Si + 1.63 Ca + 1.94 Ti + 2.42 Fe \quad (2)$$

$$\text{Sea salt} = 2.54 Na \quad (3)$$

As would be expected, given the lack of nitrate and carbon analysis of the filters, and the possibility of unaccounted sources not represented by the equations, the reconstructed average mass concentrations are lower than those that were measured  $24.5 \mu\text{g m}^{-3}$  for PM<sub>2.5</sub> (accounting for 87.0% of measured mass), and  $63.4 \mu\text{g m}^{-3}$  for PM<sub>10</sub> (or 72.2% of measured mass). Despite the missing speciation data, this calculation of reconstructed mass verifies that we had sufficient trace elemental data to be used in source apportionment. The difference between the actual measured PM<sub>2.5</sub> and the reconstructed mass would therefore include ammonium nitrates, carbon particulates, and any other PM component (such as biological aerosol) that had not been accounted for.

**Table 2.** Comparative concentrations of PM ( $\mu\text{g m}^{-3}$ ) and  $\text{PM}_{2.5}/\text{PM}_{10}$  ratio

Country	Site	Year	$\text{PM}_{2.5}$	$\text{PM}_{10}$	$\text{PM}_{2.5}/\text{PM}_{10}$	Reference
Saudi Arabia	Jeddah (urban - residential)	Summer 2011	28.4	87.3	0.33	This study
Spain	Barcelona (urban)	2003-2004	25.0	39.0	0.64	Pey et al. (2008)
	Tarragona (urban - industrial)	2001	22.2	37.4	0.59	Moreno et al. (2006)
	Hospitalet (urban)	1999-2000	33.9	50.0	0.69	Rodriguez et al. (2004)
	Navarra (urban)	2009	17.4	28.0	0.63	Aldabe et al. (2011)
Greece	Athens (urban)	1999-2000	40.2	76.0	0.53	Chaloulakou et al. (2003)
	Finokalia (coastal)	2004-2006	18.2	31.0	0.63	Gerasopoulos et al. (2007)
Italy	Milan (urban)	2001	45.0	63.0	0.71	Marcazzan et al. (2003)
Turkey	Izmir (urban)	2004-2005	64.0	80.0	0.80	Yatkin and Bayram (2008)
Egypt	Cairo (urban)	Summer 2002	59.0	136.0	0.43	Abu-Allaban et al. (2007)
Lebanon	Beirut (urban)	2004-2005	38.9	103.8	0.37	Saliba et al. (2010)
China	Hong Kong (urban - mixed)	2000-2001	57.4	73.1	0.78	Ho et al. (2003)
Korea	Seoul (urban)	Spring 2001	49.0	51.0	0.98	Kim et al. (2003)
India	Hyderabad (urban)	2004-2005	50.0	135.0	0.37	Gummeneni et al. (2011)

**Table 3.** All sites summary of elemental concentrations ( $\text{ng m}^{-3}$ ) and enrichment factors

	$\text{PM}_{2.5}$				$\text{PM}_{10}$			
	Mean	S.D.	% Detected	EF	Mean	S.D.	% Detected	EF
Na	430	390	99	3	1 600	830	100	3
Mg	300	510	79	2	1 400	750	100	2
Al	800	1 600	100	1	3 300	2 600	100	1
Si	2 100	4 800	100	1	11 000	8 300	100	1
P	80	43	79	18	160	62	96	5
S	3 400	1 300	100	3 000	3 500	1 100	100	490
Cl	62	200	70	51	1 600	1 400	100	430
K	190	230	100	2	710	410	100	1
Ca	540	960	100	2	4 400	2 200	100	3
Sc	4.3	2.3	49	48			ND	
Ti	55	160	83	1	290	320	100	1
V	23	12	99	40	31	14	100	8
Cr	2.1	3.9	23	4	8.8	7.8	90	2
Mn	19	41	99	2	98	81	100	3
Fe	590	1 400	100	1	3 100	2 800	100	1
Co	8.9	14	90	57	32	26	100	34
Ni	6.6	3.6	99	19	11	5.6	100	5
Cu	5.6	8.8	61	17	18	14	96	8
Zn	41	69	100	84	77	120	100	26
As	8.7	14	29	800	10	16	38	150
Se	2.9	1.9	49	14 000	3.6	2.3	58	2 400
Br	8.7	4.3	98	815	15	5.4	100	200
Rb	1.2	1.8	19	4	3.4	3	66	1
Sr	4	7.6	65	2	27	15	99	2
Cd	7.6	15	20	8 800	98	85	88	15 000
Cs	12	16	69	560	52	31	96	470
Ba	34	41	94	11	110	74	100	7
Pb	160	350	83	1 500	200	420	90	300

Atmospheric particles containing trace elements are mainly derived from either crustal sources (e.g., weathering and soil remobilization) or anthropogenic sources. To obtain preliminary information about the sources of elements in Jeddah aerosol, the crustal enrichment factors (EFs) for each element, both in PM<sub>2.5</sub> and PM<sub>10</sub>, were calculated. The enrichment factor for a generic element X with respect to a reference crustal element Y is defined as  $EF_X = (X/Y)_{air} / (X/Y)_{crust}$ , where the ratio (X/Y) is the concentration ratio of X and Y in either aerosol sample or Earth crust. In the present study, Al was used as the reference element Y, and the Earth crust chemical composition was taken from Taylor (1964) and Taylor and McLennan (1985). Our calculation of EFs was done assuming that contributions of man-made sources to Al are insignificant in Jeddah. The use of average crust values provides meaningful comparison to many other studies that commonly use this technique. The average local soil profiles are not available and will be investigated in the follow up study. The mean EF values of the elements measured in PM<sub>2.5</sub> and PM<sub>10</sub> are summarized in Table 3. The EF values less than 10 indicate that the element has a significant crustal source, while EF values higher than 10 are ascribed to elements of anthropogenic origin (Biegalski et al., 1998). In Jeddah samples, EF values lower than 10 were found for Na, Mg, Al, Si, K, Ca, Ti, Cr, Mn, Fe, Rb, Sr in PM<sub>2.5</sub> and Na, Mg, Al, Si, P, K, Ca, Ti, V, Cr, Mn, Fe, Ni, Cu, Rb, Sr and Ba in PM<sub>10</sub>. The similarities between elements in these fractions suggests that both PM<sub>2.5</sub> and PM<sub>10</sub> main sources are of a crustal type (e.g., soil and re-suspended dust), while anthropogenic sources have a lesser contribution. In fact, we found significant correlation between PM<sub>2.5</sub> and PM<sub>10</sub> mass concentrations ( $r = 0.77, p < 0.001$ ), suggesting that PM<sub>2.5</sub> and PM<sub>10</sub> in Jeddah come from similar sources, particularly elements Al, Si, K, Ti, Mn, and Fe that consistently exhibited EF values of 1. Slight EF elevations for Na, Mg, and Ca could account for the effect of sea spray, especially in coarse fraction; other traditional crustal elements (Mn and Sr) indicate additional contributions from anthropogenic and human activities such as construction, wind-blown road dust, cement factories and building material manufacturing facilities.

In Jeddah, P, S, Cl, Sc, V, Co, Ni, Cu, Zn, As, Se, Br, Cd, Cs, Ba and Pb in PM<sub>2.5</sub> and S, Cl, Co, Zn, As, Se, Br, Cd, Cs and Pb in PM<sub>10</sub> had EFs higher than 10. The vast majority of the enriched elements (e.g., S), were more concentrated in PM<sub>2.5</sub> than in PM<sub>10</sub>, which was consistent with the established fact that in an urban environment anthropogenic emissions are of combustion origin such as mobile sources (vehicle-exhaust emissions) and industrial activities (Sternbeck et al., 2002; Loughet et al., 2005; Birmili et al., 2006). High EFs for Zn and Pb, suggests traffic emissions, tire wear, incineration, fossil fuel combustion and construction activities. Elevated Ni, V, S, As and Se are usually attributed to fuel combustion (Gordon, 1988), while Cd, Zn, and Cl are due to

incinerator type emissions (Gordon, 1988; Ondov and Wexler, 1998). The presence of Cd and Cr, Cu and Ni were probably related to anthropogenic activities such as non-ferrous metal industries and chromium plating. Our consideration of EFs and the pattern of spatial variability of elements from site to site (data not shown) had enhanced our understanding of regional and local sources of pollution in Jeddah.

### 3.2. Sources of Jeddah PM<sub>2.5</sub> aerosol – factor analysis

We used factor analysis with Varimax rotation to predict five sources for PM<sub>2.5</sub> and four sources for PM<sub>10</sub>. The factor loadings for each of the source category are shown in Figures 2 and 3. We let a pattern be limited to those variables with 10% or more of their variation involved in a pattern. Factors were interpreted by comparing the factor loadings for all factors and variables. In our modeling, we also considered the communalities for elements, which indicate the proportion of a variable's total variation that is involved in the patterns, and percent total variance for each factor. The contributions of each source to overall mass were computed by each factor score regression onto daily mass, and the averages of daily mass contributions per source are shown in Table 4. These daily source category contributions were paired with wind direction to form pollution wind roses to support our interpretation of source influences at each site. Spatial analysis of data was possible by comparing data periods from Site 4 with other concurrent sampling sites, as also shown in Table 4.

Five factors were identified for PM<sub>2.5</sub>, and they explained 82.7% of total variance of the data set. The communalities for most species were high (>0.80), and the five factors identified were satisfactory. However, low communalities of Cu, Zn and Pb (0.3 – 0.5) indicated that these element concentrations cannot be entirely predicted from identified sources. The first factor that explained most of the total variance (51%) was heavily loaded with Al, Si, and Fe, classified as re-suspended soil source category (e.g., Amato et al., 2009). Although the strongest in our factor analysis, this re-suspended soil source had a mean mass contribution 5.4  $\mu\text{g m}^{-3}$  (or 8.3% of total mass), and had almost uniformly low mass contribution of less than 3%. Sites 5 and 6, located in the western part of the city did not collect significant soil amounts. The wind rose analysis at Site 4 confirmed that even under prevailing westerly winds, soil contribution was rather small. The large soil contributions to samples at Site 3 (46  $\mu\text{g m}^{-3}$ ) were driven by several days of exceptionally high PM mass under south-east winds conditions on 7/30 and 8/3/2011 (115 and 150  $\mu\text{g m}^{-3}$ , respectively). Site 3 is located in the eastern suburb of the city, and under prevailing westerly winds is influenced the most by pollution from nearby Al-Haramain Road that has daily traffic density of 300 000 cars, numerous marble and granite workshops, and

**Table 4.** Average mass source contribution (compared with concurrent samples at University Campus Site 4) to PM<sub>2.5</sub> ( $\mu\text{g m}^{-3}$ )

	Factor 1	Factor 2	Factor 3	Factor 4	Factor 5
	Soil	Heavy oil combustion	Traffic	Industry mix 1	Industry mix 2
1. Al-Muhammadiyah	0.4 (0)	12 (10)	0.7 (1.1)	0.3 (1.4)	0.1 (0)
2. Al-Rehab	0.6 (0.2)	11 (11)	0.9 (0.9)	0.9 (1.8)	0 (0.1)
3. Al-Rughama	46 (11)	18 (16)	0.7 (0.2)	3.8 (3.8)	3.2 (0)
4. University Campus	2.7	14	0.8	2.6	0.1
5. Al-Nuzlah	0 (0)	28 (16)	0.7 (0.8)	2.0 (2.9)	0 (0)
6. Pitrumin	0 (1.0)	34 (15)	0.7 (0.5)	0.5 (2.3)	0 (0.3)
7. Al-Alfiyyah	1.6 (4.2)	17 (17)	1.1 (1.4)	2.3 (3.3)	0 (0)
All sites	5.4	17	0.8	2.1	0.3

ongoing construction, with the latter two producing significant particulate concentrations in both fine and coarse fractions. However, on 7/30 and 8/3 the soil factor mass contribution was also high at the University Campus site (50 and 22  $\mu\text{g m}^{-3}$ , respectively), indicating that the whole city was experiencing an additional dust episode, that most likely was transported. The

analysis of the 48-hr back trajectories indicated the air mass origin from open land of Tokar region in East Sudan. Strong winds up to 29  $\text{km hr}^{-1}$ , 40 °C ambient temperature, and low relative humidity all contributed to increased soil suspension. Overall correlation between the University Campus and the concurrent sites for the re-suspended soil source mass contribution was high ( $r = 0.97$ ).

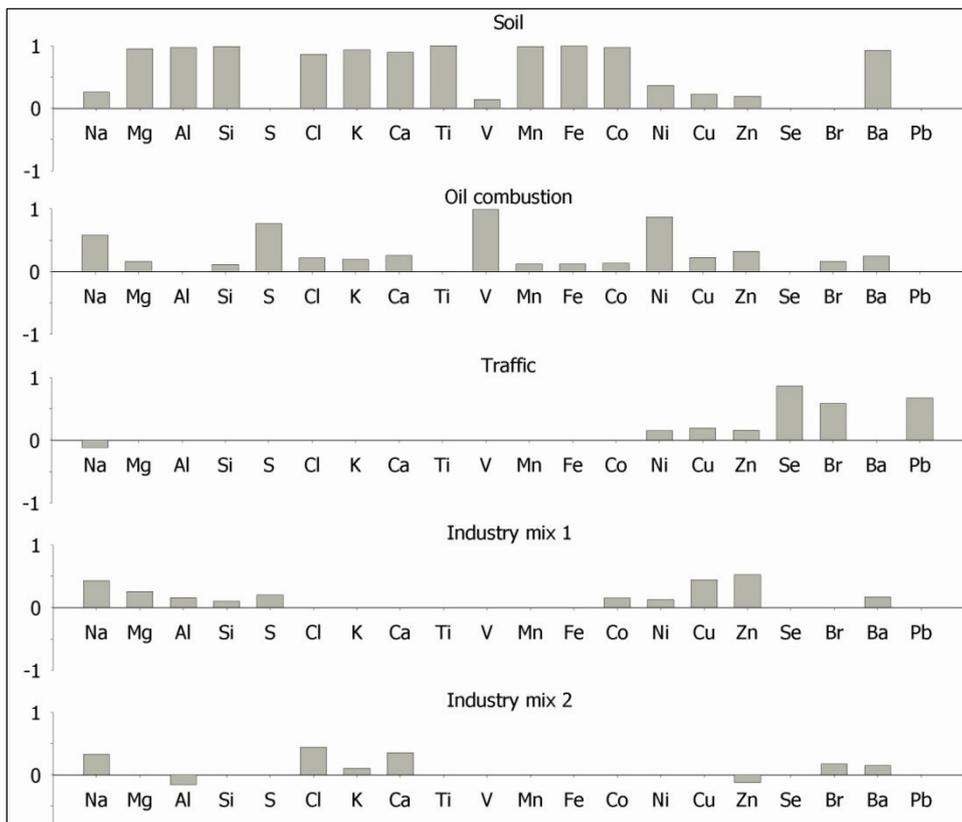


Figure 2. Varimax rotated factor loadings for chemical species of  $\text{PM}_{2.5}$  combined from seven sites.

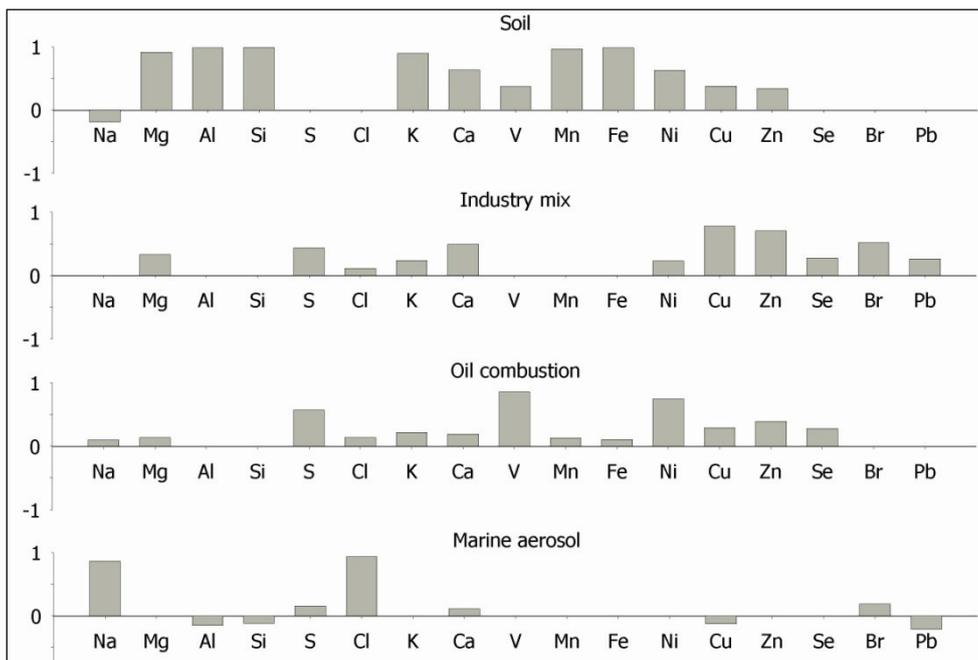


Figure 3. Varimax rotated factor loadings for chemical species of  $\text{PM}_{10}$  combined from seven sites.

The second PM<sub>2.5</sub> factor was rich in V, Ni, and S, and was assigned as emissions of oil combustion (Gordon, 1988) from the Jeddah oil refinery, electrical utilities and industrial and large commercial boilers, as well as residual oil combustion emitted from ocean-going ships docked in Jeddah's port. We did not have sufficient data to attribute some of these sources to transported emissions from oil producing Sudan and Egypt. Mass contribution from this source was 16.9  $\mu\text{g m}^{-3}$  (or 69% of PM<sub>2.5</sub> mass), and explained 15.7% of total variance. Overall, this source contribution had more spatial variation – while University Campus contributions were quite similar regardless of wind direction (10–17  $\mu\text{g m}^{-3}$  or 61–69% contribution to PM<sub>2.5</sub> mass, as shown in Table 4 and Figure 4), Sites 5 and 6, located closest to the port and refinery had the highest Factor 2 contributions of 28 and 34  $\mu\text{g m}^{-3}$ , respectively. Other sites are not immediately downwind of the refinery or the seaport, and thus had lesser contribution. Even at a concurrent Site 4 under exact westerly winds the mass contribution from oil was two-fold less. Thus, it seems that the average background concentration of oil combustion source is about 14  $\mu\text{g m}^{-3}$ , with local point sources having significant effect at immediate proximity. In fact, heavy oil combustion contributed overall 88 and almost 100% of total PM<sub>2.5</sub> mass at sites 5 and 6. Site 3 located in the eastern suburbs, farthest from the port and refinery, had third highest contribution from that source category. Interestingly, the average Ni/V ratio was fairly consistent at all sites (0.24–0.28) except for Site 3 where it was 0.35, indicating possibly another source of V that was not completely resolved by the model. Poor correlation coefficient between Ni and V indicates that these elements were not always emitted from the same source(s) (Maciejczyk et al., 2010; Peltier and Lippmann, 2010). Correlation between the University Campus and concurrent sites was poor ( $r = 0.26$ ), indicative of the local point source influences. Removing Site 3 improved that correlation to  $r = 0.57$ .

The third factor (8.4% of total variance) contained Pb, Br, and Se was assigned as traffic source. This factor mean contribution to PM<sub>2.5</sub> mass concentration was 0.80  $\mu\text{g m}^{-3}$  (or 3.7%). Atmospheric Pb and Br historically had been attributed to traffic emissions as these elements were bound to fuel additives. Although Saudi Arabia phased out the leaded gasoline consumption in January 2001, current allowable Pb content in gasoline remains at 13 mg L<sup>-1</sup>, which means that in high traffic density cities the EF for Pb still is elevated. It was estimated that total Pb in consumed fuel in Jeddah is about 83.6 ton year<sup>-1</sup> (Aburas et al. 2011). Additional vehicular Pb emissions are also caused by engine wear (Smichowski et al., 2008). Besides the motor vehicles, both Pb and Se are emitted during open garbage burning (Emsley, 2011) which happens near sites 3 and 7 as well as north of the city. Selenium, commonly attributed to the coal combustion in Eastern U.S., is also present in oil (ATSDR, 2003). However, here Se did not correlate with heavy oil combustion source but rather with traffic, and thus, here serves as tracer for fossil fuel combustion. Bromine is present in multitude of consumer products, and volatilizes easily when heated during combustion. Elements such as Br, As and Zn are considered as indicators of emission from fossil fuel combustion process, including vehicle exhaust (Gordon, 1988; Pacyna and Pacyna, 2001). Despite the worldwide abatement measures for fuel additives, both Pb and Br are persistently present in both urban and rural environment in Europe and U.S. For example, Pb and Br in the daily TSP and PM<sub>10</sub> samples from 1998–2000 collected in Germany were correlated, however not strongly with  $r(\text{Pb}, \text{Br}) = 0.56$  (Lammel et al., 2002). Similarly,  $r = 0.61$  was found in rural NY samples (Maciejczyk and Chen, 2005; Maciejczyk et al., 2010). Here, we found  $r(\text{Pb}, \text{Br}) = 0.56$  in PM<sub>2.5</sub> and  $r(\text{Pb}, \text{Br}) = 0.51$  in PM<sub>10</sub>. Although Pb has been banned in petrol for several years in Saudi Arabia, the levels of lead in the street dust of the urban areas can still reflect the significant degree of lead contamination from the past. We also noticed that the pollution rose for traffic source was virtually identical with overall wind rose, therefore, this can be considered as a regional source,

further confirmed by the high inter-site correlation ( $r = 0.94$ ) between University Campus and concurrent sites.

The fourth factor (Industry Mix 1) was characterized by high Cu and Zn representing the combined sources of industry and some additional mix of motor vehicle emission. This source category explained 4.5% of the total variance. Although Zn mostly originates from tire wear, it is also attributed to incinerator emissions and metal working (e.g., Ondov and Wexler, 1998; Morishita et al., 2011). The metallurgical processes could produce the largest emissions of Cu, Ni, and Zn (Pacyna, 1998), while exhaust emissions from road vehicles could also contain various amounts of Cu, Zn, and Ni (Pacyna, 1986; Thorpe and Harrison, 2008). There is no typical metallurgical plant in Jeddah; however, welding, fine sanding, and other metal cutting activities occur in the Jeddah port and shipyard on the west side of the city. Spatial and temporal variation of this source was variable, with a mean mass contribution of 2.1  $\mu\text{g m}^{-3}$  (or 8.2%), and poor concurrent site correlation ( $r = 0.31$ ).

The last factor identified for PM<sub>2.5</sub> was rich in Na and Cl, the main elements in sea salt. However, presence of Ca but not Mg, and absence of S indicates that this source is not a marine aerosol, and was named Industry Mix 2. The highest contribution from that source was at the Site 3 which is furthest from the seashore. This was a weak source explaining 2.8% of total variance, with contribution to mass of 0.3  $\mu\text{g m}^{-3}$  (or 0.4%), which was also, spatially, highly variable. Our analysis of wind directions indicate this source contributed to Site 3 exclusively under west and south-west winds, yet during the same time period the contribution to Site 4 was null. We concluded that there were some other sources located in between Sites 4 and 3, and these include fine particulates from polishing (Na and Ca), open air plastic burning (Cl), that all contributed to the significant enrichment of these elements. Further investigation of this source is possible only under favorable wind conditions.

### 3.3. Sources of Jeddah PM<sub>10</sub> aerosol – factor analysis

In PM<sub>10</sub> factor analysis, four factors explained 75% of total variance. The loading for this PM fraction are shown in Figure 3. The strongest Factor 1 explained 39.1% of variance and was rich in Al, Si, and Fe, and, similarly to PM<sub>2.5</sub>, was identified as re-suspended soil. Overall, this factor contributed to 64% of PM<sub>10</sub> mass (or 55  $\mu\text{g m}^{-3}$ ), although there were significant spatial and temporal variations (Table 5). Sites 3 and 7 were affected the most by soil, but, we believe, for different reasons as confirmed by comparison to concurrent sampling at Site 4 (Table 5). Site 3 was downwind of a major highway, open dirt areas, numerous marble and granite workshops, and ongoing construction projects, and, therefore, had significantly higher soil factor contribution (128  $\mu\text{g m}^{-3}$ , or 87% of overall PM<sub>10</sub> mass) than at Site 4 (37  $\mu\text{g m}^{-3}$ , or 49% mass). However, while at Site 7 soil contribution was 78  $\mu\text{g m}^{-3}$ , the concurrent Site 4 samples were also of comparable 82  $\mu\text{g m}^{-3}$ , thus indicating regional dust episode rather than influence of local source. There is no spike for soil under south-east winds at site 4 soil pollution rose because we did not collect the PM<sub>10</sub> samples on 7/30 and 8/3 (the days for high soil mass in PM<sub>2.5</sub> samples). We believe significant contribution from re-suspended soil comes from traffic transporting goods from the seaport, sugar plant, also located near the seaport, and refinery.

Factor 2 (named mixed industrial) in PM<sub>10</sub> showed high loadings of Cu, Zn, Br, S and Ca, and it explained about 13% of total variance. This was the only PM<sub>10</sub> factor positively correlated with Pb. High contributions of Zn and Cu were assigned to metallurgical industries and to diesel powered vehicles. Zn was present in tire wear dust as well as in tailpipe emissions due to its use in motor oil, and Cu was present in brake wear dust (Lopez et al., 2011). This factor was likely a mixture of coarse emission of industrial and

**Table 5.** Average mass source contribution (compared with concurrent samples at University Campus Site 4) to PM<sub>10</sub> (μg m<sup>-3</sup>)

	Factor 1	Factor 2	Factor 3	Factor 4
	Soil	Industry mix	Heavy oil combustion	Marine aerosol
1. Al-Muhammadiyah	21 (39)	3.6 (6.9)	13 (12)	9.1 (5.9)
2. Al-Rehab	42 (46)	7.3 (7.1)	6.8 (8.3)	6.8 (7.6)
3. Al-Rughama	128 (37)	4.5 (5.7)	8 (16)	4.2 (3.2)
4. University Campus	51	9.0	13	6.7
5. Al-Nuzlah	43 (58)	5.8 (11)	19 (9)	8.8 (10)
6. Pitrumin	46 (47)	7.4 (13)	31 (12)	5.5 (7.1)
7. Al-Alfiyyah	78 (82)	10 (10)	11 (24)	5.6 (3.3)
All sites	55	7.8	14	6.7

traffic sources similar to Factor 3 and 4 for PM<sub>2.5</sub>, however, we were not able to further split this source. Contribution of this source was somewhat variable, on average 7.8 μg m<sup>-3</sup> (or 10% of PM<sub>10</sub> mass), and at the Site 4 it followed the wind rose, and the concentrations there were almost higher than at any other concurrent other sites.

The third factor had high loadings of V, Ni and S, and this clearly represented oil combustion similarly to PM<sub>2.5</sub> Factor 2. The average mass contribution of this heavy oil combustion source was 14 μg m<sup>-3</sup> (or 18% mass). Sites 5 and 6 in immediate proximity and downwind of Jeddah refinery and sea port had the largest contribution of 19 and 31 μg m<sup>-3</sup> (or 26 and 29% PM<sub>10</sub> mass). Site 1, which is also close to the seashore, was likely affected by ships' exhaust. Temporal variation of oil source contributions to Site 4 confirms that this is like a point source, and thus highly affected by wind direction. For example, during concurrent sampling at Sites 4 and 7, the overall contribution of oil combustion source to Site 4 was twice as high (24 μg m<sup>-3</sup>), quite possibly because Site 7 (with 11 μg m<sup>-3</sup>) was not downwind of the refinery and port emissions. Notably, the northwest contribution is missing in Site 4 PM<sub>10</sub> pollution rose, compared to PM<sub>2.5</sub>. The fourth factor was loaded with Na and Cl, which we attributed to coarse sea salt of marine aerosol which contributed on average 6.7 μg m<sup>-3</sup> of PM<sub>10</sub> mass (or about 9% mass). This source was reasonably expressed under favorable winds from the sea. Site 1 had larger overall contribution from the marine aerosol due to its proximity to the seashore.

#### 4. Conclusions

This is the first study of elemental composition and sources of PM<sub>10</sub> and PM<sub>2.5</sub> in Saudi Arabia, and it reveals that airborne particulate matter in Jeddah is a serious problem. Using factor analysis, we determined that five and four source categories contributed to the mass concentration of PM<sub>2.5</sub> and PM<sub>10</sub>, respectively, and estimated daily levels of source-apportioned PM mass concentrations for each of these categories. The PM<sub>2.5</sub> mass in Jeddah was dominated by contribution from heavy oil combustion (69%), followed by resuspended soil (8.2%), industrial mix 1 (8.2%), traffic (3.7%), and industrial mix 2 source (0.4%). While mass contributions from soil (64%), heavy oil combustion (18%), mixed industrial sources (18%) and marine aerosol (9.3%) were found in PM<sub>10</sub>. Mass reconstruction accounted for 87.0% of measured PM<sub>2.5</sub> mass, and 72.2% of measured PM<sub>10</sub> mass. Due to the sampling limitations our source apportionment did not account for primary and secondary organic aerosol, and primary and secondary marine aerosol. Our modeling determined the contributions from individual sources within each size fraction, identified the need for further sampling, and could be used to (1)

better focus control strategies and (2) in a subsequent time-series analysis of health effects, in particular morbidity outcomes.

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