Health risk associated with airborne particulate matter and its components in Jeddah, Saudi Arabia

Harrison, Roy M.; Bousiotis, Dimitrios; Mohorjy, A. M.; Alkhalaf, A. K.; Shamy, M.; Alghamdi, M.; Khoder, M.; Costa, M.

DOI:
10.1016/j.scitotenv.2017.02.216

HEALTH RISK ASSOCIATED WITH AIRBORNE PARTICULATE MATTER AND ITS COMPONENTS IN JEDDAH, SAUDI ARABIA

Roy M. Harrison[a,d,e*], Dimitrios Bousiotis[a], A.M. Mohorjy[b], A.K. Alkhalaf[c], M. Shamy[d], M. Alghamdi[d], M. Khoder[d,e], M. Costa[f]

[a] Division of Environmental Health and Risk Management
School of Geography, Earth and Environmental Sciences
University of Birmingham
Edgbaston, Birmingham B15 2TT
United Kingdom

[b]Department of Civil Engineering, Faculty of Engineering
King Abdulaziz University, Jeddah, Saudi Arabia
(email: amohorjy@kau.edu.sa)

[c]Department of Meteorology, Faculty of Meteorology
Environment and Arid Land Agriculture
King Abdulaziz University, Jeddah, Saudi Arabia
(email: akhalaf@kau.edu.sa)

[d]Department of Environmental Sciences, Faculty of Meteorology
Environment and Arid Land Agriculture
King Abdulaziz University, Jeddah, Saudi Arabia
(email: mshamy@kau.edu.sa; mans99@gmail.com;)

[e]Center of Excellence in Environmetal Studies
King Abdulaziz University, Jeddah, Saudi Arabia
(email: khoder_55@yahoo.com)

[f]Department of Environmental Medicine
New York University School of Medicine
New York, NY, USA
(email: max.costa@nyumc.org)

[*] To whom correspondence should be addressed.
Tel: +44 121 414 3494; Fax: +44 121 414 3709; Email: r.m.harrison@bham.ac.uk

ABSTRACT

Samples of $PM_{2.5}$ and PM_{10} have been collected in all of four seasons at seven sites within the city of Jeddah, Saudi Arabia. The samples have been analysed for a range of trace elements. There is a large loading of wind-blown dust and the majority of elements are predominantly associated with coarse particles. Enrichment factors, however, show that some elements are markedly enriched above crustal abundance. Using mean data for the $PM_{2.5}$ and PM_{10} fractions from each of the seven sampling sites, health risks have been estimated for particulate matter mass, the elements Cr, Mn, Ni, Pb, As, Cd and V measured in this study, and polycyclic aromatic hydrocarbons using data from an earlier study within Jeddah. Cancer risks are calculated from mean airborne concentrations and cancer slope factors for the carcinogenic metals and PAH, but the cancer risks are relatively modest compared to the lifetime risk of mortality due to $PM_{2.5}$ exposure. The risks associated with exposure to V and Mn are considered to be small, while concentrations of cadmium far exceed the European Union Limit Value and World Health Organisation guideline. Cadmium shows a very high crustal enrichment factor but is present predominantly in the coarse particle fraction suggesting that local soils and surface dusts are unusually enriched in Cd relative to the global average. Using national data for mortality rates, the excess mortality due to $PM_{2.5}$ exposure has been calculated and amounts to over 1100 deaths annually for the city of Jeddah.

Keywords: Particulate matter; $PM_{2.5}$; PM_{10}; health risk

1. INTRODUCTION

Saudi Arabia is a country with a fast growing population enumerated as 30.8 million in 2014. The population is heavily focussed on the major cities and especially Riyadh and Jeddah. The city of Jeddah is located on the Red Sea coast of Saudi Arabia and has a population of 3.98 million (in 2014). In addition to its resident population, the sea port and airport of Jeddah act as a gateway for pilgrims entering Saudi Arabia for the traditional Hajj and Umrah in the Holy City of Makkah. Jeddah extends considerably further from north to south than from east to west (see Figure 1) with the Red Sea on its western border. However, to the north, south and east of Jeddah lie large areas of desert which provides an extensive source for wind-blown dusts.

While there have been air quality studies in the inland city of Makkah (Al-Jeelani, 2009; Simpson et al., 2014), and the coastal town of Yanbu to the north of Jeddah (Khalil et al., 2016), these have focussed largely on gas phase pollutants and only the latter study provides limited data for particulate matter concentrations. Mean concentrations of PM_{10} and $PM_{2.5}$ in Yanbu based on six years of observations are reported as 70 µg m^{-3} and 60 µg m^{-3} respectively (Khalil et al., 2016). The small differential between $PM_{2.5}$ and PM_{10} measured between 2000 and 2005 in Yanbu is rather surprising and diverges from the experience of many other sites in western Saudi Arabia (e.g. Khodeir et al., 2012).

There have been a number of studies within and close to the city of Jeddah. Kadi (2014) reports measurements of total suspended particulate matter (TSP) collected with high volume samplers together with analyses of Al, Ba, Ca, Cu, Mg, Fe, Mn, Zn, Ti, V, Cr, Co, Ni, As and Sr. These were made at seven sites within Jeddah, and concentrations of the various metallic components and crustal enrichment factors are reported. Enrichment factors of elements at the more polluted sites range approximately from 10-60 whilst for Cu and Zn, these are much higher at some of the sites with a peak value of over 700 for Cu at a site influenced by light industry and road transport

activities. The data show very large inter-site differences for the majority of the elements analysed. In another paper, the same author (Kadi, 2009) also determined soil composition and reports a strong elevation in lead and zinc content at heavily trafficked sites.

Khodeir et al. (2012) report data from seven sampling sites within Jeddah from samples collected in 2011. They report overall mean mass concentrations of 28.4 ± 25.4 μg m^{-3} for $PM_{2.5}$ and 87.3 ± 47.3 μg m^{-3} for PM_{10} with considerable spatial and temporal variability. The average ratio of $PM_{2.5}$ to PM_{10} of 0.33 appears typical of data from western Saudi Arabia but is very different from the pattern of behaviour reported above from Yanbu. Khodeir et al. (2012) provide a factor analysis model with Varimax orthogonal rotation to determine the sources contributing to concentrations of $PM_{2.5}$ and PM_{10}. These include heavy oil combustion, resuspended soil and a mixed industrial source for both $PM_{2.5}$ and PM_{10}, and for $PM_{2.5}$ road traffic and a second industrial source, and for PM_{10}, marine aerosol. The main contributor to $PM_{2.5}$ was identified as heavy oil combustion while for PM_{10} it was wind-blown soil. Crustal enrichment factors relative to Fe in $PM_{2.5}$ were very high for S (average 3000), Se (14000) and Cd (8800). The same elements were enriched in PM_{10}, with Se (2400) and Cd (15000) showing the highest enrichment.

Alghamdi et al. (2015a) sampled PM_{10}, $PM_{2.5}$ and PM_1 fractions and measured the elemental composition of $PM_{2.5}$ in Jeddah during March 2012. The data were disaggregated into dust storm and non-dust storm periods. Based upon enrichment factors, it was concluded that in both non-dust storm and dust storm periods, the main sources of Na, Mg, Si, K, Ca, Ti, Cr, Mn, Fe, Rb and Sr are of a crustal type whereas S, Cl, Co, Cu, Zn, Ga, As, Pb and Cd as well as V and Ni are predominantly anthropogenic. The conditions giving rise to dust storms were also considered. Crustal enrichment factors relative to aluminium in $PM_{2.5}$ were highest for S (average 2792), As (2581), Cd (28,699) and Pb (5879). Enrichment factors were highest during non-dust storm conditions but the same elements also showed enrichment during dust storm conditions. From

samples collected in Riyadh, Alharbi et al. (2015) found that concentrations were considerably higher in summer than winter which was attributed to dust storm activity. Crustal species such as Fe, Mn, Ti, Ca and Mg were found at appreciably higher concentrations in summer.

Porter et al. (2014) analysed PM_{10} data collected in 2010-2011 in sites in and around Jeddah and at a remote background site for comparison. Data were collected with automated beta gauges making diurnal variations in concentrations available. The PM_{10} concentrations do not show a very consistent seasonal pattern with major differences between the various sites. PM_{10} showed a reduced concentration at weekends relative to weekday concentrations clearly indicating an anthropogenic influence. Data from Yanbu (Khalil et al., 2016) showed marked diurnal variations that do not link clearly with road traffic activity and appear more likely to be influenced by the speed of local winds.

Shaltout et al. (2013, 2015) have reported concentrations of $PM_{2.5}$ and trace elements in the city of Taif in western Saudi Arabia. In the more recent study (Shaltout et al., 2015) they report $PM_{2.5}$ concentrations of 50, 57 and 37 µg m^{-3} respectively at traffic, industrial and residential sites.

Measurements of particulate matter made at a rural background site (Hada Al-Sham) about 60 km east of the Red Sea coast and the city of Jeddah are reported by Lihavainen et al. (2016). Mean PM_{10} concentrations were 109 ± 89 µg m^{-3} and $PM_{2.5}$, 38 ± 68 µg m^{-3} hence showing a clear dominance of coarse mode particles. PM_{10} concentrations were markedly higher in January to June than in July to December, but given the limited duration of sampling, it is difficult to attach any significance to this. The mass fraction of $PM_{2.5}$ was around 0.35 and showed maxima in February and December with minimum concentrations in March, June and July. PM_{10} and $PM_{2.5}$ showed diurnal variations which appeared to be related to traffic activity with reduced concentrations at the weekend. The strength of the diurnal variation, apparently connected with traffic, is rather

surprising given that it was a rural site. However, the authors speculate that the strong sea breeze circulation with diurnal changes in wind direction and speed may have been an important influence. The fact that black carbon showed a very different diurnal pattern peaking at night suggests that road traffic was probably not the cause.

Two studies have reported concentrations of particulate and vapour phase polycyclic aromatic hydrocarbons (PAH) sampled at sites within and north of Jeddah (Alghamdi et al., 2015; Harrison et al., 2016). Alghamdi et al. (2015) carried out a source apportionment study reporting that the major identifiable sources of PAH were gasoline vehicles (17%), industrial sources, particularly the oil refinery (33%) and diesel/fuel oil combustion (50%).

2. EXPERIMENTAL

Full details of the sampling sites and analytical methods are given by Khodeir et al. (2012). In the interests of completeness, brief details are provided here. The sampling sites and brief details of their characteristics are provided in Table 1, while Figure 1 shows their location within the city of Jeddah and in relation to major local sources. In particular, the desalination plant is notable as it burns heavy fuel oil and emits through two elevated chimneys. The older parts of Jeddah lie to the south, where there is a concentration of light and heavy industries, mainly concentrated around the port and refinery area (see Figure 1). In contrast, the north of Jeddah is more recently developed, with less industry and lower population density. Hussein et al. (2014) provide a valuable map which shows the distribution of light and heavy industries and major facilities in the city.

$PM_{2.5}$ and PM_{10} were sampled using an automated cartridge collector unit (ACCU) sampler in the period June 2011 to June 2012. Daily samples of 24 hours duration were collected on alternate days on 37 mm, 0.2 µm pore size Gelman Teflo filters. Chemical analysis was by energy dispersive x-

ray fluorescence after filter mass had been determined on a micro-balance (Mettler-Toledo Model MT5).

Samples were collected through all seasons of the year, the number at each site appearing in Table 2.

3. RESULTS AND DISCUSSION

The sampling sites used in this study were the same as those used by Khodeir et al. (2012) for their receptor modelling work, but the study now reported differs in important ways from the work of Khodeir et al. (2012). Firstly, a substantially larger number of samples was collected, and at all sites, samples were collected in all of the four seasons of the year. This was in order to estimate annually-averaged exposure, rather than that in just one season of the year. The seasonal variations have not been analysed as they are not directly relevant to the topic of this paper, which focusses on the consequences of long-term exposure. Secondly, Khodeir et al. (2012) pooled their data and did not look at it on an individual site basis. We now look at site-specific information for PM mass and selected health-relevant trace constituents.

A large number of elements was analysed and the data are summarised for individual sites in relation to means and standard deviations in Tables S1 to S7 in the Supplementary Information. As this study is focussed upon the health risk associated with particulate matter exposures within the city of Jeddah, the data analysis has focussed upon $PM_{2.5}$ and PM_{10} mass and a number of specific chemical constituents for which there are significant health concerns, and for which regulatory guidelines and standards are available. Those elements are chromium (Cr), nickel (Ni) and arsenic (As), which are of concern because of their carcinogenicity, lead (Pb) which is a potent neurotoxin, manganese (Mn) which can affect neuro-behavioural function, vanadium (V), which is a potent respiratory irritant, and cadmium (Cd) which leads to an increased risk of renal dysfunction.

Regulatory standards and guidelines relating to chronic exposure to these constituents and to $PM_{2.5}$ and PM_{10} mass appear in Table 3. This contains both concentration guidelines (listed as a concentration) and cancer slope factors (presented as incremental lifetime risk per unit of concentration). In the case of chromium, Cr(VI) is a potent respiratory carcinogen, while Cr(III) is relatively benign, hence the cancer slope factors and concentration guideline relate only to the former oxidation state.

Mean concentrations of the health-related particle size fraction masses and chemical species at the seven sampling sites and the overall mean of all sites appear in Table 4. There has been no attempt to elucidate seasonal patterns because of the limited number of samples collected in each season at each of the sites (see Table 2). It is clear from Table 4 that PM_{10} mass far exceeds $PM_{2.5}$ mass at all of the sites and that this is also the case for many of the elemental constituents. The split between fine particles ($PM_{2.5}$) and coarse particles ($PM_{2.5\text{-}10}$) is shown for all constituents in Figure S1 and for the elements of health concern, in Figure 2. It may be seen from Figure S1 that the typical crustal elements, Ca, Ti, Fe, Si and Al are 90% or more in the coarse fraction consistent with a large input of crustal dust to the samples as has been observed in earlier studies (Rushdi et al., 2013; Hussein et al., 2014). Those elements of health concern which show a larger contribution from the fine fraction indicative of anthropogenic sources are Ni, As, V and Pb, for which 40-60% lies in the fine fraction. The question of anthropogenic contribution to concentrations has been further examined through the calculation of crustal enrichment factors according to the method described by Pant et al. (2015) and Table 5 shows averaged crustal enrichment factors for the elements of concern calculated separately for the PM_{10} and $PM_{2.5}$ size fractions. If the enrichment factor of 5 is taken as the threshold for a significant enrichment above crustal ratios, then in the PM_{10} fraction, V shows a slight enrichment, with As appreciably enriched, and Pb and Cd showing very large enrichments. In the $PM_{2.5}$ fraction, Ni now shows significant enrichment with a larger enrichment of V and substantial enrichments of As, Pb and Cd.

Referring to Table 4, site 2 has the highest concentration of PM_{10}, but the second lowest of $PM_{2.5}$, suggesting a local source of coarse dust, possibly resuspension from the dense traffic at this site. Site 3 is located in a predominantly residential area, with substantial light industry locally, and a visit to the site revealed recent tyre and waste oil burning. There is a marked contrast to site 1, also in a residential area, but without intense vehicle traffic. Site 1 shows markedly lower concentrations of both PM_{10} and $PM_{2.5}$ mass than the other sites as well as appreciably lower concentrations of many of the trace elements, especially Pb and As. The site showing the highest concentrations of Ni and V is site 6 which is located most closely to the oil refinery and the port and shipyard, although with the prevailing winds coming predominantly from the NNW, it is unlikely to have a high exposure to emissions, especially from the oil refinery. Nonetheless, the elevated concentrations of these elements are indicative of a fuel oil combustion source influencing this site. Some evidence of this is also seen in elevated concentrations at site 5 which is also in the area of Jeddah closest to the oil refinery and port. Concentrations of Pb and As are elevated at all sites except for rather lower concentrations at site 1 which is in a residential area in the north of Jeddah and remote from major industrial activity. The highest concentrations of Pb in both the PM_{10} and $PM_{2.5}$ size fraction appear at sites 3 and 4, and since leaded additives are not used in gasoline in Saudi Arabia, must result from one or more local industrial sources.

Hussein et al. (2014) measured particle mass and number (D_p 0.25-32 µm) through the year 2012 at a sampling site on the campus of King Abdulaziz University in Jeddah. The diurnal variation of both PM_{10} and $PM_{2.5}$ on workdays showed a pattern typical of an influence of traffic emissions. The concentration of total particle number, but not of $PM_{2.5}$ or PM_{10} mass showed a marked elevation in a wind sector centred on 250°, leading Hussein et al. (2014) to infer that the industrial city in the south of Jeddah is the main source of particulate matter. They also report that the PM_{10} concentration shows a clear U-shaped dependence upon wind speed, which is characteristic of a contribution of wind-blown dust at high wind speeds, with dilution of emissions below the

threshold for dust resuspension (Harrison et al., 2001). The occurrence of dust storm events in Saudi Arabia is well documented (Alharbi et al., 2013; Kutiel and Furman, 2003).

Aburas et al. (2011) measured lead concentrations in the air of Jeddah in 2008-9, seven years after the phase-out of leaded gasoline in Saudi Arabia. The mean lead content of $PM_{2.5}$ was 73 ng m^{-3} (range 4-446 ng m^{-3}), with crustal enrichment factors (relative to K) at four sites of 761 to 15080. Concentrations were markedly higher at two sites in the south of Jeddah (King Abdulaziz University campus and Alfayhaa district) which was attributed to very high traffic density and the proximity to the industrial zone. Rushdi et al. (2013) report concentrations and enrichment factors (relative to Al) for Na, Mg, Al, Si, P, S, K, Ca, Mn, Fe Ni, Cu, Zn and Ba. The mean enrichment factor for Ni was 16.3. The only other analyte showing an elevated enrichment factor was S, suggesting fuel oil combustion as the source. Measurements from Taif in western Saudi Arabia made on samples collected in 2011-12 showed average concentrations of Mn of 34-52 ng m^{-3}; Ni of 3.5-4.0 ng m^{-3}; and Pb of 6.3-8.5 ng m^{-3} across traffic, industrial and residential sites. These are in all cases lower than those measured in our study and suggest that Jeddah is subject to greater levels of pollutant emissions.

The results for Cd are quite surprising. This shows substantial enrichment relative to crustal abundance in both the PM_{10} and $PM_{2.5}$ size fractions (Table 4) and the predominant presence in coarse particles (see Figure 1) seems to suggest either that the local soils have an abnormal geochemical enrichment of Cd or that there is a widespread source of coarse Cd arising from an industrial process. However, such a process would need to be widespread in order to cause such an extensive enrichment across all of the sites. It is notable that Alharbi et al. (2015) measured concentrations of Cd in PM_{10} in Riyadh of ca. 180 ng m^{-3} during dust storm periods which exceeded the non-dust storm concentrations by a factor of 2.3-fold. Such concentrations are broadly consistent with those in our measurements from Jeddah which strongly suggests an abnormal

enrichment of cadmium in surface soils in Saudi Arabia. Alghamdi et al. (2015a) also report very high enrichment factors for Cd in $PM_{2.5}$ sampled in western Saudi Arabia with average concentrations in this size fraction in excess of 10 ng m^{-3}, and appreciably higher on dust storm than non-dust storm days. Unfortunately, Cd concentrations were not reported by Kadi (2014) and Cd was not included in the factor analysis conducted by Khodeir et al. (2012), and consequently that work does not shed light on the likely sources of Cd.

3.1 Health Risk Assessment

Comparing the mean concentrations in Table 4 with the standards and reference concentrations in Table 3, it is clear that concentrations of V are not a matter of concern. However, concentrations of Cd, even those in the $PM_{2.5}$ fraction, exceed the recommendation of WHO (2000) and the EU Limit Value of 5 ng m^{-3}. The likely health consequences of such an exceedence are very hard to estimate particularly as there are no quantitative exposure-response functions relating airborne concentrations of Cd to the progression of kidney disease. There seems to be ample evidence for high concentrations of Cd in the atmosphere of Saudi Arabia and this warrants further study in relation to potential risks for human health. Concentrations of PM_{10} far exceed the WHO and EU requirements for this size fraction, and those for $PM_{2.5}$ exceed the WHO (2006) recommendation at all sites and exceed the EU recommendation of 25 µg m^{-3} at site 3, but not the other sampling sites.

In Table 6, health risks associated with the mean exposures have been calculated for those pollutants for which there are quantitative exposure-response functions available. In addition to the pollutants in Table 4, polycyclic aromatic hydrocarbons have been included using the cancer slope factor recommended by WHO (2000) and a mean concentration from three sites within Jeddah reported by Alghamdi et al. (2015b). The concentration used is for benzo(a)pyrene, which following the guidance of WHO (2000), is taken as a marker compound for the PAH mixture. As recommended by WHO the unit risk has been applied to the concentration of this compound, but

the risk estimation applies to the entire PAH mixture. Considering the chemical carcinogens, the highest risk appears to apply to Cr, but the value is an upper limit which assumes that all of the Cr exposure is in the form of Cr(VI) which is very improbable. This therefore represents an upper limit to the risk associated with Cr exposure. The risk is quite high and studies of the oxidation state of Cr in local airborne dusts would be well justified. Risks associated with exposure to As and PAH are of somewhat lesser magnitude but still exceed those calculated for Ni exposure.

In the case of $PM_{2.5,}$ a coefficient for all cause mortality has been taken from WHO (2006), and rather than the usual mortality burden calculation, an incremental risk has been estimated for the mean concentration exposure assuming a mean life expectancy of 74.5 years. This reveals a risk associated with $PM_{2.5}$ exposure which substantially exceeds the risks associated with the chemical carcinogens, which is logical as the $PM_{2.5}$ exposure includes exposure to the associated chemical carcinogens which present a subset of the mortality risks associated with $PM_{2.5}$ exposure. The work of Pope et al. (2002) and Lepeule et al. (2012) has shown a significant association between $PM_{2.5}$ exposure and lung cancer mortality in the ACS cohort, but the lung cancer risk is only a component of the overall all cause mortality risk. Harrison et al. (2004) considered whether exposure to the chemical carcinogens within $PM_{2.5}$ could explain the carcinogenicity demonstrated by Pope et al. (2002). Their conclusion was that it was quite plausible that the chemical carcinogens present could explain the observed carcinogenicity, which serves to confirm the view that the risk associated with exposure to the specific chemical carcinogens is only one part of the overall risk to health from $PM_{2.5}$ exposure which has been associated with a range of cardiopulmonary diseases. The mean concentration of Ni in the Jeddah samples falls significantly below the EU and EPAQS recommendation of 20 ng m^{-3} serving to confirm that the cancer risks associated with Ni exposure are not excessively high. However, the recommendations of the EU and EPAQS for As of 6 ng m^{-3} and 3 ng m^{-3} respectively are appreciably exceeded in Jeddah and there is a good case for further investigating the source of emission of this element and seeking to take action to mitigate the risk.

The USEPA IRIS reference concentration of 50 ng m^{-3} for manganese is a highly precautionary value designed to protect against impairment of neuro-behavioural function. It is exceeded by a factor of up to almost four-fold at the Jeddah sampling sites, but this factor is relatively small compared to the large in-built margin of safety and it seems unlikely that manganese presents an important risk to public health. The crustal enrichment factors in Table 5 show little evidence for anthropogenic emissions and hence the majority of exposure is from crustally-derived material in which the manganese may be significantly less bio-accessible than in the industrial exposures used as the basis for setting the reference concentration. In the case of lead, the exposure concentrations in PM_{10} at many of the sites exceed the USEPA (1996) and EPAQS (1998) recommendations. Air quality standards for lead are designed to protect the developing infant from neuro-developmental effects which have been shown to lead to a reduction in IQ. The fact that the concentrations in Jeddah exceed the regulatory guidelines from these jurisdictions is a matter of some concern. The lead concentrations reported by Aburas et al. (2011) are considerably exceeded by the recent measurements suggesting that there is a significant industrial source or sources in the south of Jeddah which is responsible for the substantial elevation of concentrations at sites in this part of the city. It is notable that lead concentrations at the most northerly site (site 1) are very much lower and within the acceptable range.

Very few studies have provided data on the effects of mixtures of pollutants, and it is not possible to comment on the possible interactions. It can reasonably be expected that the effects of the chemical carcinogens are additive, but as noted above this effect is included in the overall toxic effect of $PM_{2.5}$ as an exposure metric.

3.1.1 *Premature mortality due to $PM_{2.5}$ exposure*

The latest demographic information available for Jeddah relates to the year 2014. It lists populations for 60 areas of the city, which have been classified according to their similarity to the

areas represented by the sampling sites in Figure 1 and Table 1. The land use types are shown diagrammatically in Figure 3, and are listed in Table S8, both of which include population data. Table 7 shows a calculation of premature mortality according to the different district types for Jeddah, using both the 2014 population data for the districts listed in Table S8, as well as the total Jeddah population, including districts beyond the boundaries shown in Figure 3, making the questionable assumption that growth in the population is distributed in the same way as the 2014 population within Table S8.

In the approach used to estimate the burden of premature mortality, the burden is linearly related to both the exposure concentration and the population exposed. Thus for a similar concentration of $PM_{2.5}$, the overall number of premature deaths will be greater in a larger city, while in a city of similar size to Jeddah, the burden will be greater if the mean $PM_{2.5}$ concentration is higher. The World Health Organization has recently reviewed air quality data from around the world (WHO, 2016). While measured data for PM_{10} are plentiful, measurements of $PM_{2.5}$ are far less abundant, and in many cases have been crudely estimated from the PM_{10} data. Measured concentrations vary greatly between countries. Australia reports some of the lowest $PM_{2.5}$ concentrations, with annual means ranging from 5-10 µg m^{-3}. European concentrations are typically a little higher with annual means mostly in the range of 10-20 µg m^{-3} in western Europe and 20-40 µg m^{-3} in eastern Europe. Concentrations in China and India are typically higher, with most in the range of 30-100 µg m^{-3} and some exceeding 100 µg m^{-3}. The mean of 10 sites in Delhi in 2013 was 122 µg m^{-3}. The WHO data for $PM_{2.5}$ concentrations in Saudi Arabia are all estimated from PM_{10} measurements and range from 65 to 156 µg m^{-3} in 2014. These include a mean for Jeddah of 68 µg m^{-3}. This concentration well exceeds those reported for Jeddah in Table 4, but this may be due, at least in part, to the calculation method of WHO as opposed to direct measurement. This wide range of $PM_{2.5}$ data implies that many countries will have considerably lower mortality rates per million of population

due to $PM_{2.5}$ exposure than calculated in this work for Jeddah, while in others the rates will be higher.

A further factor to be considered is that almost all buildings and cars in Jeddah are air conditioned. Janssen et al. (2002) have shown that in the United States there appear to be lower rates of some diseases associated with PM_{10} exposure in areas with a high percentage of homes with air conditioning, as this can reduce exposures. This implies that the estimated premature mortality shown in Table 7 may be an over-estimate, but this effect has not to date been established for $PM_{2.5}$ exposure.

4. CONCLUSIONS

It is clear from the high concentrations of the crustally-related elements such as Ca, Fe and Si that crustal material in the form of wind-blown soil and dust makes up a substantial proportion of particulate matter in Jeddah. It is predominantly in the coarse ($PM_{2.5\text{-}10}$) size fraction, but a significant proportion lies also in the fine fraction. Of the health-related elements, only Ni, Pb, As, Cd and V show significant anthropogenic enrichment which is most marked in the fine particle fraction, except for Cd. Comparison with health-related guidelines suggest that the risks associated with exposure to Mn and V are very modest or wholly negligible, while the chemical carcinogens Ni, Cr and As present a smaller risk from chronic exposure than does exposure to $PM_{2.5}$. This is unsurprising as $PM_{2.5}$ exposure has been shown to be associated with a range of cardiopulmonary diseases, including lung cancer which is only a sub-component of the overall health impact of $PM_{2.5}$ exposure. Inclusion of data for PAH from an earlier study shows that these also do not represent a large risk in relation to the overall risk of $PM_{2.5}$ exposure. The largest potential risk from the chemical carcinogens relates to Cr, but the calculated risk is an upper limit which makes the pessimistic and probably unrealistic assumption that all of the Cr is present in the Cr(VI) oxidation state. Were this oxidation state to make up only a small proportion of the Cr content, then the

estimated risk would be greatly reduced. The enrichment factor for Cr is very small indicating that most Cr arises from the local soils and dusts and could not readily be abated. One unexpected finding is the very high enrichment factor for Cd. The concentrations measured in this study are not dissimilar from those reported in earlier studies from both Jeddah and Riyadh, and the fact that the enrichment is broadly similar in both the fine and coarse particle fractions suggests that there is an abnormal geochemical abundance of Cd in local surface soils which could be readily verified by chemical analysis. The calculated Cd exposures exceed health-based guidelines by a small factor in the $PM_{2.5}$ size fraction and a much larger factor in PM_{10}. However, if the Cd is associated with surface soils, it seems likely that its bio-accessibility is limited and hence the risk to health may be relatively modest.

The health risk associated with chronic exposure to $PM_{2.5}$ has been estimated in the form of premature mortality. This shows that total deaths influenced by chronic exposure to $PM_{2.5}$ exceed 1100 for the 2014 population of Jeddah, making this a very significant public health problem.

ACKNOWLEDGEMENT

This work was funded by King Abdulaziz University (KAU), Jeddah, under grant number 4/00/00/252. The authors thank NYU and KAU for technical and financial support.

REFERENCES

Aburas, H.M., Zytoon, M.A., Abdulsalam, M.I., 2011. Atmospheric lead in $PM_{2.5}$ after leaded gasoline phase-out in Jeddah City, Saudi Arabia. Clean – Soil, Air, Water 39:711-719.

Alghamdi, M.A., Almazroui, M., Shamy, M., Redal, M.A., Alkhalaf, A.K., Hussein, M.A., Khoder, M.I., 2015a. Characterization and elemental composition of atmospheric aerosol loads during springtime dust storm in western Saudi Arabia. Aerosol Air Qual. Res. 15:440-453.

Alghamdi, M.A., Alam, M.S., Yin, J., Start, C., Jang, E., Harrison, R4.M., Shamy, M., Khoder, M.I., Shabbaj, I.I., 2015b. Receptor modelling study of polycyclic aromatic hydrocarbons in Jeddah, Saudi Arabia, Sci. Tot. Environ. 506-507:401-408.

Alharbi, B.H., Maghrabi, A., Tapper, N., 2013. The March 2009 dust event in Saudi Arabia. Precursor and Supportive Environment. BAMS, 515-528, doi: http://dx.doi.org/10.1175/BAMS-D-11-00118.1

Alharbi, B., Shareef, M.M., Husain, T., 2015. Study of chemical characteristics of particulate matter concentrations in Riyadh, Saudi Arabia. Atmos. Pollut. Res:6, 88-98.

Al-Jeelani, H.A., 2009. Air quality assessment at Al-Taneem area in the Holy Makkah City, Saudi Arabia. Environ. Monit. Assess. 156:211-222.

EPAQS, 2009. Metals and Metalloids, Expert Panel on Air Quality Standards, Department for Environment, Food and Rural Affairs, London.

EPAQS, 1998. Lead, Expert Panel on Air Quality Standards, Stationery Office, London.

EU, 2016. Air Quality Standards, European Commission, http://ec.europa.eu/environment/air/quality/standards.htm

Harrison, R.M., Yin, J., Mark, D., Stedman, J., Appleby, R.S., Booker, J., Moorcroft, S., 2001. Studies of the coarse particle (2.5-10 µm) component in UK Urban Atmospheres. Atmos. Environ. 35:3667-3679.

Harrison, R.M., Smith, D.J.T., Kibble, A.J., 2004. What is responsible for the carcinogenicity of $PM_{2.5}$? Occup. and Environ. Med:61, 799-805.

Harrison, R.M., M.S. Alam, Dang, J., Basahi, J., Alghamdi, M.A., Ismail, I.M., Khoder, M., Hassan I.A., 2016. Influence of petrochemical installations upon PAH Concentrations at Sites in western Saudi Arabia, Atmos. Pollut. Res. 7:954-960.

Hussein, T., Alghamdi, M.A., Khoder, M., AbdelMaksoud, A.S., Al-Jeelani, H., Goknil, M.K., Shabbaj, I.I., Almehamadi, F.M., Hyvarinen, A., Lihavainen, H., Hameri, K., 2014. Particulate matter and number concentrations of particles larger than 0.25 µm in the urban atmosphere of Jeddah, Saudi Arabia. Aerosol and Air Qual. Res. 14:1383-1391.

Index Mundi, http://www.indexmundi.com/saudi_arabia/death_rate.html

Janssen, N.A.H., Schwartz, J., Zanobetti, A., Suh, H.H., 2002. Air conditioning and source-specific particles as modifiers of the effect of PM_{10} on hospital admissions for heart and lung diseases. Environ. Health Perspect. 110:43-49.

Kadi, M.W., 2009. Soil pollution hazardous to environment: Case study on the chemical composition and correlation to automobile traffic of the roadside soil of Jeddah city, Saudi Arabia. J. Hazard. Mater. 168:1280-1283.

Kadi, M.W., 2014. Elemental spatiotemporal variations of total suspended particles in Jeddah City, Sci. World J. 2014, Article ID 325492, doi.org/10.1155/2014/325492.

Khalil, M.A.K., Butenhoff, C.L., Porter, W.C., Almazroui, M., Alkhalaf, A., Al-Sahafi, M.S., 2016. Air quality in Yanbu, Saudi Arabia. JAWMA 66:341-355.

Khodeir, M., Shamy, M., Alghamdi, M., Zhong, M., Sun, H., Costa, M., Chen, L.-C., Maciejczyk, P., 2012. Source apportionment and elemental composition of $PM_{2.5}$ and PM_{10} in Jeddah City, Saudi Arabia. Atmos. Pollut. Res. 3:331-340.

Kutiel, H., Furman, H., 2003. Dust storms in the Middle East: Sources of origin and their temporal characteristics. Indoor & Built Environ. 12:419-426.

Lepeule, J., Laden, F., Dockery, D., Schwartz, J., 2012. Chronic exposure to fine particles and mortality: An extended follow-up of the Harvard Six Cities Study from 1974 to 2009. Environ. Health Perspect. 120:965-970.

Lihavainen, H., Alghamdi, M.A., Hyvarinen, A.-P., Hussein, T., Aaltonen, V., Abdelmaksoud, A.S., Al-Jeelani, H., Almazroui, M., Almehmadi, F.M., Al Zawad, F.M., Hakala, J., Khoder, M., Neitola, K., Petaja, T., Shabbaj, I.I., Hameri, K., 2016. Aerosols physical properties at Hada Al Sham, eastern Saudi Arabia. Atmos. Environ. 135:109-117.

Pant, P., Baker, S.J., Shukla, A., Maikawa, C., Godri Pollitt, K.J., R.M. Harrison, 2015. The PM10 fraction of road dust in the UK and India: Characterization, source profiles and oxidative potential. Sci. Tot. Environ. 530-531:445-452.

Pope, C.A. III, Burnett , R.T., Thun, M.J., Calle, E.E., Krewski, D., Ito, K., Thurston, G.D., 2002. Lung cancer, cardiopulmonary mortality, and long-term exposure to fine particulate air pollution. JAMA 287, 287:1132-1141.

Porter, W.C., Khalil, A.K., Butenhoff, C.L., Almazroui, M., Al-Khalaf, A.K., Al-Sahafi, M.S., 2014. Annual and weekly patterns of ozone and particulate matter in Jeddah, Saudi Arabia. JAWMA 64:817-826.

Rushdi, A.I., Al-Mustlaq, K.F., Al-Otaibi, M., El-Mubarak, A.H., Simoneit, B.R.T., 2013. Air quality and elemental enrichment factors of aerosol particulate matter in Riyadh City, Saudi Arabia. Arab J. Geosci. 6:585-599.

Shaltout, A.A., Boman J., Al-Malawi, D.-A.R., Shehadeh, S.F., 2013. Elemental composition of $PM_{2.5}$ particle sampled in industrial and residental areas of Taif, Saudi Arabia.Aerosol Air Qual. Res., 13:1356-1364.

Shaltout, A.A., Boman, J., Shehadeh Z.F., Al-Malawi, D.-A.R., Hemeda, O.M., Morsy, M.M., 2015. Spectroscopic investigation of $PM_{2.5}$ collected at industrial, residential and traffic sites in Taif, Saudi Arabia. J. Aerosol Sci. 79:97-108.

Simpson, I.J., Aburizaiza, O.S., Siddique, A., Barletta, B., Blake, N.J., Gartner, A., Khwaja, H., Meinardi, S., Zeb, J., Blake, D.R., 2014. Air quality in Mecca and surrounding holy places in Saudi Arabia during Hajj: Initial survey. Environ. Sci. Technol. 48:8529-8537.

USEPA, 1996. Manganese: CASRN 7439-96-5, Chemical Assessment Summary, Integrated Risk Assessment System (IRIS), US Environmental Protection Agency.

WHO, 2000. Air Quality Guidelines for Europe, Second Edition, World Health Organization, Copenhagen.

WHO, 2006. Air Quality Guidelines Global Update 2005, World Health Organization, Copenhagen.

WHO, 2016.Global Urban Ambient Air Pollution Database (update 2016, http://www.who.int/mediacentre/news/releases/2016/air-pollution-estimates/en/ [last accessed 23/02/2017].

World Health Rankings, http://www.worldlifeexpectancy.com/country-health-profile/saudi-arabia [last accessed 23/02/2017].

TABLE CAPTIONS

Table 1: Characteristics of sampling sites.

Table 2: Seasonal distribution of sample numbers at the seven sites.

Table 3: Air quality standards, reference concentrations and cancer slope factors for chronic respiratory exposure to relevant aerosol components.

Table 4: Mean concentrations of health-relevant size fractions and chemical species at the seven sites.

Table 5: Average crustal enrichment factor for the elements of concern.

Table 6: Health risk associated with the mean exposures.

Table 7: Estimated premature mortality due to $PM_{2.5}$ exposure in Jeddah.

FIGURE CAPTIONS

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

Figure 2: Average coarse and fine percentages of the health-relevant elements and particulate matter (PM) mass.

Figure 3: Map of Jeddah, showing the districts according to land use type (colour), population (circles) and the air sampling sites (stars).

Table 1: Characteristics of sampling sites.

	District	Population	Type	Characteristics
1	Al-Muhammadiyah	28315	Residential	Typical residential with no intense traffic
2	Al-Rehab	43400	Residential	Influenced by heavy traffic from the nearby highway and the crowded Tahleya street
3	Al-Rughama	38437	Suburban	Heavy traffic, open burning of batteries, electric wires, and tyres. Some significant marble workshops
4	University	141277	Urban	Dense traffic
5	Al-Nuzlah/Al Yamaneyyah	53602	Urban	Dense traffic, some car repair workshops
6	Pitrumin	41774	Urban	Refinery emissions
7	Al-Alfiyyah	43037	Residential	Refinery emissions (less affected than Pitrumin)

Table 2: Seasonal distribution of sample numbers at the seven sites.

Site/Season	**1**	**2**	**3**	**4**	**5**	**6**	**7**	**All sites**
Spring	7	8	7	6	7	7	50	92
Summer	7	2	5	7	6	7	37	71
Autumn	6	7	7	7	7	7	37	78
Winter	6	9	5	7	7	5	47	86
Totals	**26**	**26**	**24**	**27**	**27**	**26**	**171**	**327**

Table 3: Air quality standards (annual mean), reference concentrations and cancer slope factors for chronic respiratory exposure to relevant aerosol components.

Constituent	WHO (2000)	WHO (2006)	USEPA (1996)	EU (2016)	EPAQS (1998; 2009)
$PM_{2.5}$		10 µg m^{-3}	12 µg m^{-3}	25 µg m^{-3}	
PM_{10}		20 µg m^{-3}		40 µg m^{-3}	
Cr(VI)	4 x 10^{-5}/ng m^{-3}		1.2 x 10^{-5}/ng m^{-3}		0.2 ng m^{-3}
Mn			50 ng m^{-3}		
Ni	3.8 x 10^{-7}/ng m^{-3}			20 ng m^{-3}	20 ng m^{-3}
Pb			150 ng m^{-3}	500 ng m^{-3}	250 ng m^{-3}
As	1.5 x 10^{-6}/ng m^{-3}		4.3 x 10^{-6}/ng m^{-3}	6 ng m^{-3}	3 ng m^{-3}
Cd	5 ng m^{-3}			5 ng m^{-3}	
V	1000 ng m^{-3}				

Table 4: Mean concentrations of health-relevant size fractions and chemical species at the seven sites.

Site/Analyte	1	2	3	4	5	6	7	All sites
PM_{10} fraction								
PM_{10} mass (µg m^{-3})	69.8	143	120	112	110	104	94.0	108
Cr (ng m^{-3})	4.9	12.1	14.2	10.3	10.5	7.4	8.1	9.6
Mn (ng m^{-3})	56.4	153	137	105	85.6	100	95.7	105
Ni (ng m^{-3})	6.6	12.1	12.6	12.6	12.4	15.0	11.6	11.7
Pb (ng m^{-3})	38.6	595	695	695	84.6	379	440	450
As (ng m^{-3})	3.3	26.5	19.7	19.7	5.8	11.4	15.2	15.2
Cd (ng m^{-3})	80.2	194	145	145	100	231	98.4	140
V (ng m^{-3})	20.6	32.6	27.8	27.8	34.2	43.5	26.8	30.7
$PM_{2.5}$ fraction								
$PM_{2.5}$ mass (µg m^{-3})	14.2	17.5	21.6	21.6	23.4	24.2	21.8	20.7
Cr (ng m^{-3})	0.5	1.1	1.7	1.7	1.6	0.9	1.2	1.2
Mn (ng m^{-3})	5.3	9.7	12.2	12.2	9.8	9.5	8.4	9.4
Ni (ng m^{-3})	2.6	2.8	3.6	3.6	4.6	7.2	3.6	4.1
Pb (ng m^{-3})	31.3	256	443	443	59.8	137	209	248
As (ng m^{-3})	0.8	15.6	10.3	10.3	1.8	5.3	6.9	8.4
Cd (ng m^{-3})	9.5	13.5	9.4	9.4	9.8	11.9	6.7	11.0
V(ng m^{-3})	9.1	8.8	9.5	9.5	16.3	25.8	11.7	13.4

Table 5: Average crustal enrichment factor for the elements of concern* .

Element/ Size fraction	**Cr**	**Mn**	**Ni**	**Pb**	**As**	**Cd**	**V**
PM_{10}	1.4	3.0	3.4	810	88	20,200	5.9
$PM_{2.5}$	1.8	2.7	12.0	4533	491	16,100	26

* Relative to Al

Table 6: Health risk associated with the mean exposures.

Pollutant	**End Point**	**Coefficient**	**Mean Concentration**	**Lifetime Risk**
$PM_{2.5}$[a]	Mortality (all cause)	4%/10 µg m^{-3}	22.5 µg m^{-3}	1.2×10^{-3}
Cr[b]	Cancer	4×10^{-5}/ng m^{-3}	9.6 ng m^{-3}	3.8×10^{-4}
Ni	Cancer	3.8×10^{-7}/ng m^{-3}	11.7 ng m^{-3}	4.4×10^{-6}
As	Cancer	1.5×10^{-6}/ng m^{-3}	15.2 ng m^{-3}	2.3×10^{-5}
PAH (B(a)P)[c]	Cancer	8.7×10^{-5}/ng m^{-3}	0.23 ng m^{-3}	2.0×10^{-5}

Notes:

(a) Calculation based upon a life expectancy of 74.5 years (World Health Rankings, 2016)

(b) Calculation assumes all Cr is present as Cr(VI) which is extremely unlikely, and hence this is an upper limit to risk

(c) B(a)P concentrations measured in Jeddah by Alghamdi et al. (2015); mean of particulate concentration at three sites

Table 7: Estimated premature mortality due to $PM_{2.5}$ exposure in Jeddah.

District Type	$PM_{2.5}$ (µg m^{-3})	Total Population (2014) (thousand)[a]	Premature Deaths[b,c]	Total Premature Deaths[b,d]
Residential (1)	14.2	274	53	77
Residential (2)	17.5	392	94	136
Residential (3)	21.8	211	63	91
Suburban	21.6	173	51	74
Urban (1)	21.6	1153	341	494
Urban (2)	23.4	307	98	142
Urban (3)	24.2	234	77	112
TOTAL		2744	777	1126

Notes:

[a] Population data from Jeddah Council (personal communication).

[b] Based upon a crude death rate of 3.42 per 1000 in 2015 (Index Mundi, 2016).

[c] Based on population for 2014 from districts listed in Table S8.

[d] Extrapolated to total population of Jeddah in 2014.

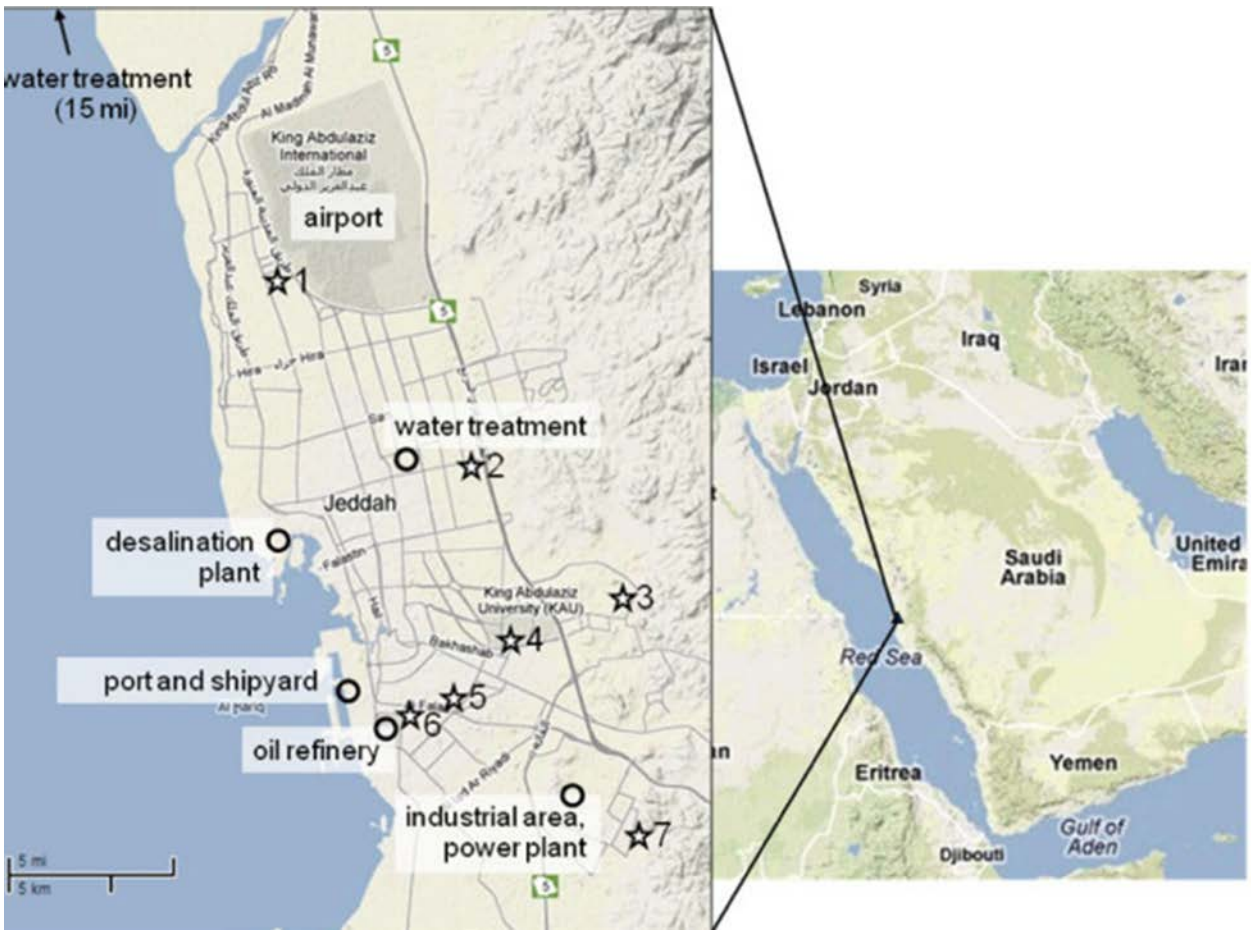

Figure 1: Location of sampling sites (stars) and major industrial sources (circles) in Jeddah, Saudi Arabia.

Figure 2: Average coarse and fine percentages of the health-relevant elements and particulate matter (PM) mass.

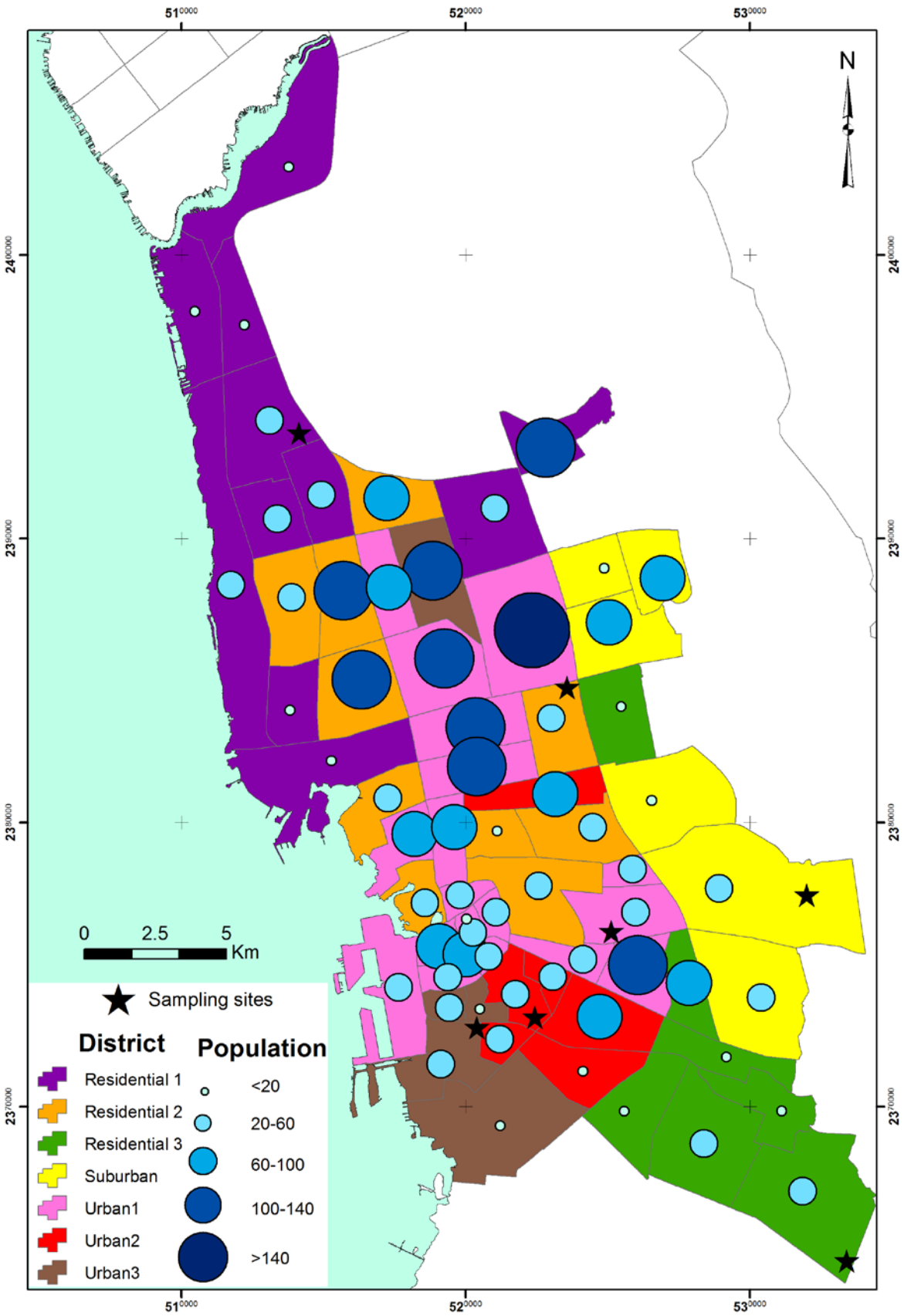

Figure 3: Map of Jeddah, showing the districts according to land use type (colour), population (circles) and the air sampling sites (stars).