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Original Research

Concentration Level and Health Risk Assessment of Heavy Metals in $PM_{2.5}$ in Ambient Air of Makkah City, Saudi Arabia

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Abstract

Exposure to air pollution in general and Particulate Matter (PM) and its constituents, in particular, can be extremely harmful to human health. Given the importance of PM and its constituents concerning human health, this study aimed to evaluate the levels of $PM_{2.5}$ and some HMs in the atmosphere of Makkah City, Saudi Arabia, and assess the health risks associated with exposure to HMs. The 24-h mean concentration of $PM_{2.5}$ in Makkah city was 38.0±13.5 µg/m^3 which exceeded the WHO air quality guidelines and national ambient air quality standard of Saudi Arabia. The average concentrations of HMs were Mn (54.3±13.3 ng/m^3)>Pb (40.3±9.2 ng/m^3)>As (27.8±4.1 ng/m^3)>Ni (20.5±12.9 ng/m^3)>Cr (18.2±6.2 ng/m^3)>Cd (13.0±3.1 ng/m^3). The non-carcinogenic risks (non-CRs) and carcinogenic risks (CRs) associated with exposure to measured HMs were analyzed using hazard quotient (HQ) and incremental lifetime cancer risk (ILCR), respectively. Arsenic was found to be the major contributor to health risk and oral ingestion was found to be the most detrimental pathway of exposure to HMs. For future research, it is recommended to evaluate the HMs and their health risks in various environmental media in different Saudi cities.

Keywords: $PM_{2.5}$, heavy metals, health risk assessment, Makkah, Saudi Arabia

Introduction

Environmental pollution is a serious global concern that is responsible for premature death and disability worldwide [1]. The Lancet Commission on pollution and health has attributed 9.0 million premature deaths (16% of all deaths globally) to diseases caused by environmental pollution [2].

Amongst environmental pollutants, air pollution is the most hazardous to human health. The World Health Organization (WHO) estimates that 7.0 million premature annual deaths worldwide are caused by air pollution. Among air pollutants, Particulate Matter (PM) poses the greatest threat to human health compared to other air pollutants [3]. PM has been classified as a Group 1 carcinogen by the International Agency for

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Research on Cancer (IARC) [4]. The toxicity of PM depends on its size, chemical characteristics, and surface properties [5]. PM with an aerodynamic diameter ≤2.5 µm ($PM_{2.5}$) has received substantial attention recently. Its small size is what makes it worthy of researchers' attention as it could have an environmental impact (i.e. it can stay suspended for long periods and travel great distances) and human health impact (i.e. it can get to the deep parts of the lung and might end up in the bloodstream) [6].

According to the WHO, exposure to $PM_{2.5}$ is more hazardous to human health than exposure to coarse PM [7]. Ambient $PM_{2.5}$ was ranked as the fifth mortality risk factor, being responsible for 4.2 million deaths and 103.1 million disability-adjusted life-years (DALYs) [8]. Considering its harmful effects, the maximum concentrations of $PM_{2.5}$ in the ambient atmosphere have been established by different governmental and non-governmental organizations. The United States Environmental Protection Agency (USEPA) has set the daily average at 35.0 µg/m^3 and the annual average at 12.0 µg/m^3 as maximum limits for $PM_{2.5}$ concentration in the air. The concentration of $PM_{2.5}$ should not exceed 25.0 µg/m^3 (daily average) and 10.0 µg/m^3 (annual average) as per WHO air quality guidelines. In Saudi Arabia, the ambient air quality standards defined the daily average of 35.0 µg/m^3 and the annual average of 15.0 µg/m^3 as the maximum concentrations of $PM_{2.5}$ in the atmosphere.

PM represents a mixture of organic and elemental carbon, sulfate, ammonium, nitrate, trace metals, and water [9]. HMs may be found at trace concentrations in PM, but they warrant a great consideration as they can pose serious threats to human health [10]. Exposure to HMs has been linked to several health problems such as liver and kidney damage, nervous system disorder, respiratory diseases, muscular dystrophy, cardiovascular diseases, diabetes, anemia, and cancer [11-13]. The IARC has classified arsenic (As), chromium (Cr), cadmium (Cd), and nickel (Ni) as group 1 carcinogens, whereas lead (Pb) and cobalt (Co) were classified as group 2A carcinogens [14]. Because of their health effects, the WHO has recommended the target values for mean concentrations of As, Cd, Ni, and Pb in the ambient atmosphere as 6.0, 5.0, 25, and 500 ng/m^3 respectively. The main sources of HMs atmospheric pollution include combustion of fossil fuels, industrial activities, waste incineration, metal manufacturing, cement production, traffic, and resuspension of dust [15]. Irrespective of their sources, heavy metals (HMs) are found to be bounded to PM which facilitates their transportation in the atmosphere [16, 17]. The concentrations and size distributions of PM and HMs in the atmosphere are governed by the nature of emission, air mass trajectory, rate of dry and wet deposition, exchange of air between the free troposphere and boundary layers, and chemical transformation [18].

Makkah is the Holiest city of the Islamic world where millions of Muslims visit the city throughout the year for Pilgrimage (Hajj and Umrah). Hajj is an annual pilgrimage whereas Umrah is performed daily over the year. In 2019 (before the emergence of COVID-19), the number of people who performed Hajj was around 2.5 million whereas the number of people who performed Umrah was more than 19 million [19]. This is accompanied by an increase in the number of vehicles in the city for transportation. Vehicular emissions have been considered as the major source of air pollution in the city [20, 21]. Recently, the Holly city witnessed considerable construction projects including the most significant expansion in the Grand Mosque. Being in a desert region, Makkah suffers from frequent dust storms. Dust storms, construction projects, and dust resuspension from automobiles contribute significantly to PM air pollution in the city [20]. Moreover, the air quality in Makkah can be influenced by long-range transports of air pollutants from surrounding industrial cities [22].

Given the hazardous nature of $PM_{2.5}$ and its constituents, the main objective of this study is to evaluate the levels of $PM_{2.5}$ and HMs contamination in the ambient air of Makkah city. Furthermore, the carcinogenic and non-carcinogenic health risks associated with exposure to HMs will be assessed.

Experimental

Study Area

Makkah (21.40 N, 39.820 E) is the capital city of Makkah Al-Mukarramah Region in Saudi Arabia. The Holy city is located about 80 km inland from the Red Sea and lies on the Western slopes of the Sarwat Mountains at an elevation of 277 m above sea level [22]. According to the General Authority of Statistics in Saudi Arabia, the city's population was 1.58 million in 2010. The city's climate is characterized as a hot and dry, desert climate. The city has very low annual precipitation (10-33 mm), the temperature in summer can exceed 40ºC while in winter can be below 18ºC, the humidity ratio is in the range of 45 to 53 % [23]. The wind is predominately West and Northwestern with moderate speeds throughout the year. Sandstorms occur during the spring and end of the fall which is the major source of air pollution by PM in the region [24].

Sampling Procedures

Samples of $PM_{2.5}$ were collected on pre-weighed 46.2 mm PTFE filters (Whatman, UK) for 24 hours using a combo dust sampler (GTI-CDS-401, Greintech) at a flow rate of 16.7 L/m. Sampling was performed at four sites in Makkah city, Al-Haram (site 1), Al-Azizyah (site 2), Batha Quraish (site 3), and Rea Azakher (site 4) (Fig. 1). Site 1 represents the central area of Makkah city which includes the Grand Mosque where pilgrims perform their Islamic rituals. Site 2

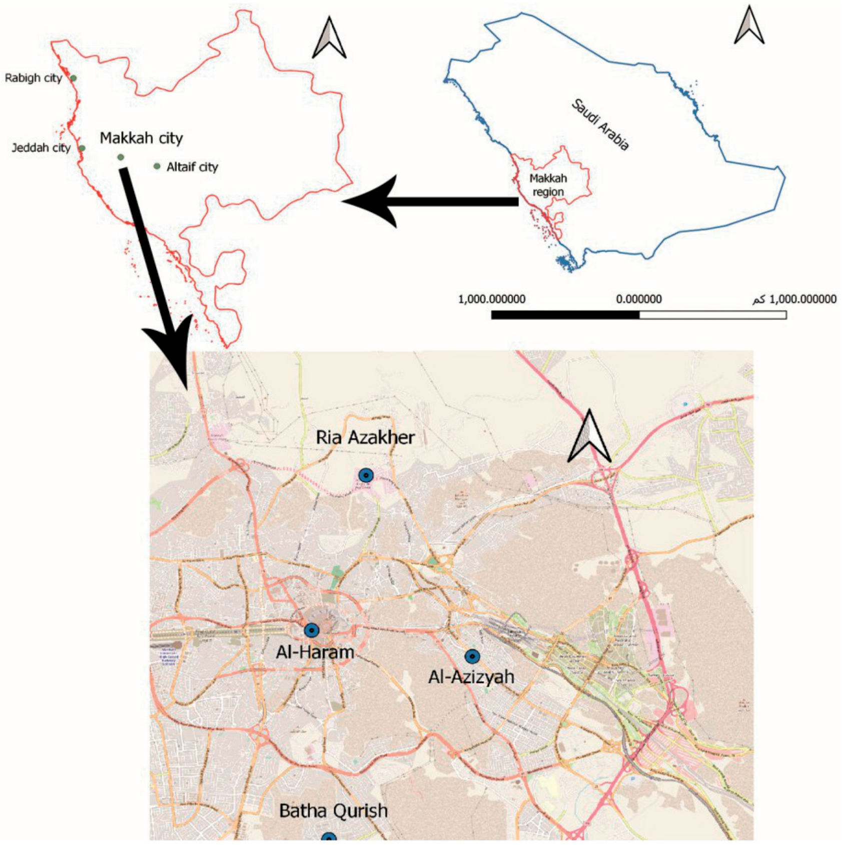

Fig. 1. Location of sampling sites in Makkah city, Saudi Arabia.

is a residential/commercial area located to the south of the Grand Mosque. Site 3 is a residential area that is influenced by traffic from the 3rd and 4th ring roads in the city. Site 4 contains small metallurgy and auto repair workshops. The samples were collected from the 14th of April to the 2nd of July 2021 (this period covered the Umrah season during the Holy month of Ramadan 2021).

After sampling, the filters were placed in 47 mm Petri slide dishes and transferred to the laboratory where the samples were preserved in a refrigerator at 6ºC until the chemical analysis. Field blank filters were included in each sampling set and handled in a resemble manner to the collected samples. The filters were weighed before and after sampling using an electronic microbalance (Mettler Toledo, XPE26 model), and the total mass of $PM_{2.5}$ (µg) was determined by calculating the difference in filter mass before and after sampling.

Extraction and Analysis of HMs

A temperature-controlled Milestone ETHOS easy advanced microwave digestion system was used to digest the filters for metal analysis. Before digestion, the filters were cut into small pieces and then digested with 10 ml of nitric acid and hydrogen peroxide mixture (9 ml of 65% HNO_3 and 1 ml of 30% H_2O_2). The samples were digested using the two-stage microwave digestion program: Stage-1, heating to 230ºC over 20 min; Stage-2, incubation at 230ºC for 15 min. After cooling, the samples were filtered and diluted with Milli Q water. The blanks were also prepared in triplicate by adopting the same procedure. The quantification of metal content was performed in triplicate by using the inductively coupled plasma spectrometer (ICPE-9000, Shimadzu).

Before analyzing the samples, the instrument was calibrated with a standard blank and the multi-element calibration standard. The analysis was started after getting the best linear regression correlation coefficient ($r^2 \geq 0.99$) from the calibration plot. All the analytical reference multi-element standards were purchased from AccuStandard, USA. The field blank filters were also analyzed in parallel with sample filters using the same procedures.

Enrichment Factor (EF)

EF provides information on HMs contamination level from anthropogenic sources by comparing the concentration of HMs in PM samples with reference elements in Earth's crust as shown in Eq. (1) [25].

$$EF = [(C_i / C_{ref})\ \text{sample}] / [(C_i / C_{ref})\ \text{earth crust}] \quad (1)$$

Where (C_i) is the concentrations of investigated HMs in $PM_{2.5}$ and Earth crust and (C_{ref}) is the concentration of reference elements in $PM_{2.5}$ and Earth crust. In the present study, Mn was used as a reference element and HMs concentrations in Earth's crust were obtained from Taylor [26]. Mn has been used as a reference element in previous studies [27-31]. According to previous studies, the EF value of ≤10 suggests the source of the metal from the natural source of Earth's crust whereas EF value of >10 indicates significant enrichments from anthropogenic sources [32].

Health Risk Assessment

Health risk assessment is the process of estimating the nature and the probability of occurrence of adverse health effects in humans due to exposure to environmental contaminants. In the present study, the non-carcinogenic health risk and lifetime cancer risk were evaluated based on inhalation, ingestion, and dermal routes of exposure to HMs. Human exposure is measured in terms of average daily dose (ADD) via ingestion (mg/kg-day) (Eq. 2), exposure concentration via inhalation (EC) $\mu g/m^3$ (Eq. 3) and dermal absorption dose (DAD) (mg/kg-day) (Eq. 4). [16, 33]

$$ADD = (C_i \times IngR \times EF \times ED \times 10^{-6}) / (BW \times AT) \quad (2)$$

$$EC = (C_i \times ET \times EF \times ED) / (AT_n) \quad (3)$$

$$DAD = (C_i \times SA \times AF \times ABF \times EF \times ED \times 10^{-6}) / (BW \times AT) \quad (4)$$

The definitions and the constant factors included in Eqs. (2, 3), and (4) are given in Table 1.

Non-Carcinogenic Health Risk

Non-carcinogenic health risk assessment of $PM_{2.5}$-bound HMs was estimated using hazard quotient (HQ) which is calculated based on Eqs (5-7) [16, 33].

$$HQ_{ing} = ADD / (RfD) \quad (5)$$

$$HQ_{inh} = EC / (RfC \times 1000) \quad (6)$$

$$HQ_{der} = DAD / (RfD) \quad (7)$$

Where HQ_{ing}, HQ_{inh} , and HQ_{der} are the hazard quotient via ingestion, inhalation, and dermal contact respectively. RfD is the reference dose (mg/kg-day) and RfC is the reference concentration of the HM (mg/m^3). The values of RfD and RfC for investigated HMs were obtained from Zhang et al. [33]. HQ values ≤1 indicate no significant or acceptable risk, while HQ values >1 indicate the potential for adverse health effects [36]. Hazard index (HI) is used for the estimation of health risks associated with exposure to multiple metals. Hazard index (HI) is the summation of hazard quotients (HQ_k) of individual metal "k" which can be calculated using the following equation [16],

$$HI = \sum HQ_k \quad (8)$$

Table 1. Definitions and values of parameters used for carcinogenic (CRs) and non- carcinogenic risk (non-CRs) assessment.

Parameter	Definition	Unit	Values		References
			Children	Adults	
C_i	The mean concentrations of HMs	($\mu g/m^3$) for EC, (mg/kg) for ADD and DAD	From the present study	From the present study	This study
IngR	Ingestion rate	mg/day	200	100	[34]
EF	Exposure frequency	Days/year	180	180	[33]
ED	Exposure duration	Years	6	24	[35]
ET	Exposure time	Hours/day	24	24	[35]
AT	Average lifetime	Days	ED×365 (non-carcinogens)	ED×365 (non-carcinogens)	[35]
			70×365 (carcinogens)	70×365 (carcinogens)	[35]
AT_n	Average lifetime	Hours	ED×365×24 (non-carcinogens)	ED×365×24 (non-carcinogens)	[35]
			70×365×24 (carcinogens)	70×365×24 (carcinogens)	[35]
BW	Body weight	kg	15	70	[35]
SA	Skin surface area	cm^2	2800	5700	[35]
AF	Adherence factor	mg/cm^2	0.2	0.07	[35]
ABF	Absorption factor	-	0.1 (Pb), 0.03 (As), 0.001 (Cd), 0.01 (others)		[33]

Lifetime Cancer Risk

The probability of developing cancer because of human exposure to these carcinogens over the lifetime (ILCR) can be estimated using Eqs (9, 10), and (11) for ingestion, inhalation, and dermal contact respectively [37].

$$ILCR_{ing} = ADD \times SF \ (9), \ ILCR_{inh} = EC \times IUR \ (10)$$

$$ILCR_{der} = DAD \times SF \qquad (11)$$

where $ILCR_{ing}$, $ILCR_{inh}$, $ILCR_{der}$ are incremental lifetime cancer risks via ingestion, inhalation, and dermal contact respectively. SF is the slope factor $(mg/kg\text{-}day)^{-1}$ and IUR is the inhalation unit risk $[(\mu g/m^3)]^{-1}$. The values of SF and IUR for carcinogenic metals were taken from the California Office of Environmental Health Hazard Assessment [38]. The ILCR can be classified as very low ($ILCR \leq 1 \times 10^{-6}$), low ($10^{-6} \leq ILCR < 10^{-4}$), moderate ($10^{-4} \leq ILCR < 10^{-3}$), high ($10^{-3} \leq ILCR < 10^{-1}$) and very high ($ILCR \geq 10^{-1}$) [33]. The cumulative ILCR for different carcinogenic metals (i) is given by Eq. (12). [39]

$$\text{Cumulative ILCR} = \sum ILCR_i \qquad (12)$$

The cumulative ILCR for different carcinogenic metals should be maintained below 10^{-4} [40].

Results and Discussion

Mass Concentration of $PM_{2.5}$

The 24-h average concentrations of $PM_{2.5}$ in Makkah city was 38.0±13.5 μg/m^3 during the whole period of study (Table 2). This concentration exceeded the WHO air quality guidelines of 25 μg/m^3 and the national ambient air quality standard of Saudi Arabia (35.0 μg/m^3). The mean concentrations of $PM_{2.5}$ were 26.7±12.8 μg/m^3 (range 13.2-62.3 μg/m^3), 25.9±8.2 μg/m^3 (range 10.6-41.8 μg/m^3), 49.5±16.3 μg/m^3 (range 25.3-74.1 μg/m^3) and 49.9±30.0 μg/m^3 (range 20.3-137.7 μg/m^3) at sites 1, 2, 3 and 4 respectively. The average mass of 24-concentrations of $PM_{2.5}$ at individual sites exceeded the WHO air quality guidelines (25 μg/m^3). It was observed that 70% of sampling days have exceeded the permissible levels of WHO.

The air quality index (AQI) of USEPA has been used to assess the air quality for $PM_{2.5}$. The breakpoints for this index are as follows: good (0.0-12.0 μg/m^3), moderate (12.1-35.4 μg/m^3), unhealthy for sensitive groups (USG) (35.5-55.4 μg/m^3), unhealthy (55.5-150.4 μg/m^3), very unhealthy (150.5-250.4 μg/m^3) and hazardous (250.5-500 μg/m^3). The results showed that the air quality of Makkah city ranged from moderate to unhealthy for $PM_{2.5}$. Most of the samples (60%) showed moderate air quality which indicates acceptable air quality in the city throughout period of study. Unhealthy air quality was observed in 18.3% of samples whereas 21.7% of samples showed unhealthy air quality for sensitive groups.

Previous studies on the air quality of Makkah city showed elevated concentrations of $PM_{2.5}$ at Al-Haram (site 1) during the holy month of Ramadan which was attributed to a significant increase in the number of vehicles used for transporting millions of Muslims who visited The Holy city for prayer and Umrah [20]. The concentration of $PM_{2.5}$ was higher than 240 μg/m^3 during the Holy month of Ramadan in 2014 as noticed by Nayebare et al. [20]. A similar trend was observed by Shaltout et al. [41] who reported a remarkable increase in the concentration of $PM_{2.5}$ during the Holy month of Ramadan in the year of 2013. In the present study, the mean 24-concentration of $PM_{2.5}$ during the Holy month of Ramadan was 31.9 μg/m^3 at Al-Haram (site 1), which is below the limit specified by ambient air quality standards of Saudi Arabia and U.S.EPA (35 μg/m^3). The value of AQI for $PM_{2.5}$ showed moderate air quality at Al-Haram (site 1) during Ramadan 2021 whereas poor air quality was observed during Ramadan of 2013 and 2014 [20, 41].

It is noteworthy that the prayer and Umrah at the Grand Mosque during the Holy month of Ramadan 2021 were only permitted for people immunized against COVID-19. These restrictions have been adopted by Saudi authorities to control the spread of COVID-19. The decrease in the concentration of $PM_{2.5}$ during Ramadan of 2021 could be due to the limited number of Muslims who were allowed to perform prayer and

Table 2. The mean concentrations of $PM_{2.5}$ (μg/m^3) and HMs (ng/m^3) (mean ± std).

Location	$PM_{2.5}$	As	Cd	Cr	Pb	Mn	Ni
Site 1	26.7±12.8	32.4±13.4	10.3±4.8	12.9±3.2	33.4±10.4	42.2±10.5	39.6±46.3
Site 2	25.9±8.2	28.6±12.2	10.5±3.9	12.9±5.6	33.0±19.0	43.4±22.5	13.4±5.5
Site 3	49.5±16.3	23.4±4.2	16.8±4.1	22.1±2.9	52.5±17.6	67.3±3.8	12.2±3.1
Site 4	49.9±30.0	26.2±17.5	14.2±4.7	25.0±11.3	42.2±15.9	64.2±19.2	16.8±3.1
All	38.0±13.5	27.8±4.1	13.0±3.1	18.2±6.2	40.3±9.2	54.3±13.3	20.5±12.9
Enrichment factor (EF)		198.2	556.5	3.4	14.9	1.0	4.5

Umrah at the Grand Mosque, therefore reducing the number of vehicles used for their transport.

Recent studies have reported improvements in urban air quality due to significant decreases in traffic and industrial emissions as a result of implementing COVID-19 prevention measures [42]. The complete lockdown in Lima, Peru (March 16 to April 30, 2020) has resulted in a significant reduction in the concentrations of $PM_{2.5}$, PM_{10}, and NO_2 as compared to the concentrations of these pollutants over the same period in 2019 [43]. Altuwayjiri et al. [42] have reported a significant decrease in nitrogen dioxide and benzene during a full lockdown in Milan, Italy compared to the same time in 2019. The concentrations of $PM_{2.5}$, NO_2, SO_2, O_3, and HMs were reduced significantly during a partial lockdown in Hanoi, Vietnam in comparison with historical data obtained in 2017 [44].

In Makkah city, a significant decrease in the concentrations of PM_{10}, CO, NO_2, SO_2, O_3 was reported during the COVID-19 lockdown, compared with concentrations in the pre-pandemic period [45]. The reduction in the concentrations of air pollutants (PM_{10}, NO_2, and CO) during COVID-19 lockdown in the ambient air of Makkah was also recorded by Farahat et al. [22].

Previous studies have considered the construction projects surrounding Al-Haram as an additional source for $PM_{2.5}$. During the period of the present study, most of the construction projects were finished or in their final finishing stages which may explain the lower levels of $PM_{2.5}$ found compared to the levels previously reported by Nayebare et al. [20] and Shaltout et al. [41].

The air mass backward trajectories were analyzed using HYbrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT) transport model to determine the origin and transport pathways of $PM_{2.5}$ to the sampling sites. In the present study, the highest concentrations of $PM_{2.5}$ were found during the first week of June whereas the lowest concentrations were recorded during the third week of May. After the application of the HYSPLIT model (Fig. 2) to these two periods, it was found that the source of backward trajectories during the first week of June was from an area inland with air mass circulation observed around the sampling sites which lead to an increase in the concentration levels of $PM_{2.5}$. In contrast, the backward trajectories in the third week of May originated from the west over the Red Sea with lower loads of $PM_{2.5}$.

Concentrations of HMs in $PM_{2.5}$

The present study focused on the health risk associated with the inhalation of $PM_{2.5}$-bound HMs. Therefore, some of the HMs which are classified as hazardous air pollutants (HAPs) by U.S. EPA were taken into consideration. These included arsenic (As), cadmium (Cd), chromium (Cr), manganese (Mn), nickel (Ni), and lead (Pb). The 24-h average concentrations of these metals in $PM_{2.5}$ from Makkah city were ranked as: Mn (54.3±13.3 ng/m^3)>Pb (40.3±9.2 ng/m^3)>As (27.8±4.1 ng/m^3)>Ni (20.5±12.9 ng/m^3)>Cr (18.2±6.2 ng/m^3)>Cd (13.0±3.1 ng/m^3) (Table 2). The mean concentrations of As and Cd exceeded the limits of WHO air quality guidelines of 6.6 ng/m^3 and 5.0 ng/m^3 respectively while the concentrations of Ni, Mn, and Pb were below the WHO recommended values of 25 ng/m^3,

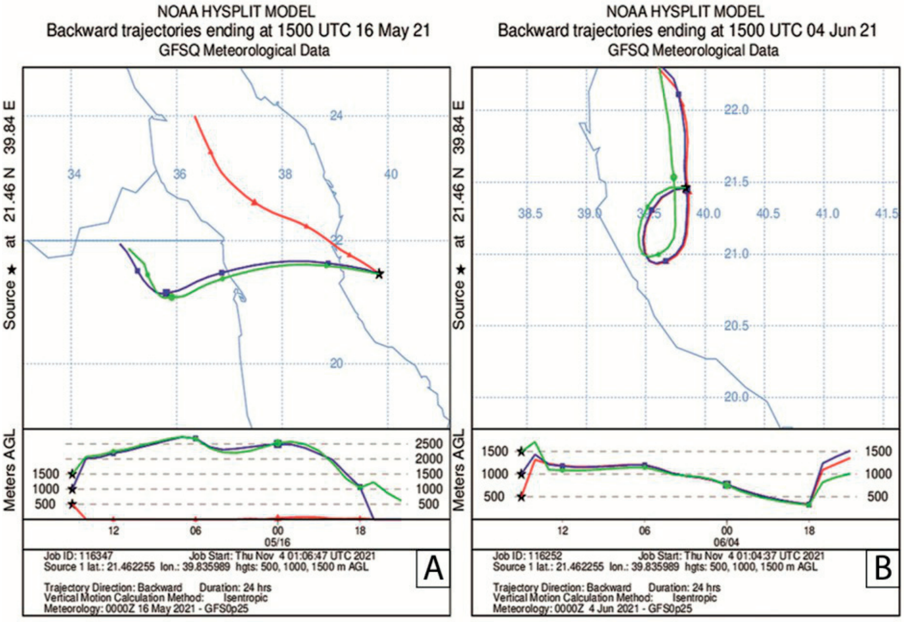

Fig. 2. Air mass backward trajectories at sampling sites in Makkah city (A) during the third week of May (B) during the first week of June.

150 ng/m^3,and 500 ng/m^3, respectively. The average concentration of Cr exceeded the limit associated with an excess lifetime risk of 1:10^4 which is defined as 2.5 ng/m^3 by WHO air quality guidelines [46].

Concentrations of Cr and Ni were comparable to those reported by Nayebare et al. [20] for Makkah city during the spring and summer of 2014 whereas the mean concentrations of Mn and Pb were lower in the present study. It was found that the mean concentrations of As, Cd, and Pb were much lower than the annual mean concentrations described by Habeebullah [21] for the Holy city from August 2012 to September 2013. This difference could be attributed to the short period of sampling in the present study compared to the annual averages reported by Habeebullah [21]. Reduction in transportation and other human activities during the period of the present study, which was a result of the restrictions that were imposed to control the spread of COVID-19 during the year of 2021, could be another factor that resulted in the decrease of concentrations of As, Cd, Mn, and Pb in $PM_{2.5}$ compared to previous studies. The mean concentrations of HMs investigated in the present study were compared with those reported in other cities of different countries (Table 3). It is shown in Table 3 that the levels of HMs in $PM_{2.5}$ in Makkah city were much lower than those determined in other cities.

Anthropogenic Contamination of HMs in $PM_{2.5}$

The calculated EF values were used to evaluate the contamination levels of HMs in the atmosphere of Makkah city (Table 2). The EFs for Mn, Ni, and Cr were lower than 10 indicating that they are of a natural origin from Earth's crust whereas the EFs of Pb, Cd, and As were higher than 10 suggesting they are significantly enriched from anthropogenic sources. The EFs of Pb, Cd, and As from the present study are consistent with those measured for Jeddah city [55].

Although the EF values of Pb showed significant enrichment of this metal from anthropogenic activities, it was found that the mean concentration of Pb in $PM_{2.5}$ from Makkah city was below the permissible limit of the WHO air quality guidelines. Before 2001, which is the year leaded gasoline has been phased out in Saudi Arabia [56], the average concentration of lead in the atmosphere of Riyadh City was twice that of international standards [57]. The change in fuel standards has resulted in a significant reduction of lead in the ambient atmosphere of Saudi Arabia. Lead is not degradable naturally and its presence in the atmosphere of Makkah city reflects the high level of historical contamination and its persistence in the environment [58, 59]. Resuspension of road dust and wheel weight can serve as sources for atmospheric contamination with lead [60]. Although the lead has been banned in petrol, the acceptable limit of lead remains as 13 mg/L which means that the high level of gasoline consumption can contribute to Pb atmospheric pollution [61]. Long-range transportation of this metal from other cities can be a source for Pb in the atmosphere of Makkah city [56].

The major sources of As and Cd atmospheric pollution are industrial activities such as the smelting of metals and burning of coal [62]. Khodeir et al. [61], Harrison et al. [55], and Alghamdi et al. [63] found extremely high enrichment of Cd and As in $PM_{2.5}$ from Jeddah city with EFs>100. Emission of cadmium into the atmosphere can be due to brake and tire wear [58]. The long-range transport of industrial emissions from surrounding industrial cities can be a source for As and Cd contamination in the atmosphere of Makkah as observed from air mass backward trajectories.

Human Health Risk Sssessment of $PM_{2.5}$-bound HMs

The health risk assessment model was proposed by the U.S.EPA to evaluate health risks associated

Table 3. Comparison of HMs concentrations (ng/m^3) in $PM_{2.5}$ in different cities of other countries.

Location	HMs concentrations in $PM_{2.5}$ (ng/m^3)						Reference
	As	Cd	Cr	Pb	Mn	Ni	
Makkah/Saudi Arabia	27.8	13.0	18.2	40.3	54.3	20.5	This study
Algiers, Algeria	93.0	-	45.0	371	3299	-	[47]
Agra, India	35	8	192	128	-	108	[48]
Guangzhou, China	40	20	70	450	150	-	[49]
Bangkok, Thailand	11	113	39	297	201	32	[50]
Xinxiang, China	100	20	140	400	90	70	[51]
Central Andes, Peru	32	14	-	242	-	38	[52]
Shalu, Taiwan	-	8.26	-	105.2	-	40.98	[53]
Thessaloniki, Greece	-	-	96	73	291	87	[54]

with exposure to toxic chemicals [64]. This model is used to assess the non-carcinogenic risks (non-CRs) and carcinogenic risks (CRs) from inhalation, ingestion, and dermal contact with toxicants. The present study estimated the non-CRs and CRs of As, Cd, Cr, Mn, Ni, and Pb using the aforementioned model.

Non-Carcinogenic Risks (non-CRs)

The results on non-CRs associated with ingestion, inhalation, and dermal contact with HMs in Makkah are presented in Table 4. The results showed that the EC>ADD>DAD indicated that the most exposure to HMs in Makkah was via inhalation. The HQ_{inh} for investigated HMs was below the safe level (HQ = 1) suggesting no significant non-CRs for adults and children in Makkah from inhalation. None of the studied HMs showed the ability to induce non-CRs for adults through dermal contact. However, HQ_{der} for children showed the possibility of non-CRs from exposure to As and non-harmful effects from exposure to other HMs. The results showed that Cd, Cr, Pb, Mn, and Ni have no harmful effects for adults through ingestion while As may cause non-CRs. The HQ_{ing} of As, Cd, Cr, and Pb suggested their ability to produce non-CRs for children whereas HQ_{ing} for Mn and Ni indicated the safe level of children exposed to these two metals via ingestion route.

The cumulative non-CRs, which is expressed in terms of the hazard index (HI), were calculated for inhalation, ingestion and, dermal routes using Eq.8 and presented in Table 4. The results of HI for adults showed the likelihood of non-CRs via inhalation (HI = 1.7) and ingestion (HI = 2.3) and no-significant health effects via dermal exposure (HI = 0.29). The HI values of all six metals were >1.0 for children through inhalation (HI = 1.7), ingestion (HI = 21.7), and dermal contact (HI = 1.9) suggesting the possible cumulative non-CRs from children's exposure to these metals. It was observed that the HQ_{inh} values for individual metals have not exceeded the safe level whereas exposure to these metals collectively surpassed the safe level. It can be noticed from Table 4 that arsenic (As) has the major contribution to HQ_{inh} (53.7% for adults and children), HQ_{ing} (74.7% for adults and 73.9% for children), and HQ_{der} (70.9% for adults and 69.1% for children) among the six metals investigated in this study. These results imply that oral ingestion was the most harmful exposure pathway of HMs and arsenic was the main contributor to hazard quotients in $PM_{2.5}$ in Makkah city.

Carcinogenic Risks (CRs)

Among the six investigated HMs, The International Agency for Research on Cancer (IARC) has categorized Cd, Cr (VI), As, and Ni as group 1 carcinogens and Pb as group 2A carcinogens. The CRs of Cd, Cr (VI), As, Ni, and Pb via inhalation were calculated and presented in Table 5. The $ILCR_{inh}$ values for Pb and Ni were $\leq 1 \times 10^{-6}$ indicating very low CRs from these metals whereas the ILCR values for As, Cd, and Cr were in the range of ($10^{-6} \leq ILCR < 10^{-4}$) suggesting low carcinogenic risks from inhalation of these metals through $PM_{2.5}$. Carcinogenic effects could occur through ingestion of As, Cr, Pb, and Ni as depicted from $ILCR_{ing}$ values while Cd showed no carcinogenic effects via this route of exposure for both adults and children. Arsenic has the highest carcinogenic risk from dermal contact followed by Cr whereas Cd and Ni have no CRs for both age groups. Pb has low carcinogenic effects for children and cannot induce carcinogenic effects on adults via dermal exposure.

The cumulative ILCR values for the studied HMs in $PM_{2.5}$ were lower than the acceptable level of (1×10^{-4}) for adults and children via inhalation and dermal routes (Table 5) suggesting no significant carcinogenic risks from exposure to the investigated metals collectively via those pathways. The cumulative ILCR values for ingestion of HMs were higher than the acceptable level indicating the moderate CRs from ingestion of $PM_{2.5}$ containing these metals together by children and adults. It was found that arsenic contributed for the most of cumulative ILCR (80.3 % for adults and 81.4% for children) through the ingestion route followed by Cr with 17.5 % for adults

Table 4. Non-CRs assessment of HMs in $PM_{2.5}$ via ingestion, inhalation, and dermal exposure

HMs	ADD		EC		DAD		HQ_{ing}		HQ_{inh}		HQ_{der}	
	Adults	Children	Adults	Children	Adults	Children	Adults	Children	Adults	Children	Adults	Children
As	5.1×10^{-4}	4.8×10^{-3}	1.3×10^{-2}	1.3×10^{-2}	6.2×10^{-5}	4.0×10^{-4}	1.7	16.0	9.1×10^{-1}	9.1×10^{-1}	2.1×10^{-1}	1.3
Cd	2.4×10^{-4}	2.2×10^{-3}	6.4×10^{-3}	6.4×10^{-3}	9.6×10^{-7}	6.2×10^{-6}	2.4×10^{-1}	2.2	3.2×10^{-4}	3.2×10^{-4}	9.6×10^{-4}	6.2×10^{-3}
Cr	3.3×10^{-4}	3.1×10^{-3}	8.9×10^{-3}	8.9×10^{-3}	1.3×10^{-5}	8.8×10^{-5}	1.1×10^{-1}	1.0	8.9×10^{-2}	8.9×10^{-2}	4.4×10^{-3}	2.9×10^{-2}
Pb	7.4×10^{-4}	6.9×10^{-3}	1.9×10^{-2}	1.9×10^{-2}	2.9×10^{-4}	1.9×10^{-3}	2.1×10^{-1}	1.9	5.6×10^{-3}	5.6×10^{-3}	8.5×10^{-2}	5.5×10^{-1}
Mn	1.0×10^{-3}	9.4×10^{-3}	2.6×10^{-2}	2.6×10^{-2}	4.0×10^{-5}	2.6×10^{-4}	2.1×10^{-2}	1.9×10^{-1}	5.4×10^{-1}	5.4×10^{-1}	8.5×10^{-4}	5.5×10^{-3}
Ni	3.8×10^{-4}	3.5×10^{-3}	1.0×10^{-2}	1.0×10^{-2}	1.5×10^{-5}	9.9×10^{-5}	1.9×10^{-2}	1.7×10^{-1}	1.1×10^{-1}	1.1×10^{-1}	7.5×10^{-4}	4.9×10^{-3}
(HI)							2.3	21.7	1.7	1.7	0.29	1.9

Table 5. CRs assessment of HMs in $PM_{2.5}$ via ingestion, inhalation, and dermal exposure.

HMs	ADD		EC		DAD		$ILCR_{ing}$		$ILCR_{inh}$		$ILCR_{der}$	
	Adults	Children	Adults	Children	Adults	Children	Adults	Children	Adults	Children	Adults	Children
As	1.7×10^{-4}	4.1×10^{-4}	4.7×10^{-3}	3.3×10^{-3}	3.0×10^{-5}	1.7×10^{-5}	2.7×10^{-4}	6.2×10^{-4}	1.6×10^{-5}	1.1×10^{-5}	4.5×10^{-5}	2.6×10^{-5}
Cd	8.3×10^{-5}	1.9×10^{-4}	2.2×10^{-3}	1.5×10^{-3}	3.3×10^{-7}	5.4×10^{-7}	1.6×10^{-9}	3.9×10^{-9}	9.2×10^{-6}	6.5×10^{-6}	6.6×10^{-12}	1.1×10^{-11}
Cr	1.1×10^{-4}	2.7×10^{-4}	3.1×10^{-3}	2.2×10^{-3}	4.6×10^{-6}	7.6×10^{-6}	5.8×10^{-5}	1.4×10^{-4}	4.6×10^{-5}	3.3×10^{-5}	2.3×10^{-6}	3.8×10^{-6}
Pb	2.6×10^{-4}	6.0×10^{-4}	6.8×10^{-3}	4.8×10^{-3}	1.0×10^{-4}	1.7×10^{-4}	2.2×10^{-6}	5.1×10^{-6}	8.2×10^{-8}	5.8×10^{-8}	8.7×10^{-7}	1.4×10^{-6}
Ni	1.3×10^{-4}	3.0×10^{-4}	3.5×10^{-3}	2.5×10^{-3}	5.2×10^{-6}	8.5×10^{-6}	2.6×10^{-6}	6.1×10^{-6}	9.0×10^{-7}	6.4×10^{-7}	1.0×10^{-7}	1.7×10^{-7}
(∑ ILCR)							3.3×10^{-4}	6.7×10^{-4}	7.2×10^{-5}	5.1×10^{-5}	4.8×10^{-5}	3.1×10^{-5}

and 17.8 % for children while other metals showed low CRs.

This brings us to the conclusion that exposure to $PM_{2.5}$-bound arsenic through ingestion is the most health-threatening exposure in terms of CRs and non-CRs in Makkah. Therefore, controlling arsenic contamination in $PM_{2.5}$ in Makkah city should be given the highest consideration due to its non-carcinogenic health risk.

Compared to adults, children were at higher CRs and non-CRs to HMs from ingestion as can be inferred from HI and ∑ILCR values. This can be attributed to their hand-to-mouth and objects-to-mouth behavior, which could result in the direct consumption of dust-containing HMs [65]. Moreover, children are more vulnerable to the absorption of HMs from the gastrointestinal tract and their blood hemoglobin sensitivity to these metals is also higher than that of adults because of their lower body weight [66].

Health Risk Mitigation Measure

Health risk assessment (HRA) provides information to the concerned agencies helping them to take the best decision for abatement of environmental pollution and reducing its effects. Although there are uncertainties associated with using the health risk assessment model [37], HRA is important for the development of regulations and strategies to reduce chemical exposure for the protection of public health.

In 2020, The Ministry of Environment, Water and Agriculture of Saudi Arabia has made amendments to air quality regulations. These amendments include the ambient air quality standards which involve the permissible limits for several contaminants including Pb, As, Cd, Mn, and Ni. Many regulations have been adopted to obligate operators (mobile and stationary sources) to utilize the best available technologies to meet the emissions standards. Enforcement of these standards is the most important step in reducing the emissions of HMs into the atmosphere. Recently, many initiatives have been adopted by the Government of Saudi Arabia as an effort for protecting the environment. This includes the Saudi Green Initiative and the Middle East Green Initiative which aim at reducing the emissions of hydrocarbon industries by 60% and planting billions of trees in the region. Effective implementation of these initiatives will play an important role in mitigating hazardous air pollutants including $PM_{2.5}$ and HMs. Controlling air pollution needs regional collaboration because of its transboundary nature.

Education and dissemination of information regarding the contamination levels and health risks associated with exposure to air pollutants is important for raising awareness among the public on the effects of atmospheric pollution and the available measures to reduce exposure to such contaminants. Children who are at a higher health risk from exposure to HMs must be educated on food safety and not consume food and water that has been exposed for long periods. Also, they should be educated on the importance of hand hygiene and how to develop the habit of handwashing after touching contaminated surfaces.

Conclusions

In the present study, the concentration level and health risk assessment associated with exposure to $PM_{2.5}$-bound HMs in Makkah city were evaluated. The mean concentration of $PM_{2.5}$ in Makkah (38.0±13.5 $\mu g/m^3$) was above the value recommended by WHO and ambient air quality standards of Saudi Arabia. The AQI for $PM_{2.5}$ showed moderate air quality in the city during 60% of the sampling period.

The EF values showed extremely high enrichment of $PM_{2.5}$ with As and Cd. The mean concentrations of As and Cd have exceeded the limits of WHO air quality guidelines. Their presence in the air of Makkah can be due to the long-range transport from surrounding industrial cities.

Arsenic was the major contributor to HQ_{inh}, HQ_{ing}, and HQ_{der} among the six metals. Oral ingestion was the most detrimental pathway of HMs in Makkah. The HI values for children suggest the likelihood of non-CRs from exposure to several metals via inhalation, ingestion, and dermal contact whereas the HI values for adults showed that the exposure to the investigated metals collectively can induce non-CRs through inhalation and ingestion only. The cumulative ILCR values showed moderate CRs from ingestion of $PM_{2.5}$-bound HMs by children and adults in Makkah with arsenic being the major contributor for cumulative ILCR.

The present study recommends that future research should evaluate the contamination levels and health risks of HMs and other toxic chemicals present in various environmental media in different cities of Saudi Arabia.

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Conflict of Interest

The authors declare no conflict of interest.

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