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OUTDOOR AIR CONTAMINANTS-HEAVY METALS AND ASSOCIATED HEALTH RISKS IN DUHOK-IRAQ

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ABSTRACT

Toxic elements in the atmosphere can cause a variety of health and environmental problems. The current work aims to assess the concentrations of heavy metals associated with the air in the Duhok city-Kurdistan Region of Iraq. For the first time a low volume sampling pump with Glass Microfiber Filter paper was used. Forty samples of air filter aerosol particles were collected from several areas in the Duhok city, during the dry and rainy seasons. Samples of air filters were prepared by a microwave digestion system and assessed by inductively coupled plasma Optical-Emission Spectrometry (ICP-OES). The average concentrations of Cr, Zn, Ni, Cd, Fe, and Pb found in 20 sites in the study area for the dry season were 0.004 mg/m^3, 0.007 mg/m^3, 0.003 mg/m^3, 0.002 mg/m^3, 0.142 mg/m^3, and 0.402 mg/m^3 respectively. Also, the average levels of the mentioned elements were 0.003 mg/m^3, 0.004 mg/m^3, 0.012 mg/m^3, 0.003 mg/m^3, 0.138 mg/m^3, and 0.645 mg/m^3 respectively in wet season. Seasonal results confirmed no significant differences in the concentrations of elements measured at all study sites, with the exception of lead, which doubles during the rainy season. Results showed that the levels of metal concentrations in the present study exceeded the standard limits of WHO. The results of the health risk assessment showed that the population in Duhok city had a higher lifetime chance of developing cancer as a result of these air concentrations and their heavy metal content.

KEYWORDS: heavy metals, air pollution, inhalation, air filter, ICP-OES, Duhok city.

1. INTRODUCTION

Today, the ecosystem of the organism and humans is at risk due to exposure to pollution in the atmosphere. It is a serious human health concern. People's life has attracted more attention to researchers to study the effect of pollutants emitted from industrial activity and other technologies on human health [1].

Wet and dry separation processes continuously separate air particles. Factors such as the energy of the dust-carrying wind and the characteristics of dust concentration in the atmospheric deposition environment influence the rate of air pollution deposition [2]. Other factors include the nature of the release source, the specific weather conditions, the location of the sampling site, and the distance [3]. A helpful foundation for assessing the characteristics of dust in particular places is provided by understanding the spatial and temporal variability of dust deposition rates [4]. Beryllium, cadmium, chromium, and nickel and their compounds, arsenic and inorganic arsenic compounds are classified as Group 1 by the International Agency for Research on Cancer (IARC, 2012) [5], (Carcinogenic to humans). Many other metals and metal compounds are classified as Group 2B (probably carcinogenic to humans), while inorganic lead compounds are classified as Group 2A (possibly carcinogenic to humans). Trace elements, on the other hand, are known to be specific to their source. Trace metals are transported with equally hazardous aerosols without volatilization or decomposition. Therefore, measuring trace elements to elucidate their health effects and identify the origin of dangerous chemical species is of great importance [6]. The earth's crust contains a lot of the metal zinc (Zn). It enters the environment from both anthropogenic and natural sources. Releases from man-made sources, however, are greater than releases from natural sources. The usage of commercial products containing zinc as well as zinc mining and metallurgical processes are the main anthropogenic sources of zinc in the environment (air, water, and soil) [7].

According to the International Agency for Research on Cancer (IARC), cadmium (Cd) falls within Group I and cadmium in ambient air represents the majority of total airborne cadmium. The quantities and processes of human infection vary greatly, but different categories of cadmium can have an impact on human health and Cd uptake. Contrarily, cadmium from cigarette smoke and cadmium from work situations directly infect people. Cadmium from the air typically settles in the water and soil, and then moves on to animals and plants before it finally makes its way into people's bodies through the food chain. Because it targets the kidneys and leads to the formation of kidney stones, cadmium is extremely harmful [8].

More than 2 million premature deaths each year can be linked to the impacts of indoor and outdoor urban air pollution (from fuel burning), according to estimates of the burden of illness from air pollution from the World Health Organization (WHO) [2]. And more than half of these disease encumbrances are borne by the inhabitants of developing countries. Since 1987, air quality guidelines have been available in awareness of the need for clean air among humans. In order to manage danger assessments and hazard determinations, local and national authorities are meant to get background information and direction from these recommendations [9].

The main goal of this study is to establish a base-level data set of the concentration of different heavy metals and health risks in air particles, in addition to the statistical evaluation of the outcomes. This set could be used for the development of relevant guidelines or standards for taking corrective or regulatory actions in cases of high or needless concentration values.

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2. MATERIALS AND METHODS

2.1. Study Area

Duhok Governorate is located in the far northwest of Iraq and forms the Western Governorate of Iraq's Kurdistan Region. The county lies in a tolerable range between rolling and alpine areas. At 36.7°N, 42.3°E and 37.4°N, 44.2°E, it lies between the two lines. Duhok district: with an area of 1092 km^2, has a population of 340,000 and is located in the center of the administrative district. In general, the Kurdistan Region of Iraq has a semi-arid climate, with temperatures ranging from around 40°C in summer to below freezing degree in winter and low relative humidity. Precipitation is concentrated in winter, with an annual rainfall of around 400 mm, especially from November to April. Northwest winds from Turkey are dominant all year round[10].

2.2. Sample Collection

The sub-sites or locations for the total suspended particulate (TSP) samples were selected in four different sites in the Duhok governorate according to four criteria such as traffic lights, streets, residential areas, and industrial areas. A topographic map of the study area was used to identify suitable locations for air samples for each site. The studied four sites and the number of samples was identified and coded accordingly as shown in Table 1 and Figure1.

A total of 40 air samples were collected on a high collection efficiency glass fiber filter (Whatman 934-AH Glass Microfiber Filter 25 mm) at a constant flow rate of 2.5 LM^{-1} for 12 hours by a low volume air samplers pump (SENSIDYNE, USA). The first 20 air samples were collected from all sites during the dry season for 2 months and the remaining 20 air samples were collected from the same sites during the wet season for 2 months.

Table1: The four air sampling locations and number of samples

Location	Samples code					
Traffic	A01	A02	A03	A04	A05	
Street	A06	A07	A08	A09	A10	A11
Residential	A12	A13	A14	A15	A16	
Industrial	A17	A18	A19	A20		

Figure 1: Air sample sites in Duhok city

The set of air sampling systems (filtered pumps) shown in Figure 2 was designed and installed outdoors 1.5 m above the ground. The equipment is housed in a small, portable metal cabinet with open sides to protect the system from the elements. This device was used to collect air samples from various sites around the city of Duhok. Before and after each sampling day, the pump was calibrated to ± 5% of the true reading. The calibration of the air pump was performed using an accurate flowmeter. Before and after sampling, a sensitive balance with a 0.0001 mg scale was used to obtain an accurate weight of the contaminant on the filter and store it in a polyethylene bag for weighing.

Figure 2: Air sampler system [11]

2.3 Samples Analysis

Following a standard protocol [9], the air pump filters (air samples) digested in 10 ml concentrated nitric acid [12]. The resulted solution of each sample is composed in a PTFE digestion vessel and adding 2ml of HCl to break hydrogen bonds. The solution is mixed and left standing for 2hrs. A microwave digestion system is used to digest samples (Multiwave Go, Software Version 2.00, Austria).

The microwave digestion system was heated at 1200 W for 0.5 hours and the pressure increased from 5.5 bar to 10.4 bar. The digested sample was diluted with 50 mL of deionized water and passed through a 0.45-micron pore size membrane filter to remove solid residues [13].

Inductively coupled plasma optical emission spectroscopy (ICP-OES) was used to analyze the total elemental composition of various heavy metals in all samples (Thermo Scientific, ICAP 7000 Plus Series ICP-OES, Cambridge, UK). The accuracy of each sample's analysis is checked using concurrent analysis of standard reference materials.

2.4 Health Risk Assessment of the Selected Metals

Heavy metals in outdoor air can exacerbate human health risks. To uncover additional health intimidations from those pollutants, equation 1 was used to determine the average amount of specific metals exposed by inhalation (D_{inh}) for children and adults over a given length of time depending on body weight [14].

$$D_{inh} = \frac{C \times InhR \times EF \times ED}{BW \times AT} \ldots\ldots\ldots\ldots\ldots\ldots (1)$$

Here D_{inh} is exposed via respiratory tract inhalation *(mg/kg.day); C (mg/m³)* is the concentration of the desired metal in air calculated from the upper 95% confidence interval of the mean; *InhR* is the inhalation rate (7.6 and 20 m^3/day for children and adults, respectively) [14]; *EF* (350 day/year) is the exposure frequency [12].; *ED* is the duration of exposure (6 and 24 years for children and adults, respectively); *BW* is the mean weight (15 kg and 70 kg for children and adults, respectively) and *AT* is the mean time *AT (day) = ED × 365* for non-cancer risk and AT (day) for cancer risk = 70 × 365 [14].

The lifetime average daily dose (*LADD*) of the selected metals is used to assess cancer risk as shown in Equation 2 [15].

$$LADD = \frac{C \times EF}{AT} \times \left[\left(\frac{InhR \times EF}{BW}\right)_{child} + \left(\frac{InhR \times EF}{BW}\right)_{adult}\right] \ldots\ldots\ldots\ldots\ldots\ldots (2)$$

The following formula was used to estimate the non-carcinogenic risk hazard index (*HQ*) for each metal by inhalation. This was achieved by dividing the D_{inh} from the inhalation route of exposure by a specific reference dose (*RfD*) [15].

$$HQ = \frac{D_{inh}}{RfD} \ldots\ldots\ldots\ldots\ldots (3)$$

$$HI = \sum HQ_i \ \ldots\ldots\ldots\ldots\ldots (4)$$

Where *RfD* is the dose reference (mg/ kg. day); *HI* is hazard Index calculated by summing up the individual *HQ* to estimate the total health risks of all heavy metals considered. *RfD* values were used for Cr(2.86×10^{-5}), Zn(3.01×10^{-1}), Ni(2.06×10^{-2}), Cd(1×10^{-3}), Fe(7×10^{-1}) and Pb(3.52×10^{-3}) [12,14]. If the computed HI is less than 1, then exposure is not anticipated to have any negative impacts on health. On the other hand, if HI is greater than 1, adverse health effects may result. [14].

The cancer risk of a certain element (Re) and the Cancer risk of total elements (Rt) are calculated using equations 5 and 6 [14].

$$R_e = LADD \times SF_a \quad \ldots\ldots\ldots\ldots\ldots (5)$$

$$R_t = \sum R \ \ldots\ldots\ldots\ldots\ldots (6)$$

Where SF_a is a slope factor (mg/kg. day). The SF_a values used for Cr, Ni, Cd, and Pb are 42, 0.84, 6.4, and 0.042 respectively.

Internationally accepted precautionary or threshold values for cancer risk are 10^{-6}, above which the risk is unacceptable [14].

3. RESULTS AND DISCUSSION

3.1 Metal Concentrations

The descriptive statistics results of trace elements of samples in dry and wet seasons were presented in Table 2. General view shows there were no significant differences in concentration of measured elements for the four sites in and around Duhok city in each season. However, a higher concentration is observed for Pb and Fe in both seasons.

From the ratio of the standard deviation to the mean, the values of the coefficient of variation (CV %) (Given in Table 2) varied from lowest value 14% for Pb for dry season to highest value 62% for Cr in wet season. It was clear from the table that the coefficient of variation for lead and iron was the lowest and increased for the rest of the elements. This means that the concentration of Pb and Fe elements in different locations had the lowest level of dispersion around the mean and the more precise the estimation.

As carcinogenic in humans, the mean concentration values of chromium (Cr) in both seasons exceeded by 10 times the maximum limit values established by WHO regulation 0.0004 mg/m^3 [16]. A higher variation in the concentration of zinc was found in summer 0.003 mg/m^3 to 0.010 mg/m^3 with the coefficient of variation at 47%, and decreased to 33% in winter.

Table 2. Concentration of heavy metals in dry and wet air samples

Season	Element	Con.* (mg/m^3)				Min*	Max*	Mean*	S.D.*	CV%
		Traffic	Street	Residential	Industrial					
Dry	Cr	0.006	0.004	0.003	0.002	0.002	0.006	0.004	0.002	50
	Zn	0.003	0.010	0.005	0.010	0.003	0.010	0.007	0.003	47
	Ni	0.004	0.003	0.001	0.003	0.001	0.004	0.003	0.001	39
	Cd	0.002	0.002	0.002	0.001	0.001	0.002	0.002	0.001	28
	Fe	0.150	0.115	0.168	0.135	0.115	0.168	0.142	0.022	16
	Pb	0.320	0.441	0.433	0.416	0.320	0.441	0.402	0.056	14
Wet	Cr	0.002	0.003	0.006	0.002	0.002	0.006	0.003	0.002	62
	Zn	0.006	0.004	0.004	0.003	0.003	0.006	0.004	0.001	33
	Ni	0.011	0.012	0.015	0.011	0.011	0.015	0.012	0.002	16
	Cd	0.002	0.003	0.004	0.002	0.002	0.004	0.003	0.001	43
	Fe	0.101	0.162	0.123	0.165	0.101	0.165	0.138	0.003	22
	Pb	0.692	0.597	0.828	0.462	0.462	0.828	0.645	0.015	24

* Con.: Concentration of the heavy metals (mg/m^3); Min: minimum concentration value of the different location (mg/m^3); Max: maximum concentration value of the different location (mg/m^3); Mean: average concentration (mg/m^3); S.D.: Standard deviation (mg/m^3); CV%: Coefficient of variation.

As shown in Table 3, the variation of Zn concentration in the present study from other locations might be due to the geological formations and anthropogenic sources.

The higher average nickel (Ni) concentration found was 0.003 mg/m^3 in the dry season with a higher coefficient of variation standing at 39% and was 0.012 mg/m^3 with lowest CV at 16% for wet season (Table 2). The results observed in the current study were higher than the WHO's maximum allowed limit of 0.00007 mg/m^3 however, the maximum value (0.04) was lower than the nickel concentration in some earlier works except for Babylon.

The average concentration of Cd in Duhok found were 0.002 mg/m^3 and 0.003 mg/m^3 observed in the dry and wet season respectively as it is clear in Table 2. The 28% and 43% for dry and wet seasons respectively, which showed a little higher degree of effect in wet weather on its coefficient variation of Cd concentration was 28% and 43% for dry and wet seasons respectively, which showed a little higher degree of effect in wet weather on its concentration. As carcinogenic in humans, the main values for this metal exceed by about 100 times the maximum limit values established by WHO regulation 0.00003 mg/m^3 [16]. Sludge treatment is an important source of Cd

disposal [13]. This higher value needs improvement by sludge processing system in Duhok city.
In both seasons, comparatively high concentrations of iron (Fe) and lead (Pb) were found in all sampling sites. The Fe concentrations ranged from 0.101 mg/m^3 to 0.168 mg/m^3 with a higher average value 0.142 mg/m^3 observed in dry season. The coefficient of variation was 16% and 22% for dry and wet seasons respectively. Higher iron content readings could have attributed to local geological phenomena such as iron-manganese nodules and Carrstones. High iron concentrations in the region's soils and water provide a significant problem [17].
The average concentration of lead (Pb) varied from 0.402 mg/m^3 in dry season to 0.645 mg/m^3 in wet season with the coefficient of variation standing at 14% and 24% for both seasons respectively as shown in Table 2. It can be seen that the concentrations of lead in the atmosphere of Duhok were almost constant for all locations in the summer season. It was assumed that this is due to two reasons; the first of which is the poor quality of the gasoline used that contains a high percentage of lead in the means of transportation of more than 235,000 different vehicles in use per day [18]. The second is the huge number of local electric power stations within the cities of the Kurdistan region. There are over 1814 generators in Duhok city to provide residences and business establishments with electricity during a countrywide power outage [19]. These stations operate throughout the entire day at more than 12 hours intermittently. This act pollutes the city's air with toxic pollutants such as lead. On the other hand, we note that the highest concentration of lead in the winter season was found in residential areas 0.828 mg/m^3. The reason could be attributed to the rain, which leads to the deposition of toxic substances in the same area and prevent their separation to neighboring areas. Values recorded in the present study were much above the average permissible limit of 0.0005 mg/m^3 (WHO).
Another possible source of air pollution in the area of Duhok city is the Duhok gas power plant (Kwashi) located in the northwest of the city. It is about 30 km away from the city center. Pollutants emitted by this industry include gases from petroleum refineries and coal combustion, trace elements in calcareous materials, dust from grinding, organic waste in raw materials, petroleum and petroleum combustion, toxic elements and organic contaminants, and organic waste emitted from raw materials [20]. As shown in Table 3, the concentrations of Fe and Pb in the present study were less than the concentration measured by previous work in Duhok, which depend on the collection of the deposited dust samples by other techniques [20]. While the values of most elements in this study contrast or agree with some studies conducted on other locations in Iraq.

Table 3: The average concentration of heavy metals in air samples reported from some other locations of the Iraq

Country		Con.* (mg/m^3)						Reference
		Cr	Zn	Ni	Cd	Fe	Pb	
Duhok	DS	0.008	0.019	0.007	0.009	0.324	3.411	Present study
	WS	0.007	0.010	0.030	0.011	0.314	5.464	
Duhok		-	-	-	-	7.04	14.48	[20]
Basrah		3.56	-	3.92	0.08	-	7.51	[21]
Baghdad		-	-	0.0981	0.0223	-	0.213	[22]
Basrah		0.0712	-	0.078	0.0016	-	0.1502	[3]
Maysan		-	-	4.93	2.01		4.99	[23]
Baiji		0.9	-	0.65	0.13	-	4.90	[24]
Al- Diwaniyah		0.001	0.002		0.036		0.003	[25]
Babylon				0.002			0.007	[26]
Permissible limits		0.0004	0.0006	0.00007	0.00003	0.0043	0.0005	[10]

* Con.: Concentration of the heavy metals (mg/m^3); DS: Dry season; WS: Wet season.

3.2 Health Risk Assessment

Table 4 showed the average dose of of Cr, Ni, Zn, Cd, Fe, and Pb through the inhalation exposure system, *LADD, HQ and HI* of non-carcinogenic metals, as well as the risk values of carcinogenic heavy metals in Duhok air.
The current study's findings demonstrated that, with the exception of Pb, the likelihood of exposure to heavy metals causing non-cancer consequences was higher during the dry season for Cr, Zn, Ni, Cd, and Fe than during the rainy season.
In both seasons, the non-carcinogenic effect (*HQ*) through the inhalation routes for both children and adults of Cr and Pb was greater than unity. It is acceptable to note that the non-cancerous effects of these six non-carcinogenic heavy metals affect children more than adults as the HQ children values were nearly twice as high as the HQ adult values in both seasons. Due to the fact that children's hand-to-mouth actions are a significant conduit for chemical exposure, this is caused by their mouthing behaviours [27].
In previous studies, epidemiological studies reported associations between exposure to elevated airborne concentrations of some metals, such as Cd and Ni, and markers of cardiovascular disease [15]. Epidemiological studies have established a link between Cd exposure and the development of hypertension leading to atherosclerosis and myocardial infarction. Toxicological studies report hyperglycemia, insulin resistance, and glycemic dysregulation due to Ni exposure [28].
Moreover, the hazard index (HI) of all the heavy metals in the present study was counted as the sum of all HQs for individual metals in inhalation [14], the safe limit of unity was observed to be exceeded by the cumulative non-carcinogenic effects of all heavy metals through inhalation exposure pathways. This indicates that there is a higher likelihood of non-carcinogenic effects from exposure to the synergy of heavy metals in the air than there is from exposure to individual metals. This implied that the metals might have an overall, non-cancer influence on people of all ages. Metals' HI values for the two separate age

groups vary seasonally, with the HI value in both seasons being larger than 1. There have also been reports of HI occurring when the safe limit for multiple elements is exceeded [15].

The study found a consistent pattern across age groups for non-cancer effects. For example, Cr and Ni might have the greatest non-cancer effects in children via the inhalation route. It was previously established that children are more susceptible to the adverse effects of pollutants than adults [15]. Reasons for this include children breathing more air per body weight and their immature immune systems being unable to cope with environmental contaminants [1].

It was clear that carcinogenic heavy metals had the following risk levels in that order: Cr > Pb > Cd > Ni. All metals had carcinogenic risks that were greater than 10^{-6}. The U.S. Environmental Protection Agency considers any cancer risk lower than 10^{-6} to be insignificant (USEPA) [14]. These findings showed that the lifetime cancer risks of Cr, Pb, Cd, and Ni clearly surpass the cut-off and may put Duhok inhabitants at an increased risk of developing cancer.

Table 4: The exposure and health risks of heavy metals in air via inhalation for dry and wet season

Season	Element	Mean Con.* (mg/m^3)	D_{inh} of children	D_{inh} of adult	*LADD*	*HQ* children	*HQ* adult	R_e
Dry	Cr	0.004	8.10E-05	4.57E-05	2.26E-05	2.83E+00	1.60E+00	9.50E-04
	Zn	0.007	1.42E-04	8.00E-05	3.96E-05	4.71E-04	2.66E-04	
	Ni	0.003	6.08E-05	3.43E-05	1.70E-05	2.95E-03	1.66E-03	1.43E-05
	Cd	0.002	4.05E-05	2.29E-05	1.13E-05	4.05E-02	2.29E-02	7.24E-05
	Fe	0.142	2.88E-03	1.62E-03	8.03E-04	4.11E-03	2.32E-03	
	Pb	0.402	8.14E-03	4.59E-03	2.27E-03	2.31E+00	1.31E+00	9.55E-05
	∑	0.560	1.13E-02	6.40E-03	3.17E-03	5.20E+00	2.93E+00	1.13E-03
Wet	Cr	0.003	6.69E-05	3.77E-05	1.87E-05	2.34E+00	1.32E+00	7.84E-04
	Zn	0.004	8.71E-05	4.91E-05	2.43E-05	2.89E-04	1.63E-04	
	Ni	0.012	2.49E-04	1.41E-04	6.96E-05	1.21E-02	6.82E-03	5.84E-05
	Cd	0.003	5.67E-05	3.20E-05	1.58E-05	5.67E-02	3.20E-02	1.01E-04
	Fe	0.138	2.80E-03	1.58E-03	7.81E-04	3.99E-03	2.25E-03	
	Pb	0.645	1.31E-02	7.37E-03	3.65E-03	3.71E+00	2.09E+00	1.53E-04
	∑	0.806	1.63E-02	9.21E-03	4.56E-03	6.12E+00	3.45E+00	1.10E-03

4. CONCLUSIONS

The seasonal results confirmed that there is a non-significant difference in the concentrations of the measured elements in all study sites. In all locations, the concentrations of the most toxic heavy metals, such as lead, chromium, and cadmium, nearly doubled in the rainy season. The highest concentration of these metals, specifically lead was observed in residential areas. This might be due to the huge number of local electric power stations within the city that spew these toxins into the atmosphere, in addition to the poor quality of the gasoline used, which contains a high percentage of lead in these facilities and means of transportation.

A health hazard assessment concluded that inhalation is the primary route of human exposure to particle-bound trace elements, followed by absorption through the skin. The multi-element hazard index (HI) showed an exacerbation of health hazards when exposure to mixtures of elements was taken into account. The potential carcinogenic risk of each element (Cr, Ni, Zn, Cd, Fe, and Pb) observed via the inhalation route far exceeds the acceptable carcinogenic levels, and both adults and children are at very high cancer risk. The study also found that children in the study area were more susceptible than adults to both non-carcinogenic and carcinogenic effects. This demonstrates the need for mitigation and control measures against the sources that release these elements into the environment.

More epidemiological research is needed to better understand these exposure routes and levels. It should also be noted that this is a preliminary health risk assessment study based on observed concentrations of various elements and various health risk assessment models based on certain assumptions. As a result, the health risks obtained are only estimates of potential consequences. In addition, we assumed that all elements of the ambient dust were bioavailable, which is not possible in practice. Human resistance development is also not taken into account. Despite some uncertainties in estimating health risks, this exposure assessment model is an effective means of assessing human health risks from airborne trace elements in the atmosphere.

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