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Evaluation of Cancer Risk of Heavy Metals in the Air of a High Traffic Urban Region and Its Source Identification

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ABSTRACT

Background: Sampling was conducted on particles smaller than ten microns (PM_{10}) in a high-traffic urban region once a week for two years in which fifteen heavy metals were measured.
Methods: positive matrix factorization (EPA-PMF_5), was used for source apportionment and characterization of the collected PM10. Assessment of cancer risk resulting from metals including arsenic, cadmium, chromium, nickel and lead was conducted in three concentration ranges of maximum, average and minimum.
Results: Results for children and adults living in the region indicated that cancer risk indexes at different concentration ranges of carcinogenic metals were between 10^{-4} to 10^{-6} for adults and children. Since EPA recommendations suggest that planning should be conducted if cancer risk is in the range of 10^{-4} to 10^{-6}, using PMF_5 model, source characterization of pollutants was implemented by all measured heavy metals.
Conclusion: It was found that 41.5% of PM_{10} resulted from fuel and combustion, 12% from waste dump soil of lead and zinc industries, 35.7% from suspended open soil and 11% from industrial activities. It was also found that cadmium, nickel and, chromium have higher cancer risk than other metals and, suspended open soil, industrial activities and industrial fuel and combustion are the main sources of these metals respectively.

1. Introduction

International Agency for Research on Cancer (IARC), has classified the outdoor air pollution and particulate matter as carcinogenic to humans (Group 1) (1). Heavy metals in airborne particles enter the body through breathing. The emission of heavy metals due to their absorption, accumulation and lack of decomposition and carcinogenicity in human body is one of the noteworthy points which has been introduced as the main urban air pollutants (2, 3). Usually in areas prone to heavy metal pollution, health risk assessment is implemented in terms of their confrontation with

vulnerable people (4, 5, 6). However in most cases especially when several pollutants are present in the region, improvement would not be conducted in risk status because the source which caused the emission of health risk pollutants would not be identified.

The present study was conducted in Zanjan, which is the center of lead and zinc production of Iran. Iran is the fourth largest producer of lead and zinc in Asia after China, Kazakhstan and India. In addition to the common urban pollution sources including traffic, fuel combustion, suspended open soil etc. Zanjan possesses more than a hundred factories around the city associated with lead and zinc industry. Anguran as Iran's largest zinc and lead mine is also located in this province. The studies have proved that heavy metals are present in concentrations larger than standard levels in the air (7), water (8, 9), soil (10, 11), and regional products (12). Several researches have been done to remove the heavy metals existing in this area (13), but health risk assessment and source routing of air pollutants have not been conducted in this area.

In this study, after measurement of heavy metals in air particles smaller than and equal to ten microns (PM_{10}) in this area, cancer risk assessment was conducted for both children and adult residents. Then regional pollution sources and their share of pollution were identified using PMF_5 source routing model. Afterwards the contribution of identified sources to emission of carcinogenic metals was determined.

2. Material and Methods

2.1. Description of Study Area

Zanjan is located in North West Iran with coordinates 36° 41' N, 48° 27' E and average height of 1620 meters. The area of this city is 81 km^2 and its population was 480000 people in 2015. The area has semi-arid cold climate with hot dry summers and cool wet winters. The annual precipitation is about 295 mm and annual average air temperature is 10 ° C. There is evacuation site for waste soil on the south side of the city near the zinc industrial complex with almost one kilometer area. More than three million tons of waste soil from about 100 factories inside the area is daily discharged in the site without any environmental concerns.

Moreover, there is a site with an area of almost one Km^2 on the east side of town within 16 km distance near Zanjan lead and zinc factory allocated to the discharge of waste soils. Location of industrial complexes surrounding the city is shown in Figure 1.

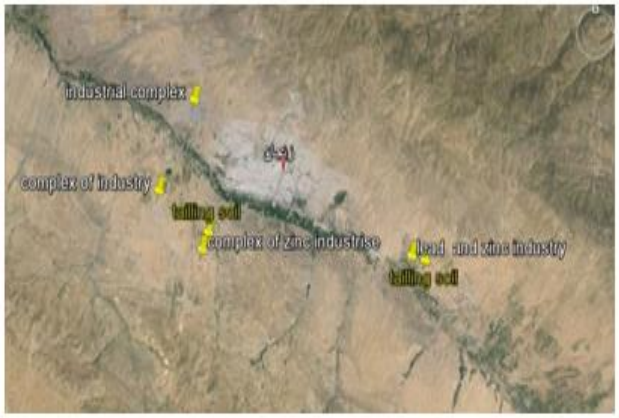

Fig.1: Zanjan map and surrounding sources of emission.

2.2. Data collection

Sampling station was selected in a high traffic region in the city center. The sampling was conducted randomly once a week with high-volume sampling device (TCR-Tecora) to collect environment PM_{10} sample. The quartz-fiber filter was used for sampling. Sampling was performed for 24 hours at a flow rate of 16.7 L/m. 96 samples were collected from June 2013 to 2015. The samples were digested by microwave digester (Sineo, MDS-10 model) based on USEPA-IO-3.1microwave method. Then ICP-OES device was used for analysis of metals including arsenic, aluminum, cadmium, calcium, chromium, copper, iron, manganese, nickel, lead, antimony, vanadium, titanium, zinc, and mercury. Parameters required for measuring metals using ICP device are shown in the following table.

Table 1: Parameters for ICP-OES operation.

Parameter	Values
RF generator (W)	1400
Plasma torch	auxiliary
Nebulizer gas	Argon
Plasma gas flow rate (l/min)	14.5
Auxiliary gas flow rate (l/min)	0.9
Nebulizer gas flow rate (l/min)	0.85
Sample uptake time (S)	240 total
Rinse time of (S)	60
Initial stabilization time (S)	Preflush:60
Measurement replicate	3
Element (λ/nm)	As below
Frequency of RF generator (MHz)	resonance frequency: 27.12 MHz
Type of detector Solid state	CCD
Type of spray chamber Cyclonic	Modified Lichte

Table 2: Concentration of heavy metals in PM10 particles in µg/m³.

Species	MIN	AVERAGE	MAX
W	1.67E+01	6.30E+01	1.00E+02
AL	1.05E-01	2.69E-01	4.85E-01
As	8.30E-05	1.60E-04	2.41E-04
Ca	4.59E-01	1.15E+00	2.19E+00
Cd	1.67E-04	1.82E-03	3.94E-03
Cr	9.72E-03	1.39E-02	2.38E-02
Cu	7.08E-03	1.04E-02	1.65E-02
Fe	3.34E-01	7.61E-01	1.29E+00
Mn	1.25E-02	3.10E-02	5.16E-02
Ni	1.09E-03	7.23E-03	2.39E-02
Pb	1.59E-02	4.76E-02	1.02E-01
Sb	1.89E-02	2.68E-02	4.36E-02
Ti	5.00E-04	2.40E-03	4.99E-03
V	4.20E-05	8.54E-05	1.32E-04
Zn	7.38E-02	1.99E-01	3.58E-01
Hg	1.45E-03	3.87E-03	9.13E-03

2.3. Health Risk Assessment

Metals such as arsenic, cadmium, chromium, nickel and lead have been selected as cancer risk metals (14). Because chromium 6 is carcinogenic and chromium 3 is safe and chromium 6 is in the air with a ratio of 1 to 6, chromium value was divided by 7 in risk calculations (15, 16, 17).

Cancer risk assessment method was calculated by the method recommended by the EPA using formulas (1) and (2) for minimum, average and maximum concentrations. The parameters used in this study are shown in Table (3). In health risk assessment with carcinogens, there is a linear relation between higher concentration of pollutants and increment of cancer risk. Health risk assessment was conducted based on the formulas (1) and (2). (18, 19, 20).

$$AD_{inh} = (C \times IR_{inh} \times ET \times EF \times ED) / (BW \times AT) \quad (1)$$

$$CANCER\ RISK = AD_{inh} \times SF \quad (2)$$

2.4. Particle emission source characterization

Positive matrix factorization (EPA-PMF_5), a multivariate receptor- based model, was used for source apportionment and characterization of the collected PM_{10} (21). A PMF model assumes that there are p factors (sources) which can be involved in a receptor site and stated with the following equation:

$$X_{ij} = \sum_{k=1}^{p} (g_{ik} \times f_{kj}) + E_{ij} \quad (3)$$

Where,

Table 3: Parameters applied in exposure assessment model.

Parameter	Definition	Unit	Children	adults
ADinh	absorbed dose of inhalation	mg/(kg×day)		
C	Consentration of metals	mg/m^3		
IRinh	Inhalation Rate	m^3/h	0.4	0.6
ET	Exposure Time	h/d	24	24
EF	Exposure Frequency	d/year	350	350
ED	Exposure Duration	year	8	35
AT	Average Time	d	70×365	70×365
BW	Body Weight	kg	20	65
SFinh	slope factor	(kg×day)/mg		

Table 4: Results for children living in the region.

METALS		con.(mg/m^3)	AD $_{inh}$(mgr/Kg.d)	SF(Kg.d/mgr)	CANCER RISK
As	Max	2.41E-07	1.27E-08	15.1	1.91E-07
	Min	8.30E-08	4.37E-09	15.1	6.59E-08
	MEAN	1.59E-07	8.37E-09	15.1	1.26E-07
Cd	Max	3.94E-06	2.07E-07	6.1	1.26E-06
	Min	1.67E-07	8.78E-09	6.1	5.36E-08
	MEAN	1.80E-06	9.47E-08	6.1	5.78E-07
Cr	Max	3.40E-06	1.79E-07	41	7.33E-06
	Min	1.39E-06	7.29E-08	41	2.99E-06
	MEAN	1.97E-06	1.04E-07	41	4.25E-06
Ni	Max	2.39E-05	1.26E-06	0.84	1.06E-06
	Min	1.09E-06	5.74E-08	0.84	4.83E-08
	MEAN	7.06E-06	3.71E-07	0.84	3.12E-07
Pb	Max	1.02E-04	5.38E-06	0.042	2.26E-07
	Min	1.59E-05	8.36E-07	0.042	3.51E-08
	MEAN	4.71E-05	2.48E-06	0.042	1.04E-07

Table 5: *Results for adults living in the region.*

ETALS		con.(mg/m^3)	AD $_{inh}$(mgr/Kg.d)	SF(Kg.d/mgr)	CANCER RISK
As	Max	2.41E-07	2.56E-08	15.1	3.87E-07
	Min	8.30E-08	8.82E-09	15.1	1.33E-07
	MEAN	1.59E-07	1.69E-08	15.1	2.55E-07
Cd	Max	3.94E-06	4.18E-07	6.1	2.55E-06
	Min	1.67E-07	1.77E-08	6.1	1.08E-07
	MEAN	1.80E-06	1.91E-07	6.1	1.17E-06
Cr	Max	3.40E-06	3.61E-07	41	1.48E-05
	Min	1.39E-06	1.47E-07	41	6.03E-06
	MEAN	1.97E-06	2.09E-07	41	8.58E-06
Ni	Max	2.39E-05	2.54E-06	0.84	2.13E-06
	Min	1.09E-06	1.16E-07	0.84	9.74E-08
	MEAN	7.06E-06	7.50E-07	0.84	6.30E-07
Pb	Max	1.02E-04	1.09E-05	0.042	4.56E-07
	Min	1.59E-05	1.69E-06	0.042	7.09E-08
	MEAN	4.71E-05	5.00E-06	0.042	2.10E-07

X_{ij} : Concentration of species J in ith sample

g_{ik} : Contribution of kth factor to the ith sample

f_{kj}: Fraction of kth factor that is species j or chemical composition profile of factor K

E_{ij}: Residual for the jth species on the ith sample

The contributions of factor (g_{ik}) and source profiles (f_{kj}) are estimated by the PMF model by minimizing the objective function:

$$Q = \sum_{i=1}^{n} \sum_{j=1}^{m} \left(\frac{x_{ij} - \sum_{r=1}^{p} g_{ik} \times f_{kj}}{u_{ij}} \right)$$

u_{ij}: Uncertainty of species Jth of the sample

Q: Objective function a critical parameter for PMF model.

The main aim of EPA PMF is to minimize the sum of squares of standardized residuals or Q. In EPA PMF_5, two versions of Q are applied and displayed for the model runs .

1) Q_{true} is the goodness-of-fit parameter calculated including all points.

2) Q_{robust} is the goodness-of- fit parameter calculated excluding points not fit by the model which are the samples with uncertainty-scaled residual greater than 4. The difference between these two Qs is the degree of the impact of the data points with high-scaled residuals.

The main step in PMF modeling is determination of the uncertainty for each of the measured data. In PMF, the weight of missing and below-detection-limit data would decrease with appropriate uncertainty (21). PMF can decrease the weight of the missing data and values below

the detection limit and low S/N (signal to noise), using robust mode (22). The S/N calculation in PMF_5 has been revised which is described in the EPA PMF_5 user guide in details. In order to reduce the weights of the species with low S/N in the solution, the species with S/N ratio less than 1 were categorized as weak variables (23). Most of the species have S/N higher than 8, the reason for this high S/N is that the species were analyzed in PM_{10}, hence, most of the concentrations were high.

The uncertainties of the species were determined according to the recommended methods and equations in the EPA-PMF_5 manual (24). The data with concentrations below MDL, were substituted by 1/2 MDL and 5/6 MDL was used as the corresponding uncertainty value (25). If the concentration was greater than MDL, the following equation was used (23).

$$UNC = \sqrt{(Error\ fraction \times concentration)^2 + (MDL/2)^2}$$

As described by Nooris et al (2014) and detailed in Paatere et al (2014, 2015), EPA PMF has 2 main error estimation methods: displacement (DISP), Bootstrapping (BS), as well as a useful tool for rotation that is named F peak (23, 26).

DISP includes the effects of rotational ambiguity and does not affect random errors in the data. BS includes the effects of random errors and partially-rotational ambiguity. In this study, the number of factors was determined on the basis of variations in values of Q_{true} and Q_{robust} and $Q_{expected}$.

3. Results and Discussion

3.1. Cancer Risk Assessment

Cancer risk assessment calculation results are shown in Tables 4 and 5. According to the EPA recommendation (27). In case that risk index is less than 10^{-6} emission source is negligible. If the amount is larger than 10^{-4}, the intended pollutant is extremely harmful to human health and if it ranges between 10^{-4} to 10^{-6}, the index is accepted, however, a risk management to reduce the and can reduce the influence of extreme values carcinogens would be necessary. The results showed that from 30 calculated risk indices of five heavy metals in three concentration ranges and for both children and adult groups, risk index was higher than 10^{-6} in eleven cases. It was observed that the highest risk was related to chromium in maximum concentration equal to (1.48E-5). Since 11 indices were between 10^{-6} and 10^{-4}, the source apportionment of pollutants is necessary to determine the emission source of the metal for further management and emission control measures.

3.2. Source characterization

The PMF_5 with 96 samples in which 15 heavy metals were measured was implemented for four-factors, five-factors and six-factor emission status.

The conditions for all modes have been selected based on the following status. PM_{10} was selected as weak due to its lower level of arsenic and vanadium and enrichment in S/N.

In four-factor run, factor 1 is associated with traffic and fuel combustion which contributed to 41.5% of emission. The presence of mercury, vanadium and titanium shows the species which are entered into the atmosphere from fuel combustion. Stationary and mobile combustion sources are the origin of the species and release of factor 1. Location of sampling station in the center of high traffic city would justify this condition (28, 29, 30).

The second factor with a contribution of almost 11% contains different species which mostly have industrial origin. The presence of nickel, chromium and copper has associated this factor with industrial activities and small factories within the city. There are electroplating, knife and copper workshops in Zanjan for making copper containers both decorative and utilizable which could be the source of air particles. The third factor with emission share of almost 12% is related to dominant lead and zinc species. The existence of lead and zinc industries, waste and tailing soil piling up and filter cake waste of lead and zinc

factories surrounding Zanjan are the origin of this emissions in the region. Waste soil of lead and zinc factories are stacked in two points of Zanjan which are shown on the map (Fig. 1). One of these areas is located in the vicinity of the specialized zinc complex which is in the southeast of Zanjan.

Filter cake waste and soils of all factories in specialized zinc complex which are almost 100 units are daily discharged. The soils which contain high levels of lead, zinc and other heavy metals are piled up at this point without any environmental considerations. Now more than 3 million tons of soil has been stacked in this area.

Similarly, another area is located in East of Zanjan, the in vicinity of lead and zinc plant in which the waste and soils of aforesaid factory are discharged. Factor 4 with emission share of 35.7 % is related to re-suspended surface soil particles.

The presence of cadmium, aluminum, calcium and iron is the main reason for the association of these factors with soil particles. This element has shown its highest part in this factor. Cadmium is one of the specific species which has shown its maximum part in this factor. In other words, this element has been increased with increment of species related to soil. The main reason is indiscriminate use of chemical fertilizers in the surrounding areas on agricultural lands which has significantly increased. The study by Zanjan Agricultural Research Center indicated that concentration of cadmium in agricultural soils of lands surrounding Zanjan is exceeding the standard limit (31).

The main identified factors in runs with five and six factors are surface soil, fuel combustion, industrial activities and, lead and zinc tailing soil. Other identified sources are not interpretable with regard to physical observation of the study area.

As an overall result of these solutions it could be said that with regard to Q/Q_{exp} values which was 0.6290 in four-factor solution, 0.4414 in five-factor solution and 0.3318 in six-factor solution, the four-factor solution brings about better results while fuel and traffic, suspended open soil, lead factor and one of the major sources of particulate and zinc, waste and industrial activities are the most important factors identified in this area. Furthermore, we could compare the solutions based on the error estimation parameters.

The results of the base runs were tested by error estimating tools of bootstrap and displacement moods. These tools showed that four-factor solution is the best answer with minimum error.

3.3. Main Source of Emission and Percent of Affective Heavy Metals in Cancer Risk

In four-factor solution as the best solution, share of the intended metal in cancer risk could be seen in each of the pollutant factors in Figures (2) (3) and (4). Cadmium with the share of 83.1% released from factor 4 exhibited cancer risk in maximum concentration for children and in maximum and average concentration for adults. It was associated with suspended open soil.

Chromium had also cancer risk in three concentration ranges for children and adults and is released from factor 1 with share of 31.7% and is associated with fuel and combustion. Nickel which has cancer risk for children and adults in maximum concentration range is released from factor 2 with share of 84.3% and is associated with industrial activities.

4. Conclusion

The cancer risk assessment of metals including arsenic, cadmium, chromium, nickel and lead concentrations in three maximum, minimum and average ranges was conducted in two groups of children and adults living in the area. Results for children and adults living in the area indicated that cadmium, chromium, and, nickel have the main role in cancer risk in the studied area. According to the EPA recommendations planning should be conducted to reduce cancer risk in this area. Source characterization of all heavy metals using PMF_5 model showed that fuel combustion, waste dump soil of lead and zinc industries, re-suspended open soil and activities are the main sources of PM_{10} in the studied area. It was also

found that more than 83.1% of the cadmium is released from re-suspended open soil, 31.7 % of the chromium is released from fuel combustion and 84.3% of nickel is released from industrial activities. To reduce the risk of cancer, management and pollution control of these sources especially cadmium in the soil should be prioritized.

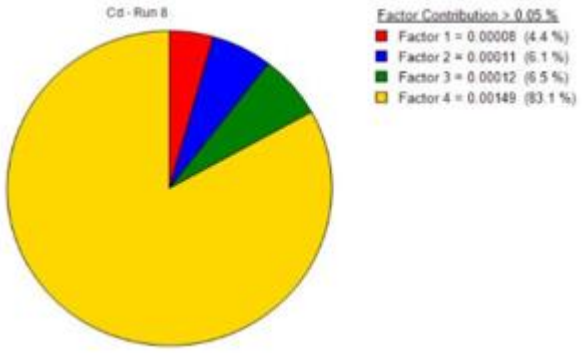

Fig. 2: Share of cadmium in emission sources.

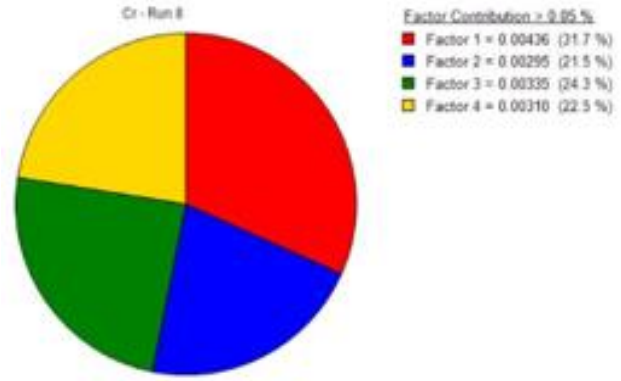

Fig. 3: Share of chromium in emission sources.

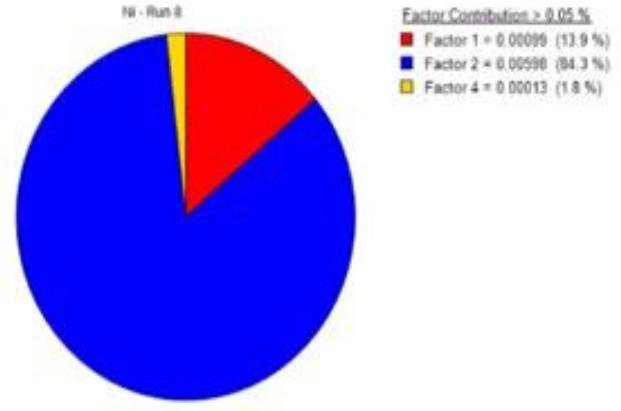

Fig. 4: Share of nickel in emission sources.

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