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Chemical Characterization of Particulate Matter at Traffic Prone Roadside Environment in Agra, India

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ABSTRACT: In the present study, size-segregated samples of PM were collected from urban and semi-urban traffic junctions in Agra, India. PM samples were collected during the monsoon season (July to September 2015) using Grimm portable aerosol Spectrometer. The recorded mean concentration of PM_{10} at urban site was 137.09±61µg/m^3 and at semi-urban site was 270.14±21µg/m^3, which were higher than the suggested limits by WHO and NAAQS India. Mean concentrations of $PM_{2.5}$ were 41.45±40µg/m^3, 48.88±34µg/m^3 at the urban and semi-urban site, respectively. Whereas, mean concentrations of $PM_{1.0}$ were 30.35±64µg/m^3, 12.64±4µg/m^3 and $PM_{0.25}$ were 0.06±0.05µg/m^3, 0.17±0.06µg/m^3 at the urban and semi-urban site, respectively. It was estimated that $PM_{10,}$ $PM_{2.5}$ and $PM_{0.25}$ values were higher at semi-urban site than urban sampling site but in case of $PM_{1.0}$ concentrations were higher at urban site. The surface morphology of $PM_{2.5}$ was studied using Scanning Electron Microscope (SEM). The results show flaky, branched chain like aggregates of carbon bearing spheres at the urban site while cluster, branched, spherical and fluffy particles at semi-urban site. The presences of carbonaceous particles were enhanced due to use of fuel combustion. Chemical analysis was done using ICP-AES. Concentrations of Zn and Cu were found higher while Ni was least in comparison to other metals. Elemental composition present in $PM_{2.5}$ was used to calculate the health risk assessment to identify the possible health effect on human health, hazards quotient (HQ) values was found higher as Ingestion to inhalation pathways while ECR values found higher as Cr(VI)>Ni>Pb for both medium (Air and Dust).

Keywords: *$PM_{2.5}$, Heavy metals, APM-550, SEM, Hazard Quotient.*

INTRODUCTION

Air pollution can be defined as any malicious substance present in the ambient air which imparts deleterious effects on the health and environment. Vehicular emissions are one of the prime sources contributing to a wide range of particulate matter (PM), hydrocarbons, carbon monoxide, oxides of sulphur and oxides of nitrogen. Airborne microscopic solid or liquid particles are generally described by the term Particulate Matter, which are further classified depending upon their size. Particles ranging ≤10 micrometers in diameter are referred as PM_{10}. Particles ≤2.5 micrometers in diameter as $PM_{2.5}$ while Particles ≤0.1 micrometers in diameter are referred as ultrafine particles (UFPs) (Khillare & Sarkar, 2012; Pandey et al., 2014). The deterioration of the air quality in India has been significantly escalated due to a peak increase in motor vehicles and small-scale industries in the past few years

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(Roy et al., 2016). Epidemiological studies confirmed that this increase in air pollution has resulted in increased levels of respiratory disorders and mortality (Yin et al., 2010). In worldwide during 2012, 3.7 million deaths were recorded which occurred due to ambient air pollution (WHO, 2014). Many studies including those of (Chan *et al*., 2009; Colbeck *et al*., 2011) have been carried out to determine mass concentrations of different fractions of PM in heavy traffic areas along the roadside. Humans are left to risks of high respiratory and cardiovascular problems due to the presence of high concentrations of fine particulate matter (PM_{10} and $PM_{2.5}$) (Lee et al., 2011). Myocardial infarction, cardiovascular diseases, lung cancer, and other respiratory diseases are causes of these small particles (Woodruff et al., 2006). Izhar et al., 2016 studied the excess cancer risk in IIT-Kanpur, India and it was found that Ingestion is the major exposure pathway for heavy metals. Several epidemiological studies (Schwartz et al., 1996; Pope, 2000) have suggested a statistical association between health effects and ambient fine particle concentrations, prominently for the particulate matters consisting of sub-micron sizes as they have the tendency to penetrate deep into the alveolar region of the lungs and remain there for long period of time and then eventually enters the circulatory system of the blood. Many inclusive studies have been put forth on the toxicity of metals present in air and soil in various parts of Europe and America (Popov et al., 2014; Diaz & Dominguez 2009; Zhao et al., 2014), while a sparse study is done in Asia, prominently in the countries like China and India which are rapidly developing. Cao et al., 2012 studied that Noncarcinogenic assessments of health risks for Pb, Cd and Cr (HI>) showed high deleterious impact chances on children rather than adults as they were under the safe limits. Also, decreasing order of cancer risk was observed in adults and children for the metals Cr(VI)>Cd>Ni>As>Pb. For adults and children, it was found to be higher than the permissible limits of (10^6). It has been studied that the particle sizes of particulate matters have a significant influence on the human risk assessment. From most of the studies conducted on $PM_{2.5}$ and their sources (Du et al., 2013; Zhou et al., 2014); a very few have been addressed on the Heavy metals bound to particulate matters and their related health risks (Hua et al., 2012; Du et al., 2013). Also, very few studies have been conducted on soil heavy metals toxicity and their associated health risks (Dumitrescu et al., 2012). Metal contaminated soil contributes to one of the most environmental pollution and exposure of humans to them and other related problems (Dao et al., 2010). Recent studies demonstrated on the human health risk assessment of metals (Pena-fernandez et al., 2014; Abechi et al., 2010), Parveen et al., 2018, determined the health risk caused by carcinogenic and non-carcinogenic effects of metals and metalloid on the health of children as well as on adults ensuing the methods implemented by USEPA. In indoor and outdoor environments, complexity in the size distribution of metals in airborne particles in Agra was done by Rohra et al., 2018 where the study revealed that exposure from PM in different sizes through inhalation was not negligible causing health risks. Furthermore as the degree of toxicity is determined by the size of particles, the obtained results highlighted that finer sizes possessed higher non-carcinogenic risks (~2.5 times) and carcinogenic (~14.8 times) risk than coarser particles. The study proposed that health risk due to exposures from PM in different particle sizes is not negligible. Pena-fernandez et al., 2014 carried out as study on human health risk assessment for metals and metalloids at Alcala de Henares, Spain in urban environments. Du et al., 2013 calculated Health Risk Assessment from Heavy metals in road dusts at urban parks of Beijing, China, where the health risk was estimated by carring out Hazard Quotient Index (HQ) and Health Index (HI). The

output of the study revealed that there were three major exposure pathways in people viz., Ingestion, dermal Contact and through Inhalation. The study indicated that HQ and HI lower than the safe level (=1). On the contrary, HI values for children were found to be higher in children compared to adults, Thus the present study, undertaken is focused to give an insight of the concentration levels of PM at two different traffic junction sites of Khandari, Agra (Urban sampling site) and Iradatnagar (Rural traffic site) of Agra. The major objectives for carrying out the study are 1) To provide quantified information based on the concentration levels of the fine particulate matters at different traffic junctions. (2) To determine concentration levels of metal constituents in $PM_{2.5}$. (3) To carry-out health risk assessment for carcinogenic and non-carcinogenic metals through three different exposure routes viz., ingestion, inhalation, and dermal contact pathways conspicuously. This putforth study will help city planners and the governing authorities in the developing world, to know about the status of pollutants and their concentrations at traffic junctions. Thus, the results of this study will give a better insight of the air quality that would give an idea to health officials and to the general public elicit a sight of awareness regarding the toxicity of metal bound particles with a better understanding to control them.

MATERIALS AND METHODS

Agra (27˚10' and 78˚2' E, 169 A.S.L) also referred as the city of 'Taj Mahal' is present in Uttar Pradesh state in the North central part of India and is one of the tourist attraction place in India (Fig. 1). The city is situated along the bank of river Yamuna.

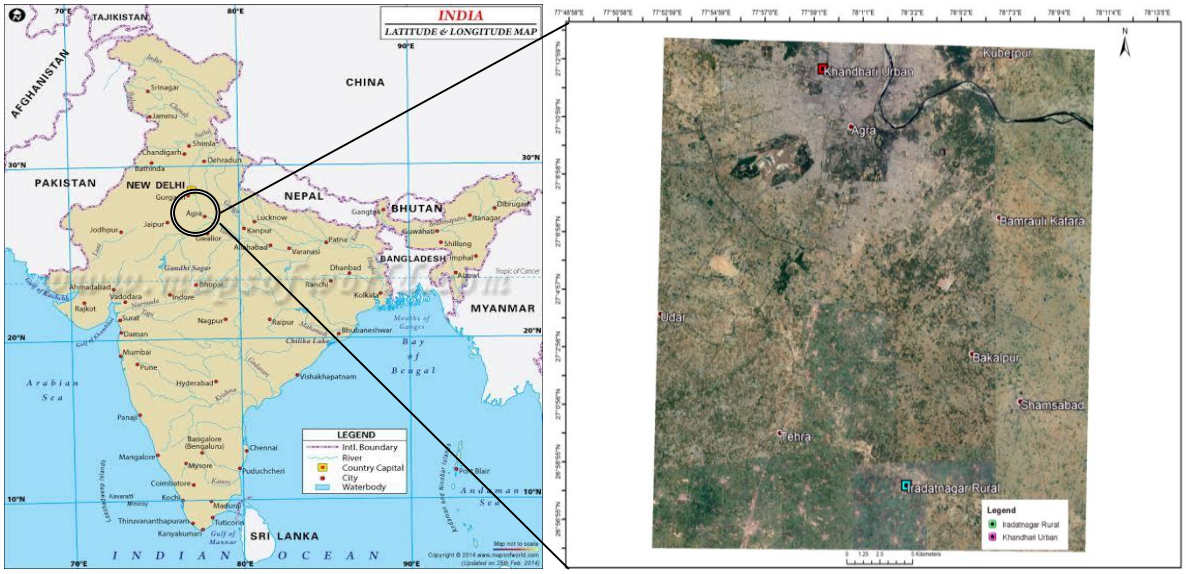


Fig. 1. Map of India showing sampling sites.

The city is located almost 200 km to the South from New Delhi (Capital of India). Accordingly it is also considered as the fourth most populated city in state with 4027 persons/km^2 density having a total population ranging 44,18,797 in the year 2011 as per Directorate of Census operation, 2011. The city has a semi-arid tropical type of weather as it is surrounded by the Thar Desert of Rajasthan. Meteorologically the city has three prominent seasons (summer, monsoon and winter). The start of summer is marked from the month of April to June while Monsoon from July to September followed by winter in the month of October to February. The temperatures in summer are between 25.6°C to 47°C with a relative

humidity ranging between 16.6% and 76.4%. Winters are with falling low temperatures marked up to 4°C to 26°C. Rainy season are hot and humid where temperature ranges between 22°C to 37°C while the relative humidity ranges between 57% and 89% with a wind speed varying from 0.1 to 19.8 km/h with the highest values recorded during summer and monsoon and lowest in winter. The city is also connected with four national highways namely; NH-2, 3, 11 and 93 which passes through the city making the condition even outrageous. Due to high traffic and extreme emission loads the city suffers high atmospheric pollution. Sampling sites are shown in Map of Agra city (Fig. 1) 8 hrs monitoring from 9 A.M. to 5 P.M. was done with the help of APM- 550 and 24 samples were collected in the month of July to September 2015. Collection of PM samples was carried out at Khandari crossing (27.2014^{o} N,77.9915 ^{o} E), Agra (Urban site), which is one of the busiest roads with National highway-2 (Agra-Delhi) and at Iradatnagar town (26.9665^{o} N,78.0438 ^{o} E) (Semi-Urban site) which was located 32 kms away from Agra city. The Site witnessed normal traffic, unpaved sidewalks with green grass and plants. Sampling site is covered with agricultural farms agricultural practices like field plowing, irrigation and biomass burning are done at this site. Construction activity like offloading of brick and cement, cutting and pilling of iron roads for frame work are the major activities carried out on this site.

Road dust was collected by sweeping with brooms in a polyethylene zip lock bag and it was desiccated at room temperature for a week. Samples were sieved through a 2-mm sieve to remove debris (Pena-Fernandez et al., 2014; APHA et al., 1998). $PM_{2.5}$ samples were collected using APM-550 (High volume air sampler) which is deliberate to lock fine particles (between 2.5-10 microns) at constant flow rate 16.67 L/min (recommended by USEPA). Oil less rotary pump is used to maintain suction pressure. Silicon oil immersed 37-mm glass fiber (GF/A) paper was used in the WINS impactor to trap the coarse size particles. After every 72 hrs. of sampling filter must be changed or when the filter gets clogged or depending as per the operator's judgment (Chow & Watson, 1998). 47-mm-diameter polytetrafluoroethylene (PTFE) filter papers used to collect $PM_{2.5}$. Periodic cleaning of sampler was done to make it dust free so that the dust on the sample should not be counted along with the concentration of the sample. Real-time monitoring of $PM_{10,}$ $PM_{2.5,}$ $PM_{1.0}$ and $PM_{0.25}$ was done with the help of GRIMM 1.109 aerosol spectrometer at constant flow rate (1.2L/min) of pump. The physical principal of GRIMM is light scattering outcomes are united to get an absolute mass assortment allotment with 31 channels.

All Glassware which were used in experimental set up were washed with acid. Analytical Grade Reagent Merck, Darmstadt, Germany used to prepare the entire standard and double distilled water was used in the experiment. Approximately 1g triplicate samples were weighed on analytical balance and then transferred into digestion tubes. Aqua-Regia ($1HNO_3$:3HCl) method was used to extract the samples on hot plate at 30-35^{0} C in 50-ml measuring beaker (Demirezen & Aksoy, 2004; Abechi et al., 2010; Tug & Duman 2010). Pre-washed Whatman filter paper was used to filter the extracted samples and at the end extract was diluted up to 40ml de-ionized water and stored at 4^{o} C in refrigerators. Six heavy metals viz., Cu, Cd, Cr, Zn, Ni and Pb were analyzed by using Inductively Coupled Plasma-Atomic Emission Spectrometer. The information regarding the minimum detection limit and the accuracy (%) of the elements is provided in Table 1. The recovery range varied from 96-98%. The minimum detection limit was 10 ppb and all samples are taken in triplicates.

Table 1. Quality assurance/Quality control of ICP-AES for the analysed metals.

Metals	Cu	Zn	Cr	Ni	Pb
Accuracy (%)	95%	95%	95%	95%	95%
Precision(%) at 0.2mg ml−1	< 1% RSD	< 1% RSD	< 1% RSD	< 1% RSD	< 1% RSD

The risk assessment was calculated to find out the magnitude and the type of exposure compared to the chemical elements which are present in air and soil/dust. Exposure assessment is the determination of the frequency, magnitude, duration and exposure pathways (Dumitrescu et al., 2012). According to USEPA, (2004a) Methodology exposure magnitudes are determined by estimation or measurement by the concentration/quality of the chemical element during a certain period. During the assessment, different pathways, population, exposed were identified. Exposure evaluation is done for each identified pathway. The main exposure pathways are following: (1) Ingestion (2) Inhaling and (3) Dermal contact (Zhao et al., 2014). In air/soil studies, considering that the chemical elements like Ni, Cr, Pb, Zn, Cd and Cu were deposited in the soil through different activities which were only ingested through food and oral mucus, and exposure through dermal contact have been considered for the human health risk assessment (Perveen et al., 2018).

Exposure through Ingestion, inhalation and dermal contact trail in the form of exposure to the average daily dose (ADD) (mg/ kg/ day) of the pollutant, through different paths was estimated using Eq. (1), (2) and (3) According to USEPA Exposure Factors Handbook (1997). Exposure factors for dose models are given in Table 2 (USEPA 1989, Parveen, et al., 2018).

Table 2. Exposure factors for dose models

Factors	Definition	Unit	Value of Average daily dose	
			Child	Adult
c	Concentration of the contamination in dust / air	mg/kg	-	-
$R_{ing.}$	Ingestion rate of soil	mg/kg	200	100
EF	Exposure Frequency	days/year	350	350
ED	Exposure Duration	Years	6	24
BW	Average bodyweight	Kg	15	55.9
AT	Average time	days	365×ED	365×ED
CF	Conversion factors	Kg/mg	1×10^{-6}	1×10^{-6}
$R_{inh.}$	Inhalation rate	m3/day	5	20
SA	Surface area of the skin that contacts the dust	cm^2	1800	5000
SL	Skin adherence	Mg/cm^2	1	1
PEF	Particle emission factors	M^3/kg	1.32×10^{6}	1.32×10^{9}
ABS	Dermal absorption factors (chemical specific)	-	0.001	0.001

$$ADD_{ing} = \frac{c \times Ring \times CF \times EF \times ED}{BW \times AT} \quad (1)$$

$$ADD_{inh} = \frac{c \times RInh \times EF \times ED}{PEF \times BW \times AT} \quad (2)$$

$$ADD_{derm} = \frac{c \times SA \times CF \times SL \times ABS \times EF \times ED}{BW \times AT} \quad (3)$$

HQ (Hazard Quotient) is based upon non-cancer risk which can be calculated by using the given formula (USEPA 1989; Parveen et al., 2018).

$$HQ = \frac{ADD}{RFD} \quad (4)$$

Here, RfD is an assessment of highest permissible risk on human population through daily exposure, considering a sensitive group during lifetime. If ADD value found lower than R*f*D, there would not be any adverse health effect; whereas, the ADD value found higher than RfD, it shows adverse health effect (USEPA 1993). Overall HQ>1 indicates unfavorable health effects on human and HQ<1 reveals the safe level (USEPA 1986).

ECR's prediction is done in human life to detect the possibility of cancer which increases over time; ECR is calculated by using following approach. (USEPA 2011; Hu et al., 2012)

$$ECR = \frac{c \times ET \times EF \times ED \times IUR}{AT} \quad (5)$$

where, C is the pollutant's concentration (µg/m^3), ET is exposure time which was 8h/day in this study. IUR is inhalation unit risk (µg/m^3)$^{-1}$, AT is the average time for carcinogens (70 year × 365 days/year × 24h/day), Carcinogens are non-thresholds, carcinogens are substances that cause cancer and the safe amount of carcinogen is "Zero". The data of carcinogens and IUR values of metals are taken from the USEPA data base for IRIS (Integrated information risk system) (Khanna et al., 2015). USEPA not provided the reference value of dermal exposure and ingestion, only carcinogenic risk of metals through inhalation pathway is considered. If the risk assessment ranges between 10^{-6}-10^{-4}, contamination does not cause carcinogenic exposure risk (Hu et al., 2012).

RESULTS AND DISCUSSION

Mean particulate concentrations were measured at two different traffic junction's urban and semi-urban sites which are shown in Figure 2. PM_{10} (270.14µg/m^3), $PM_{2.5}$ (48.88µg/m^3), $PM_{0.25}$ (0.17µg/m^3) concentrations were higher in semi urban site in comparison to urban site (139.09µg/m^3), (41.45µg/m^3) and (0.06µg/m^3), respectively.

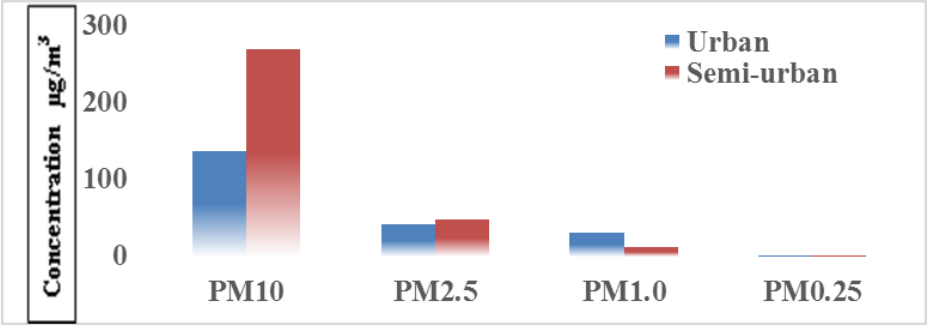


Fig. 2. Particulate Matter concentrations at urban & semi-urban traffic junctions.

India is largely dependent on biomass to fulfill their need of fuel for cooking, heating and other activities (Venkataraman et al., 1999; Ram et al., 2008). Other than biomass burning, India has large demand of gasoline and diesel due to steadily increasing vehicular market in India. Hence, vehicular emissions and biomass burning are mainly responsible for higher concentrations of PM at urban site. PM concentrations at semi-urban site were found elevated due to use of heavy diesel vehicle, such as tractors, generators for irrigation activities, incineration of solid waste, coal and wood burning. Construction activities and re-suspended road dust/refuse dust due to unpaved roads are the main cause to this elevated PM concentration (Schneidemesser et al. 2010; Sahu et al., 2018; Gautam et al., 2016). The concentration of $PM_{1.0}$ was higher at urban site (30.35µg/m^3) comparison to semi-urban site (12.56µg/m^3) this may be validated due to higher vehicular emissions and industrial emissions (nearly 3-5 km away). It may be augmented by anthropogenic emissions by a variety of

local sources like biomass burning in various small-scale industries (petha, iron casting, electroplating etc.). Average concentration of $PM_{2.5}$ and PM_{10} compared with 24-hrs guideline of WHO (World Health Organization, 2007) of $PM_{2.5}$ (25µg/m^3) and PM_{10} (50µg/m^3), in this study it was found that $PM_{2.5}$ and PM_{10} concentration exceeded about 2 times and 8.6 times respectively. Results of this study are compared to other studies carried out over India (Table 3). Meteorological parameters play a major role in governing the attentiveness disparity of PM. Meteorological parameters were acquired from the meteorological station (Met-One) instrument located at the Uttar Pradesh Pollution Control Board (UPPCB), Agra's site, which recorded the mean atmospheric temperature (AT) of 30.4±2.84ºC, relative humidity (RH) of 69.5±9.56%, wind speed (WS) of 3.1±1.68m/s and wind direction (WD) of 208.8±74.05^0 during the study period. PM concentration depends on the source/sink mechanism which is generally influenced by metrological conditions.

Table 3. Comparison of PM concentrations in different parts of India.

Place	$PM_{0.25}$	$PM_{1.0}$	$PM_{2.5}$	PM_{10}	References
Urban site	0.06±0.05	30.35±64	41.45±40	137.09±61	Present study
Semi-urban site	0.17±0.06	12.64±4	48.88±34	270.14±21	Present study
Hydrabad	-	-	72.6±18	105.2±28.6	Guttikunda and Kopakka (2014)
Raipur	-	-	115±36	426±68	(Giri et al. 2013)
Pune	-	-	72.3±31.3	113.8±51.6	Yadav and Satsangi (2013)
Agra	-	93±22	125±42	219±72	Massey et al. (2012)
Kanpur		31.1±13.7–199±66.0	-	-	Chakraborty and Gupta (2010)
Agra	-	-	91.2±50.4-104.9±47.1	148.4±69.4-154.2±68	Kulshrestha et al., (2009)

Fig. 3 and Fig. 4 show the metallic concentration of $PM_{2.5}$ at both sampling sites. Concentration of Cd was found below the detection limit for both medium (Air and Dust). Trend of metals found at urban site for air samples were followed as Zn (0.0035µg/m^3) > Cu(0.00018µg/m^3) > Cr(0.00063µg/m^3) > Pb(0.0004µg/m^3) > Ni(0.0001µg/m^3), whereas at semi-urban site the trend was observed as Zn (0.0015µg/m^3) > Cu(0.00069µg/m^3) > Pb(0.0006µg/m^3) > Cr(0.00025µg/m^3) > Ni(0.0002µg/m^3). While for dust samples the values of metals for Urban Site were Zn(38.74mg/kg) > Cu(18.13mg/kg) > Pb(10.42mg/kg) > Cr(7.83mg/kg) > Ni(2.99mg/kg) whereas for semi urban site it were observed as Zn(34.8mg/kg) > Cu(7.07mg/kg) > Pb(6.9mg/kg) > Ni(4.15mg/kg) > Cr(2.29mg/kg). The sources of Cu, Zn, Pb, Cr and Ni are vehicular exhaust, industrial emission, fossil fuel combustion and road dust (Celo et al., 2010; Lee & Hieu 2011).

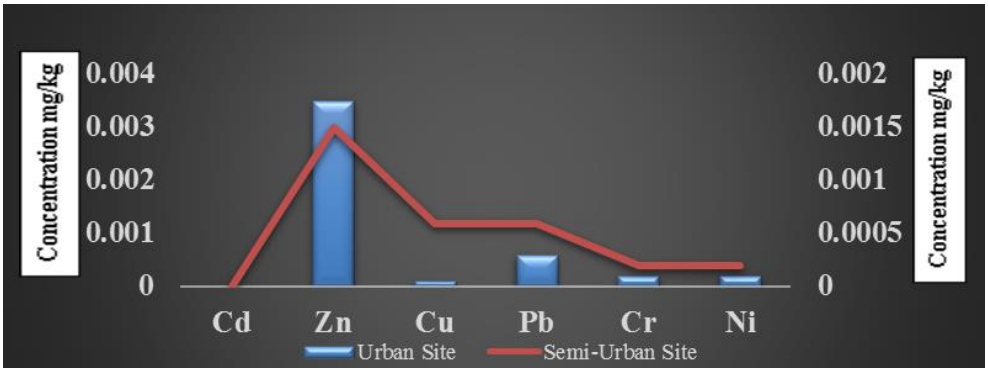


Fig 3. Comparison of Metals in Air at urban and semi-urban site.

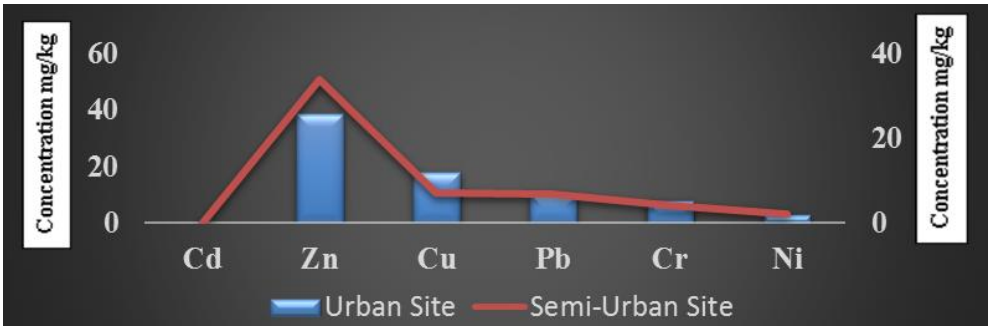
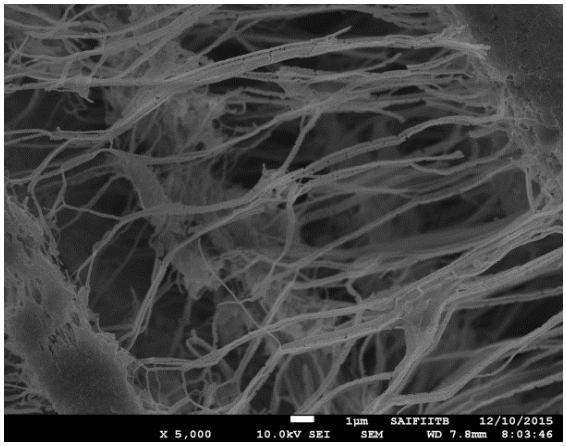
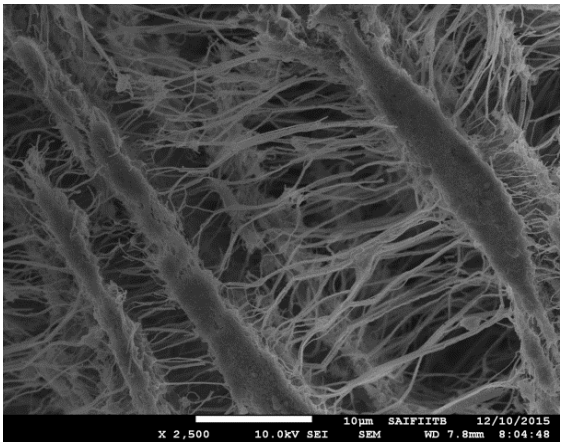


Fig 4. Comparison of Metals in dust at urban and semi-urban site.

Zn is also associated with brake wear and tire wear and it is also used as a vulcanization agent in tires, are the most likely sources, particularly associated with oil spills on road surface (Jaradat & Momani 1999). Non-exhaust emissions from vehicular sources also result in release of metallic elements, (e.g. wearing of tires, brakes and other parts of vehicles) (Sternbeck et al., 2002; Adachi & Tainosho 2004). Zn and Cr attributed to cement used for construction which contains these metals. Cr is present in steel and cutting tools. Ni and Cu are found to have higher concentration in cement and clay. Cement and exhaust of vehicles are the main source of Pb at urban site and combustion of diesel fuel (Razanica et al., 2014). pH of soil was also measured and it was found highly alkaline at both sites. Urban (7.89) soil was highly alkaline in nature in comparison to semi-urban (8.11) site. This is the obvious as the roads are uncemented/unpaved at semi urban site which enhance re-suspension of the road dust at ground level. The sampling site in our study was near ground sampling located close to road intersection that enables PM collection and metal concentration in air at semi-urban sampling site. The morphology of $PM_{2.5}$ collected from traffic sites indicates a set of carbonated substances (Fig. 5: a, b) (Fu et al., 2006).



5 (a)

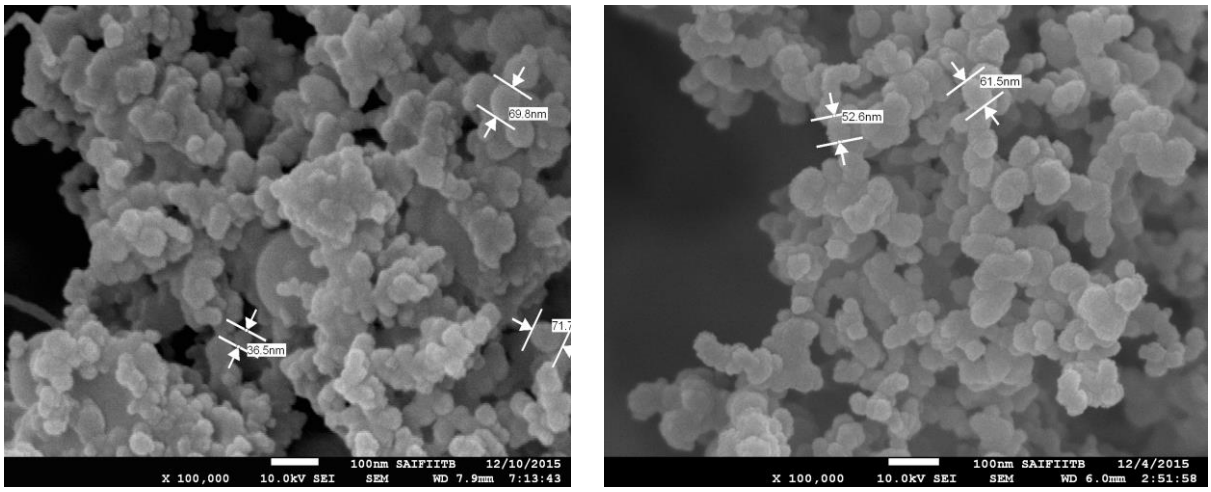


5(b)

Fig. 5. (a) and (b)-Blank Quartz fiber filter

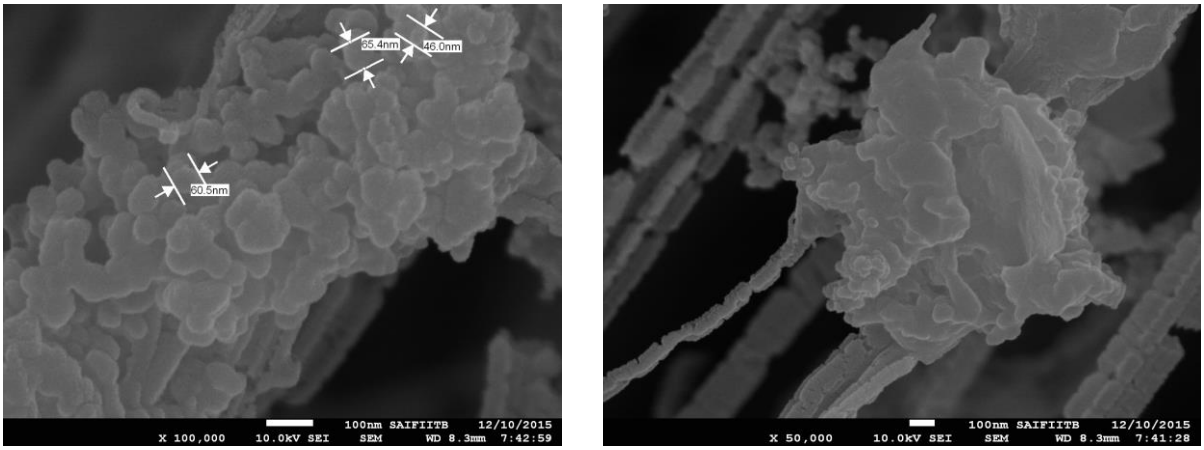
Blank quartz fiber filter itself contains silica, which is identified by the presence of Si and O particles. Spherical to roughly spherical particles of carbon were identified at both sites along with the presence of O and Cl. These carbon rich particles indicate the presence of bio-fuel burning. High temperature burning processes like burning

of fuel, coal, wood and garbage at the urban site produced clustered and branched carbonated particles (Bang et al., 2004). Therefore, it can be said the traffic junction site vehicular activities and garbage burning were the main contributors in $PM_{2.5}$ chemical composition. On further calculating the combustion process, groups of amorphous branches of the amorphous flurries obtained by the diesel excreta (diesel particulate matter) were also observed. Various researchers also acknowledged this phenomenon of carbon chain agglomerates derived from fossil fuels combustion (Murr & Bang, 2003; Sachdeva & Attari, 2008) and reported that the morphology of carbon particles arising from the combustion process fuel and its burning condition. (Posfai et al., 2010; Touolwa et al., 2010). It differs from soot chains to complex structures, there branched clusters made of interconnection of hundreds of carbonaceous spherules which attach collectively through a blend of adhesive surface forces and particle coalescence which occur at high temperature during combustion (Pachauri et al., 2013). Analysis of individual particles collected from urban and semi-urban sites was differentiated into two types of particles: carbon rich particles and mineral dust. Some spherical shaped particles of Al-Si-O were also noticed in the SEM images which indicated the presence of fly ash. In our study, the concentrations of these particles of metals such as Ni and Pb have been confirmed by ICP-AES. This indicates that the origin of these particles is likely to be from anthropogenic activities, the process of small industries around this region shows the emission of vehicles emission. The results show that aluminosilicate particles obtained from dust/soil are mainly made of C and Al oxide (Cong et al., 2010). Aluminum is the most common source of particles; the crystal originates from windblown dust. In the major metal particles there are marks of Cr, Mn and Ni with Fe, Si and O because these particles are like their rough texture (Lu et al., 2008; Satsangi & Yadav, 2014) Therefore, the common source credited to these metals is re-suspension of road dust due to vehicular activity. Ni, Cr are found rich in wear and tear force of tires, oil burning, abrasion of mechanical parts of road vehicles and oil lubricants. Earlier study by Adachi & Tainosho, 2004 has also reported that anthropogenic metal elements are usually embedded into re-suspended dust particles. The results indicate spherical, cluster and flaky appearance of particles was found in urban site (Fig 6: a,b) whereas reticular, branched chain like aggregates and fluffy appearance was found at semi-urban site (Fig 7: a, b).



6 (a) 6 (b)

Fig 6 (a), (b). SEM Micrograph of $PM_{2.5}$ at urban Site.



7 (a) 7 (b)

Fig 7 (a),(b). SEM Micrograph of $PM_{2.5}$ at semi-urban Site.

Difference of ADD for Ni, Cu, Zn, Cr, Pb and Cd in $PM_{2.5}$ at all three dissimilar exposure pathways including ingestion, inhalation and dermal exposure are calculated. Table 4 and Table 5 show the results of ADD in air and dust samples respectively at urban and semi-urban site. The average daily dose through different exposure pathway for child follows the trend as $ADD_{ing}>ADD_{inh}>ADD_{der}$,whereas for adult the trend was observed as $ADD_{ing}>ADD_{der}>ADD_{inh}$ in both air and dust sampling site. On comparing our results with Perveen et al., 2018, we found the similar trend. Results of this study show that great amount of heavy metals enters human body via ingestion. Maximum ADD concentration of Ni, Pb and Zn were observed in comparison to other metals in both (air and soil).

Table 4. Average daily dose in three models (Ingestion, inhalation and dermal) in air sample at urban and semi-urban site.

		Cu			Zn			Cr			Ni			Pb		
Urban site		**Min**	**Max**	**Mean**	**Min**	**Max**	**Mean**	**Min**	**Max**	**Mean**	**Min**	**Max**	**Mean**	**Min**	**Max**	**Mean**
ADDing µg/(g.d)	Child	1.53E-09	3.19E-09	2.39E09	1.53E-09	3.19E-08	1.22E-08	2.17E-09	3.19E-09	2.55E-09	1.40E-09	1.15E-08	4.15E-09	4.47E-09	7.41E-09	6.36E-09
	Adult	2.91E-10	8.75E-10	4.89E-10	2.05E-10	4.28E-09	1.63E-09	2.91E-10	4.28E-10	3.43E-10	1.88E-10	1.54E-09	5.56E-10	6.00E-10	1.54E-09	9.90E-10
ADDinh µg/(g.d)	Child	2.90E-11	6.05E-11	4.54E-11	2.90E-11	6.05E-10	2.31E-10	4.11E-11	6.05E-11	4.84E-11	1.40E-09	1.54E-08	4.15E-09	4.47E-09	7.41E-09	6.36E-09
	Adult	3.11E14	5.45E-11	1.37E-11	3.11E-14	6.49E-13	2.48E-13	5.45E-14	8.20E-14	7.09E-14	1.88E-10	1.54E-09	5.56E-10	6.00E-10	1.54E-09	9.90E-10
ADDder µg/(g.d)	Child	1.38E-14	1.27E-10	4.61E-11	1.38E-11	2.87E-09	7.55E-10	1.95E-11	2.87E-11	2.30E-11	1.40E-09	1.15E-08	4.15E-09	4.47E-09	7.41E-09	6.36E-09
	Adult	1.02E-11	2.14E-11	1.60E-11	1.02E-11	2.14E10	8.18E-11	1.45E-11	2.14E-11	1.73E-11	1.88E-10	1.54E-09	5.56E-10	6.00E-10	1.54E-09	9.90E-10
Semi- Urban site																
ADDing µg/(g.d)	Child	1.54E-09	2.42E-09	1.98E-09	1.78E-08	2.04E-08	1.91E-08	2.68E-09	3.70E-09	3.19E-09	1.78E-09	3.96E-09	2.87E-09	3.83E-09	1.27E-08	8.27E-09
	Adult	2.05E-10	3.25E-10	2.65E-10	2.40E-10	2.74E-09	2.57E-09	3.60E-10	4.97E-10	4.29E-10	2.40E-10	5.31E-10	3.86E-10	5.14E-10	1.71E-09	1.11E-09
ADDinh µg/(g.d)	Child	2.90E-11	4.60E-11	3.75E-11	3.93E-11	3.83E-10	3.63E-10	5.08E-11	7.02E-11	6.05E-11	3.93E-11	7.50E-11	5.45E-11	7.26E-11	2.42E-10	1.57E-10
	Adult	3.11E-14	4.93E-14	4.02E-14	3.63E-13	4.15E-13	3.89E-13	5.45E-14	7.53E-14	6.49E-14	3.63E-14	7.79E-14	5.71E-14	7.79E-14	2.59E-13	1.68E-13
ADDder µg/(g.d)	Child	1.38E-11	2.18E-11	1.78E-11	1.61E-10	1.84E-09	7.73E-10	2.41E-11	3.33E-11	2.87E-11	1.61E-11	2.41E-11	2.01E-11	3.45E-11	1.15E-10	7.48E-11
	Adult	1.02E-11	1.62E-11	1.32E-11	1.20E-10	1.37E-10	1.29E-10	1.80E-11	2.48E-11	2.14E-11	1.20E-11	2.65E-11	1.93E-11	2.57E-11	8.57E-11	5.57E-11

Table 5. Average Daily Dose in dust sample at urban and semi-urban sit

		Cu	**Zn**	**Cr**	**Ni**	**Pb**
Urban sie						
ADDing	Child	0.00023	.00049	0.00010	3.82E-5	0.00013
µg/(g.d)	Adult	3.11E-5	6.64E-5	1.34E-5	5.12E-5	1.78E-5
ADDinh	Child	4.39E-6	9.38E-6	1.89E-6	7.24E-7	2.52E-6
µg/(g.d)	Adult	4.71E-9	1.00E-8	2.03E-9	7.77E-10	2.70E-9
ADDder	Child	3.47E-7	7.42E-6	1.50E-7	5.73E-8	1.99E-7
µg/(g.d)	Adult	1.55E-7	3.22E-7	6.71E-8	2.54E-8	8.93E-8
Semi-urban						
ADDing	Child	9.03E-5	0.0004	8.82E-5	5.30E-5	2.92E-5
µg/(g.d)	Adult	1.31E-5	5.96E-5	1.18E-5	7.11E-5	3.92E-5
ADDinh	Child	1.71E-6	8.25E-6	1.89E-6	7.24E-7	2.52E-6
µg/(g.d)	Adult	1.83E-9	8.85E-8	1.07E-9	1.07E-10	5.95E-9
ADDder	Child	8.13E-7	3.92E-6	7.93E-7	4.77E-7	2.63E-7
µg/(g.d)	Adult	6.06E-7	2.92E-7	2.95E-8	3.55E-8	1.96E-8

The calculated results of this study for HQ_{Air} and HQ_{soil} are listed in Table 6 and Table 7 for both sites respectively. The HQ through different exposure pathway for child followed the trend as $HQ_{ing}>HQ_{inh}>HQ_{der}$ whereas for adult the trend follows $HQ_{ing}>HQ_{der}>HQ_{inh}$ in both sampling medium (air and dust). Pathways for adult and child was found similar with (Izhar et al., 2016) but for child inhalation values found higher compare than dermal pathways. In comparing with child and adult cases, the higher values found for children, results agreements with other studies (Kong et al., 2011; Liu et al., 2015; Xu et al., 2015). The average HQ values found under the safe limit, it shows non-carcinogenic effect for both receptor (adult and child). HQ value of Pb was found near to 1 in air sample while HQ value of Cr was found near to 1 in dust sample. For adults and child HQs are mostly inferior to safe level (=1).

The HQs value of these metals for children is quite higher in comparison with adults. The overall result states that children faced larger potential health risk from the soil. Overall, in this study the values of health risk are in the receivable range, the results indicate that exposure to heavy metals in air solely would not cause severe health effect on child and adult during monsoon seasons. Soil toxic risk assessment exposes on risk-oriented approach to inform residents/government about possible risks exposed to metals (USEPA 2011). The ECR of metals was calculated using the Eq (5) through the inhalation path which results are shown in Table 8 and Table 9.

Table 6. Health Risk Assessment for heavy metals in air sample at urban and semi-urban site.

		Cu			Zn			Cr			Ni			Pb		
Urban site		**Min**	**Max**	**Mean**	**Min**	**Max**	**Mean**	**Min**	**Max**	**Mean**	**Min**	**Max**	**Mean**	**Min**	**Max**	**Mean**
HQing µg/(g.d)	Child	3.84E-08	7.99E-08	5.99E-08	5.11E-09	1.07E-09	4.07E-08	7.25E-07	1.07E-06	8.52E-07	7.03E-08	5.75E-07	2.08E-07	3.19E-05	5.29E-05	4.54E-05
	Adult	7.29E-09	2.18E-08	1.22E-08	6.86E-10	1.43E-08	6.31E-09	9.72E-08	1.43E-07	1.14E-07	9.43E-09	1.11E-08	2.78E-07	4.28E-06	1.10E-05	7.07E-06
HQinh µg/(g.d)	Child	7.26E-10	1.51E-08	1.13E-08	9.68E-11	2.01E-09	7.68E-10	1.37E-08	2.01E-08	1.61E-08	1.33E-09	1.08E-08	3.91E-09	8.99E-08	1.00E-06	6.57E-07
	Adult	7.79E-13	1.34E-09	3.36E-10	1.03E-13	2.16E-10	6.91E-11	1.81E-11	2.73E-11	2.36E-11	1.42E-12	1.16E-11	4.19E-12	6.49E-10	1.07E-09	9.21E-10
HQder µg/(g.d)	Child	3.45E-10	3.20E-09	1.16E-09	4.60E-11	9.59E-09	2.52E-09	6.52E-09	2.68E-06	6.77E-07	6.33E-10	5.18E-09	1.87E-09	2.88E-07	4.77E-07	4.09E-07
	Adult	2.57E-10	5.36E-10	4.02E-10	3.43E-11	7.14E-10	2.73E-10	4.86E-09	7.15E-09	5.72E-09	4.72E-10	3.85E-09	1.39E-09	3.31E-10	3.55E-07	1.75E-07
Semi- Urban site																
HQing µg/(g.d)	Child	3.88E-08	6.07E-08	4.98E-08	5.96E-08	6.81E-08	6.39E-08	2.68E-09	1.23E-06	6.16E-07	8.94E-08	1.98E-07	1.44E-07	9.13E-06	1.22E-05	1.07E-05
	Adult	5.14E-09	8.14E-09	6.64E-09	8.00E-09	9.14E-09	8.57E-09	1.65E-07	1.23E-06	6.98E-07	1.20E-08	2.65E-08	1.93E-08	3.67E-06	2.73E-05	1.55E-05
HQinh µg/(g.d)	Child	7.26E-10	1.15E-09	9.38E-10	1.13E-09	1.29E-09	1.21E-09	1.70E-08	2.34E-08	2.02E-08	1.70E-09	3.75E-09	2.72E-09	5.19E-07	1.73E-06	1.12E-06
	Adult	1.65E-13	7.79E-13	4.72E-13	1.21E-12	1.32E-12	1.30E-12	1.81E-11	2.52E-11	2.16E-11	1.82E-12	3.90E-12	2.86E-12	1.86E-09	1.88E-09	1.87E-09
HQder µg/(g.d)	Child	3.45E-10	5.46E-10	4.46E-10	5.37E-10	6.14E-10	5.75E-10	8.05E-09	1.11E-08	9.59E-09	8.05E-09	8.05E-09	8.05E-09	2.38E-07	8.22E-07	5.30E-07
	Adult	2.57E-10	4.07E-10	3.32E-10	4.00E-10	4.57E-10	4.29E-10	6.00E-09	8.29E-09	7.15E-09	6.00E-10	1.32E-09	9.60E-10	1.84E-07	6.13E-07	9.60E-10

Table 7. Health Risk Assessment for heavy metals in dust sample at urban and semi-urban site.

Urban sie		**Cu**	**Zn**	**Cr**	**Ni**	**Pb**
HQing µg/(g.d)	Child	0.00023	0.00049	0.00010	3.82E-5	0.00013
	Adult	3.11E-5	6.64E-5	1.34E-5	5.12E-6	1.78E-6
HQinh µg/(g.d)	Child	0.00017	0.00037	7.58E-5	2.89E-5	0.0001
	Adult	2.33E-9	5.03E-8	1.01E-8	3.88E-9	1.35E-8
HQder µg/(g.d)	Child	2.08E-6	4.45E-6	9.00E-7	0.00034	1.19E-6
	Adult	1.55E-6	3.32E-6	6.71E-7	2.56E-8	8.93E-7
Semi-urban						
HQing µg/(g.d)	Child	0.0022	0.0014	0.0294	0.0026	0.2091
	Adult	0.0003	0.0001	0.0039	0.0003	0.0280
HQinh µg/(g.d)	Child	4.27E-5	2.75E-5	6.32E-5	3.62E-5	0.0180
	Adult	4.59E-8	2.95E-8	3.59E-8	5.39E-9	4.21E-8
HQder µg/(g.d)	Child	8.69E-6	2.47E-5	5.00E-5	2.38E-5	0.0018
	Adult	1.51E-6	9.74E-7	9.86E-5	1.77E-6	0.00014

Table 8. Comparison of cancer risk in air at urban and semi- urban site.

METALS	**IUR ($\mu g/m^3$)$^{-1}$ (Izhar et. al (2016))[62]**	**Urban Site**		**Semi-Urban Site**	
		Child	**Adult**	**Child**	**Adult**
Cr	0.012	2.07E-8	8.28E-7	8.21E-8	3.28E-7
Ni	0.00024	1.57E-9	2.36E-8	2.63E-9	1.05E-8
Pb	0.000012	1.61E-10	6.44E-10	1.97E-10	5.89E-10

Table 9. Comparison of cancer risk in dust at urban and semi-urban site.

Metals	Urban Site		Semi-Urban Site		Rural Site	
	Child	Adult	Child	Adult	Child	Adult
Cr	0.0025	0.010	0.0022	0.0090	0.0019	0.0079
Ni	3.93E-5	0.0001	5.45E-5	0.0002	4.94E-6	0.0001
Pb	3.42E-6	1.37E-5	7.52E-7	3.011	ND	ND

Three metals including Cr (VI), Ni and Pb are known Carcinogens through inhalation pathway. Some literature also revealed that Cr (VI) and Cd contributing to cancer risks than any other metals (Hieu & Lee, 2010). Ratio of carcinogenic between Cr (VI) to non-carcinogenic Cr (III) is 1:6, so one seventh concentration of Cr (VI) to computing the ECR value (Hieu & Lee 2010). The Inhalation Unit Risk value was taken from Integrated Risk Information System (USEPA 2004b; Rohra et al., 2018). In air sample the highest value of metals for ECR was found as Cr(VI)>Ni>Pb for both sampling site. Values of Cr and Ni were found higher for child while Pb was higher for adult at both site (urban and semi urban). All ECR values of metals were found higher for adult at urban site while the value for child was higher at semi-urban site. ECRs value was found higher in dust in compare to air, the trend of ECR value for metal was followed as Cr(VI)>Ni>Pb in dust sample at both sites. The results of this study go well with (Izhar et al., 2016). In dust samples value of ECR was higher for adult comparison to child. Trend of higher value of ECRs was observed as semi-urban>Urban. ECR value of all metals in air samples were found below the permissible limit while values were exceeded in soil samples.

CONCLUSION

The onset of the traffic and industrial revolution has marked a drastic increase in the rate of heavy metal pollution in our environment. Advancements in the sectors of Industry and Technology have resulted in the increased use of heavy metals. Heavy metals are introduced in the environment through many different routes like Industries being the prominent one followed by agricultural activities, transportation activities, Wastewater runoffs from household activities, atmospheric deposits or during the processes of extraction of natural resources. Heavy metals bound to $PM_{2.5}$ present in dust are among the crucial factors for human health problems. In the present context of the study, we have focused our attention on the measurements of the concentrations of heavy metals present in the study area environments. The presence of such metals causes severe health effects on human health as they may be carcinogenic or non-carcinogenic. The concentration of heavy metals in air samples were followed as Pb>Cr>Ni>Cu>Zn whereas in dust samples it was followed by Zn>Cu>Pb>Cr>Ni at urban sampling sites. At the semi-urban site the trend of metals in air and dust sample was found the same as Pb>Cr>Ni>Cu>Zn. Health risk assessment of all-metal pollutant was found to be below the threshold levels. For this study the ADD and HQ for child pathways decrease in the order of ingestion>dermal>inhalation whereas for adult the pathways followed the trend as Ingestion>Inhalation>dermal. It was found that ingestion is a primary pathway to harming human healthiness, ECR values of all metals were below the permissible limit at both sampling sites. $PM_{10,}$ $PM_{2.5,}$ $PM_{0.25}$ were found higher in semi-urban site whereas $PM_{1.0}$ was found higher in the urban site. Overall the mass concentration is substantially higher than NAAQS and WHO. The study showed that health risk due to exposure to PM is not negligible to pedestrians, commuters as well as residents living in that area. In the present study, the influence of PM_{10} and $PM_{2.5}$ were higher at the semi-urban site due to re-suspended dust

while the finer particle $PM_{1.0}$ was higher at the urban site the reason for the same is due to rise in many small industries and emissions from the vehicle at urban sites. The morphological study and elemental analysis of PM provide information for determining their physical properties and diverse sources. Appearances like spherical, cluster and flaky were found in urban site whereas at Semi-urban site reticular, branched chain-like aggregates and fluffy appearance was found. As this study is one of its kinds to report the excess cancer risk at traffic junctions within the aforementioned area, the results should be treated as suggestive rather than definite. As the duration of the study was carried out for a short period of time, hence more thorough studies are required to strengthen the databases and investigate the range of outdoor air pollutants. All these results obtained in this study shows the necessity of the continuous monitoring of $PM_{2.5}$ as it is now recognized as an important factor in the field of air pollution which causes several diseases.

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CONFLICT OF INTERSET

The authors declare that there is not any conflict of interests regarding the publication of this manuscript. In addition, the ethical issues, including plagiarism, informed consent, misconduct, data fabrication and/or falsification, double publication and/or submission, and redundancy has been completely observed by the authors.

LIFE SCIENCE REPORTING

No life science threat was practiced in this research.

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