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ORIGINAL PAPER

Non-carcinogenic and Carcinogenic Risk Assessment of Trace Elements of $PM_{2.5}$ During Winter and Pre-monsoon Seasons in Delhi: A Case Study

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Abstract

This study focuses on exposures of metal constituents of particulate matter (PM) in the ambient air sample collected at Indian Institute of Technology Delhi (IIT Delhi), India, which might lead to occurrence of non-cancerous events and cancer events. A step-wise construction of risk assessment framework for estimating risks due to exposures of $PM_{2.5}$ presented. Samples from winter and pre-monsoon seasons of Delhi region (28.5450°N, 77.1926°E) (duration 1: December 2013–February 2014; duration 2: March 2014–May 14) were collected. More than 12 samples were collected using $PM_{2.5}$ sampler on an 8-h basis and analysed gravimetrically for determining mass content and chemically for determining metal content of $PM_{2.5}$. Twenty-eight metals in samples were detected using Energy Dispersive X-Ray Fluorescence (ED-XRF). Using these values, health risks of hypotheticals exposures of $PM_{2.5}$ in ambient air samples were estimated either in terms of hazard quotient (i.e. ratio of daily inhaled dose to daily acceptable dose) for exposures of non-carcinogenic metals or lifetime excess risk of cancer for exposures of carcinogenic metals. Dose–response information of different metals was taken from the U.S. EPA IRIS database. Among metals, S content was highest followed by Cl, Si, K, Ca and Fe, Zn and Pb. High S can be attributed to vehicular emission or particles generated from abrasion of tyres of vehicles. High contents of Si, Ca, Fe in PM samples may be contributed from resuspension road dust, while source of K may be local biomass burning for space heating in winter. Zn comes from vehicle and coal burning probably used by local residents for space heating. Chlorine used in lubricants and diesel fuel could be a source of high Cl content in samples collected in the present work. Small traces of Pb in samples might be coming from brake and tyre wear or resuspension of road dust contaminated with lead used earlier in diesel and/or petrol to improve combustion. Estimates of potential risk due to hypothetical exposure of adults and children to four carcinogenic metals of $PM_{2.5}$ were found to be more than 1/10,000,000, indicating chance of cancer risks. Among metals, exposures to PM-associated Cd resulted in consistent cancerous risk in both seasons, whereas exposures to PM-associated Cr resulted in HQ value > 1 indicating chance of non-carcinogenic risks.

Keywords Ambient PM ($PM_{2.5}$, PM_{10}) sampling · Inhalation risks · Carcinogenic metals · Cancer risk

Introduction

Exposure of pollutants to human can occur through different pathways which include ingestion, dermal and inhalation. Exposure through air-pollution-related sources has been seen to be more in recent studies compared to that from other two pathways (ASTDR 1999; Hu et al. 2012; Kong et al. 2012). Exposure of particulate matter (PM) has been recognized as a well-known human health risk factor (Zheng et al. 2010). However, its quantification for various regions, such as India, is still not well established (Brauer et al. 2016). PM is considered to be among the six criteria pollutants, and it comprises heterogeneous mixture of different

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toxic elements and compounds (Delfino et al. 2002). Epidemiologically, it is tested that PM imposes mutagenic acute and chronic effects. However, the intensity of such effects strongly depends upon pollution sources, their spatial and temporal distributions (Wang et al. 2006). The International Agency for Research on Cancer (IARC 2012) classified outdoor air pollution as Group 1 carcinogen to humans in the year 2013 (IARC 2013). Among various trace elements present in PM, iron, vanadium, nickel, chromium, copper and zinc have been cited to be most toxic elements on the basis of their abilities to support electron exchange (Contini et al. 2016; Santos and Fernández-Olma 2016; Sen et al. 2016), and catalyse and generate reactive oxygen species (Sun et al. 2012; Das et al. 2016). These potential effects indicate the need for conducting quantitative analysis of health risks due to carcinogenic components of PM, including trace elements and PAHs, for various developing regions of the world, such as cities in India where air pollution levels are at alarming stage.

Toxicological effects of $PM_{2.5}$ on lung have been reported through epidemiological testing and laboratory-based toxicity studies. Different regulatory bodies had already given the permissible limit of $PM_{2.5}$; however, such limits have not been advised for its chemical toxic constituents (Das et al. 2016). Very few studies have incorporated these aspects in conducting health risk assessment (Massey et al. 2013; Khanna et al. 2015) where detailed characterization of $PM_{2.5}$ for its toxic components and estimation of associated carcinogenic risk were conducted. The PM concentration and composition in developing countries vary largely due to spatial and temporal distributions of energy sources which are governed by energy demand patterns and available technology options (Jain and Khare 2008; Stone et al. 2010). Regionally dominated sources, such as solid fuels combustion in cooking stoves, open burning of agricultural residue in field after harvesting, use of coal and biomass fuel in brick kilns, open solid waste burning and fossil fuel combustion in vehicles, have been identified as threats to worsening air quality in India (Reddy and Venkataraman 2001; Jain et al. 2014) Exposures of PM might result in lung disease, which has been recognized as one of the major burdens for diseases in India (Khanna et al. 2015). However, studies on health risk due to exposures of PM in India are very limited (Das et al. 2006). Indian epidemiological studies showed correlation of respirable particulate matter (PM_{10}) with respiratory and pulmonary symptoms (Srivastava et al. 2004, 2008). However, few studies have reported the estimated cancerous and non-cancerous risk due to exposures of ambient fine PM ($PM_{2.5}$) and its toxic components to children and adults (Curtis et al. 2006).

The Capital of India (Delhi) has been recognized as one of the highly polluted cities of the world and ranked as 3rd in list (WHO Global Urban Air Pollution Database 2016). Factors, such as high population ranging more than 14 million, increase in industrialization, housing and infrastructure, increased vehicular traffic, congested streets, poorly maintained vehicles and lack of effective air pollution control programmes (Basha et al. 2010), have been shown to aggravate the problem of human health risk due to exposures of particulate matters and its toxic constituents. In Delhi, some studies have focused on characterizing PM particles (Sharma and Maloo 2005; Khanna et al. 2015; Izhar et al. 2016; Srivastava et al. 2004, 2008) and some studies have focused on assessing health impacts, either through theoretical calculation or through epidemiologically based approach (Kumar et al. 2011, 2014; Sharma et al. 2010; Aggarwal and Jain 2015; Khanna et al. 2015; Izhar et al. 2016). However, very few studies appear to consider (i) effect of seasonal variation on risk due to exposures of PM-associated components and (ii) toxicity of mixture of PM constituents. This represents a big data gap for regulatory agencies in formulating health-protective guidelines considering all realistic exposure considerations, such as exposure of PM with different constituents (metals and polycyclic aromatic hydrocarbon contents).

To understand the risk of getting cancer- and non-cancer-based effects due to exposures of PM in Indian cities, this study analysed metal contents in $PM_{2.5}$ of ambient air samples in Delhi (India) and estimated health risks to adults and children for a hypothetical exposure scenario of inhalation of PM-contaminated ambient air samples of winter and pre-monsoon seasons. Risk estimates were reported either in terms of hazard quotient (i.e. ratio of daily inhaled dose to daily acceptable inhaled dose) for exposures of non-carcinogenic metals or lifetime excess risk of cancer (ECR) for exposures of carcinogenic metals.

Methodology

Aerosol Sampling and Chemical Characterization

Site Description

The aerosol samples were collected at ~ 10 m height in the Department of Civil Engineering, Indian Institute of Technology, Delhi, India (28.5450°N, 77.1926°E). Delhi is situated 160 km south of the Himalayas. The climatic conditions of Delhi include extreme hot in summer (45–47 °C), harsh winter (1–2 °C) and average rainfall of 797.3 mm (Ministry of Statistics and Program Implementation, GOI 2016). The detailed information about the site has been given elsewhere (Jaiprakash and Kumar 2016).

Ambient Monitoring

Ambient air monitoring was conducted for 10 sampling days in a month for 3 months in winter (December, January, February) and 3 months in pre-monsoon season (March, April and May). $PM_{2.5}$ sample was collected on a 47-mm-diameter Teflon Filter (Whatmann; WHA7582002). PTFE membrane filters were conditioned at controlled environment of 25 °C and 50% RH for 24 h before and after sampling events as per the previously used standard procedure (Singh et al. 2015). The pre- and post-weight of the filter papers were recorded using microbalance (CPA-2PF, Sartorius, Germany; least count = 1 µg, accuracy = 3%). Each weight measurement was taken thrice and average of the three weights was used in calculation. After pre-weighing, the filter papers were stored in sealed filter cassettes and kept in freezer at − 4 °C. The filter papers were loaded in impactor of Envirotech, APM-550 EL sampler which was operated at 16.7 L per minute rate. Samples were collected for 8 h, as 8 h is the considered working hour when human being could be exposed to PM (Izhar et al. 2016). Particle-loaded filter papers were again conditioned in the same environment (25 °C and 50% RH) as before and then weighed. Then, the filter papers were stored at − 4 °C before conducting further chemical analysis (Jaiprakash and Kumar 2016).

Trace Element Determination

The trace elements of the samples were determined by energy dispersive X-ray fluorescence (ED-XRF) using a Shimadzu EDX-700HS spectrometer (Shimadzu Corp., Japan) at IISER, Bhopal, India. X-ray fluorescence analysis is a non-destructive analytical method, often used for determining elemental concentrations of various heavy and toxic metals. A total of 28 metals were detected from the sample: Mg, Al, Si, S, Cl, K, Ca, Sc, Ti, V, Cr, Mn, Fe, Co, Ni, Cu, Zn, As, Se, Br, Sr, Mo, Cd, In, Sn, Sb, Ba and Pb. For a given month, monthly average concentration of metals was calculated using metal content information of individual filter papers for a given month.

Quality Control and Quality Assurance

The quality control/quality assurance of trace element analysis of collected aerosol samples and blank filter mass of each elements are summarized in Table 1. The mass of various elements on field and dynamic blank filters ranged from 2 to 5% of the sample mass (Table 1). The method detection limit (MDL) for trace elements was calculated as three times the standard deviation of 10 replicates of the blank filter (Raman and Kumar 2016).

Table 1 Quality assurance and quality control (QA/QC) parameters of trace elements species of $PM_{2.5}$

Species	MDL (µg/m^3)	Repeatability (%)[a]	Blank/sample mass (%)
Si	0.084	< 15	3.0
Ti	0.005	< 10	5.0
V	0.005	< 10	3.0
Cr	0.004	< 10	2.0
Mn	0.008	< 10	4.0
Fe	0.007	< 12	0.5
Ni	0.005	< 15	3.5
Cu	0.006	< 10	4.0
Zn	0.004	< 10	0.6
Sr	0.007	< 13	0.4
Cd	0.038	< 10	2.0
Sn	0.043	< 10	1.0
Sb	0.033	< 14	3.0
Pb	0.011	< 15	3.0

MDL method detection limit

[a]10% samples in a batch of 20 samples were analysed in duplicate

Health Risk Assessment

This study used a four-step human health risk assessment framework consisting of hazard identification, exposure assessment, dose–response assessment and risk characterization steps, developed as per the USEPA methodology (U.S. EPA 2009a, b) and previously published studies (Hu et al. 2012; Wei et al. 2015; Han et al. 2016). This assessment simplifies the study by representing the population of any area exposed to the airborne metals. Due to differences in respiration, behaviour and lifestyle, the risk values have been estimated for two subgroups, i.e. adults and children. Exposure of PM-associated metals can happen through three major pathways: (a) inhalation of the airborne PM through nose and mouth, (b) absorption of the adhered metal contents of PM by dermal layer, and (c) ingestion of the food items which might have received particles during their settling in ambient environment (Choudhury and Mudipalli 2008). In this study, only $PM_{2.5}$-associated metal was considered as hazard and risk estimation was conducted for a hypothetical exposure scenario (Tables 2, 3, 4).

Exposure Dose Determination

The US Environmental Protection Agency (EPA) had proposed a model to compute the risk posed by metals to humans. Average daily dose (ADD) (mg/kg/day) is calculated for the exposure of individual metal content from $PM_{2.5}$ samples using the following Eq. (1):

Table 2 $PM_{2.5}$ concentration in few Asian countries (Brauer et al. 2016, for the Global Burden of Disease Study 2015) (http://data.worldbank.org/indicator/)

Country	Annual mean $PM_{2.5}$ concentration (μg/m^3)		
	1990	2015	Standard (WHO)
China	48.5	58.4	10
India	59.8	74.4	10
Bangladesh	63.9	89.4	10
Sri Lanka	30.0	28	10
Pakistan	68.0	65.0	10
Nepal	57.1	74.9	10
Myanmar	43.7	54.3	10

Table 3 RfD and IUR values of the metals (U.S. EPA 2009a, b)

Heavy metals	Cd	Cr	Ni	Pb
RfD Inhalation	1.00E−04	2.86E−05	3.52E−03	3.52E−03
IUR	0.0018	0.012	0.00024	0.000012

RfD inhalation reference dose in mg/kg/day, *IUR* integrated risk information taken in μg/m^3

$$\mathrm{ADD} = \frac{C \times InhR \times \mathrm{EF} \times \mathrm{ED}}{\mathrm{BW} \times \mathrm{AT}}, \quad (1)$$

where C is the concentration of $PM_{2.5}$-associated metal content in this study, obtained through XRF analysis (μg/m^3); *InhR* is the inhalation rate (m^3/day) (assumed to be 7.63 and 20 m^3/day for adults and children, respectively); EF is the exposure frequency (days/year) (assumed value = 180 days/year); ED is the exposure duration (years) (assumed value = 24 for adults and 6 for children); BW is the body-weight (kg) (assumed to be 70 kg for adults and 15 kg for children); AT is averaging time, equal to ED × 365 days/year, i.e. 24 × 365 (days/year) for adults and 6 × 365 (days/year) for children (U.S. EPA 2004b, c, 2007, 2009a, b).

Estimation of Risk of exposures of Non-carcinogenic PM-Associated Metals

Exposure of One Type of Metal At-a-time Non-carcinogenic health risk was estimated by calculating Hazard Quotient (HQ) and Hazard Index (HI) value. The HQ was estimated by dividing the ADD with Reference Dose (RfD) values (Eq. 2) (U.S. EPA 2009a, b; Hu et al. 2012).

$$\mathrm{HQ} = \frac{\mathrm{ADD}}{\mathrm{RfD}}. \quad (2)$$

HQ value smaller than 1 indicates that non-carcinogenic PM-associated metal do not possess any risk to human health (Zheng et al. 2010; Li et al. 2013; Du et al. 2013). For a given month, monthly average value of HQ was calculated using related HQ value of exposure of $PM_{2.5}$-associated metals, detected on 10 filter papers.

The total hazard values of exposures of PM-associated metals in two seasons were calculated by adding HQ of each metal of each month for the tenure of 3 months using the previously used methodology (Xu et al. 2015; Liu et al. 2015; Greene and Morris 2006). The formula given below is used for the same; '*a*' is various metals for which HQ is calculated and '*b*' is the month for which it is calculated. If the total hazard for the season is found to be more than 1 for the non-carcinogens, exposures of PM-associated non-carcinogenic metals might cause adverse effects to the human health.

$$\text{Total HQ(season)} = \mathrm{HQ}(a_1, b_1) + HQ(a_2, b_2) + \mathrm{HQ}(a_3, b_3). \quad (3)$$

Exposure of More Than One Type of Metal At-a-time Risk estimate was also calculated for hypothetical exposure of

Table 4 Summary of parameters required for calculating both cancerous and non-cancerous risk

Parameter	Description	Value and source
C (concentration) (μg/m^3)	C of metals	Obtained from ED-XRD analysis of collected $PM_{2.5}$ samples
ED (exposure duration) (years)	Duration of acute and chronic effect	1 year (U.S. EPA 2009a, b)
EF (exposure frequency) (days/year)	Average days of annual exposure	180 days/year (U.S. EPA 2009a, b)
ET (exposure time) (day)	Duration of exposure	8 h/day (U.S. EPA 2009a, b)
IUR (inhalation unit risk) [1/(μg/m^3)]	An estimate of the increase cancer risk from inhalation exposure to a concentration of 1 mg/m^3 for a lifetime; it can be multiplied by an estimate of lifetime exposure in (mg/m^3) to estimate the lifetime cancer risk	IRIS (Integrated Risk Information System), U.S. EPA
AT (average time) (days)	Average time for exposure.	ED × 365 days/year (U.S. EPA 2009a, b)
BW (body weight)	The average body of human being taken.	70 kg (adult), 15 kg (children) (U.S. EPA 2009a, b)
InhR (inhalation rate)	Rate of inhaling air in m^3/day	7.63 (adult), 20 (children) (U.S. EPA 2009a, b)

mixture of PM-associated metals, which is a realistic exposure scenario. In this regard, risk estimate was represented as hazard index which is defined in terms as "weighted sum of exposures of the metals in mixtures". Two sensitive cases were considered: (i) no interaction of effects due to metals, (ii) interaction of effects due to metals.

(i) No interaction of effects due to metals (dose additivity approach) Hazard index for exposure of mixture of PM-associated metals was calculated by adding hazard quotient (HQ) values related to exposures of individual PM-associated metals using Eq. (4) where values of HQ_j (i.e. Hazard quotient for *j*th metal), reference dose and exposure-related parameters are required.

$$\mathrm{HI} = \sum_{j=1}^{n} \mathrm{HQ}_j. \tag{4}$$

(ii) Interaction of effects due to metals (the weight-of-evidence approach) When chance of interaction of toxic effects of mixture of PM-associated metals exists, the "Weight-of-evidence" approach can be used. In this method, HQ obtained from Eq. (4) is modified by multiplying it with some modification factor (representing effects of mixture toxicity) to calculate interaction-based HI values ($HI_{interaction}$) (Parsai and Kumar 2016) (Eqs. 5–7).

$$\mathrm{HI}_{\mathrm{interaction}} = \sum_{i=1}^{n} (\mathrm{HQ}_i) \times \sum_{j \neq 1}^{n} f_{ij} M_{ij}^{B_{ij}\theta_{ij}}. \tag{5}$$

$$f_{ij} = \frac{\mathrm{HQ}_j}{\mathrm{HI}_{\mathrm{add}} - \mathrm{HQ}_i} \tag{6}$$

$$\theta_{ij} = \frac{\left(\mathrm{HQ}_i \times \mathrm{HQ}_j\right)^{0.5}}{\left(\mathrm{HQ}_i + \mathrm{HQ}_j\right) \times 0.5}, \tag{7}$$

where HQ_i is the hazard quotient of *i*th metal, f_i the toxic hazard of the *j*th metal relative to the total hazard from all metals potentially interacting with *i*th metal (where *j* is not equal to *i*), M_{ij} the interaction magnitude, i.e. the influence of *j*th metal on the toxicity of *i*th metal, B_{ij} the score for the strength of evidence that *j*th metal will influence the toxicity of *i*th metal (as per the criteria given in Table 5), and θ_{ij} is the degree to which *i*th chemical and *j*th chemical are present in equitoxic amounts.

Estimation of Excess Cancer Risk (ECR) Due to Exposures of Carcinogenic PM-Associated Metals

Risk estimates of exposure of carcinogenic PM-associated metals were calculated using lifetime excess cancer risk metric, which indicates the incremental probability of a person developing cancer over a lifetime due to total exposure to the potential carcinogen. ECR is calculated using following Eq. (8) (Hu et al. 2012; Izhar et al. 2016).

$$\mathrm{ECR} = \frac{C \times \mathrm{ET} \times \mathrm{EF} \times \mathrm{ED} \times \mathrm{IUR}}{\mathrm{AT}}, \tag{8}$$

where *C* is the concentration of the metal pollutant in ($\mu g/m^3$), IUR is the Inhalation Unit Risk ($1/(\mu g/m^3)$), AT is the average time for exposure to carcinogens (i.e. (70 years × 365 days/year × 24 h/day) for adults and (15 years × 365 days/year × 24 h/day) for children), ET is exposure time which is considered to be 8 h in this study, EF is the exposure frequency (i.e. assumed to be 180 days/year), and ED is the exposure duration (i.e. assumed to be 24 years for adults and 6 years for children). Data on the carcinogenic types and the inhalation unit risk of the metals are obtained from the USEPA 2011 database for IRIS (Integrated Information Risk System) (www.epa.gov/IRIS/). If the ECR value of exposure of carcinogens exceeds the 10^{-6} value, the exposure may produce carcinogenic health risk. For a given month, monthly average value of ECR was calculated using related ECR value of exposure of $PM_{2.5}$-associated metals of

Table 5 Classification and default weighting factors for the modified weight of evidence

Category	Description	Direction	
		Greater than additive effect	Lesser than additive effect
I	The interaction has been shown to be relevant to human health effects and the direction of the interaction is unequivocal	1	−1
II	The direction of the interaction has been demonstrated in vivo in an appropriate animal model, and the relevance to potential human health effects is likely	0.75	−0.5
III	An interaction in a particular direction is plausible, but the evidence supporting the interaction and its relevance to human health effects is weak	0.5	0
IV	The assumption of additivity has been demonstrated or must be accepted	0	0

10 different collected samples of that month. The calculation of cumulative risk (ECR) of exposures of $PM_{2.5}$-associated metals for 3 months was done by adding ECR data of individual months (assuming no interaction during toxic effects).

Results and Discussion

Variation in $PM_{2.5}$ Concentration During Winter and Pre-monsoon Seasons

Concentration values of $PM_{2.5}$ collected for consecutive months of winter (i.e. December 2013, January 2014 and February 2014) and pre-monsoon (i.e. March 2014, April 2014 and May 2014) are presented in Table 6. The average concentration values were found to be higher in the months of winter (December 2013: 216.5 μg/m^3, January 2014: 256.7 μg/m^3, February 2014: 188.5 μg/m^3) compared to months of pre-monsoon seasons (March 2014: 76.2 μg/m^3, April 2014: 70.5 μg/m^3, May 2014: 54.7 μg/m^3) ($p = 0.005$; pair-wise statistical t test). The observed difference could be attributed to the combined effects of $PM_{2.5}$-producing sources and weather conditions. Firstly, additional $PM_{2.5}$-producing sources, such as burning of fire crackers in Diwali and burning of agricultural waste after crop harvesting, increase during winter season than in pre-monsoon season (Khanna et al. 2015; Srivastava et al. 2008). Secondly, the stagnant air during winter slows down the movement of PM and keeps pollution close to the ground than dry weather condition of pre-monsoon season which helps in reduced concentration of industrial and vehicle-induced $PM_{2.5}$ sources (Srivastava et al. 2004).

Table 6 $PM_{2.5}$ concentration of IIT Delhi

Sampling months (year)	Concentration of $PM_{2.5}$ (μg/m^3) (average ± 1 SD) ($n = 10$)
December 2013	216.5 ± 116.9
January 2014	256.7 ± 94.6
February 2014	188.5 ± 111.2
March 2014	76.6 ± 42.4
April 2014	70.4 ± 33.3
May 2014	54.6 ± 30.5

Concentration of PM-Associated Heavy Metals

In this study, a total of 28 trace elements (Mg, Al, Si, S, Cl, K, Ca, Sc, Ti, V, Cr, Mn, Fe, Co, Ni, Cu, Zn, As, Se, Br, Sr, Mo, Cd, In, Sn, Sb, Ba, Pb) in $PM_{2.5}$ samples were analysed using the ED-XRF method according to the instrument's maximum capacity of detection. Out of these metals, major metals, such as As, Cd, Cr, Pb, Ni, Zn, Cu, were detected. For winter season, monthly average concentrations of major metals were found to range between 0.0095 and 0.68 μg/m^3. Among major metals, concentrations of Pb and Ni were found to be highest followed by that of Cd and Cr. For pre-monsoon season, the monthly average concentrations of heavy metals were found to range between 0.0082 and 0.44 μg/m^3. The comparison of concentrations of individual metals showed that concentration of Pb was found to be highest followed by that of Ni, Cd, Cr and As.

Concentrations of Si, Ca, K and Fe were found to be high which could be attributed to their widespread presence in

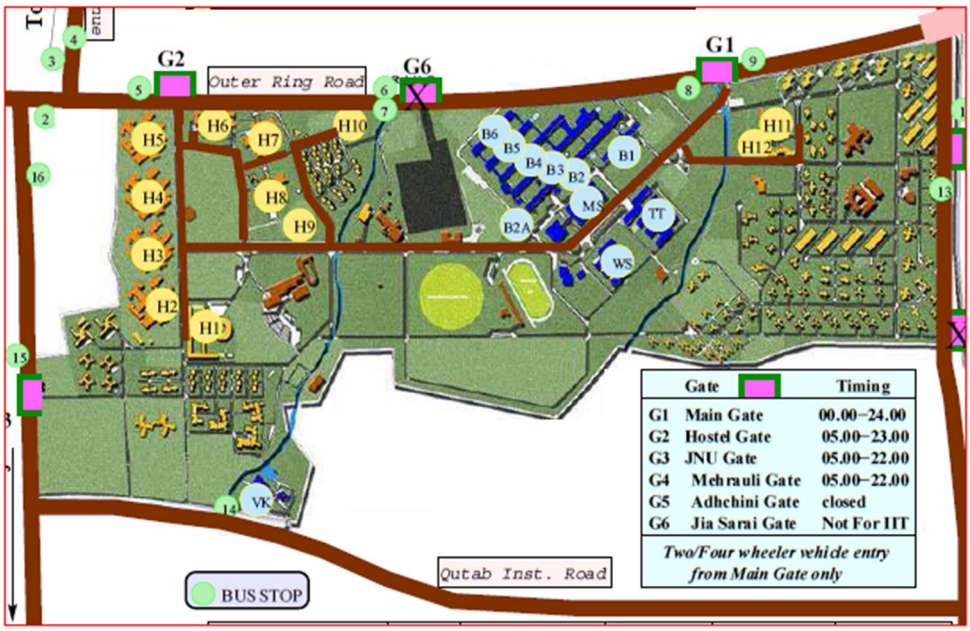


Fig. 1 Campus Area of IIT Delhi showing ambient air sampling site (Department Of Civil Engineering Block IV). *Source* http://web.iitd.ac.in/~vvbuwa/contact.html

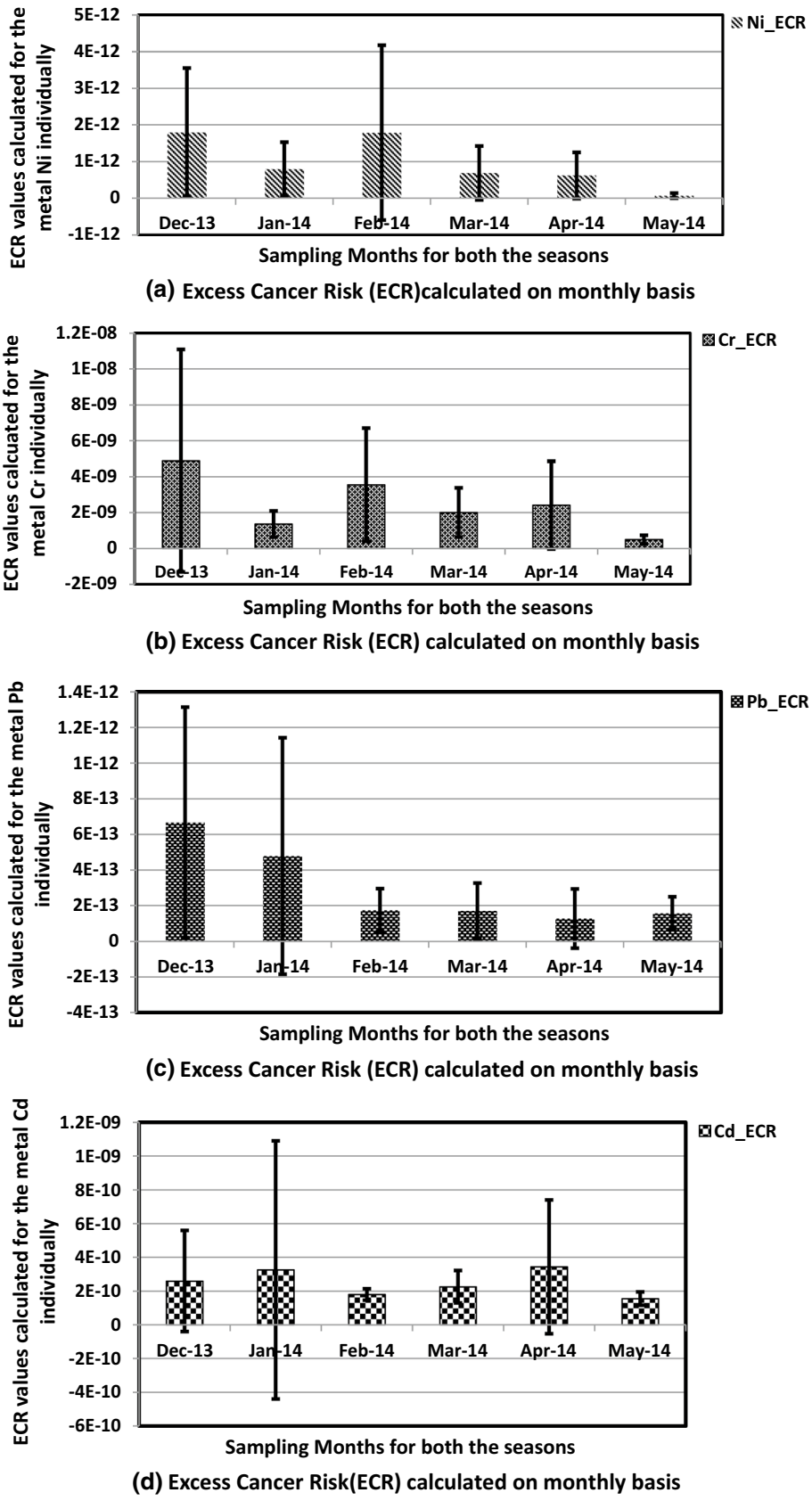


Fig. 2 Excess Cancer Risk (ECR) calculated for adults for both the seasons (Winter and Pre-monsoon): **a** ECR for metal Ni, **b** ECR for metal Cr, **c** ECR for metal Pb, **d** ECR for metal Cd. (i) *Dec-13* month of December in the year 2013; the trend is followed for all the months in the series. (ii) *Y*-axis denotes average ECR value calculated using data of 10 filter papers for a given month. (iii) Error bar denotes one standard deviation around mean value

Table 7 Summary of assumptions for different parameters

Parameters	Values considered for calculations	Reason (calculation or assumption with reference)
f_{ij} = toxic hazard of the jth metal relative to the total hazard from all metals potentially interacting with metal i (where j is not equal to i)	1 (Eq. 3)	Exposure factor f. Sum must reduce to unity (1) when dose addition is assumed so must be normalized in some fashion to avoid double counting the individual hazard quotients. (U.S. EPA 2009a, b; Supplementary Guidelines)
M_{ij} = interaction magnitude, the influence of metal j on the toxicity of metal i	5	The default interaction magnitude is set at 5, as the weight of evidence suggests an interaction but the magnitude of the interaction cannot be quantified. The magnitude of the deviation from additivity for the mixtures tested was about a factor of 5 in either direction (U.S. EPA 2009a, b; Supplementary Guidelines)
B_{ij} = score for the strength of evidence that metal j will influence the toxicity of metal i (To be obtained from table below)	B_{ij} values are obtained by assumption (see Table 1; i.e. classification and default weighting factors for the modified weight of evidence And effect of one metal on other (Antagonistic or Synergistic) is decided referring Table 5	Assumption (U.S. EPA 2009a, b, Supplementary Guidelines; Choudhury and Mudipalli 2008)
θ_{ij} = degree to which chemicals i and j are present in equitoxic amounts	Values are calculated using Eq. 4	The 2 chemicals (metals for this case) are considered to be equal amount (U.S. EPA 2009a, b; Supplementary Guidelines).

earth crust (Khanna et al. 2015). On the other hand, concentrations of S and Pb in air were found to be high due to their presence in exhaust of vehicles or in materials coming out from tyres during their abrasion (Sternbeck et al. 2002). The presence of Pb in air could be related to its presence in dust from previous emissions of vehicles using Pb-based oils (Khanna et al. 2015). High presence of Ni in $PM_{2.5}$ samples could possibly be attributed to the presence of large number of heavy duty diesel-based vehicles (Xu et al. 2017).

As metals, such as Cd, Cr, Pb and Ni, were found to be in considerable amounts in all of the samples tested and have been shown to pose risks (Li et al. 2015), this study estimated risks of getting non-cancerous effects and cancerous effects from exposures of these $PM_{2.5}$-associated metals under a hypothetical exposure scenario. These metals have been placed in the IARC carcinogenic groups and classes (Greene and Morris 2006) and have been termed as carcinogens through inhalation pathways. Risk estimation of exposure of Cr(VI) was also considered in this study as it has been reported to be carcinogenic in nature and is more prevalent in nature than Cr(III) (Choudhury and Mudipalli 2008). Following sections present findings on calculated risk estimates (Figs. 1, 2).

Estimates of Non-cancer Risks

The non-cancer risk estimated (i.e. HQ value) was calculated for the metals Cd, Cr, Ni, Pb. Risk estimation of exposures of observed high concentrations of metals, like Fe, Ca and Mg, Se (i.e. essential elements for growth and metabolism of human body) could not be done due to non-availability of their RfD values.

HQ values, irrespective of sub-population type and season, were found to be smaller than 1(range: 3.6×10^{-6}–0.17), indicating no risk to adults and children. The following order of HQ values of exposures of PM-associated metals to adults was observed: (smallest HQ value) Ni < Cd < Pb < Cr (highest HQ value) during winter season and (smallest HQ value) Cd < Pb < Ni < Cr (highest HQ value) during pre-monsoon season. The similar trend of order of HQ values of exposures of PM-associated metals to children was also observed (Tables 7, 8).

The total hazard values of exposures of PM-associated metals in two seasons were calculated by adding the HQ values for each month. HI for exposure of Cr to children in IIT Delhi campus was found to be maximum in winter (HI value = 17.4), indicating that children are at great health risk in winter season. Further, health impacts on children in IIT Delhi campus were found to be higher than that on adults (Figs. 3, 4). Findings of this study were found to be comparable with that reported for Beijing, Huludao City, China (Du et al. 2013; Zheng et al. 2010) and Nanjing (China) (Wu and Sheng 2011; Hu et al. 2012).

Table 8 Interaction Matrix table of the metals

Binary mixture components	Metal (*i*)	Metal (j)	Synergistic/antagonistic effect (effect on total additive effect)	Score suggested
Cr + Ni	Cr	Ni	Interaction seen positive, deficient in evidence. (Choudhury and Mudipalli 2008)	0.5
Cr + Pb	Cr	Pb	Interaction seen positive, deficient in evidence.(Choudhury and Mudipalli 2008)	0.5
Cr + Cd	Cr	Cd	Interaction is positive and evidence shows its likely chances to occur	0.75
Ni + Pb	Ni	Pb	Synergistic effect found with greater additive effect	1
Ni + Cd	Ni	Cd	Interaction is positive and evidence shows its likely chances to occur	0.75
Cd + Pb	Cd	Pb	Antagonistic effect found	− 1

During calculation of risk estimate of exposure of mixture of PM-associated metals, the HI (Interaction) values for most of the above cases were found to be lesser than 1 (Table 9), except for the case of exposure of mixture of metals involving Cr to children. These findings further indicate that efforts are required for reducing exposures of Cr to children. These findings are important, however, have been obtained using different assumptions. Although some studies have investigated combined toxicity of Cd, Pb, Ni, Cr and other metals to rat models (alone or in mixture) (Choudhury and Mudipalli 2008), the information could not be used for estimating health risks due to non-availability of toxicity benchmark for human receptor. Very few or no information is available for the risk estimation of toxic interactions of the metals in binary or ternary or tertiary sub-mixtures. Relevant conclusions were not provided for mixture toxicity both for short-term cases and long-term cases. So the effects of one metal on the toxicity due to other are not discussed properly. Synergistic effects or antagonistic effects have not been addressed properly. Such relative effects have been reported only in few medical field-related peer-reviewed journals which indicate that metals interact with each other and interfere with toxic effects. For example, Pb has been observed to show antagonistic effects in Zn absorption in children (Choudhury and Mudipalli 2008). However, detailed information on different metal pairs is not available. Further, the existing USEPA 2004a database does not contain the reference dose values of mixture of components (Cd–Pb, Ni–Cd, Cd–Pb, etc.), making it difficult to do risk assessment in the context of exposure of mixture of $PM_{2.5}$-associated metals. A detailed study is required for obtaining information on (i) interaction of effects of different metals, and (ii) guidelines data representing dose–response data of mixture of metals to human health so that uncertainty associated with risks estimate of exposures of mixture of PM-associated metals could be reduced (Fig. 5).

Lifetime Excess Cancer Risk (ECR)

The monthly average ECR values of exposure of Cr(VI) to both adult and children for both seasons were found to exceed the 1×10^{-6} value, indicating a cause of potential concern. For adults, the monthly average ECR values of Cr(VI) were found to be greater than 12×10^{-6} (winter seasons) and greater than 11.3×10^{-6} (pre-monsoon seasons). For children, the above-mentioned values were found to exceed 3×10^{-6} for winter season and 2.7×10^{-6} for pre-monsoon seasons. The calculation of cumulative risk (ECR) of exposures of $PM_{2.5}$-associated metals for 3 months indicated that adults have higher chances of getting risk of cancer than children of IIT Delhi campus which could be attributed to higher values of ADD for adults than for children. ECR value of exposures of $PM_{2.5}$-associated Cd or As was observed to exceed the limit value for certain months. However, the ECR value of combined exposure of two metals was observed to exceed the limit value for all months. Similar kind of observations have also been reported by the Izhar et al. (2016) study which estimated risks of getting cancer from PM_1-associated metals (Cr(VI) and Cd) in Kanpur (India).

Summary and Conclusions

The study presented a structured approach in incorporating $PM_{2.5}$-associated metal information in estimating the risk. Calculations were done to find the non-cancerous risk as well as cancerous risk of exposures of metals (alone or in binary mixture). Important findings of this study are present below:

1. Concentration values of $PM_{2.5}$ for three consecutive months of winter (i.e. December 2013, January 2014 and February 2014) and pre-monsoon (i.e. March 2014, April 2014 and May 2014) were determined. The average concentration values were found to be higher in the months of winter (188.5–256.7 $\mu g/m^3$) compared to months of pre-monsoon seasons (54.7–76.2 $\mu g/m^3$).
2. A total of 28 trace elements (Mg, Al, Si, S, Cl, K, Ca, Sc, Ti, V, Cr, Mn, Fe, Co, Ni, Cu, Zn, As, Se, Br, Sr, Mo, Cd, In, Sn, Sb, Ba, Pb) were detected on $PM_{2.5}$ samples using the ED-XRF method. Major metals, such as As, Cd, Cr, Pb, Ni, Zn and Cu, were detected (range

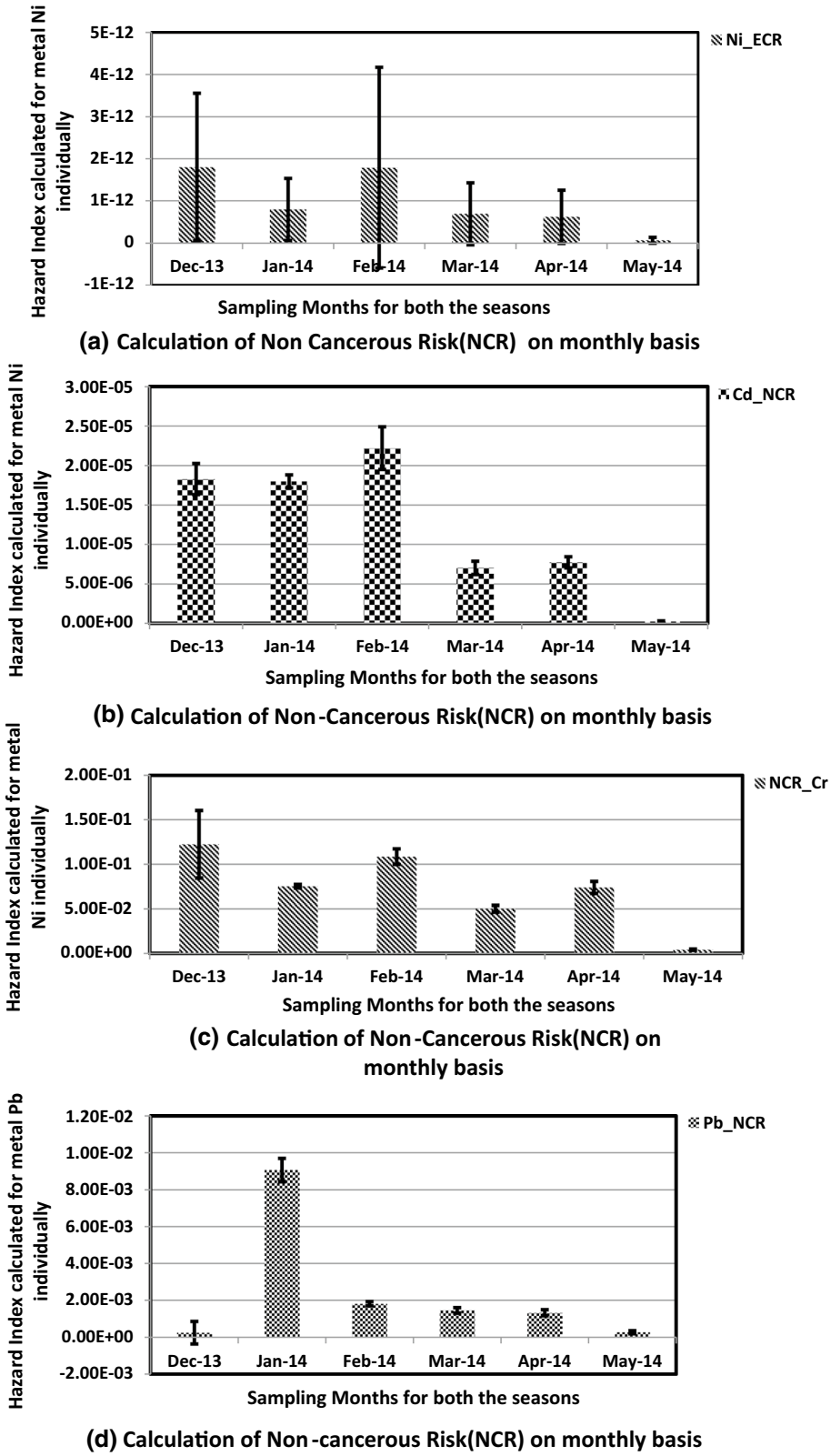
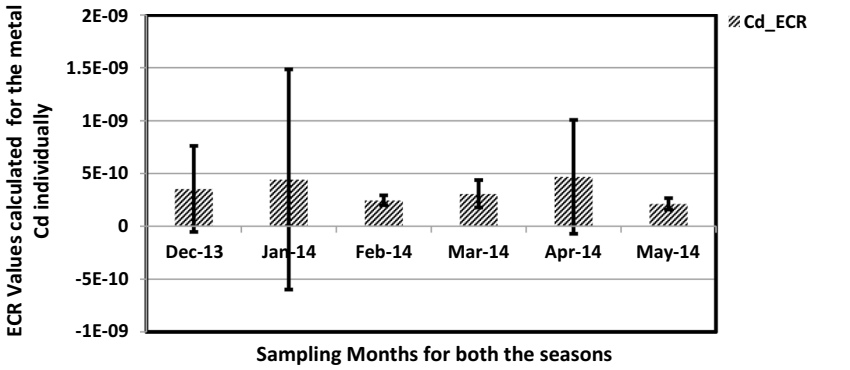
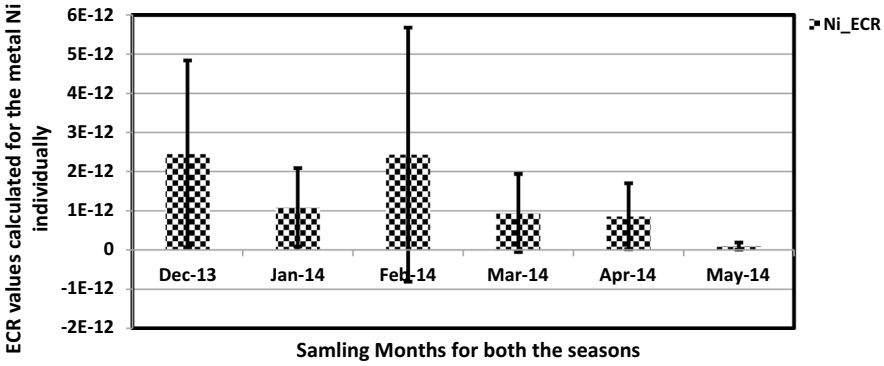


Fig. 3 Non-Cancerous Risk (NCR) calculated for adults for both the seasons (Winter and Pre-monsoon). **a** NCR for metal Ni, **b** NCR for metal Cd, **c** NCR for metal Cr, **d** NCR for metal Pb. (i) *Dec-13* month of December in the year 2013; the trend is followed for all the months in the series. (ii) *Y*-axis denotes average HI value calculated using data of 10 filter papers for a given month. (iii) Error bar denotes one standard deviation around mean value

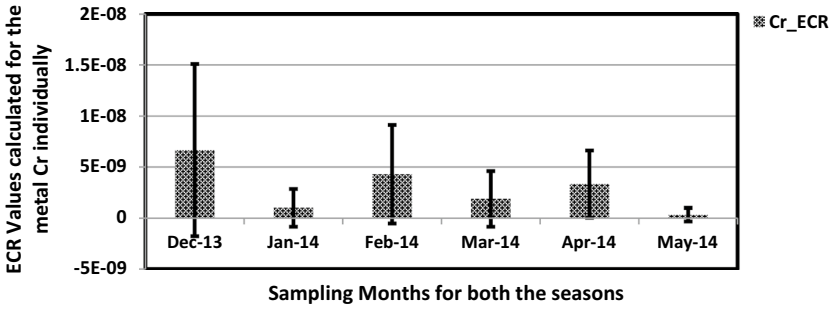
of concentration: 0.0095–0.68 μg/m^3 (winter season); 0.0082–0.44 μg/m^3 (pre-monsoon season). As major metals, such as Cd, Cr, Pb and Ni, were found to be in considerable amounts in all of the $PM_{2.5}$ samples tested and may pose health risks, this study used concentration values of these metals for estimating risks of getting non-cancerous effects and cancerous effects from expo-



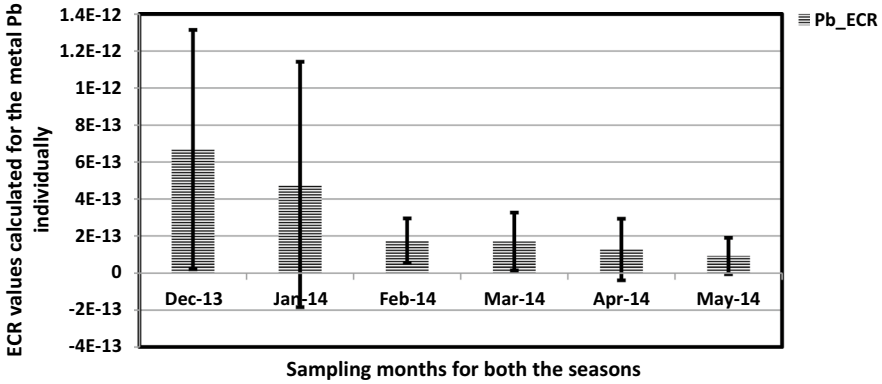
(a) Excess Cancer Risk(ECR) calculated on monthly basis



(b) Excess Cancer Risk (ECR) calculated on monthly basis



(c) Excess Cancer Risk (ECR) calculated on monthly basis



(d) Excess Cancer Risk (ECR) calculated on monthly basis

Fig. 4 Excess Cancer Risk (ECR) calculated for children for both the seasons (Winter and Pre-monsoon). **a** ECR for metal Pb, **b** ECR for metal Ni, **c** ECR for metal Cr, **d** ECR for. (i) *Dec-13* month of December in the year 2013; the trend is followed for all the months in the series. (ii) *Y*-axis denotes average ECR value calculated using data of 10 filter papers for a given month. (iii) Error bar denotes one standard deviation around mean value

Table 9 Results of calculation for the weight-of-evidence approach (HI value exceeding 1 are shown as black shaded texts)

Metals (binary mixture)	HQ_1	HQ_2	θ_{ij} $\theta_{ij}=\theta_{ji}$ (Eq. 7)	B_{ij} (score) ($B_{ij}=B_{ji}$) or $B_{ij}\neq B_{ji}$	HI (interaction)
Adult (winter)					
Cr + Ni	3.13E−01	5.98E−05	2.77E−02	0.5	3.13E−01
Cr + Pb	3.78E−01	6.80E−03	2.63E−01	0.5	3.85E−01
Cr + Cd	3.28E−01	2.88E−04	5.91E−02	0.75	3.29E−01
Ni + Pb	8.12E−05	7.64E−03	2.04E−01	1	7.72E−03
Ni + Cd	1.48E−04	6.76E−04	7.67E−01	0.75	8.24E−04
Cd + Pb	1.36E−04	2.79E−03	4.21E−01	− 1	2.93E−03
Adult (pre-monsoon)					
Cr + Ni	1.30E−01	1.52E−05	2.16E−02	0.5	1.30E−01
Cr + Pb	1.39E−01	3.58E−04	1.01E−01	0.5	1.39E−01
Cr + Cd	1.34E−01	5.62E−05	4.09E−02	0.75	1.34E−01
Ni + Pb	2.88E−05	3.30E−04	4.07E−01	1	3.59E−04
Ni + Cd	4.06E−05	1.45E−04	8.27E−01	0.75	1.86E−04
Cd + Pb	4.05E−05	1.08E−04	6.93E−01	− 1	1.26E−04
Children (winter)					
Cr + Ni	1.78E+01	3.39E−03	2.75E−02	0.5	1.79E+01
Cr + Pb	1.96E+01	1.04E−01	1.45E−01	0.5	1.97E+01
Cr + Cd	1.88E+01	1.78E−02	6.15E−02	0.75	1.88E+01
Ni + Pb	5.96E−03	1.67E−01	3.65E−01	1	1.73E−01
Ni + Cd	8.15E−03	4.07E−02	7.45E−01	0.75	4.89E−02
Cd + Pb	5.23E−03	2.93E−02	7.16E−01	− 1	3.46E−02
Children(pre-monsoon)					
Cr + Ni	7.42E+00	8.52E−04	2.14E−02	0.5	7.43E+00
Cr + Pb	8.37E+00	6.18E−02	1.71E−01	0.5	8.43E+00
Cr + Cd	7.67E+00	3.21E−03	4.09E−02	0.75	7.67E+00
Ni + Pb	6.70E−05	8.00E−02	2.45E−01	1	8.01E−02
Ni + Cd	2.26E−03	8.23E−03	8.22E−01	0.75	1.05E−02
Cd + Pb	1.48E−03	2.61E−02	4.51E−01	− 1	2.76E−02

sures of these $PM_{2.5}$-associated metals under a hypothetical exposure scenario.

3. The calculated hazard quotient (HQ) values of exposures of non-cancer causing $PM_{2.5}$-associated metals (Cd, Cr, Ni and Pb) were found to be smaller than 1 (range: 3.6×10^{-6}–0.17), irrespective of sub-population type and season studied, indicating no potential health risk. The total hazard values of exposures of PM-associated metals, calculated by adding HQ values of each month, indicated that exposure of $PM_{2.5}$-associated Cr in winter season pose more health risk to IIT Delhi children than IIT Delhi adult. During calculation of risk estimate of exposure of mixture of PM-associated metals, the HI (Interaction) values for most of the above cases were found to be lesser than 1, except for the case of exposure of mixture of metals involving Cr to children.
4. Estimates of excess cancer risk for exposures of four $PM_{2.5}$-associated metals (Cd, Cr, Ni and Pb) were calculated. Among different metals, the ECR values of exposure of Cr(VI) were found to exceed the 1×10^{-6} value irrespective of sub-population type and season studied, indicating no potential health risk. The calculation of cumulative risk (ECR) of exposures of $PM_{2.5}$-associated metals for 3 months indicated that adults had higher chances of getting risk of cancer than children in IIT Delhi campus. Exposures of $PM_{2.5}$-associated Cd or As resulted in ECR values, exceeding the limit value for certain months. However, the ECR value of combined exposure of these two metals exceeded the limit value, indicating a cause of concern during exposure of more than one type of PM2.5-associated metals.

Overall, findings of this study are important due to the following two important reasons. Firstly, it provided information on $PM_{2.5}$-associated metals and estimated risks of getting cancerous and non-cancerous health effects to adults and children for a longer period (i.e. 6 months) as compared to previously reported study by Khanna et al. (2015) which provided data only for 1-month period. Secondly, this study provided information on risk estimate of

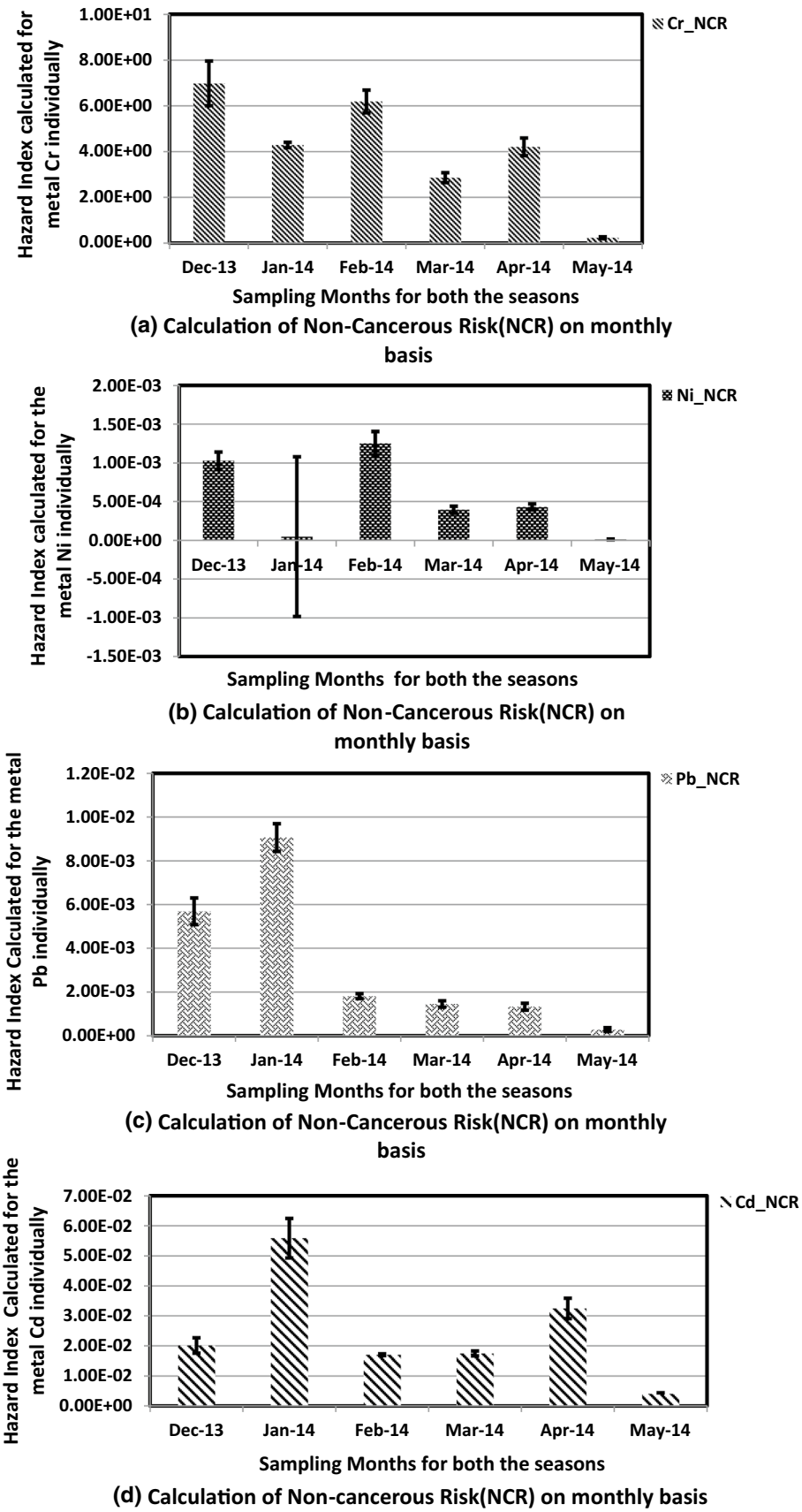


Fig. 5 Non-Cancerous Risk (NCR) calculated for children for both the seasons (Winter & Pre-monsoon). **a** NCR for metal Cr, **b** NCR for metal Ni, **c** NCR for metal Pb, **d** NCR for metal Cd. (i) *Dec-13* month of December in the year 2013, the trend is followed for all the months in the series. (ii) *Y*-axis denotes average HI value calculated using data of 10 filter papers for a given month. (iii) Error bar denotes one standard deviation around mean value

exposure of more than one $PM_{2.5}$-associated metals which is important for developing health protective policies using realistic exposure scenario. These two types of information are useful in providing insight about seasonal variation of $PM_{2.5}$-associated metals and its resulting impacts on health of adults and children. More efforts are required for systematically obtain information on relationship of toxicity of PM with its components (for example, PM-associated metals) in estimating risk of cancerous and/or non-cancerous base health effects. In this regard, hazard index (i.e. summation of hazard quotient values for inhalation exposure of different metals) can be calculated and used to estimate allowable concentrations of PM as per maximum allowable value of heavy metals. Field and laboratory studies are required for collecting above-mentioned information so that effect of mixture of $PM_{2.5}$-associated constituents can explicitly incorporated in the risk estimation process.

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