Assessment of PM2.5 bound Heavy Metals and Associated Health impact- A case study in Warangal City, India

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Abstract

$PM_{2.5}$ in the atmosphere is widely reported, but few studies have focussed on the metal-bound particle concentration of $PM_{2.5}$ in urban areas. The present study aims to quantify Heavy Metals (HM) concentration associated with $PM_{2.5}$ for the urban region of Warangal in the State of Telangana, India. A Respirable Dust sampler was used to collect the air samples during the study period and the samples were analysed for specific heavy metals [Iron (Fe), Zinc (Zn), Nickel (Ni), Cadmium (Cd) and Copper (Cu)] using Microwave Plasma Atomic Emission Spectroscopy (MP-AES). Average $PM_{2.5}$ values were found to be above the annual limit (40 $\mu g/m^3$) set by the National Ambient Air Quality Standard (NAQS) of India and range from 41.58 to 58.21 $\mu g/m^3$for the region studied. Slightly higher concentrations of Zn and Fe were observed as compared to Cu, Ni and Cd. The order of heavy metal based on the concentration levels was as follows: Zn>Fe>Cu>Ni>Cd (higher to lower concentration). Analysis of enrichment factor showed that the Zn, Cu and Cd fall in the highly enriched element category.

Based on health risk assessment for intake pathways, the risk of exposure was found to be in the following order: Ingestion>Dermal>Inhalation. Non-carcinogenic and carcinogenic risks for children and adults were found to be negligible. Source identification studies based on Concentration Weighted Trajectory (CWT), Potential Source Contribution Function (PSCF) and cluster analysis results showed that the western and north-western regions of India are more dominant contributors. Furthermore, cluster 4 was reported to have high significance (27.11%) and dominant contributions of trajectories were observed from the regions of Maharashtra, Chhattisgarh, Rajasthan and Madhya Pradesh. The results of the study can help policy makers implement mitigation measures and formulate suitable regulations to reduce the health risk associated with $PM_{2.5}$ and heavy metals in the atmosphere.

Keywords: $PM_{2.5}$, Heavy metal concentrations, Enrichment factor, Health impact, Trajectory Analysis.

Introduction

Air pollution assessment based on $PM_{2.5}$ concentration focuses on particles with an aerodynamic diameter of less than 2.5 micron[79]. The $PM_{2.5}$ sources include both anthropogenic and natural sources. Millions of people are affected every year as a consequence of air quality. India is one of the country air pollution accounts for a major impact on human health. The ever-increasing anthropogenic activities being carried out in the developing economy of India are leading to high levels of air pollution. Factors such as the number of vehicles, low-level fuel use, poorly identified transport systems, low land use pattern and inefficient environmental legislation[11,33] are contributing to the poor air quality.

However, few studies on particulate matter (PM) transport and chemical composition have been reported in the literature pertaining to different atmospheric regions[18,36,65,73]. Identification of sources with the help of receptor models are more popular in India as compared to the use of other models like trajectory based models[4]. Trajectory based approaches and models are gaining popularity because of their advantages due to the identification of the potential regions more accurately at receptor location[43,50,80,103].

For most of the Indian cities with a population above 2 million, the concentration of air pollutants has been reported to be above the limits prescribed by World Health Organization (WHO). Furthermore, heavy metals in Kanpur were almost 5–10 times higher than levels in European cities[13,81]. Finer particles in ambient air are uniformly distributed compared to coarse particles and the higher in metal concentation[8]. Literature review shown that the seasonal variations in PM10 concentrations indicate higher levels in winter compared to that of other seasons[1,45,61,69,77,82].

Metals like Fe, Zn, Cu, Mn, etc. are reported in the ambient atmosphere depending on the major activities in nearby regions and sometimes by long-range transport due to winds[5,92]. The wind speed and wind direction are major contributors to the dispersion of pollutants from the sources to the receptor[66] and their re-suspension in air[41,101]. A study on blood samples revealed the presence of Cu in people working in related industrial areas[20].

A trace of $PM_{2.5}$bound HM causes serious health impacts even at low-level concentrations[2,54]. HM's are non-biodegradable and can enter the food chain to cause

substantial damage to the ecosystem[39,82]. Most of the heavy metals are associated with industrial activities and anthropogenic sources[19]. These metals are mainly found in industrial areas and heavy traffic locations. Heavy metals affect human health and cause diseases like lung cancer, asthma, diabetes, autoimmune disease, pulmonary diseases etc. Metals are identified and reported by many researchers[6,22,30,35,68]. Evaluation of health risk associated with HM is important in order to assess cause-effect relationships and the source regions[7]. The fact that the health problems in India due to HMs are constantly increasing over the year[21], it was the one of the main motivations for the present study.

The present study focus on the assessment of $PM_{2.5}$concentrations and heavy metal species bound to $PM_{2.5}$ in the study region. The knowledge of heavy metals bound to $PM_{2.5}$ is critical for formulating air pollution control strategies and framing regulation[31]. Hence, assessment of the variation in $PM_{2.5}$ and heavy metals in an urban environment has also been attempted in the present study. Subsequently, the enrichment factor analysis was carried out to indicate whether the sources were of natural or anthropogenic origin. The health impact analysis was carried out to reveal the carcinogenic and non-carcinogenic impacts on children and adults due to heavy metals. Source regions were also identified based on back trajectory analysis.

Material and Methods

Study area: The Warangal city located in the southern part of India. The population of 811,844 by the 2011 census and is the second-largest municipality in Telangana, next to the State capital of Hyderabad. The air sampling location was setup at the latitude of 17.982715 °N 18.0 °N 79.58 °E. The Warangal city records an average temperature of 34.5°C during summer and an average temperature of 22.4°C during winter. Tropical climate prevails in the study area with normal annual average precipitation of 945 mm. Though no streams pass through the city, however, Warangal has three historic lakes namely, Bhadrakali lake, Dharma Sagar lake and Waddepally lake. Air samples were collected at the institute campus by adopting prescribed procedures. The index map of the study area with sampling location is presented in fig. 1.

The sampling location was situated on the roof (~15 m above ground level) of the Chemistry Department building which is located in a mixed educational and residential area with no major industrial sources surrounded by 3-4 km radius. Warangal - Hyderabad National Highway is about 300m to North of the sampling site.

Sample Collection: $PM_{2.5}$ particles were gathered using a respirable dust sampler. For proper control of $PM_{2.5}$, flow win impactor and silica gel were used and filter paper was used to control the coarse particles. The dust sampler flow rate was 16.67 ± 5% liters per minute (LPM) and an accuracy of ±2% was maintained throughout the sampling period. The daytime and night time (12 hours) samples were collected separately. A total of 130 samples were collected (September 2018 to February 2019) the duration of six months. A glass-fiber filter paper was used for filtration of samples[6,12,76]. The weight of dry filter paper before and after sampling was recorded with a help of mass balance and subsequently, $PM_{2.5}$ concentrations were calculated. A desiccator was used to control the influence of atmospheric moisture levels on filter papers. The collected samples were stored in the refrigerator before the metal analysis using the MP-AES technique.

MP-AES heavy metals analysis: The glass-fiber filter papers were subjected to acid digestion with 20 ml concentrated HNO_3 solution for 2hours on the hot plate[9,44].The solution was maintained at 180°C until the acid got evaporated completely. The residual liquid was then filtered through a 0.22µm Teflon filter and diluted to 100 mL with Milli-Q water (resistivity 18.2MΩ) for subsequent elemental analysis. A blank filter was digested following the same procedure as the sample filters. The blank filters were measured separately and then these values were subtracted from the total weight after filtration to get the weight of the retentate. The reference standard solutions for MP-AES were prepared for calibration to find the levels of metals[44] in the samples.

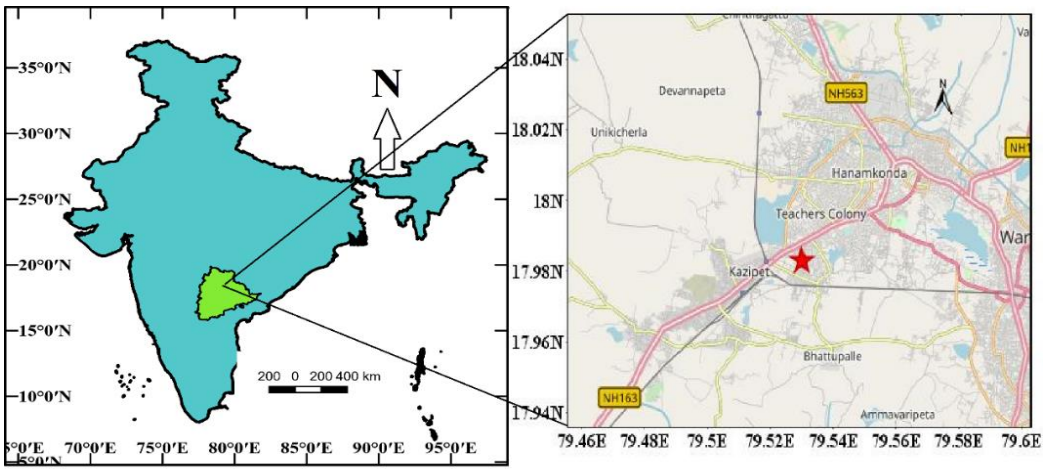


Fig. 1: Study area

Enrichment Factor: The enrichment factor (EF) is generally used to identify the pollutant originating from the earth crust and non-crustal sources. Equation 1 may be used for the determination of EF. In most studies[22,102], Fe and Al are used as the reference elements as these elements exhibit stable chemical properties and are associated with anthropogenic activities. Therefore, in this study, Fe was used as a reference element. The standard crustal composition reported by Rudnick and Gao[72] was used in the present study. However, there is no thumb rule for selection of reference element.

$$EF = \frac{((\frac{X}{ref})sample)}{((\frac{X}{ref})crust)} \quad (1)$$

where X is the concentration of the element being examined and "ref" is reference element with respective sample and crust. Relation between EF and the level of enrichment is as follows: EF < 2 minimal enrichment, EF = 2–5, moderate, EF = 5–20, significant enrichment, EF = 20–40, very high enrichment, EF > 40, extremely high enrichment [102].

Health Risk Assessment: The health risk assessment for adults and children was analysed based on heavy metals associated $PM_{2.5}$. Exposure to ambient metals affects inhalation, ingestion and skin. In the present study, exposure assessment methodology developed the U.S. EPA[90,91] has been adopted. Ambient heavy metals are inhaled through the nose and mouth, ingested through food and absorbed through skin pores. Risk analysis methodologies have been reported in a few studies[37,40,96] and the health risk assessment framework includes the identification of pollutants, the exposure assessment based on dose-response assessment.

(i) Exposure dose assessment: U.S. EPA considers Average Daily Dose (ADD) (mg/kg/day) for exposure dose assessment of the risk posed by metals to mankind. The present study computed the potential exposure through three different pathways for each metal separately with the help of three equations (Eq. 2, 3, 4) to estimate ADD[26,40,49].

$$ADD_{ing} = \frac{C \times IngR \times EF \times ED \times CF}{BW \times AT} \quad (2)$$

$$ADD_{der} = \frac{C \times SA \times AF \times ABS \times EF \times ED \times CF}{BW \times AT} \quad (3)$$

$$ADD_{inh} = \frac{C \times InhR \times EF \times ED}{PEF \times BW \times AT} \quad (4)$$

where ADD_{ing} is average daily dose units ingestion (mg/kg/day), ADD_{der} is average daily dose dermal contact (mg/kg/day), ADD_{inh} is average daily dose inhalation (mg/kg/day), C is heavy metal concentration (mg/kg), IngR is ingestion rate (mg/day) 30_adults, 60_children[89], EV is Events frequency occurring every day, EF is exposure frequency, 180 days for a year, ED is exposure duration for 24 years for adults, 6 years for Children, CF is conversion factor 10^{-6} kg/mg, BW is Body weight - 70kg for adults, 15kg for children, SA is Skin surface area - 5700 cm^2 for adults, 2800cm^2 for children, AF is adherence factor of soil to skin - 0.07 (mg/cm^2/event) for adults, 0.2 (mg/cm^2/event) for children, ABS is dermal absorption fraction - 0.001[87], InhR is inhalation rate_- 7.63m^3/day for adults, 20m^3/day for Children, PEF is particle emission factor - 1.36 X 10^9 m^3/kg[90] and AT is averaging time for Non_carcinogens (AT = ED X 365 days/year) and Carcinogens (AT = 70 year X 365 days/year)[24].

In the present study, standard parameter values as given by U.S. EPA[87-91].

(ii) Non-carcinogenic health risk: The non-carcinogenic health risk assessment was calculated based on hazard quotient (HQ) and hazard index (HI). HQ and HI were determined using eq. 5 and 6 respectively. The reference dose (Rfd) of metals is presented in table 1[24,49,52]. HQ value <1 indicate that there was no significant health impact while HQ value of >1 indicates an adverse effect on human health[104]. Hazard index (HI) is the sum of all the hazard quotients[26,104]. HI value of less than 1 indicates that there is no significant non-carcinogenic impact while if the HI value was >1, then there are significant chances of non-carcinogenic impact[104].

$$HQ = \frac{ADD}{RfD} \quad (5)$$

$$HI = \sum_1^n HQ_i \quad (6)$$

(iii) Excess Cancer Risk (ECR): Excess cancer risk (ECR)is a measure of the incremental cancer risk over the lifetime[40]. ECR is calculated by eq. 7. The inhalation unit risks of the metals are provided by U.S. EPA IRIS (Integrated Risk Information System)[86]. Zero value indicates that there is no cancer risk while higher values indicate the higher chances of cancer risk. The U.S. EPA methodology provides only the inhalation unit risk; however, other pathways and associated risks are not provided. When the value falls within the range of 10^{-6}-10^{-4}, carcinogenic risk was minimal[40,70].

$$ECR = \frac{C \times ET \times EF \times ED \times IUR}{AT} \quad (7)$$

where C is pollutant levels in mg/m^3, ET is the exposure time taken as 8 h/day, EF is exposure frequency 180 days for a year, ED is exposure duration: 24 years for adults, 6 years for children, IUR is inhalation unit risk in mg/m^3 and AT is average time for carcinogens 70 year_365 days/year 24 hours/day.

Hybrid Single-Particle Lagrangian Integrated Trajectory model (HYSPLIT) Backward Trajectory: Global Data Assimilation System (GDAS) from the National Oceanic and Atmospheric Administration (NOAA)[93] data was used as input to the HYSPLIT model[23]. The 7-day back trajectories at heights of 500 m,1000 m and 1500 m were considered as a surface. HYSPLIT modeling systems are very effective for atmospheric trajectory and dispersion

calculations[83]. These back trajectory model also compute the pollutant transport, chemical transformation and deposition of pollutants and hazardous materials.

Concentration Weighted Trajectory (CWT)Analysis: CWT analysis shows the long-range pollutants at the receptor site and the strength of the source[15]. CWT model can locate the regional sources that can affect the receptor region. In CWT analysis, each cell in the grid is assigned a weight by averaging the pollutant concentration[78]. The trajectory endpoint time in the grid cells has been weighted by the corresponding $PM_{2.5}$ trajectory. The concentration of each grid cell was calculated using eq. 8[14]. However, the spatial resolution was taken as 0.5×0.5° to find the source paths.

$$CWT_{ij} = \frac{\sum_{l=1}^{L} c_l \tau_{ijL}}{\sum_{l=1}^{L} c_l \tau_{ijl}} \quad (8)$$

where C_l is the observed mean concentration of pollutant, l denotes the associated backward trajectory, τ_{ijl} each segment end point in 0.5x0.5 grid cells (i, j) and L presents the total number of backward trajectories considered in this study.

Potential Source Contribution Function (PSCF) Analysis: The Potential Source Contribution Function (PSCF) was used to indicate the probability of impact of sources on the receptor[59]. PSCF values may be calculated using the following equation (Eq. 9).

$$PSCF_{ij} = \frac{M_{ij}}{N_{ij}} W_{ij} \quad (9)$$

where $M_{ij}$is the total number of back trajectories with grid cell (i, j), N_{ij} is the total number of back trajectories with respective each grid cell (i, j) and W_{ij} denotes the weighting function of back trajectory segment endpoints in a grid cell (i, j)[27]. A weight function (W_{ij}) was established for each grid to overcome the uncertainty in N_{ij}[100].

Cluster Analysis: The clustering technique shows the average trajectory paths for each cluster. K-mean cluster technique is extensively used for studying air mass trajectories representing the pollutant pathways[32]. In the present study, K-mean cluster was adopted for clustering the back trajectories.

The source identification was done based on CWT, PSCF and cluster analysis. MeteoInfo tool, a GIS-based software, was used for meteorological data visualization and analysis[95]. The PSCF, CWT and cluster analysis were analyzed using the plugin TrajStat[95] for conducting source analysis.

Results and Discussion

Variation of PM2.5 Concentration: The monthly mean mass concentrations of $PM_{2.5}$ are presented in fig. 2(a). During the study period, the monthly mean $PM_{2.5}$ concentrations were found to be in the range of 8.3-29.6 µg/m^3 with a highest daily concentration of 58.3 ug/m^3 and the lowest daily concentration of 4.7 ug/m^3. During the monsoon period, maximum, minimum and mean concentrations of $PM_{2.5}$ were 41.6, 24.9 and 29.6 µg/m^3 respectively. Concentrations of $PM_{2.5}$ were observed to be higher during the weekend as compared to the concentration during weekdays. This is perhaps due to the proximity of the highway to the monitoring location and the fact that the highway caters to higher volumes of traffic during the weekends compared to weekdays.

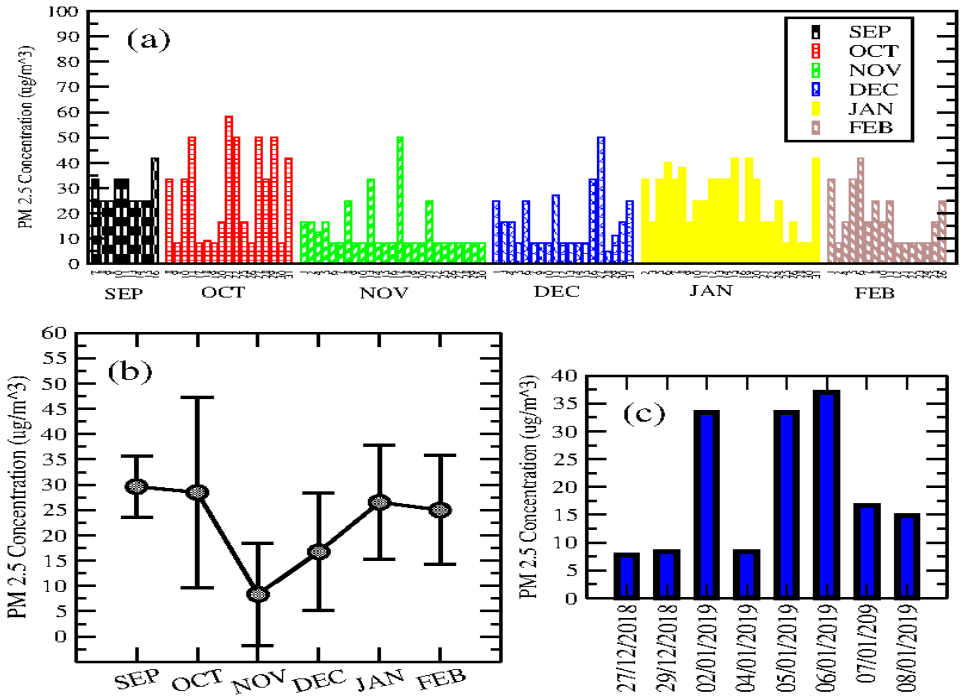


Fig. 2: (a) Diurnal Concentration of PM2.5 (b) Monthly variation of $PM_{2.5}$ (c) Night time concentration of $PM_{2.5}$

During the post-monsoon period, maximum, minimum and mean concentrations of $PM_{2.5}$ were 58.3, 8.33 and 19.5 µg/m^3 respectively. During the winter, maximum, minimum and mean concentrations of $PM_{2.5}$ were 49.9, 4.7 and 21.2 µg/m^3 respectively (Fig. 2b). As per the National Ambient Air Quality Standards (NAAQS), the $PM_{2.5}$ concentrations were below the standard levels (40 µg/m^3 for $PM_{2.5}$ annually and 60 µg/m^3 on a 24-hour period). The $PM_{2.5}$ concentration during the night-time concentration is usually considered a background levels as presented in fig. 2c. The night-time concentrations were observed to be lower than daytime concentrations. The night maximum, minimum and mean concentrations were 38.45, 8.33 and 20.51 µg/m^3 respectively. Wind profiles and temperature influence the movement of particles while the traffic volumes are generally minimum during the night.

The results of the present study were similar to the reported values in other places dominated by vehicular and urban activities[22,46,61]. In Nagpur city, the concentration $PM_{2.5}$ value was 52 µg/m^3 due to road dust on highways[12]. At Dongargarh, Central India, the $PM_{2.5}$ concentration was reported to be 64 µg/m^3 mostly due to vehicular emissions[1]. In Kolkata city, $PM_{2.5}$ concentration was reported as 83 µg/m^3 at a location where construction activities and road dust were major contributors to air pollution[22]. There were 45 µg/m3 of $PM_{2.5}$ in Hyderabad region[35].

In Agra city, $PM_{2.5}$ was 104.9 µg/m^3 mainly due to industrial emission and anthropogenic activities[51]. While all the cities have different pollution levels arising from different sources, the common sources of $PM_{2.5}$ in the urban atmosphere are road dust, construction activities, small industries and vehicular emissions.

Table 1
Heavy metals bound to $PM_{2.5}$ in air reported in literature for Indian cities

S.N.	Region	Heavy metals
1.	Agra, India[51]	Pb,Zn,Ni,Fe,Cr,Mn,Cu
2.	Dongargarh, Central India[1]	Fe > Zn >Pb> Cu > Ni > Cr > Cd
3.	Nagpur[12]	Zn > Fe >Pb> Ni > Cd > Cr 2006
4.	Hyderabad[44]	Al,As,B,Ba,Ca,Cr,Cu,Fe,K,Mn,Na,Ni,Pb,Zn
5.	Kolkata[21]	Zn Cr,Ni,Mo,Cu,Sn,Sb,V,Co,Cd and Pb (Ca,Al,Mg,Sc,Ti,Mn and Fe pm10)
6.	Tamilnadu[92]	Zn, Fe, Cu, Pb, Ni and Cr) Cd
7.	Agra[55]	Fe,Pb,Ni,Cr,Cd,Cu,Mn
8.	New Delhi[65]	Si,Ca,Fe,Ti,Mn.Ni,Cu,Zn,Pb
9.	Lucknow[64]	Fe>Pb>Ni>Cu>Cr>Cd
10.	Bolpur[31]	Mn, Zn, Cd, Pb, Ni, Co,

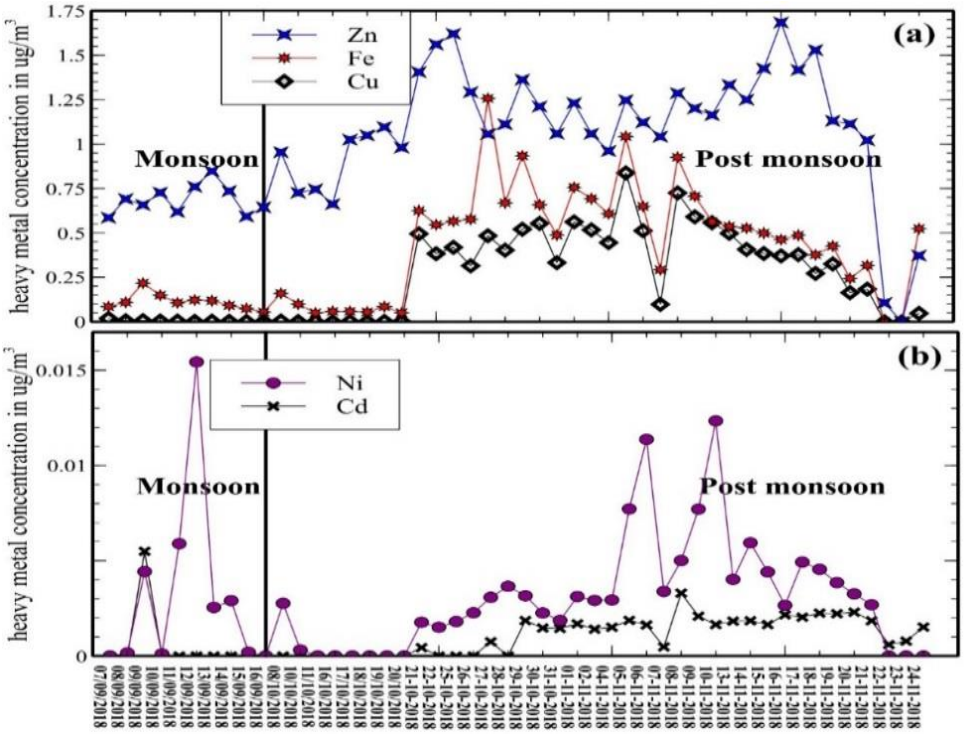


Fig. 3: Variation in heavy metal concentration

Concentration of Heavy Metals: The samples collected during the study period were analyzed for heavy metals using MP-AES and are presented in fig. 3(a) and (b). The concentrations of heavy metals bound with $PM_{2.5}$ were in the order: Zn>Fe>Cu>Ni>Cd (high to low concentrations). Some metals like Zn, Cu and Fe were higher when compared with other metals. Average concentrations of Zn, Cu and Fe were 1.68, 0.83 and 1.25 µg/m^3 respectively. As shown in table 1, several researchers for investigations carried out in India observed metals in ambient air bound to PM 2.5. For obvious reasons, the concentrations of metals vary with location depending on the sources dominating in that location. Metallic contaminants like Fe, Cu, Ca, Zn, Pb etc. are generally released predominantly from anthropogenic sources in inland regions[58].

Long-range transport of polluted air masses with possible changes at receptor location on trace metal concentrations[84] have been reported. The overall variations of heavy metals during the study period are presented in fig. 4. These metals are perhaps released from automobiles, construction activities and other urban activities. Literature suggests that Fe, Si, Al and Ti originate from the earth's crust[65]. Zn and Pb concentration levels have been shown to correlate well with non-exhaust traffic emissions[61,67], industrial sources[103] and solid waste burning[94].

Ni et al[60] concluded in their study that open biomass burning and industrial pollution result in Fe, Zn, Pb and K emissions. Cd, Cr, Ni and Pb are associated with industrial sources whereas Zn and Cu are associated with traffic emissions. Road dust[55,85] is commonly associated with high concentrations of Cd and Pb. Mn, Zn, Pb, Fe and Cu emissions from lubricants oil, brake pads and tyres are the main sources[29,34,62,63,94]. The concentration of Zn and Fe is attributed to industrial emission, crustal trace element concentrations and fluxes due to re-suspension of dust released during traffic activities and soil erosion[28]. Increased concentration of Cd was reported to be contributed by solid waste combustion, refinery and fossil fuel burning[3,17]. Further studies are needed to fingerprint the sources and their apportionment.

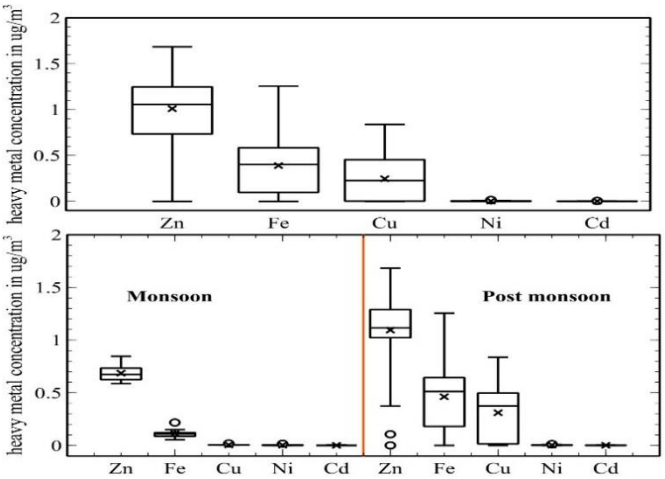


Fig. 4: Monsoon and Post monsoon Seasonal change in heavy metal concentration

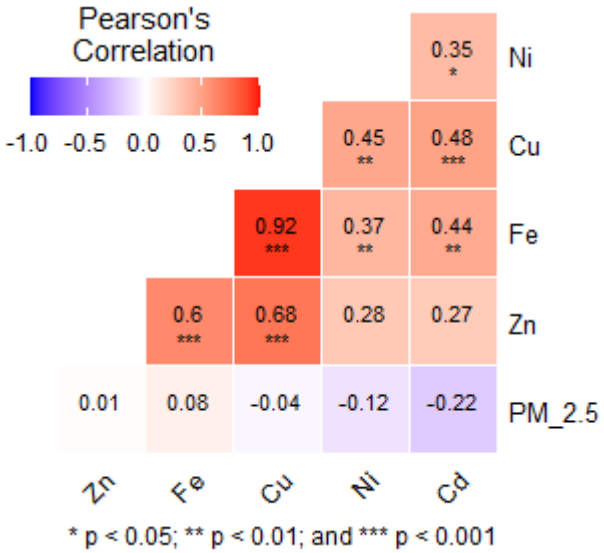


Fig. 5: Pearson's correlation between metals and PM $_{2.5}$

The Pearson's correlation analysis shown in fig. 5 indicates that Fe and Cu were strongly correlated in the present study when compared with other metals. Other significant correlations exist between Zn and Cu and Zn and Fe and are all predominantly related to traffic emissions. Fang et al[25] have reported similar findings. The main source was identified as vehicular pollution[10]. Fe is generally associated with rock weathering and dust from minerals[16,97], however, in the present study, it was not a dominating source.

Enrichment Factor (EF): The enrichment factor indicates whether the source of emission is natural or anthropogenic. EF values for the present study have been depicted in fig. 6 and were all found to be above 10. EF value for Ni falls in the moderately enriched bracket indicating nearby industries as possible sources. Zn, Cu and Cd have EF value greater than 100 and hence fall in the highly enriched category. Zhang et al[101,102] reported similar results of significant enrichment for Zn. These emissions are perhaps due to combustion and related activities originating from the automobile and industrial sectors. Ambade[1] had reported that the Ni, Cu and Cr are emitted from anthropogenic activity while Fe and Zn are generally emitted from natural sources.

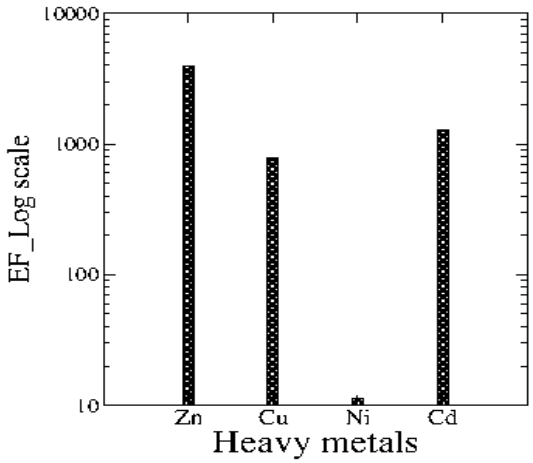


Fig. 6: Enrichment Factor for heavy metals

Health Risk Assessment

(i) Exposure Dose Assessment: The exposure assessment evaluation based on ADD of all pathways is presented in fig. 7. The results indicated a similar variation in ADD for all exposure pathways in both children and adults. The metal Zn exhibited higher values for all the three exposure pathways, while Cu and Fe showed moderate ADD values. Ni and Cd exhibited negligible ADD values for all the pathways. The total average daily dose is presented in fig. 8(a) and the order of impact of exposure of metals may be observed to be as follows: Ingestion> Dermal > Inhalation. Literature reports significant impact of resuspension of dust particles and construction activities on ADD values[48,56]. The ingestion pathway of HMs is a dominant route of exposure followed by dermal contact[104].

(ii) Non-Carcinogenic Health Risk: The HI index shows the sum of HQs and the values obtained in the study are presented in fig. 8. As RfD values for Fe, Se and Ca metals have not been specified by USEPA, HQ and HI for Fe were not calculated. The HQ results obtained were in the following order: HQ_{ing}>HQ_{der}>HQ_{inh}. The similar trend in reported on health risks posed by particle-bound metals at IIT Khanpur[42]. For HQ_{der}, the following order of metals was observed: Cd > Zn > Cu>Ni, whereas for HQ_{inh}, order of metals was: Cu > Zn > Ni > Cd for both children and adults. Though the values were slightly different from one another, the HQ_{ing} trend in both adults and children (Fig. 8b) was observed to be as follows: Cu> Zn >Cd> Ni. HQ index was observed to be below 1 for all pathways.

The results signify that negligible non-carcinogenic threat is posed to children and adults since HI (Fig. 9) values are below 1. However, higher risk has been reported when we consider injection pathway. Also, the risk for children was more compared to the adults. Izhar et al[42] came to the conclusion that there was no appreciable health risk since they found similar HI values (below 1) in their study. Ni, Cd, Co, Cr and Pb are considered carcinogenic metals while Fe, Cu, Zn and Mn are considered non-carcinogenic metals and generally originate from anthropogenic activities[64].

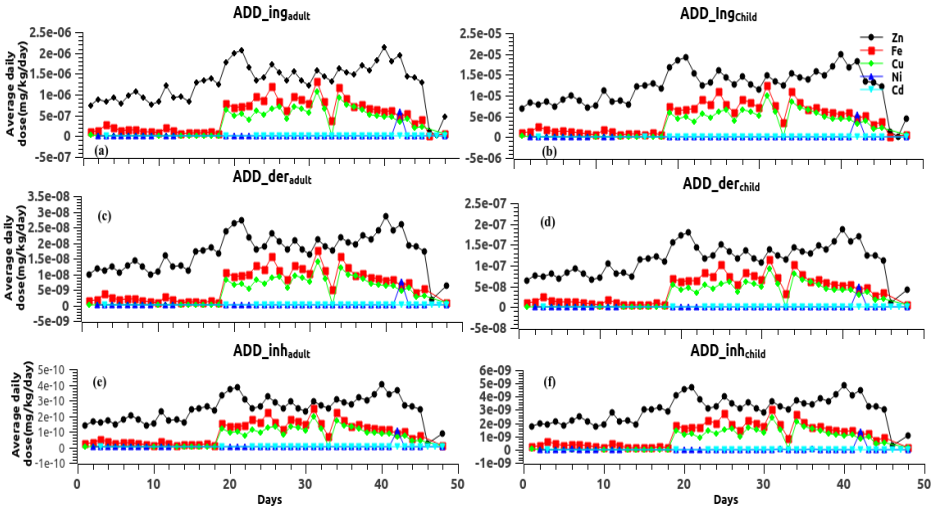


Fig. 7: ADD for children and adults for ingestion, dermal and inhalation pathways

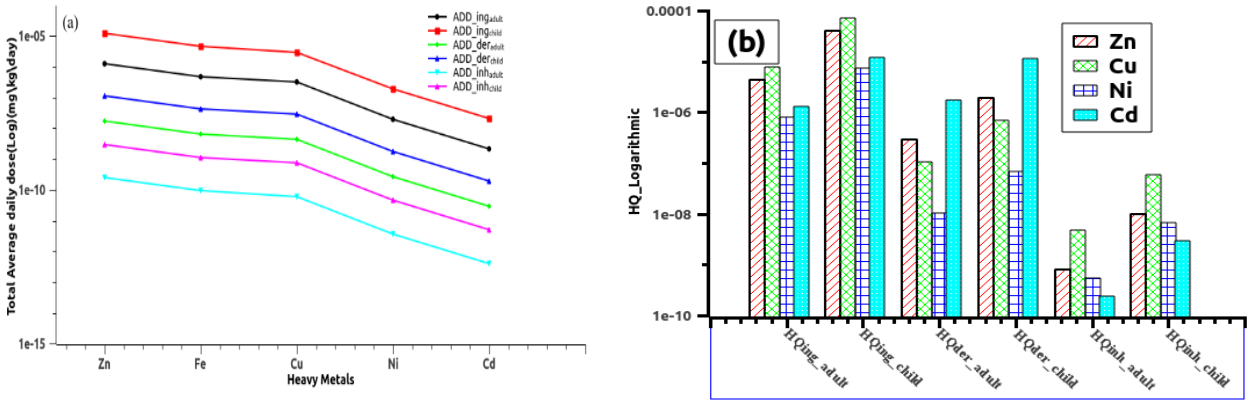


Fig. 8: (a) Total Average Daily Dose, (b) HQ_index for heavy metals

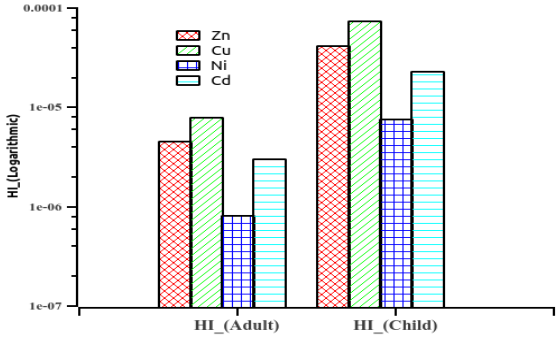


Fig. 9: Hazard index associated with heavy metals for adults and children

(iii) Excess Cancer Risk Assessment: Results of the study show that the concentration of Ni was higher than Cd when considering the two carcinogenic elements for children and adults. The cancer risk in adults with Ni was 1.02 x10^{-6} and with Cd was 6.12 x 10^{-7}. The ECR in case of children with Ni and Cd was 1.18 x 10^{-6} and 7.15x10^{-6} respectively. The results obtained in the study were within the acceptable limits of 10^{-6} to 10^{-4}. Similar trends in non-carcinogenic and carcinogenic risk assessment for both adults and children were reported in Nanjing, China[40]. The non-carcinogenic and carcinogenic risks are in general reported to be higher for children than adults[21,40,56,75,98]. Further studies are required to establish the combined risk associated with metals bound to $PM_{2.5}$.

Source Identification: The back trajectory analysis was used in the present study to trace the path of the air mass arrivals at the receptor location. 7-days back trajectory involves accessing the source regions at the receptor location. The trajectories for September, October and November are presented in fig. 10. The trajectory for September month indicates the influence of the Western region of India, while the trajectories of October and November were influenced by Indo Gangetic Plain (IGP). Few trajectories were observed from the Bay of Bengal (BOB) in the month of October. The changes in the trajectories can be attributed to changes in season and to the variations in wind and temperature profiles.

However, most trajectories were observed to be from the north-eastern (NE), northern (N) and western (W) regions of India. HYSPLIT back trajectories analysis has also been used by other researchers for the identification of source regions at receptor location[99].

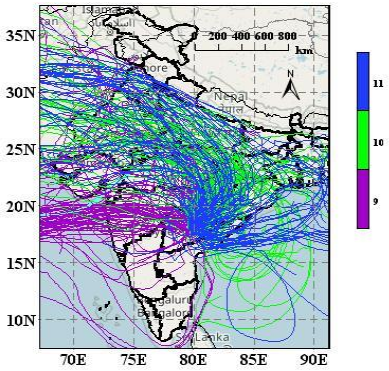
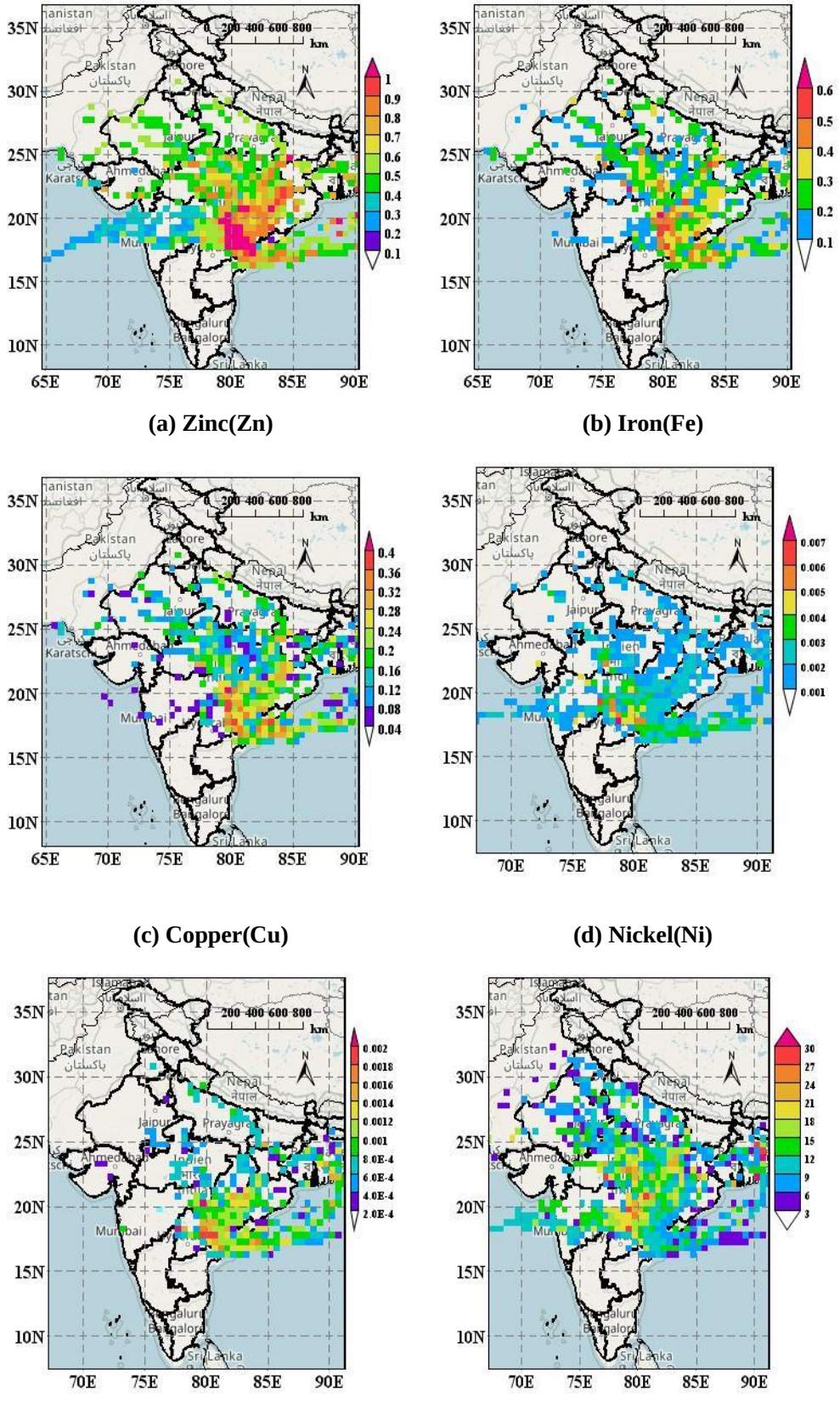


Fig. 10: HYSPLIT back trajectory for themonths of September, October and November

CWT Analysis: The CWT analysis shows the contribution of the majority of trajectory regions at receptor or sampling location. The heavy metal transformations at the receptor location may be observed in fig. 11. The trajectory when coupled with respective metals indicates the source of the trajectory. Results show that Zn is contributed from NE regions especially Odisha and Chhattisgarh and parts of Central India (mainly Madhya Pradesh). This can be attributed to significant coal mining and biomass burning in these regions. Fe, Cu, Ni and Cd are contributed by local anthropogenic activities and dust resuspension results from wind currents.

Mukherjee and Agrawal[57] reported a significant contribution to $PM_{2.5}$ from north-western (NW) regions of India using CWT and cluster analysis. Rai et al[72] identified that the pollutants moving towards the receptor, (Darjeeling in their case) originate mainly from Nepal apart from the IGP and the BOB.



(a) Zinc(Zn) **(b) Iron(Fe)**

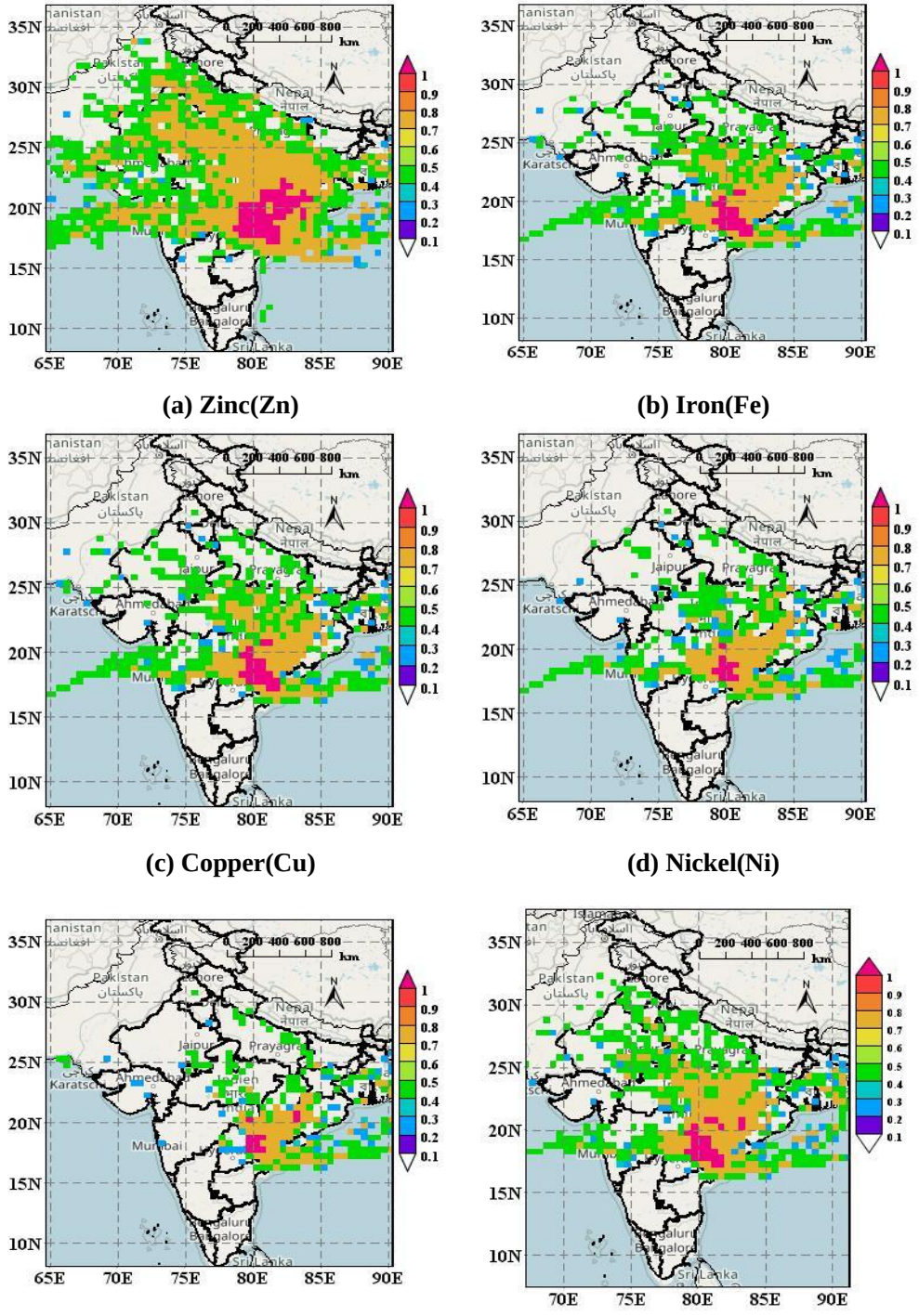
(c) Copper(Cu) **(d) Nickel(Ni)**

(e) Cadmium(Cd) **(f) PM $_{2.5}$**

Fig. 11: CWT analysis for: (a) Zn (b) Fe (c) Cu (d)Ni (e) Cd (f) PM $_{2.5}$

Potential Source Contribution Function (PSCF): NE regions (Odisha, Chhattisgarh and Jharkhand) and IGP regions contribute Zn and the same was evident in WPSCF analysis (Fig. 12). As these regions are dominated by mining activities and the burning of fossil fuels, it is likely that Zn contributions will be significant. Similar findings have been reported by Chinnam et al[17]. Fe, Cu and Ni were mainly contributed by nearby local sources. The NE coastal region was a moderate contributor (0.7-0.8 significance levels) for all metals.

The majority of the potential source regions fall upwind towards the receptor location. It may be noted that the transformation of pollutants from upwind to downwind causes a trans-boundary particles moment from most of the east coastal states and some central states. The transport and accumulation of pollutants effceted by the based on the geographical location of the existing region Chengdu region and Sichuan region[47,71].



(a) Zinc(Zn) **(b) Iron(Fe)**

(c) Copper(Cu) **(d) Nickel(Ni)**

(e) Cadmium(Cd) **(f) PM $_{2.5}$**

Fig. 12: PSCF analysis for:(a) Zn (b) Fe (c) Cu (d) Ni (e) Cd (f) PM $_{2.5}$

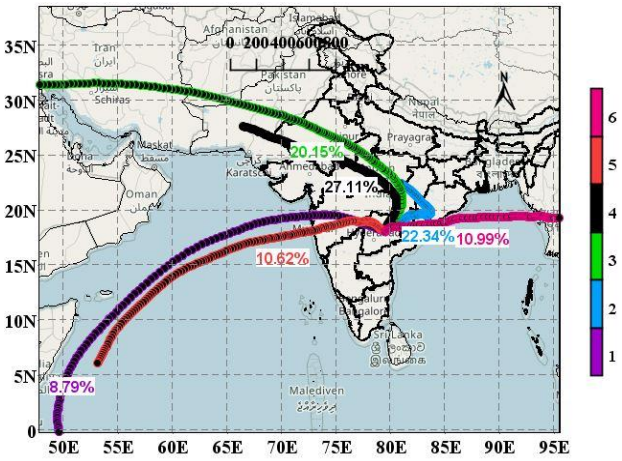


Fig. 13: Cluster analysis

Cluster Analysis: The clustering of all 7-day back trajectories at receptor location observed during the study is shown in fig. 13. Cluster 4 was observed to contribute 27.11% trajectories from the NW region of the India while cluster 2 was observed to contribute 22.34% of trajectories from Odisha, Chhattisgarh and part of Madhya Pradesh. Cluster 3 contributed about 20.15% from parts of Madhya Pradesh, Rajasthan and Western part of the Indian border. Clusters 1 and 5 contributed about 10.62 and 8.79% from part of Maharashtra and the Arabian sea mostly. Cluster 6 contributed 10.99% of trajectories from the BOB indicating the influence of sea salt origin at the receptor location.

In cluster analysis, it was noticed that Odisha and Chhattisgarh contribute significantly due to mining activities, thermal power plants and associated industries. Similar findings related to transport of pollutant in the lower layer and from nearby local regions towards the receptor location[38,50,53].

Conclusion

Based on the analysis of $PM_{2.5}$ during the study, concentrations of $PM_{2.5}$ and heavy metals are high in the Post-Monsoon season. However, the $PM_{2.5}$ concentrations observed were lower than the standards prescribed by NAAQS. Pollution due to heavy metals bound to $PM_{2.5}$ was significant due to emissions from traffic and anthropogenic activities in urban areas. It was evident that the pollution was significantly contributed by long-range transport. Zn, Fe and Cu concentrations in $PM_{2.5}$ were significantly higher compared to the concentrations of Ni and Cd. The order of occurrence of heavy metals in descending order was found to be: Zn>Fe>Cu>Ni>Cd. Long-term sampling may help in better understanding the variation in SPM and metal concentrations.

EF values indicated that Zn, Cu and Cd are high enrichment found the attribution emissions from combustion and industry. Health risk assessment showed that the ingestion pathway dominates over the dermal and inhalation pathways. Based on HQ and HI index, it may be concluded that there is no significant non-carcinogenic and carcinogenic risk from the observed metals in the region studied. However, the risk for children was higher when compared to that for adults.

From the CWT, PSCF and cluster analysis, it may be concluded that the contributions from western and North-Western regions of India dominate at the given receptor location. Since heavy metals bound to $PM_{2.5}$ analysed in this study, the presented results from health assessment and source identification can be useful in planning air pollution control strategies and for framing appropriate regulations.

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