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Metals concentration associated with respirable particulate matter (PM_{10}) in industrial area of eastern U.P. India

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Abstract: *The present study deals with the assessment of ambient air quality with respect to respirable suspended particulate matter (RSPM or PM_{10} ≤10 µm) and trace metals (Fe, Zn, Cu, Cr, Ni, Cd, Mn and Pb) concentrations in RSPM at five locations of Renukoot, an industrial area of Eastern Uttar Pradesh. The 24 hr mean concentrations of PM_{10} ranged between 69.3 to 118.9 $\mu g\ m^{-3}$, which is well within the permissible limit (150 $\mu g\ m^{-3}$) of national ambient air quality standards (NAAQS) but found higher than the prescribed annual daily limit of US EPA (50 $\mu g\ m^{-3}$). The ambient air was mostly dominated by the Fe and least by the Cd among the metal analysed. Murdhawa, a commercial place influenced by vehicular population, is found to be the most polluted area of Renukoot and Dongia nalla (forest area) the least. The ambient air of Murdhawa is rich in Cu and Ni, indicating contribution of mobile sources. The Rammandir, a residential place near the industry, is rich in Cd and Cr, suggesting contribution of point sources. The Ni concentration is found to be alarmingly high in the air at all the locations except Dongia nallah, when compared with the EC (European Commission) limit (20 ng m^{-3}). The Cd concentration is found to be higher only at Rammandir as compared with the EC limit (5 ng m^{-3}). Mean concentrations of Zn, Pb and Mn are found to be almost equal in the ambient air of all the locations, suggesting the significance of sources contributing to presence of these metals. Zn, Cu, Pb and Ni having a significant correlation with PM_{10} indicate the same source contributing these metals as well as PM_{10}. The present study has focused on the quantitative variation in different metals in the PM_{10}, which is extremely harmful due to their toxic and carcinogenic nature.*

Key words: *Air pollution, PM_{10}, Trace metals*
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Introduction

The respirable particulate matter (RSPM or PM_{10} ≤10 µm) are trapped in conducting airways and fine particulate matter ($PM_{2.5}$ or ≤2.5 µm) are trapped in respiratory airways of human lungs. Now-a-days the RSPM are receiving more attention as important air pollutants because of their deleterious effects on human health in urban as well as in industrial areas (Tripathi *et al.,* 2002; Zhu *et al.,* 2002; Hueglin *et al.,* 2005; Aybek and Arslan, 2007). The PM_{10} and $PM_{2.5}$ can also penetrate deep into the pulmonary interstitial spaces in the lungs, thus provoking inflammation due to their higher diffusion coefficient (Seaton *et al.,* 1995; Morawska *et al.,* 2002).

PMs are a complex mixture of elemental and organic carbon, ammonium, nitrates, sulphates, mineral dust, trace elements and water. Overall the effect of PMs depends on its shape, size, composition, mass and number concentrations and the receptor cells. The chemical characteristics of atmospheric PM are important for both particle toxicity and its role in climate change (Hueglin *et al.,* 2005; IPCC, 2001; Salve *et al.,* 2007).

Burning of fossil fuel by industries and automobiles are the important anthropogenic sources of particulate matter. These particles remain suspended in the air for a long time, and distribute or redistribute other pollutants absorbed with them, especially metals. Particulate matters may also account for climatic changes as they have the property of absorption of light energy. The movement and fate of PM depends on the physical nature of the PM and meteorological conditions of the surrounding areas (Fernandez *et al.,* 2000; Kim *et al.,* 2002).

Since the metals are natural constituents of the earth's crust and are widely distributed in environmental matrices, the humans are easily exposed to them even at trace levels (AMAP, 1997) by either anthropogenic activities or by necessity. When these metals are concentrated in the environment, food and water, they affect the health of humans and animals (Abulude *et al.,* 2003). At elevated concentrations all the metals are harmful to living beings including humans (Yasutake and Hirayama, 1997). Exposure can occur through a variety of routes; inhalation of particles (< 10 µm) is one of the important routes. The inorganic components constitute a small portion by mass of the particulates, however, it contains some trace elements such as As, Cd, Co, Cr, Ni, Pb and Se which are human or animal carcinogens even in trace amounts (ATSDR, 2003; Wang *et al.,* 2006). The children, elderly, smokers, people with poor health suffering from cardiovascular diseases, especially allergy and asthma and chronic respiratory difficulties have been identified as more susceptible group. The concentration of different types of metals in RSPM is important, because they influence the toxicity of a metal when inhaled.

Considering the present scenario, a study was carried out to assess the air quality with respect to RSPM (PM_{10}) and comparison of different metals associated with it and also to discuss the probable adverse effects of heavy metals and PM_{10}

on human health in an industrial area of Renukoot, District Sonebhadra, eastern UP, India.

Materials and Methods

Renukoot is an industrial area with a residential population of about twenty five thousand people. Overall this area is surrounded by different types of industries like carbon factory producing carbon anode and tyre grade carbon, an alumina plant producing aluminium, gas factory and a chemical factory producing various types of pesticides and chemicals. We considered the main market of Renukoot as the central location for our study area.

Five monitoring sites were selected. All the five locations are as: (i) Murdhawa- about 1.5 km away in north east side, (ii) Turra- about 1.0 km away in west south west, (iii) Muirpur - about 6.0 km away in south side, (iv) Rammandir about 0.5 km away in south east side and (v) Dongia nalla- about 2.0 km away in east side from centre (Renukoot market). Topographically the monitoring locations, were situated at Longitude 83^0 East and Latitude 24^{o}15' North. Four of the selected sites (*i.e.* Murdhawa, Muirpur, Rammandir and Turra) were located in suburban areas, whereas location of Dongia nalla was in a forest area and was considered as control site. The Murdhwa and Turra are located adjacent to the state highway, whereas Rammandir is near the main approach road to a chemical industry. All the three locations were also influenced by the vehicular pollution.

The study was carried out in the month of June 2002. The temperature and relative humidity was recorded hourly by auto weather station (model WM 200, Envirotech, New Delhi). During this period, the daily mean of average temperature and relative humidity ranged between 28.6 to 34.81^o and 57.8 to 91.7% respectively. Respirable dust samplers (RDS) were used for the monitoring of particulate matter at all the locations at an approximate height of 1.5 m from ground level. Monitoring was carried out weekly for 4 weeks, and the collection was done continuously for 24 hr with RDS for RSPM. The PM samples were collected at flow rate of 1.1-1.2 m^3 min^{-1} on Whatman EPM 2000 borosilicate glass micro fibre filters. The mass of collected particles was determined gravimetrically after drying. The total volume of air was calculated by multiplying average flow rate by total sampling time in minutes (Sharma *et al.,* 2006).

Thirty-two circles of 25 mm diameter were punched out from Filter papers (2 circles from each filter paper of 1st, 2nd and 3rd week of monitoring at each location and 2 circles for blank) and digested in concentrated nitric acid (Sharma *et al.,* 2006). The content was filtered through whatman filter paper no. 42 and final volume made upto 25 ml by deionised water. The filtrate was examined for the concentration of Fe, Zn, Cu, Pb, Mn, Ni, Cd and Cr by Varian Spectra AA-250 Plus (Australia) and procedure followed as per methodology described in operating manual (Methodology Spectra AA250 Plus, 1995). Fe was estimated at λ 248.3, slit 0.2 nm, Optimum working range of detection (OWRD) in μg ml^{-1} is 0.06-0.15; Zn was estimated at λ213.9, slit 1 nm, OWRD is 0.1-2.0 μg ml^{-1}; Cu was estimated at λ324 .7 slit 0.5 nm, OWRD is 0.03-10 μg ml^{-1}; Pb was estimated at λl21.7, slit 1 nm, OWRD is 0.1-30 μg ml^{-1}; Mn was estimated at λ 279.5, slit 0.2 nm, OWRD is 0.02-5 μg ml^{-1}; Ni was estimated at λ 232, slit 0.2 nm, OWRD is 0.1-20 μg ml^{-1}; Cd was estimated at λ 228. 8 slit 0.5 nm, OWRD is 0.02-3 μg ml^{-1} and Cr was estimated at λ 357.9, slit 0.2 nm, OWRD is 0.06-15 μg ml^{-1}. The final values of AAS were taken after the deducting the blank value for respective metal.

Data were statistically analyzed by one and two factor analysis of variance (ANOVA) followed by Newman Keuls post test for multiple comparison. Pearson correlation coefficient (r) was used to determine the relative association among metals and between metals and PM_{10}. A two-tailed probability less than 0.05 ($p<0.05$) was considered to be significant. All analysis were performed using the STATISTICA software ver. 7.0.

Results and Discussion

PM_{10} and corresponding trace metal concentrations of different locations are summarised in Table 1 and are shown graphically in Fig. 1, 2. Mean concentration of PM_{10} (118.95 μg m^{-3}) at Murdhawa is found to be maximum, while the Dongia nalla registered the minimum (69.33 μg m^{-3}). Similarly, mean concentration of Fe (0.988 μg m^{-3}) at Rammandir is the highest and the concentration of Cd (0.001 μg m^{-3}) in Dongia nalla was the least. PM_{10}, total mass of all the metals in each location and total mass of each metal at all the locations have been arranged in descending order as-

PM_{10} : Murdhawa > Turra > Rammandir > Muirpur > Dongia nalla
Metals : Murdhawa > Rammandir > Turra > Muirpur > Dongia nalla
Metal : Fe > Cr > Cu > Ni > Zn > Pb > Mn > Cd

In the light of value of PM_{10} and mass of metals, it was observed that Murdhawa is the most polluted and Dongia Nalla is the least polluted areas of Renukoot. The Fe concentration is the highest and Cd has the lowest value. Heterogeneous variations in the concentrations of trace metals prompted us to transform the data on metals as well as PM_{10} by using the $\log_{10}$ to make them homogenous. The transformed data are then submitted to statistical analysis such as analysis of variance (ANOVA) and correlation (Zar, 1974).

Equality of mean differences in PM_{10} concentrations of different locations have been carried out by one-way analysis of variance (ANOVA) (Table 2). Significant ($p<0.01$) difference is observed between locations (F=39.27). A pair wise comparison between locations was done by Newman Keuls post hoc test (Table 3) to know which pair differed in the mean level,. Mean concentration of PM_{10} at Murdhawa is significantly ($p<0.01$) higher and different from other locations, while mean PM_{10} in rest of the locations are not significantly ($p>0.05$) different, *i.e.* nearly equal. These results are summarised in descending order of mean PM_{10} as

Murdhawa $\neq$Turra = Rammandir = Muirpur = Dongia nalla

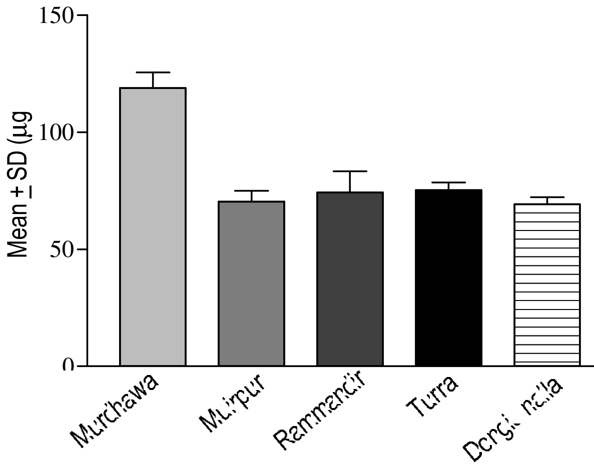

Fig. 1: Mean PM_{10} concentrations ($\mu g\ m^{-3}$) in different locations of industrial area of Ranukoot

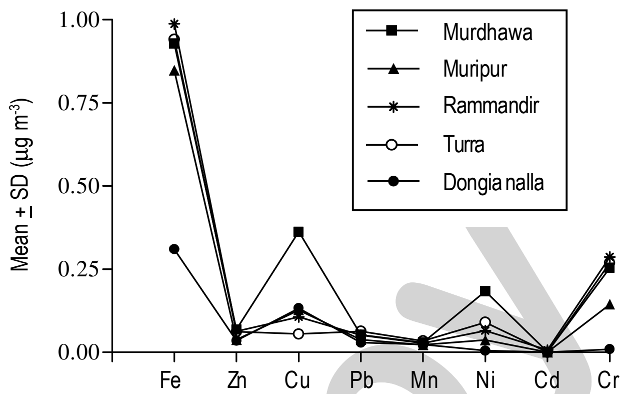

Fig. 2: Mean metals concentrations ($\mu g\ m^{-3}$) in different locations of industrial area of Ranukoot

Equality of mean differences of trace metals concentrations, between and within locations was carried out by two-factor analysis of variance (ANOVA) (Zar, 1974). This method calculates two factors (Table 1) *i.e.* rows (metals) and columns (locations) simultaneously and the obtained results were summarised in Table 4. Both the factors *i.e.* locations (F=20.37) and metals (F=175.08) and their interaction *i.e.* location x metals (F=3.70) were found to be significant ($p<0.01$).

Mean comparisons of each metal between locations are shown in Table 5. Mean concentrations of Zn, Pb and Mn at all the locations are insignificant ($p>0.05$) *i.e.* these metals were equally distributed in ambient air of Renukoot irrespective of local sources or in other words sources contributing these metals equally. But mean concentration of Fe, Cu, Ni, Cd and especially Cr show some variations among locations. Mean concentrations of Fe and Cu in Murdhawa was significantly ($p<0.05$ higher than the respective mean metals of Dongia nalla and Turra. Mean concentrations of Ni and Cr in Dongia nalla were found significantly ($p<0.01$) lower than other locations. Mean concentration of Ni at Muirpur was significantly ($p<0.05$) lower than Murdhawa and Turra. Mean concentration of Cd in Rammandir was significantly ($p<0.01$) higher than mean Cd of Murdhawa, Muirpur, Turra and Dongia nalla. No significant ($p>0.05$) difference was observed in mean concentration of Cr between Rammandir and Turra but was found significantly ($p<0.01$) higher than Murdhawa, Muirpur and Dongia nalla.

The ambient air of industrial area was enriched by the concentration of Fe the most and Cd the least. The concentrations of Cu and Ni at Murdhawa (commercial) were found significantly higher than other metals suggesting the contribution of mobile sources whereas the concentrations of Cd and Cr at Rammandir (residential) were found significantly higher than other metals indicating contribution of point sources.

As PM_{10} mass (concentration) and its trace metals concentrations depends upon so many known and unknown factors, the result suggested that PM_{10} and trace metals concentrations in the ambient air of different locations vary considerably depending upon the proximity to sources of PM_{10} emissions. That's why Murdhawa, the commercial area is found to be the most polluted and Dongia nalla (forest area) is the least polluted.

Inter correlation among metal are shown in Table 6. Correlation of Fe with Cu (r=0.41, $p<0.05$), Pb (r=0.62, $p<0.01$), Mn (r=0.83, $p<0.01$), Ni (r=0.81, $p<0.01$) and Cr (r=0.85, $p<0.01$); Zn with Pb (r=0.64, $p<0.01$) and Ni (r=0.31, $p<0.05$); Cu with Mn (r=0.53, $p<0.01$) and Ni (r=0.38, $p<0.05$); Pb with Mn (r=0.40, $p<0.05$), Ni (r=0.49, $p<0.01$) and Cr (r=0.42, $p<0.01$); Mn with Ni (r=0.61, $p<0.01$) and Cr (r=0.66, $p<0.01$) and Ni with Cr (r=0.92, $p<0.01$) are found to be positive and significant. The positive and significant association suggest that these metals are directly related to each other.

Correlation of PM_{10} with Zn (r=0.39, $p<0.05$), Cu (r=0.36, $p<0.05$), Pb (r=0.33, $p<0.05$) and Ni (r=0.36, $p<0.05$) is also found to be positive and significant (Table 6). This indicates the linear dependence of metals on PM_{10}. In other words increase or decrease in the concentrations of PM_{10} may also increase or decrease the concentrations of Zn, Cu, Pb and Ni.

Several scientific reports have revealed that PM_{10} or lesser particles size is emitted from the automobile emission depending on the speed of vehicles and type of fuel used (Morawska *et al.,* 2002; Sharma *et al.,* 2006). Higher vehicular traffic might be the cause of higher concentration of RSPM in Murdhwa, Turra and Rammandir area.

$PM_{10,}$ which include fine as well as ultra-fine particles, have greater impact on human health. The unscrupulous growth of residential areas adjacent to the sources of PM_{10} is a matter of concern. As per national ambient air quality standards (NAAQS) in India (CPCB, 1994), the annual average value for PM_{10} is 120 µg m^{-3} which is much higher than the new USEPA standard (1996). Annual daily limit value for PM_{10} is 50 µg m^{-3}, While the annual daily

Table-1: Summary-statistics (mean ± SD)- PM_{10} and metals concentrations (µg m^{-3})

	Murdhawa	Muirpur	Rammandir	Turra	Dongia nalla	Total
PM_{10}	118.95±6.55	70.25±4.78	74.33 ±8.95	75.25 ± 3.35	69.33 ± 2.99	408.10
Fe	0.929± 0.564	0.848±0.640	0.988±0.450	0.941±0.437	0.310±0.035	4.02
Zn	0.069±0.019	0.038±0.002	0.064±0.035	0.062±0.026	0.035±0.009	0.27
Cu	0.363±0.330	0.127±0.075	0.107±0.017	0.056±0.025	0.133±0.031	0.79
Pb	0.053±0.011	0.039±0.006	0.052±0. 015	0.064±0.027	0.030±0.003	0.24
Mn	0.030±0.018	0.023±0.018	0.028±0.015	0.035±0.019	0.024±0.017	0.14
Ni	0.184±0.207	0.037±0.043	0.066±0.013	0.090±0.043	0.005±0.001	0.38
Cd	0.001±0.000	0.001±0.000	0.007±0.007	0.001±0.000	0.001±0.000	0.01
Cr	0.254±0.267	0.145±0.178	0.287±0.115	0.270±0.034	0.009±0.005	0.97
Tota metal	**1.88**	**1.26**	**1.60**	**1.52**	**0.55**	**6.80**

N= 4 (PM_{10}), N=6 (metals)

Table - 2: Analysis of variance summary -PM_{10}

Source of variation	Sum of squares	DF	Mean square	F-ratio
Between locations	0.1543	4	0.0386	39.27**
Residual	0.0147	15	0.0010	
Total	0.1690	19		

** = significant (p<0.01)

Table - 3: Comparison of PM_{10} by Newman Keuls test (DF=15)

Comparisions		Significance
Murdhawa	vs. Muirpur	**
"	vs. Rammandir	**
"	vs. Turra	**
"	vs. Dongia nalla	**
Muirpur	vs. Rammandir	ns
"	vs. Turra	ns
"	vs. Dongia nalla	ns
Rammandir	vs. Turra	ns
"	vs. Dongia nalla	ns
Turra	vs. Dongia nalla	ns

ns = not significant (p>0.05), ** = significant (p<0.01)

Table - 4: Two way analysis of variance summary – metals

Source of variations	Sum of squares	DF	Mean squares	F-ratio
Locations	9.0111	4	2.2528	20.37**
Metals	135.5396	7	19.3628	75.08**
Locations x metals	11.4490	28	0.4089	3.70**
Residuals	22.1187	200	0.1106	
Total	**178.1184**	**239**		

** = significant (p<0.01)

limit value as per new European commission directive has been reduced from 50 µg m^{-3} to 20 µg m^{-3} for 24 hr, likely to be achieved by 2010 and the present limit value must not be exceeded for more than 7 days $year^{-1}$ (Querol *et al.,* 2001; Harrison *et al.,* 2004). There have been reports about the adverse health effect even when the PM_{10} concentrations are below the national standards of EPA or WHO standards (Peters *et al.,* 2001). Normally these standards ignore the synergistic effects of combinations of toxic air pollutants (Curtis *et al.,* 2006). When compared with the EC limit value, Ni concentration is found to be alarmingly high at all the locations except Dongia nalla (forest area), while concentration of Cd is found to be higher only at Rammandir (near the point source). The EC limit values for Ni and Cd are 20 ng m^{-3} and 5 ng m^{-3}, respectively (EC, 2003), and both the metals are well known for causing cancer. Mn exposure leads to increased neurological impairment. An increase in the level of Cu leads to respiratory irritance. Compounds of Cr (VI) are known to have most toxic and carcinogenic effect on the bronchial tree (Manalis *et al.,* 2005) and occupational exposure cause dermatitis, penetrating ulcers in the hands and fore arms, perforation of nasal septum and inflammation of larynx and liver (Saraswathy and Usharani, 2007). It does not matter whether people are living in residential and industrial area; the effects of RSPM depend on how long people have been exposed to the finer or inhale particles in excessive concentrations depending upon mass, number as well as their composition. It is imperative to find the threshold concentration of PM_{10} or lesser size particles and their duration of exposure to human beings.

The values of respirable particulate matter, at all the locations of industrial area, are below that described by the NAAQS, but it is almost two times higher than that of USEPA standards. The concentration of Fe is the highest and the Cd is the lowest. Metals Fe, Zn Cu, Pb and Mn are almost equally distributed among locations, but the concentrations of Ni, Cd and Cr vary depending upon the proximity to sources of their emissions. The Murdhawa commercial area is influenced by vehicular population also, so the PM_{10} is rich in Cu and Ni, and the PM_{10} from Rammandir, being nearer to industry

Table - 5: Comparisons of each metal between locations by Newman Keuls test (DF=200)

Locations		Fe	Zn	Cu	Pb	Mn	Ni	Cd	Cr
Murdhawa	vs. Muirpur	ns	ns	ns	ns	ns	*	ns	ns
"	vs. Rammandir	ns	ns	ns	ns	ns	ns	**	**
"	vs. Turra	ns	ns	*	ns	ns	ns	ns	*
"	vs. Dongia nalla	*	ns	ns	ns	ns	**	ns	**
Muirpur	vs. Rammandir	ns	ns	ns	ns	ns	ns	**	**
"	vs. Turra	ns	ns	ns	ns	ns	*	ns	**
"	vs. Dongia nalla	ns	ns	ns	ns	ns	**	ns	**
Rammandir	vs. Turra	ns	ns	ns	ns	ns	ns	**	ns
"	vs. Dongia nalla	ns	ns	ns	ns	ns	**	**	**
Turra	vs. Dongia nalla	ns	ns	ns	ns	ns	**	ns	**

ns= not significant ($p>0.05$), * = significant ($p<0.05$), ** = significant ($p<0.01$)

Table - 6: Correlation matrix (n=30, DF=28)

	Fe	Zn	Cu	Pb	Mn	Ni	Cd	Cr	PM_{10}
Fe	1.00								
Zn	0.23	1.00							
Cu	0.41*	-0.14	1.00						
Pb	0.62**	0.64**	-0.10	1.00					
Mn	0.83**	0.03	0.53**	0.40*	1.00				
Ni	0.81**	0.31*	0.38*	0.49**	0.61**	1.00			
Cd	0.18	0.18	0.07	0.09	0.03	0.22	1.00		
Cr	0.85**	0.22	0.27	0.42**	0.66**	0.92**	0.29	1.00	
PM_{10}	0.28	0.39*	0.36*	0.33*	0.16	0.36*	0.01	0.16	1.00

*= significant ($p<0.05$), ** = significant ($p<0.01$)

as well as residential place, is very rich in Fe, Cr and Cd. Metals Zn, Cu, Pb and Ni show linear dependence on PM_{10}, indicating the same sources contributing to both PM_{10} and trace metals. Since the Ni and Cd are higher than the EC limit value at some locations, one has to think of the remedial measures at point source. So, this study is useful for better understanding and proper management of ambient air quality of an industrial area.

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