The Journal of Animal & Plant Sciences, 25(3 Supp. 2) 2015 Special Issue Page: 706-712
ISSN: 1018-7081

HEAVY METAL COMPOSITION OF PARTICULATE MATTER IN RURAL AND URBAN RESIDENTIAL BUILT ENVIRONMENTS IN PAKISTAN

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ABSTRACT

Heavy metals in outdoor and indoor airborne particulate matter (PM) and dust in different residential built environmentsat two rural and one urban site in Pakistan were analysed. An eight stage non-viable impactor (Thermo Fisher Scientific Inc., USA) loaded with EMP 2000 glass microfiber filter papers (Whatman, England) was used to collect airborne PM.The indoordust samples (settled dust) were collected from different indoor surfaces (floor, cupboards) in living rooms and kitchens from houses at rural sites. The outdoor samples were collected from courtyards of the houses.At the urban site dust samples were also collected by the roads at 27 different locations around Lahoreand at a background site (University of Veterinary and Animal Sciences).Additionally, samples of dung cake, used as solid fuel, at one of the rural sites were taken.Heavy metals (Si, Al, Zn, Mn, Cu, Ni, Cd, Pb, Co and As) were determined by Graphite Furnace Atomic Absorption Spectrophotometer. At rural site I, in general, the concentrations of metals were higher outdoors than indoors, except for slightly higher indoor levels of Cu (0.85µg/m^3 indoor: 0.56 outdoorµg/m^3), Si (3.31µg/m^3 indoor: 3.17 outdoorµg/m^3) and Pb(11.99 ng/m^3 indoor: 9.32 outdoor ng/m^3). At the rural site II the mean concentration were higher outdoors than indoors, excluding Ni which was considerably higher indoors (55.68 ng/m^3) than outdoors (31.91 ng/m^3).At the urban site,outdoors, Si had the highest concentration (3.46 µg/m^3) followed by Al, Zn, Mn, Cu, Ni, Cd, Pb and Co. Similarly, the indoor levels had a maximum contribution from Si (12.30 µg/m^3) followed by Al, Mn, Zn, Cu, Cd, As, Pb, Ni and Co.With reference to dust at rural site I the top five metals outdoors were Si (708 mg/kg), Al, Cu, Zn and Pb (52 mg/kg) while, indoors Al was highest (281 mg/kg), followed by Si, Cu, Zn and Pb (57 mg/kg). At rural site II, both outdoors and indoors, Al (274 mg/kg – outdoor: 266 mg/kg - indoor), Si, Zn, Cu and Pb (61 mg/kg – outdoor: 80 mg/kg - indoor) were the five most abundant metals.The main five metals in decreasing order of their concentration in the road dust around Lahore were Si (686 mg/kg), Al, Cu, Zn and Pb (81 mg/kg). On the other hand, the dust samples from the background site showed Si (345 mg/kg) > Al >Pb> Cu > Zn (73 mg/kg). The airborne metal concentration of Pb was within the guideline value of WHO (0.5 µg/m^3) but the levels of Mn, Cd and Ni were higher at all sites than the guidelines proposed by European Commission and WHO highlighting the risk of exposure to toxic metals in non-occupational environments.

Key words: PM, dust, metals, household fuel, rural, urban, Pakistan

INTRODUCTION

Knowledge of indoor air quality is of vital importance to estimate the total human exposure to different air pollutants due to the high proportion of time spent in different indoor environments (Ott, 2007). Air pollutants in indoor air can have diverse sources originating from indoors as well as outdoors.The state of air quality in the residential built environment can have major public health impact due to amount of time spent in houses, particularly, by children and elderly who are disproportionately susceptible to the risks of exposure to air pollutants. There are numerous indoor and outdoor sources of air pollutantsin residential environments: cookingand other combustion appliances, heating, household cleaning, smoking, use of aerosol products, building materials, furnishing, plants/pets, human occupants, heating and cooling systems, outdoor anthropogenic sources (Industrial processes, automobile emissions) and natural sources (volcanic/soil/rock debris, sea salt, forests). The types and emission strength of these in different residential settings will vary largely across different regionsdepending on many socio-economic developments and air quality management capabilities.

In the developing world household use of solid fuels is the biggest source of indoor air pollution. At present 2.8 billion people still use solid fuel as household energy source, often in low efficiency traditional stoves, leading to excessive levels of indoor air pollution. The majority of these households are in low to middle income countries mainly in Asia, Africa and Latin America (WHO, 2014a).In 2012, approximately 4.3 millionpremature deathswere attributable to household

airpollution: mostly from low and middle income countries (WHO 2014b).Among the range of hazardous air contaminants, particulate matter (PM) is responsible for huge burden of disease in both developed and developing world (WHO, 2014a; Correia*et al*. 2013).

The PM generated by different sources can have diverse sizes, shapes as well as chemical composition.The concentration of indoor particle variesconsiderably and can have very complex chemical composition depending not only on their sources but also on post formation processes. The most important chemical properties of particles include: elemental composition, inorganic ions, carbonaceous compounds (organic and elemental carbon) (Morawska, 2004). Knowledge of the chemical composition of PM is of great importance in order to understand the risk associated with various components and to identify the source signatures. Indoor PM and settled dust in households, particularly, those using solid fuels and/or in urban areas with high ambient air pollution may contain sufficient quantities of such elements which pose toxicological risk to occupants. A large number of studies have highlighted that indoor dust can be an important source of metal exposure and toxic metals may accumulate in humans by way of inhalation or ingestion (Kurt-Karakus, 2012; Rasmussen *et al*. 2013). Young children have a high risk of exposure due to their various activities involving direct and indirect contact with thedust and large amount of time spent indoors(Barbieri *et al*. 2014).

A number of studies have examined the level of heavy metals, particularly, in dust from indoor and outdoor of residential built environments in different regions globally: Italy (Manno*et al*, 2006), Malaysia (Latif *et al*, 2009), Bahrain (Madany, 1994), Canada (Rasmussen *et al*, 2013), Jordan (Al-Khashman, 2007), Hong Kong (Li *et al*, 2001), China (Yang *et al*, 2015; Huang *et al*, 2007), Egypt (Rashed, 2008), Turkey (Kurt-Karakus, 2012; Pekey *et al*, 2010), Oman (Abdul-Wahab, 2006), Tibet - China (Chen *et al*. 2015; Li *et al*, 2012), Sweden (Molnár *et al*, 2006), Mexico (Tovalin-Ahumada *et al*, 2007), USA (Adgate *et al*, 2007), India (Deka and Hoque, 2015). Although the vast proportion of exposure to indoor PM is due to use of solid fuels, studies on chemical characterization of smoke produced during cooking are limited. The physical and chemical characteristicsof aerosols, including heavy metals, emitted during cooking with yak dung inthe TibetanPlateau has been examined by Chen *et al*. 2015; Li *et al*. 2012 and Kang *et al*. 2009. Lab based studies have also been carried out to assess the emission factors of chemical composition (OC, EC, different ionic species)from different types of biomass fuels (fuel wood, crop residue and dung cake) (Saud *et al*. 2012; 2013). Recently, Deka and Hoque (2015) characterized chemical composition (elements, anions, carbon) of the smoke released from different biomass fuel during cooking in rural households and found that smoke from cow dung had highest level of elements, carbon and anions than other fuel types.

With reference to Pakistan, a large fraction of the population still relies on solid fuels as a household energy source. Recent estimates have shown that overall 62% of the population use solid fuels and this proportion is considerably higher in rural areas (87%) than urban households (13%). Furthermore, only 6% of householdshave a separate space for cooking and this trend is slightly higher in urban areas (5%) than rural (7%) (NIPS and ICF, 2013). Therefore, indoor air pollution is a growing public health challenge in Pakistan. Additionally the state of ambient air quality, especially, in urban centres is alarming due to a significant growth in number of vehicles, urbanization and industrial sector along with inefficient automotive technology, use of unclean fuels, weakly regulated industrial emissions and minimal air quality management capabilities. Pakistan Economic Survey 2013-14 has reported the number of total vehicles grew by 130.3% over the period of 2001 – 02 to 2012-13 and emissions from vehicles are the biggest source of air pollution in the country (Pakistan Economic Survey, 2013 - 2014).The reported level of PM from urban centres in the country are among the highest in the world and many times higher than WHO guidelines (Sanchez-Triana*et al*. 2014: Colbeck*et al*. 2010a). Similarly the available evidence on level of indoor air pollutants in Pakistan shows that that concentration of different air pollutants (CO, PM_{10}, $PM_{2.5}$, PM_1 and NO_2) are many times higher than WHO guidelines (Nasir *et al*, 2013; Colbeck*et al*, 2010b,c; Siddiqui *et al*, 2009). Severalintervention efforts have been made by government and nongovernmental organisations at short scale butthe issue of indoor air pollution still lags behind policy maker's agenda (Colbeck *et al*, 2010b). Therefore, a large fraction of the population is exposed to excessive levels of PM in both rural and urban residential built environments. Several studies have been carried out on elemental composition of ambient PM in urban centres and these have been reviewed by Colbeck *et al*. 2010a. However, studies on heavy metal composition of indoor PM and settled dust from residential built environments are scanty (Nazir *et al*, 2011; Jabeen *et al*. 2001). The present study aims to analyse the heavy metal composition of PM and settled dust from rural and urban households using different fuels.

MATERIALS AND METHODS

Sampling Sites: The samples were collected from both indoors (kitchens and/or living room) and outdoors (courtyards) at two sites (Rural Site I - Chak NO.35/2.L and Rural Site II - Bhaun) and an urban site

(Lahore)during August – November 2007 and June 2008. Households at rural site I used solid fuel predominantly whereas at rural site II and the urban site the fuel was natural gas. Rural site I was surrounded by agricultural lands and had very low traffic density. However, burning of solid waste/crop residue was common outdoors. On the other hand rural site II was semi urban and was close to a main road, although traffic density within the village was very low. The sampled households at the urban sites were located in densely populated areas in the main city with high traffic density. Additionally, road dust samples were also taken from 27 different sites in Lahore and at a background site (university roads at University of Veterinary and Animal Sciences Lahore). The detailed description of the sampling sites has been given in Nasir *et al*, (2013).

Collection of airborne PM: AirbornePM was collected with an eight stage nonviable impactor (Thermo Fisher Scientific Inc. Waltham, MA, USA). This is a multi-stage, multi-orifice sampler and can collect particles from 9 to 0.4µm aerodynamic diameters at 28.3 l/min. The collection time at each site varied from 5 – 8 hours. Glass microfiber filters (EMP 2000 – Whatman England) were used as the collection medium. Samples were taken at a height of 1 meter at each sampling site/space.

Collection of Dust Samples: Dust samples were collected from all the sampling sites, both indoors and outdoors, and from 27 different locations on the roads and at a background site in Lahore. The indoor samples (settled dust) were collected from various indoor surfaces (floor, cupboards, etc.) in living rooms and/or kitchens from randomly selected houses at rural and urban sites. The outdoor samples were collected from randomly selected points in courtyards of the houses. The road dust samples were collected from both sides of the road, one foot away from the road edge. In all the sampling locations, the dust samples were collected by disposable plastic brushes and laminated paper pans. After collection the dust samples were packed into airtight polyethylene bags. Additionally, samples of dung were also taken. These samples were kept refrigerated before analysis. For analysis, the dust and dung samples were dried in an oven at 60ºC for 72 hours and then ground to a powder with a pestle and mortar before sieving (150 µm). These samples were then extracted for the analysis of heavy metals.

Extraction and Analysis of Heavy Metals: Both air and soil samples were extracted by the same method. For air samples the filter was placed in a 50 ml boiling test tube while in case of the dust 1 g (noting the weight to three significant figures) was placed in a 50 ml boiling test tube and 4 ml of 'primar' grade Nitric Acid (70%) was added. Then these were left at room temperature, covered with Decon washed marbles, for 36 hours followed by heating in a TeckamPTC-2 digestion block in fume cupboard. At first samples were heated at 50°C for 30 minutes and thenat 140 ºC for 8 hours. Following digestion, the samples were left to cool and 5 ml deionised H_2O was added to wash any residual solution from the test tubes and stored at room temperature. Digested samples were filtered using a glass fibre syringe filter. Finally, filtered samples were made up to 20 ml with deionised H_2O. Metals in solution were determined by Graphite Furnace Atomic Absorption Spectrophotometer (Unicam atomic absorption, Cambridge, UK).

RESULTS AND DISCUSSION

Heavy Metals in AirbornePM: The heavy metal composition of PM at the three sites (Rural I and II, Urban) is shown in Table 1. For outdoors at rural site I metal concentrations decreased in the following order: Al, Si, Zn, Mn, Cu, Ni, Cd and Pb. In the kitchens Al was again dominant followed by Si, Mn, Cu, Zn, Ni, Pb and Cd. As and Co concentrations were below the limit of detection at all sites. In general, the concentrations of trace metals were higher outdoors than indoors, except for slightly higher indoor levels of Cu, Si and Pb. At rural site II the mean concentration in the living rooms and outdoors revealed a different concentration order. Outdoors Si was dominant followed by Al, Zn, Mn, Cu, Cd, Ni and Pb (Table 1). Indoors the pattern was fairly similar: Si, Al, Zn, Mn, Cu, Ni, Cd, Pb and Co. Again As was below the detection limit in both settings (indoors and outdoors) while Co was only detectable indoors. The levels of heavy metals were higher outdoors than indoors, except for Ni which was considerably higher indoors (55.68 ng/m^3) than outdoors (31.91 ng/m^3).The heavy metal composition of PM at the urban site was determined in living rooms and outdoors. Si had the highest concentration outdoors followed by Al, Zn, Mn, Cu, Ni, Cd, Pb and Co. Similarly, the indoor levels had a maximum contribution from Si followed by Al, Mn ,Zn, Cu, Cd, As, Pb, Ni and Co (Table 1). The Si concentrations were substantially higher indoors than outdoors. This might be due to the contribution from deposited indoor dust due to the use of a ceiling fan during the sampling because of the summer weather.

Another study inPakistanby Nazir*et al*. (2011)reportedI/O relationships of trace metals in PM froman industrial area (WahCantt). Total suspended particulate matter wascollected from livingrooms and atthe rooftop with a high volume air sampler. The indoor and outdoor concentrations of different metals at the urban site in the present study are in agreement to some extent with their findings. Both studies found higher levels of Cd and Pbindoors and Zn and CO outdoors. However, the concentration of different metals both indoors and outdoors differ largely. The level of indoor Cu (0.26 µg/m^3), Zn (0.63 µg/m^3) and Pb (67.33 ng/m^3) at

urban site in the present study were lower than Nazir et al. 2011 (Cu:1.14µg/m^3; Zn:3.49µg/m^3 and Pb:272.6ng/m^3). Whereas, outdoors they reported higher concentrations of Zn(3.94 µg/m^3), Pb(137.4 ng/m^3), and Co(152.9 ng/m^3). With reference to other regions the concentration of heavy metals for both indoor and outdoor were generally higher in the present study than reported from China (Huang *et al*. 2007), Mexico (Tovalin-Ahumada *et al*, 2007), USA (Adgate *et al*, 2007)and Sweden (Molnár *et al*, 2006) apart from Cd and Co which were higher in Mexico city. The levels of Pb were comparatively higher both indoors and outdoors in China while it was only higher outdoors in Mexico.

Studies with regard to heavy metals in indoor air from households using solid fuels are very rare. The indoor levels of Cd, As and Pb reported by Kang *et al*, (2009) from Tibetan plateau during cooking with Yak dung were far higher than in the present study. Their reported concentration of Cd, As and Pb during cooking time were 4.38 µg/m^3, 48.60 µg/m^3 and 112.11 µg/m^3, respectively. A recent study (Li *et al*. 2012) from the same region has reported yet again higher levels for Al (16.39 µg/m^3), Cd (3.45 ng/m^3), Ni (157 ng/m^3), and Pb(72.79 ng/m^3) than the present study except Cu (86.79 ng/m^3) and Zn (374 ng/m^3).The comparison of the heavy metal concentration determined in present study with guideline values proposed by various agencies revealed that the concentration of Pb was within the guideline value of WHO (0.5 µg/m^3) but Mn was higher than the guideline value of 0.15 µg/m^3 (WHO, 2002). The concentrations of Cd and Ni were also higher than permissible limit values by European Commission (Cd – 5ng/m^3; Ni – 10-50 ng/m^3) (European Commission, 2000).

Table 1. The heavy metal composition of airbornePM in Pakistan. Units of µg/m^3 except for Cd, Ni, Pb, As and Co which are reported in ng/m^3

Species	Rural site I		Rural site II		Urban site	
	Outdoor (n = 5)	Indoor(Kit) (n = 5)	Outdoor (n = 5)	Indoor(LR) (n = 5)	Indoor (LR) (n = 5)	Outdoor (n = 5)
Al	7.21	6.22	7.89	4.74	3.49	3.01
Cu	0.56	0.85	0.27	0.14	0.26	0.30
Mn	0.98	0.97	0.62	0.17	0.66	0.34
Si	3.17	3.31	22.31	12.65	12.30	3.46
Zn	1.36	0.76	1.01	0.93	0.63	0.85
Cd*	35.05	0.59	79.17	5.93	85.24	31.66
Ni*	151.34	69.33	31.91	55.68	13.12	65.78
Pb*	9.32	11.99	9.53	5.77	67.33	16.24
As*	ND	ND	ND	ND	72.17	ND
Co*	ND	ND	ND	3.90	0.75	12.69

* ng/m^3, n = number of samples, ND – below limit of detection, n = number of samples. Kit: Kitchen, LR: Living Room

Heavy metals in dust: Table 2 shows the concentrations of heavy metals, in dust and dung. At rural site I the chemical analysis was carried out on indoor and outdoor dust. Outdoors, the top five metals with respect to their concentration in dust were Si, Al, Cu, Zn and Pb while, indoor it was dominated by Al, followed by Si, Cu, Zn and Pb (Table 2). Apart from Si the concentration of heavy metals were approximately the same indoors and outdoors. Si was considerably higher in outdoor dust. The heavy-metal composition of dung revealed that, out of the total 10 metals, Si was the leading constituent followed by Al, Cu, Zn, Pb and Ni. The remaining metals had lower concentrations.At the rural site II the sampling was carried out in outdoors and living rooms. Al, Si, Zn, Cu and Pb were the five most abundant metals both outdoors and indoors. The dust samples at the urban site were collected from university roads (University of Veterinary and Animal Sciences, Lahore, with very limited traffic – surrounded by a large network of high traffic roads) and from the roads at 27 different locations around Lahore. The dust samples from the university showed Si > Al >Pb> Cu > Zn. On the other hand, the main five metals in decreasing order of their concentration in the road dust were Si, Al, Cu, Zn and Pb. In general, the concentrations of As, Co, Mn and Ni were approximately the same while Si , Zn, Cu and Cd were higher in road dust. In fact, Si was almost double in the city road dust than in the University road dust.

Table2. The heavy metal composition of dust (mg/kg)

Species	Rural site I			Rural site II		Urban site	
	Outdoor (n = 5)	Indoor (n = 5)	Dung (n = 3)	Outdoor (n = 5)	Indoor (n = 5)	University (n = 8)	By road (n = 27)
Al	281.69	281.32	272.75	274.59	266.58	282.21	273.78
As	5.34	3.47	6.81	4.60	6.17	4.92	4.26
Cd	2.11	3.32	3.63	2.78	3.17	3.44	6.31
Co	11.87	12.52	22.69	11.91	17.58	28.16	26.67
Cu	80.81	84.50	82.29	80.49	85.79	78.51	86.03
Mn	11.35	11.27	11.38	11.48	11.43	11.26	11.56
Ni	32.68	32.78	32.50	32.58	32.09	33.19	31.57
Pb	52.53	57.94	35.81	61.46	80.68	89.89	81.61
Si	708.09	212.08	280.14	253.25	141.94	345.95	686.03
Zn	79.89	80.09	81.55	85.16	82.26	73.22	83.35

n= Number of samples

The concentration of heavy metals during the present study was lower than those reported by Jabeen*et al*. (2001) during a study on the concentration of heavy metals in street and house dust in Gujranwala, an industrial city of Pakistan. This might be due to differences in the location and emission sources between these sites. The concentration of heavy metals in the present study were also lower than recently reported from house dust in a Chinese town except Cd (Yang *et al*, 2015). Their reported concentrations for As, Cd, Pb, Cu and Mn were 31.97 (µg/g), 0.56 (µg/g), 300.46 (µg/g), 93.44 µg/g) and 600.51 (µg/g), respectively. However, the heavy metal loadings in the present study were higher than house and street dust reported from a semi urban area in Kuala Lumpur, Malaysia (Latif *et al*, 2009).A study on house dust from Istanbul, Turkey had a higher concentration of Cu (156 µg/g), Zn (832 µg/g), Mn (136 µg/g), and Nickel (263 µg/g) than present study while Pb (28 µg/g), Cd (0.8 µg/g)and Co (5 µg/g) were lower (Kurt-Karakus, 2012).Recently, Faiz*et al*.(2009) presented an analysis of road dust (Cd, Cu, Ni, Pb and Zn) along the Islamabad Expressway. Their reported levels of Cu (52 mg/kg) and Ni (23 mg/kg) were lower than in the road dust from Lahore city. They found a moderately higher load of Pb (104 mg/kg) and Zn (116 mg/kg) as compared to road dust in Lahore (Table 2). They also found that the concentration of heavy metals in road dust of Islamabad Highway were towards the lower end of reported levels from different parts of the world. The concentrations of most heavy metals found in this study were lower than those reported from other regions of the world in recent studies. For example concentrations of Pb, Zn, Ni, and As in road dust in a Greece town were 359 (µg/g), 137.8(µg/g), 58.2(µg/g), and 62.3(µg/g), respectively, and only Cd (0.2 µg/g) and Cu (42.7 µg/g) were lower than current study (Christoforidis and Stamatis, 2009). However, it is likely that the concentration of toxic metals,particularly, in urban centres with high traffic density will increase due to ever growing number of vehicles with inefficient automotive technology and poor vehicle emission regulation implementation.

Conclusions: The current study analysed the level of heavy metal in airborne PM and dust from two rural and one urban site in Pakistan. A large variation in concentration distribution of heavy metal in both airborne PM and dust in outdoors and indoors was found. Overall, the levels of toxic metals were higher than safe limits proposed by WHO and these were almost equal or higher in rural areas using solid fuels than the urban centre. Hence both rural and urban households are under the severe threat of poor air quality and may be equally exposed to excessive levels of toxic metals. This can have significant implications for vulnerable groups (elderly, children, pregnant women) and public health in general. The main sources of heavy metals in rural areas were use of solid fuels, indoor smoking and burning of solid waste while at theurban site vehicular and industrial emissions were additional sources highlighting venue and scenario specific risk of exposure to toxic metals. At present both indoor and ambient air quality in Pakistan is among the worst in the world and calls for urgent intervention strategies in rural areas to improve indoor air quality in households relying on solid fuels. Additionally strict implementation of legislative measures to regulate industrial and vehicular emission along with enhancement of air quality management system and transportation infrastructure in the country is required. This study was carried out only at two rural sites and one urban site and may not reflect the levels of exposure to toxic metal at the country level. Nonetheless the findings contribute to the existing state of knowledge on toxic metal emissions from household fuels in Pakistan and are of relevance to other countries/regions with predominate solid fuel use as an household energy source. Further studies from different regions are needed to gain a comprehensive understanding about the extent of heavy

metal exposure in residential built environment with solid fuel use.

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