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TECHNICAL PAPER

Characterization and health risk assessment of indoor dust in biomass and LPG-based households of rural Telangana, India

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ABSTRACT

Indoor dust is one of the key sources contributing to indoor air pollution (IAP) in rural households. It acts as a media for various toxicants like heavy metal depositions and causes severe health risks. The present study deals with investigation of metal concentrations and morphological characteristics of indoor dust generated in varied fuel types followed by estimation of health risks for women and children in rural households in Telangana, India. Indoor floor dust samples were collected from households using biomass and liquefied petroleum gas (LPG) as their cooking energy during winter to evaluate the morphological and chemical characteristics in the aforementioned dust samples. A morphological (SEM-EDX) analysis revealed the presence of carbonaceous particles in biomass-based households and mineral-rich crustal sources in LPG-based households. As observed from ICP-OES analysis, there is a significant difference in mean concentrations of Al, Co, Cr, Fe, Zn, and Ni based on fuel type, except for Mn and Pb. From Pearson's correlation analysis and principal component analysis, it was observed that the biomass households were dominated by Zn, Al, Mn, Cr, and Pb, which could have been contributed from biomass burning deposits, crustal sources, and unpaved roads, while Cr, Pb, Fe, and Mn dominated in LPG households, indicating their origin from leaded paints (Pb and Cr) and crustal sources. The health risks associated with these heavy metals to women and children were investigated using an EPA health risk model. The values from the model indicated that both non-carcinogenic and carcinogenic risks were within the safe levels for both subjects. This study not only establishes chemical and morphological characteristics of indoor dust, but also quantifies the role of fuel type.

Implications: The present study provides the latest geographical evidence of chemical and morphological characterization of indoor dust particles in varied fuels; i.e., biomass- and LPG-based households and associated health risk assessment in a sub-tropical rural site in Telangana, India. Nevertheless, further research is essential from various regions across the country for more heavy metal analysis and factors impacting these levels. One of the major limitations of the present study is the analysis of few metals and measurements in only living area locations. Future studies can include soil and road dust, as well as kitchens and bedrooms, to provide more comprehensive analysis of dust compositions in varied environments.

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Introduction

Indoor air pollution (IAP) in rural households is predominantly contributed from solid fuel combustion, cigarette smoking, paints and varnishes, infiltration of outdoor air pollutants, unpaved roads, and agricultural residue burning, resulting in terrible health implications (WHO 2007). The major constituent of IAP is particulate matter (PM) that results in increased morbidity and mortality rates due to chronic bronchitis, acute respiratory infections, lung cancer, and cardiovascular and ischemic heart diseases (Lim et al. 2012; WHO 2007). The PM deposited

on the surface environment forms indoor dust, which is source and sink for various toxicants including metals and organic contaminants; i.e., polycyclic aromatic hydrocarbons (PAHs), persistent organic pollutants, asbestos, viable biological particles, etc. (Chen, Lu, and Li 2014; Kumar and Scott 2009; Lioy and Gochfeld 2002; Zhu et al. 2013). Several investigations revealed the presence of heavy metals such as arsenic (As), cadmium (Cd), chromium (Cr), copper (Cu), lead (Pb), nickel (Ni), and zinc (Zn) in indoor dust (Chattopadhyay, Lin, and Feitz 2003;

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Lin et al. 2015; Turner and Simmonds 2006; Yoshinaga et al. 2014).

A number of studies have been reported on heavy metal concentrations in household dusts across the world, though these studies were limited to urban households, offices (Cao et al. 2016; Chattopadhyay, Lin, and Feitz 2003; Huang et al. 2014; Kang et al. 2011; Kumar and Scott 2009; Kurt-Karakus 2012; Rasmussen et al. 2011; Rasmussen, Subramanian, and Jessiman 2001; Wang et al. 2011; Yoshinaga et al. 2014), and nursery and primary schools (Chen, Lu, and Li 2014), with very few in rural households (Kurt-Karakus 2012; Lin et al. 2015; Yang et al. 2016). A study of 82 residential homes in Sydney (Australia) reported the presence of Cd, Cr, Cu, Fe, Mn, Ni, Pb, and Zn with 4% households having elevated Pb concentrations (>1000 µg/g) contributed from motor vehicle emissions and leaded paints (Chattopadhyay, Lin, and Feitz 2003). In one of the first studies on urban indoor dust in India, Pb samples from 99 households in Delhi were analyzed by Kumar and Scott (2009), who concluded that Pb contents were significantly higher than in the national data in the US. Further, Chen et al. (2014) observed greater concentrations of Cu, Pb, Zn, Co, Cr, and Ba in nursery and primary schools in Xian, China, primarily originating from vehicles and industrial activities. Also, heavy metal concentrations of Zn, Pb, Cu, Cr, V, Ni, Co, and As in rural indoor dust in Anhui, China, were higher than background soils and varied within different rural areas due to a difference in structural characteristics of houses, sweeping frequency, infiltration of outdoor air, unpaved roads, etc. (Lin et al. 2015). Thus, it is understood that heavy metal concentrations and compositions differ based on anthropogenic factors such as fuel types, cooking styles, paints, varnishes, vehicular emissions, furniture, structural characteristics, and natural sources, including crustal and geogenic origins. It is also evident that there exists considerable difference in indoor dust compositions in urban and rural households, making rural indoor dust the current focus of the present study. Furthermore, few studies reported use of scanning electron microscopy (SEM) with energy dispersive X-ray spectroscopy (EDX) in morphological analysis of indoor dust samples, along with identification of the sources (Brown, Millette, and Mount 1995; Hunt et al. 1992; Jianhua et al. 2006; Kelepertzis et al. 2019; Agarwal et al. 2019; Yang et al. 2016). However, application of SEM for source determination in rural indoor dust in households using biomass and liquefied petroleum gas (LPG) as fuel has been limited so far (Yang et al. 2016).

Children, adults, and elderly women who spend ~71–89% of their time in indoor areas are exposed to heavy metals that are highly toxic and non-degradable via dermal, ingestion, and inhalation. The prolonged

exposure to heavy metals leads to chronic accumulation in internal organs and tissues, resulting in severe health hazards (Chen, Lu, and Li 2014; Kurt-Karakus 2012). In the past, many studies were reported on non-carcinogenic and carcinogenic risks associated with the heavy metals using the USEPA (1996) model for indoor dust in homes and offices, nursery and primary schools, rural households, and for street dust in Istanbul, Turkey (Kurt-Karakus 2012), Shaanxi, China (Chen, Lu, and Li 2014), Anhui, China (Lin et al. 2015), Dhanbad, India (Masto et al. 2017), and Huludao, China (Zheng et al. 2010), respectively. It was observed that in indoor dust in schools and households, both non-carcinogenic and carcinogenic risks were within safe levels (Chen, Lu, and Li 2014; Kurt-Karakus 2012; Lin et al. 2015; Lu et al. 2014). However, the risk levels were greater for street dust (Masto et al. 2017; Zheng et al. 2010). Hence, indoor dust is one of the most common metrics used for residential quantitative exposure analysis and assessment in rural households for vulnerable people who are prone to active or passive exposures (Lioy and Gochfeld 2002).

Thus, the objective of the present study is to determine metal concentrations and morphological characteristics of indoor dust samples for varied fuels during winter (high PM concentrations) in rural India, and to estimate the health risks associated with these heavy metals for children and women.

Methodology

Study area description

The study site is Kishannagar village (17.3850° N, 78.4867° E), Mahbubnagar district, southern Telangana, India (Figure 1). The selection of the village was based on key factors like fuel usage, socioeconomic status, structural characteristics of households, and climate. This village has a total population of 3,639 of which 51% are males and 49% are females living in 737 households with a total geographical area of 1571 hectares. Agriculture is the major occupation, followed by cattle farming by 30% of the villagers. Kishannagar is characterized as having 80.5% households relying on biomass and 17.7% using LPG (Census 2011). In the present study, data from the 2011 census have been used. To have additional assurance of the applicability of the data, a questionnaire survey was distributed to collect primary data on the fuel usage during field studies in 2016 (Table S1 in Supplementary Information (SI)). Though the survey showed a decline in the percentage of houses using firewood, it coincided with the census on firewood

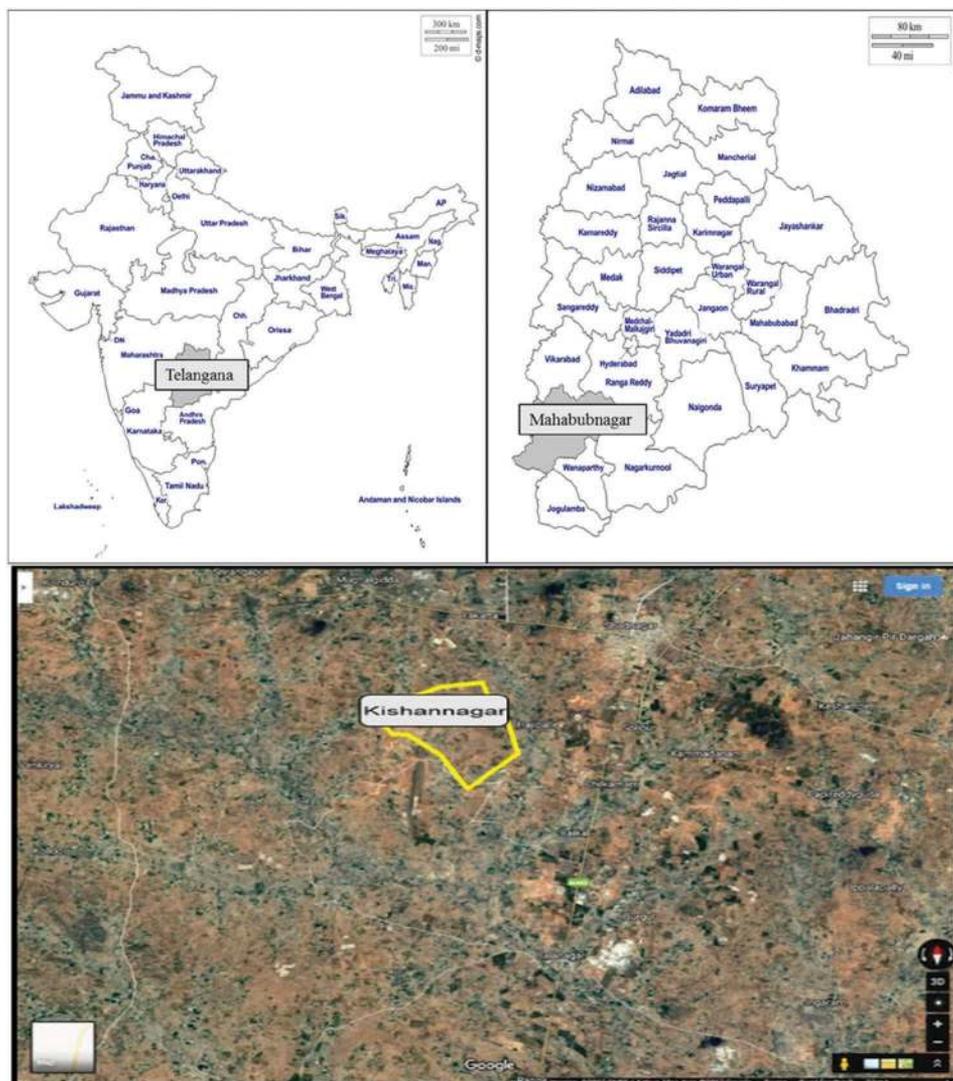


Figure 1. Study areas on map of (a) India and Telangana state and (b) aerial map of rural study site: Kishan Nagar (Image source: Google Earth).

being the primary fuel in these areas. The major soil types reported in this region are red sandy soil, followed by black cotton soil. Chemical composition of red sandy soils include non-soluble material, iron, aluminum, and organic matter, while black cotton soils are dominated by iron, lime, calcium, potassium, aluminum, and magnesium. The study site has a tropical climate; specifically, a semi-arid climate representative of south India. The Indian Meteorological Department (IMD) has characterized the winter season as taking place during the months of January and February, the summer season being between March and May, the southwest monsoon season as June to September, and the northeast monsoon season between October and December. According to the IMD, the warmest month of the year is May, with an average temperature of 33°C. December is the coolest month of the year, with an average temperature of 22.1°C.

Sampling and analytical methods

Indoor floor dust samples were collected from living area locations of various households that were selected for monitoring based on the fuel used for cooking activities. Samples were collected from 20 households during the winter season (December 20, 2016, to January 25, 2017), with 10 each of biomass and LPG-based households. The sample size was selected based on the type of fuel used for cooking, proximity to the road, structural characteristics, and economic status, along with budget constraints. Thus, the sample of 20 households representative of the study region was chosen. The indoor dust samples were collected by vacuum cleaner (Euroclean) using the ASTM standard D5438 method (1993) by estimating the dust loading over a 24 h average on an area of 1 m² floor space. The

collected samples were dried at 50°C for 24 h in an oven followed by sieve analysis to obtain a fraction of particles of less than 70 microns (ASTM 2006). The samples were kept in ziplocked plastic bags and stored in a desiccator for further analysis.

The indoor dust samples were chemically characterized for their elemental compositions following EPA method 3050B (USEPA 1996). The sieved sample dust was extracted through microwave digestion (Anton Paar Microwave Digestion System Multiwave GO). A total of 1 g of the dust samples were digested with a concentrated HCl (10 ml) – HNO₃ (5 ml) – HClO₄ (5 ml) – HF (3 ml) – H₂O₂ (3 ml) mixture in TFM vessels (50 ml capacity). For digestion of dust samples, the program was set for 10 minutes at 180°C with operating pressure of 80 bar. The collected sample filtrates were allowed to cool for 10 minutes and made up to 20 ml with milliQ water. The extracted samples were quantitatively analyzed by EPA method IO-3.1 using ICP-OES (Perkin Elmer Optima 5300 DV) for elemental compositions. The elemental compositions of aluminum (Al), cobalt (Co), chromium (Cr), iron (Fe), manganese (Mn), nickel (Ni), lead (Pb), and zinc (Zn) were analyzed. The blank samples were collected in duplicate to evaluate precision and bias for quality assurance and control (GSS-1 and GSS-3). Calibration methods and minimum detection limits of instrument and the recovery rate of each element are explained in the supplementary information (Table S2).

The morphological characteristics and quantitative identification of elements in indoor dust were analyzed by High Resolution Scanning Electron Microscope (FEI Quanta FEG 200) combined with energy dispersive X-ray spectroscopy (EDX) (Genesis 2011 Emcrafts; Oxford Instruments). The sieved dust samples were mounted on the stubs followed by gold sputter coating (Cresington sputter coater 108) to attain vacuum durability and conductivity. SEM analysis was performed by scanning the electron beam (20–30 KV accelerating voltage) over the sample and observing the subsequent secondary or back-scattered signals. The presence of the following elements was analyzed by SEM-EDX: aluminum (Al), barium (Ba), carbon (C), calcium (Ca), chlorine (Cl), iron (Fe), potassium (K), magnesium (Mg), molybdenum (Mo), niobium (Nb), oxygen (O), lead (Pb), and silica (Si).

The variations in elemental compositions in indoor dust for varied fuel types—i.e., biomass and LPG-based households—were investigated using an analysis of variance (ANOVA) test. Further, elemental compositions in biomass and LPG households were analyzed using factor analysis to determine underlying components, physical interpretations, and interrelationships among the variables. On the basis of eigenvalue and cumulative variance, the total number of factors was identified. Also, Pearson

correlation matrices were estimated to understand the relationship amongst the elements and further determine the contributing sources. This analysis was performed using R studio (version 2.14.1) and origin pro (8.5 version).

Enrichment factor

Enrichment factor (EF) analysis was employed to distinguish anthropogenic sources from those of natural origin using eq1. It is a fairly standard and described calculation used to analyze the enrichment of an element with reference to its abundance in the Upper Continental Crust (UCC) (Adimalla, Qian, and Wang 2019; Cao et al. 2008; Samiksha et al. 2017). For each element, EF was analyzed by dividing its ratio with a reference element to the ratio of baseline values; i.e., UCC values (Taylor 1964). For the present study, aluminum was used as the reference element, while the baseline values of the elements in soil were obtained from the literature (Taylor 1964; Taylor and McLennan 1995). The average crustal abundances of Al, Co, Cr, Fe, Mn, Ni, Pb, and Zn were 154,000, 17.3, 92, 50,400, 1000, 47, 17, and 67 mg/kg, respectively (Taylor 1964).

$$\text{Enrichment factor (EF)} = \frac{\left(\frac{C_n}{C_{Al}}\right)_{\text{household dust}}}{\left(\frac{C_n}{C_{Al}}\right)_{\text{background}}} \quad (1)$$

C_n and C_{Al} represents the concentration of the element and aluminum, respectively, with household dust and background values (crustal abundance).

Health risk assessment

The model developed by the EPA (USEPA 1996) was used in this study to evaluate the residential exposure of heavy metals to children and women in biomass and LPG-based households. The exposure to this dust is possible through three main pathways: dermal absorption, ingestion, and inhalation of dust particles. The risk assessment was calculated for children and adult females with 6 years and 24 years, respectively (ICMR 2009), by estimating the average daily dose (ADD) (mg/kg/day) of pollutants through the previously mentioned pathways using the following equations:

$$ADD_{\text{dermal}} = \frac{(C \times SA \times AF \times ABS \times EF \times ED)}{(BW \times AT)} \quad (2)$$

$$ADD_{\text{ingestion}} = \frac{(C \times IngR \times EF \times ED)}{(BW \times AT)} \quad (3)$$

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

where C is concentration (mg/kg) of metal in indoor dust, SA is exposed skin area (1.05 m^2 for adults and 0.35 for children) (Harinarayan et al. 2013; USEPA 2011), AF is skin adherence factor (0.001595 kg/m^2 for adults (Kissel, Richter, and Fenske 1996) and 0.0017 kg/m^2 for children (Holmes et al. 1999)), ABS is dermal absorption factor (unit less) (0.001 for all elements (Ferreira-Baptista and De Miguel 2005)), EF is exposure frequency (365 days/year for both subjects), ED is exposure duration (24 years for adult and 6 years for children (ICMR 2009)), BW is body weight (60 kg for adult and 20 kg for children (Indian situation) (ICMR 2009; Khadilkar et al. 2007)), IngR is ingestion rate (0.0001 kg/day for adult (USEPA 2011), and 0.0002 kg/day for child (USEPA 2011)), InhR is inhalation rate ($16 \text{ m}^3/\text{day}$ for adult and $10.1 \text{ m}^3/\text{day}$ for children (USEPA 2011)), PEF is particle emission factor ($1.36 \times 10^9 \text{ m}^3/\text{kg}$ (USEPA 2001)), AT is the time period over which the dose is averaged for non-carcinogens; it is $ED \times 365$ days and for carcinogens it is $70 \times 365 = 25,550$ days (USEPA 2011).

Quantified risk and hazard quotient (HQ) for non-carcinogenic effects were calculated by dividing the doses of each element and exposure pathway by their corresponding reference dose (RfD) (mg/kg/day) (Table 1), while for carcinogens, the cancer risk was estimated by multiplying the dose with corresponding slope factor (SF) (mg/kg/day)⁻¹ (Table 1) (Lin et al. 2015; Mastro et al. 2017). The sum of HQ (dermal, ingestion, and inhalation) is equal to the hazard index (HI); if $HI < 1$, there is not a significant risk from non-carcinogenic effects; if $HI > 1$, there is a greater chance that non-carcinogenic effects may arise, with a likelihood to surge as HI value increases (USEPA 2001). Further, for carcinogens, the acceptable or tolerable risk is within the range 10^{-6} – 10^{-4} for regulatory purposes (Ferreira-Baptista and De Miguel 2005). The human exposure to heavy metals present in indoor dust in living area locations of rural households using varied fuel types in sub-tropical climate conditions was assessed based on hazard index and cancer risk methods.

Results and discussion

Morphological characteristics of indoor dust in biomass and LPG-based households

The presence of particles in the indoor dust resulting from natural/crustal sources or anthropogenic sources of varied

fuel types was detailed using their morphology and elemental compositions. SEM images of indoor dust collected from biomass (Figure 2) and LPG (Figure 3) households were evaluated to differentiate various sources contributing toward it and, consequently, the impact of fuel usage on it. Figure 2(a,b) represents individual carbonaceous spherules with irregular shape, dominated by C and O along with inorganic soil-related elements like Si, Ca, Fe, K, Al, and Mg, primarily generated from anthropogenic activities like biomass combustion (Pachauri et al. 2013). Figure 2(c) also represents elongated structures with higher compositions of C and O (>90%) originating from biomass burning. However, Figure 2(d) represents elongated mineral-rich particles with C, O, Si, Al, Ca, Cl, and K, derived from crustal dust and secondary reactions arising in the atmosphere that are accumulated in the re-suspended dust (Kolluru, Patra, and Kumar 2019). Conversely, Figure 3(a,b) represents irregular and spherical morphology in LPG-based households, dominated by minerals including Si, Ca, Al, Nb, and Mo obtained from crustal sources, re-suspended dust, and paved and unpaved roads. Figure 3(c) shows irregularly shaped silica particles with high contents of Si and O originating from natural geogenic sources; i.e., soil, while Figure 3(d), reveals mineral-rich particles—i.e., Si, Al, Fe, Ca, K, Mg, Pb, and Ba—which could have been contributed from anthropogenic sources, including paints, vehicular emissions, and paved and unpaved roads, apart from crustal sources (Brown, Millette, and Mount 1995; Kelepertzis et al. 2019; Pachauri et al. 2013).

Chemical characteristics of indoor dust in biomass and LPG-based households

The metals concentrations (mg/kg) in indoor dust of biomass and LPG households are reported in Table 2. These concentrations were compared with published background values of soil in Maheshwaram (~40 km from study village), Telangana (Purushotham et al. 2012). In biomass-based households, Al was observed with highest median concentration ($54,800 \text{ mg/kg}$), followed by Fe ($27,300 \text{ mg/kg}$), Mn (300 mg/kg), Zn (90 mg/kg), Cr (35 mg/kg), Pb (32 mg/kg), Ni (20 mg/kg), and Co (5.2 mg/kg). The hierarchy of metals concentrations as observed in biomass-based households is as follows: $Al > Fe > Mn > Zn > Cr > Pb > Ni > Co$. Further, it was observed that Co, Cr, Pb, and Ni concentration levels were lower than the background values. However, Zn

Table 1. Reference doses and slope factors of heavy metals for different exposure routes.

| Elements | Co | Cr | Ni | Pb | Zn |
|---------------------------|-----------------------|-----------------------|-----------------------|-----------------------|----------------------|
| RfD _{ingestion} | 3.0×10^{-2} | 3.0×10^{-3} | 2.0×10^{-2} | 3.5×10^{-3} | 3.0×10^{-1} |
| RfD _{dermal} | 6.0×10^{-3} | 3.9×10^{-5} | 8.0×10^{-4} | 7.0×10^{-4} | 6.0×10^{-2} |
| RfD _{inhalation} | 2.86×10^{-5} | 2.86×10^{-5} | 2.57×10^{-5} | 3.52×10^{-3} | 1.0×10^{-2} |
| Slope factor | 9.8 | 4.2×10^{-1} | 8.4×10^{-1} | - | - |

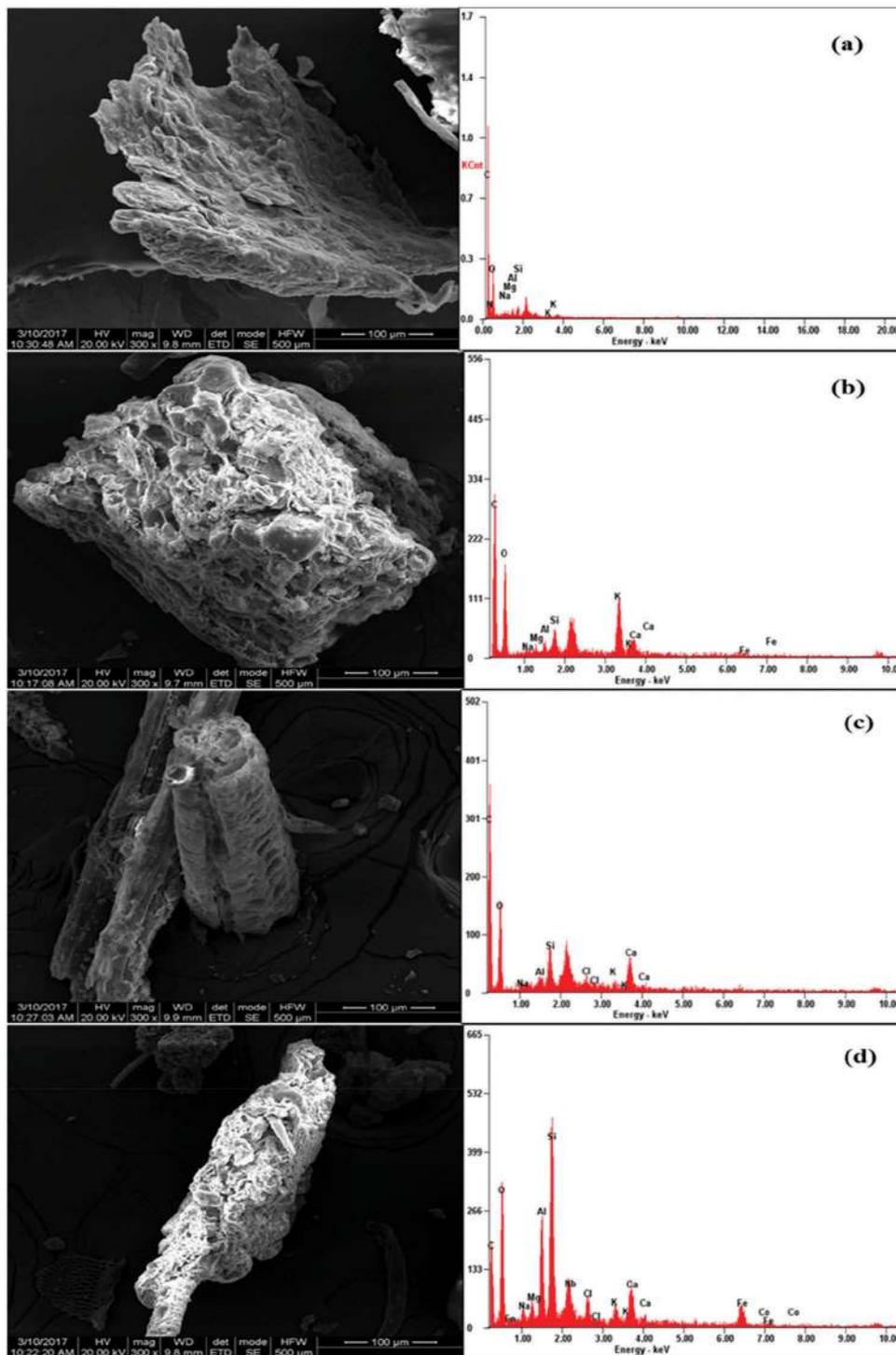


Figure 2. SEM image and EDX profile of indoor dust in biomass-based household.

levels were greater than the background levels, which could have been caused by biomass-burning deposits (Salam et al. 2013). In LPG-based households, the trend was similar to that of biomass households for Al (51,200 mg/kg) > Fe (17,400 mg/kg) > Mn (210 mg/kg), although it varied for heavy metals such as Cr (70 mg/kg), Pb (50 mg/kg), Zn (30 mg/kg), Ni (8 mg/kg), and Co

(2.5 mg/kg). Co, Ni, and Zn concentrations were within the background values, but Cr and Pb levels were found to be higher. High levels of Pb and Cr could have come from paints and unpaved roads (Lee et al. 2016). Correspondingly, the hierarchy of metals concentrations in LPG-based households is as follows: Al > Fe > Mn > Cr > Pb > Zn > Ni > Co. The ANOVA confirmed significant

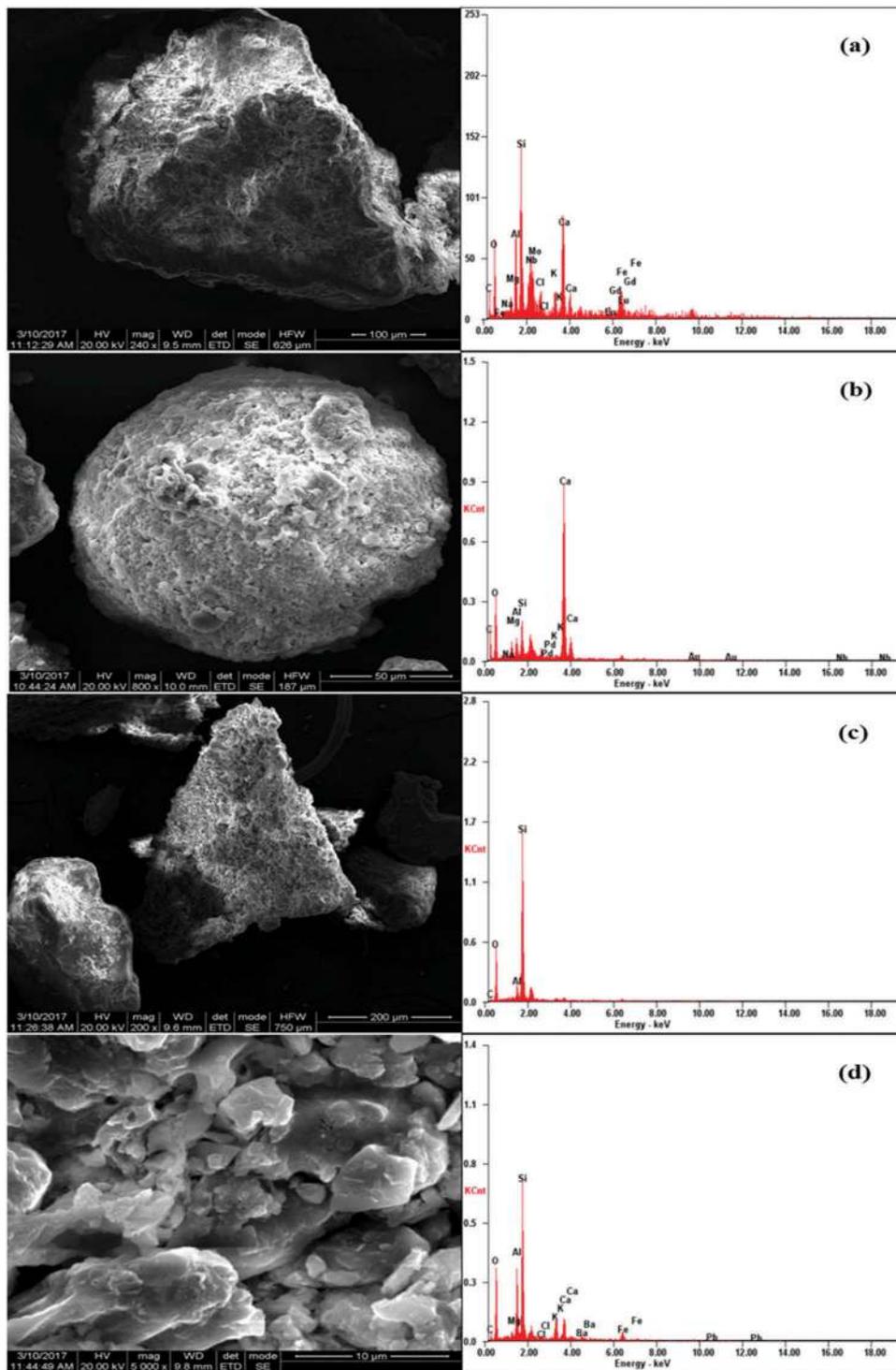


Figure 3. SEM image and EDX profile of indoor dust in LPG-based household.

difference ($p < .05$) in mean concentrations of Al, Co, Cr, Fe, Zn, and Ni for different fuel types. The results also indicated no difference for Mn and Pb, which reveals that they are coming from the same source (Table S3 in the Supplementary Information).

Further, these concentrations were compared with past investigations of indoor dust levels in households, schools,

and offices, along with soil, surface, and street dust in rural and urban areas across the globe (Table 3). From the Table 3, it is clear that Fe, Mn, Co, Cr, Ni, and Pb levels were comparable to household dust (Chattopadhyay, Lin, and Feitz 2003; Kelepertzis et al. 2019; Kurt-Karakus 2012; Yoshinaga et al. 2014), school dust (Figuroa, De la O-Villanueva, and De la Parra 2007), and street dust

Table 2. Concentrations of metals in indoor dust of biomass and LPG-based households.

| Type of fuel | Metal concentrations (mg/kg) | | | | | | | |
|--------------------------------------|------------------------------|------|----|--------|------|------|------|------|
| | Al | Co | Cr | Fe | Mn | Ni | Pb | Zn |
| Biomass | | | | | | | | |
| Minimum | 52,000 | 3.56 | 25 | 25,000 | 250 | 15 | 25 | 38 |
| 1st Quartile | 54,500 | 4.5 | 35 | 26,500 | 250 | 18 | 31 | 85 |
| Median | 54,800 | 5.2 | 35 | 27,300 | 300 | 20 | 32 | 90 |
| Mean | 55,140 | 5.57 | 39 | 27,520 | 294 | 20.6 | 35.6 | 80.8 |
| 3rd Quartile | 56,400 | 6.8 | 40 | 29,000 | 320 | 20 | 45 | 91 |
| Maximum | 58,000 | 7.8 | 60 | 29,800 | 350 | 30 | 45 | 100 |
| LPG | | | | | | | | |
| Minimum | 48,000 | 2 | 55 | 16,800 | 180 | 5 | 35 | 25 |
| 1st Quartile | 50,000 | 2.5 | 60 | 17,000 | 200 | 6 | 45 | 30 |
| Median | 51,200 | 2.55 | 70 | 17,400 | 210 | 8 | 50 | 30 |
| Mean | 50,840 | 2.91 | 68 | 17,540 | 238 | 7.6 | 49 | 33 |
| 3rd Quartile | 52,000 | 3 | 75 | 18,000 | 300 | 9 | 55 | 35 |
| Maximum | 53,000 | 4.5 | 80 | 18,500 | 300 | 10 | 60 | 45 |
| Background values (Maheshwaram soil) | n.a. | 14 | 45 | n.a. | n.a. | 33 | 52 | 44 |

Table 3. Literature on metal levels in indoor dust (household, school, office) and soil, surface, and street dust around the world.

| Reference | Type | Urban/ Rural | Location | Metal concentrations (mg/kg) | | | | | | | |
|--|---------------------------|-----------------|---------------------------------|------------------------------|------|-------|--------|------|------|-----|------|
| | | | | Al | Co | Cr | Fe | Mn | Ni | Pb | Zn |
| This study | Household dust (biomass) | Rural | Telangana, India | 54,800 | 5.2 | 35 | 27,300 | 300 | 20 | 32 | 90 |
| This study | Household dust (LPG) | Rural | Telangana, India | 51,200 | 2.55 | 70 | 17,400 | 210 | 8 | 50 | 30 |
| Akhter and Madany (1993) | Street dust | Rural | Behrain | n.a. | n.a. | 196 | n.a. | n.a. | 153 | 443 | 154 |
| Kurt-Karakus (2012) | Household dust | Rural | Gokturk, Istanbul, Turkey | n.a. | 16 | 254 | n.a. | 655 | 471 | 192 | 1970 |
| Lin et al. (2015) | Household dust | Rural | Anhui, China | n.a. | 10 | 114 | n.a. | n.a. | 39 | 349 | 427 |
| Li et al. (2016) | Surface soils | Rural | Miyun county | n.a. | n.a. | 101 | n.a. | 741 | 38 | 28 | 93 |
| Akhter and Madany (1993) | Household dust | Urban | Behrain | n.a. | n.a. | 145 | n.a. | n.a. | 110 | 360 | 64 |
| Chattopadhyay, Lin, and Feitz (2003) | Household dust | Urban | Sydney, Australia | n.a. | n.a. | 65 | 2790 | 48 | 15 | 76 | 372 |
| Kurt-Karakus (2012) | Office and household | Urban | Besiktas, Istanbul, Turkey | n.a. | 5 | 55 | n.a. | 136 | 263 | 28 | 832 |
| Yoshinaga et al. (2014) | Household dust | Urban | Japan | 16,200 | 4.43 | 66 | 10,100 | 224 | 56 | 49 | 896 |
| Kelepertzis et al. (2019) | Household dust | Urban | Greece | n.a. | n.a. | 87 | 8395 | 132 | 100 | 133 | 1505 |
| Figueroa, De la O-Villanueva, and De la Parra (2007) | School dust | Urban | Hermosillo, Sonora, Mexico | n.a. | 2.21 | 11.15 | n.a. | n.a. | 4.7 | 36 | 388 |
| Ferreira-Baptista and De Miguel (2005) | Street dust | Urban | Launda, Angola | 4700 | 2.7 | 26 | 11,100 | 238 | 9.7 | 306 | 271 |
| Du et al. (2013) | Road dust | Urban | Beijing, China | n.a. | n.a. | 69 | n.a. | n.a. | 26 | 202 | 219 |
| Yu, Wang, and Zhou (2014) | Street dust | Urban | Tianjin, China | n.a. | n.a. | 72 | n.a. | n.a. | n.a. | 61 | n.a. |
| Masto et al. (2017) | Street dust | Urban | Jharia, Dhanbad district, India | n.a. | 17 | 63 | n.a. | n.a. | 32 | 56 | 299 |
| Pathak et al. (2013) | Surface dust (Industrial) | Urban | Faridabad, India | 44,187 | 13 | 996 | 52,178 | 1385 | 74 | 57 | 502 |

(Du et al. 2013; Ferreira-Baptista and De Miguel 2005; Masto et al. 2017; Pathak et al. 2013; Yu, Wang, and Zhou 2014). Zn levels were significantly lower and similar to surface soils (Li et al. 2016) and household dust (Madany et al. 1994). However, Al levels were higher than values reported in the literature, and comparable to surface dust levels of industrial areas in Faridabad, India (Pathak et al. 2013).

The estimated median values of the enrichment factor (EF) of biomass and LPG-based households are shown in Figure 4. In both households, Pb (5.47–8.55) was higher than 5, indicating that it was significantly enriched and contaminated indoor dust. However, the median EF values of Co (0.43–0.87) and Mn (0.61–0.87) reaching 1 denotes their key contribution from nature or crustal origin (Han, Cao, and

Posmentier 2006). Further, EF values of Cr (1.1–2.21), Fe (1–1.57), Ni (0.5–1.24), and Zn (1.3–3.9) are < 2 and indicate a lack of pollution in indoor dust (Cheng et al. 2018; Han, Cao, and Posmentier 2006). The Pb and Cr levels were higher in LPG-based households than biomass households, while Co, Fe, Mn, Ni, and Zn exhibited the opposite trend. Additionally, Pb levels were higher in LPG-based households, which might have come from paints, varnishes, and unpaved roads, etc. (Hassanvand et al. 2015; Lin et al. 2015). On the other hand, Zn levels were higher in biomass-based households, which could have come from biomass burning deposits, since Zn is an essential element for plant growth (Salam et al. 2013). However, in LPG-based households, Zn could have come from outdoor dust and vehicle tire wear (Lu et al. 2014; Zhang et al. 2014).

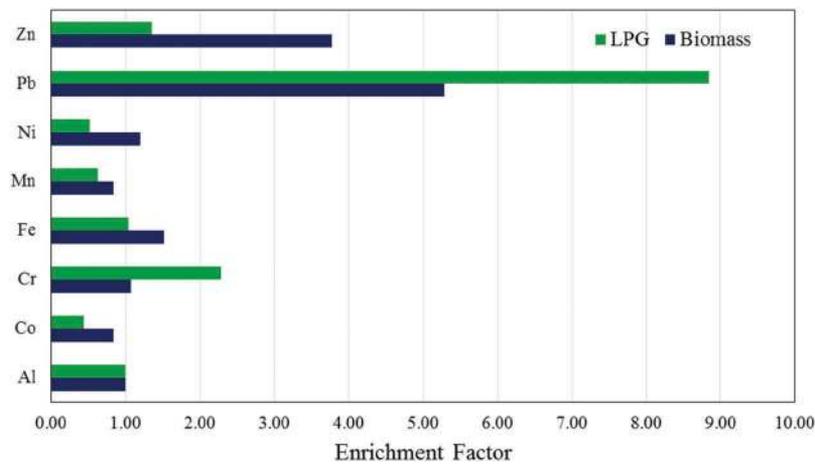


Figure 4. Enrichment factor of elements for indoor dust in biomass and LPG-based households.

Furthermore, Cr, Ni, and Zn deposits are from stainless steel utensils, metal objects, paints, and building materials (Lin et al. 2015; Lu et al. 2014).

Pearson’s correlation matrix was plotted for metals concentrations in biomass (Figure 5) and LPG (Figure 6) households to measure their degree of correlation with results obtained from EF analysis. In biomass-based households, positive correlation at 95% significance was observed for Al-Co, Al-Mn, Al-Zn, and Cr-Pb pairs; however, the Cr-Zn pair showed negative correlation. In LPG-based households, Cr-Pb and Fe-Mn had positive correlations, while Co-Cr and Co-Pb were negatively correlated. Major sources contributing to the positive correlation of Al with Co, and Mn and Fe-Mn, were natural crustal sources (Zhang et al. 2014). Moreover, Pb-Cr correlation in both households is

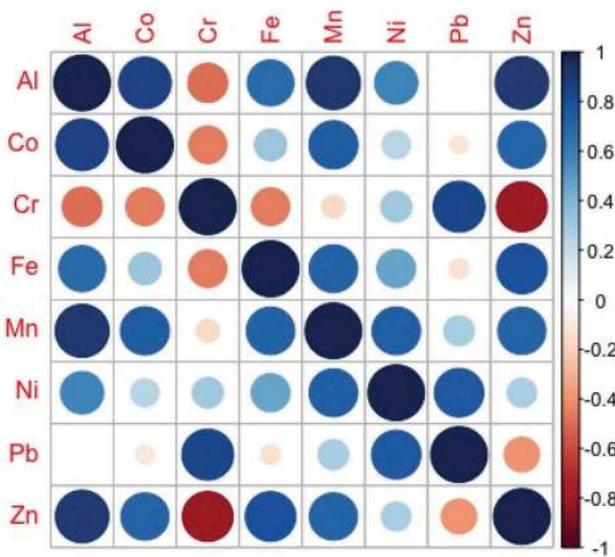


Figure 5. Correlation coefficient matrices for metal concentrations in biomass-based households.

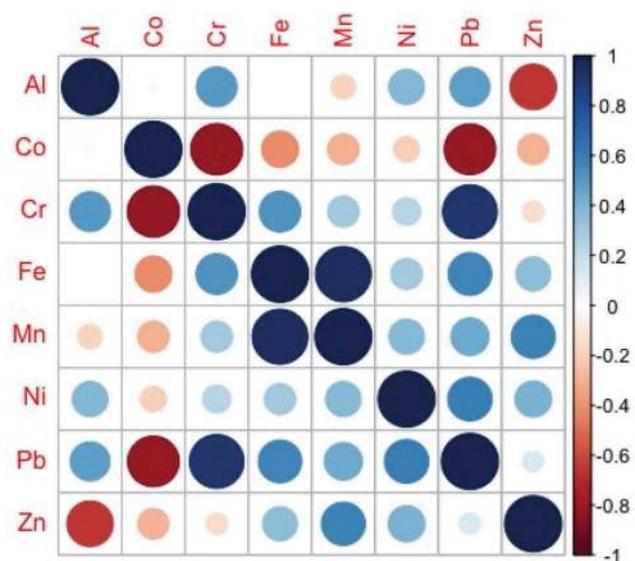


Figure 6. Correlation coefficient matrices for metal concentrations in LPG-based households.

primarily from leaded paints, apart from unpaved roads and vehicular emissions (Lee et al. 2016; Liu et al. 2014; Zhang et al. 2014).

The sources contributing to elemental concentrations in biomass and LPG-based households were identified using a PCA technique. Tables 4 and 5 depict the results of PCA analysis, such as factor loadings and eigenvalues for biomass and LPG-based households, respectively. In biomass-based households, two factors were extracted with initial eigenvalues > 1.0, which extracted 88.58% of the total variance. Factor loadings of Al, Fe, Mn, and Zn were high on Factor 1 with 56.94% of total variance, indicating that they were contributed by crustal sources dominated by laterite soils and biomass burning deposits (Salam et al. 2013; Zhang et al. 2014). On Factor 2, Cr, Pb, and Ni

Table 4. Matrix of factor analysis of metals in indoor dust of biomass households.

| Elements | Factor 1 | Factor 2 |
|-------------------------|--------------|--------------|
| Al | 0.458 | 0.065 |
| Co | 0.376 | -0.032 |
| Cr | -0.272 | 0.495 |
| Fe | 0.376 | 0.005 |
| Mn | 0.420 | 0.251 |
| Ni | 0.238 | 0.515 |
| Pb | -0.050 | 0.622 |
| Zn | 0.445 | -0.183 |
| Eigen value | 4.55 | 2.53 |
| Variance (%) | 56.94 | 31.64 |
| Cumulative variance (%) | 56.94 | 88.58 |

Table 5. Matrix of factor analysis of metals in indoor dust of LPG households.

| Elements | Factor 1 | Factor 2 | Factor 3 |
|-------------------------|--------------|--------------|--------------|
| Al | 0.136 | -0.585 | 0.378 |
| Co | -0.395 | 0.055 | 0.485 |
| Cr | 0.418 | -0.316 | -0.302 |
| Fe | 0.406 | 0.216 | -0.011 |
| Mn | 0.365 | 0.373 | 0.116 |
| Ni | 0.301 | 0.004 | 0.712 |
| Pb | 0.477 | -0.205 | -0.001 |
| Zn | 0.182 | 0.571 | 0.096 |
| Eigen value | 3.87687 | 2.20552 | 1.07754 |
| Variance (%) | 48.46 | 27.57 | 13.47 |
| Cumulative variance (%) | 48.46 | 76.03 | 89.50 |

showed great loadings with 31.64% of total variance, representing that they may have been contributed from paints, unpaved roads, and vehicular emissions (Lin et al. 2015; Lu et al. 2014; Yongming et al. 2006). However, in LPG-based households, three factors were obtained, accounting for 89.5% of the total variance. Factor 1 is dominated by Cr and Pb accounting for 48.46% of the total variance, once again reflecting the lead-chromate paints (Lee et al. 2016). Factor 2, dominated by Zn, Fe, and Mn, accounts for 27.57% of the total variance, considered to be soil or geogenic sources. Factor 3 is dominated by Ni, Co, and Al accounting for 13.47% of the total variance.

Health risk assessment

The health risk assessments of metals in indoor dust were calculated using three different pathways to

children (Table 6) and women (Table 7) for both fuel types in Kishannagar village, Telangana state, India. In terms of non-carcinogenic risk, ingestion is the dominant exposure pathway, followed by dermal contact and inhalation for children and adults, which is comparable to earlier studies (Ferreira-Baptista and De Miguel 2005; Lin et al. 2015; Lu et al. 2014). The highest ADD_{ing} value for children was 9.00E-04 for Zn (biomass); the lowest was 2.55E-05 for Co (LPG). Even amongst women, Zn (1.50E-04) had the highest ADD_{ing} value while Co (4.25E-06) had the lowest, but the levels were lower compared to children, as children are associated with hand-to-mouth activity resulting in higher values. Further, these results are consistent with earlier studies (Du et al. 2013). HQs of Co, Cr, Ni, Pb, and Zn through inhalation of dust particles were 2–5 orders lower compared to ingestion and dermal contact. HQ of Cr through ingestion was highest for children, however it was dermal in the case of women for all the other metals. The median values of HIs for all of the metals for children and women descended in the order of Cr > Pb > Ni > Zn > Co in both types of households. This study reveals that the HI values for metals due to indoor dust exposure were within safe values (USEPA 1989). Cr had the maximum HI levels, nearing 1, which may result in non-carcinogenic risk to children and women in both types of households in rural areas and cannot be disregarded. Carcinogenic risk through inhalation mode was only considered for the current assessment model, and it was observed that the levels of Co, Cr, and Ni were three orders lower than the acceptable range (10⁻⁶–10⁻⁴) for children and women in both types of households. Though the health risks associated with these heavy metals were within safe levels, they cannot be ignored. Few potential solutions to control metal levels in indoor dust can be using the paints or house building materials that have higher absorption characteristics, also using paints having a lower percentage of Pb

Table 6. Daily doses, hazard quotients, hazard indices, and cancer risks for children from metals in biomass and LPG-based households.

| | ADD _{ing} | ADD _{derm} | ADD _{inh} | HQ _{ing} | HQ _{derm} | HQ _{inh} | HI | Cancer risk |
|----------------|--------------------|---------------------|--------------------|-------------------|--------------------|-------------------|----------|-------------|
| Biomass | | | | | | | | |
| Co | 5.20E-05 | 1.55E-07 | 1.93E-09 | 1.73E-03 | 2.58E-05 | 6.75E-05 | 1.83E-03 | 1.62E-09 |
| Cr | 3.50E-04 | 1.04E-06 | 1.30E-08 | 1.17E-01 | 2.67E-02 | 4.54E-04 | 1.44E-01 | 4.68E-10 |
| Ni | 2.00E-04 | 5.95E-07 | 7.43E-09 | 1.00E-02 | 7.44E-04 | 2.89E-04 | 1.10E-02 | 5.35E-10 |
| Pb | 3.20E-04 | 9.52E-07 | 1.19E-08 | 9.14E-02 | 1.36E-03 | 3.38E-06 | 9.28E-02 | - |
| Zn | 9.00E-04 | 2.68E-06 | 3.34E-08 | 3.00E-03 | 4.46E-05 | 3.34E-06 | 3.05E-03 | - |
| LPG | | | | | | | | |
| Co | 2.55E-05 | 7.59E-08 | 9.47E-10 | 8.50E-04 | 1.26E-05 | 3.31E-05 | 8.96E-04 | 7.95E-10 |
| Cr | 7.00E-04 | 2.08E-06 | 2.60E-08 | 2.33E-01 | 5.34E-02 | 9.09E-04 | 2.88E-01 | 9.36E-10 |
| Ni | 8.00E-05 | 2.38E-07 | 2.97E-09 | 4.00E-03 | 2.98E-04 | 1.16E-04 | 4.41E-03 | 2.14E-10 |
| Pb | 5.00E-04 | 1.49E-06 | 1.86E-08 | 1.43E-01 | 2.13E-03 | 5.27E-06 | 1.45E-01 | - |
| Zn | 3.00E-04 | 8.93E-07 | 1.11E-08 | 1.00E-03 | 1.49E-05 | 1.11E-06 | 1.02E-03 | - |

Table 7. Daily doses, hazard quotients, hazard indices, and cancer risks for women from metals in biomass and LPG-based households.

| | ADD _{ing} | ADD _{derm} | ADD _{inh} | HQ _{ing} | HQ _{derm} | HQ _{inh} | HI | Cancer risk |
|----------------|--------------------|---------------------|--------------------|-------------------|--------------------|-------------------|----------|-------------|
| Biomass | | | | | | | | |
| Co | 8.67E-06 | 5.81E-07 | 1.02E-09 | 2.89E-04 | 9.68E-05 | 3.57E-05 | 4.21E-04 | 3.43E-09 |
| Cr | 5.83E-05 | 3.91E-06 | 6.86E-09 | 1.94E-02 | 1.00E-01 | 2.40E-04 | 1.20E-01 | 9.88E-10 |
| Ni | 3.33E-05 | 2.23E-06 | 3.92E-09 | 1.67E-03 | 2.79E-03 | 1.53E-04 | 4.61E-03 | 1.13E-09 |
| Pb | 5.33E-05 | 3.57E-06 | 6.27E-09 | 1.52E-02 | 5.10E-03 | 1.78E-06 | 2.03E-02 | - |
| Zn | 1.50E-04 | 1.00E-05 | 1.76E-08 | 5.00E-04 | 1.67E-04 | 1.76E-06 | 6.69E-04 | - |
| LPG | | | | | | | | |
| Co | 4.25E-06 | 2.85E-07 | 5.00E-10 | 1.42E-04 | 4.75E-05 | 1.75E-05 | 2.07E-04 | 1.68E-09 |
| Cr | 1.17E-04 | 7.82E-06 | 1.37E-08 | 3.89E-02 | 2.00E-01 | 4.80E-04 | 2.40E-01 | 1.98E-09 |
| Ni | 1.33E-05 | 8.93E-07 | 1.57E-09 | 6.67E-04 | 1.12E-03 | 6.10E-05 | 1.84E-03 | 4.52E-10 |
| Pb | 8.33E-05 | 5.58E-06 | 9.80E-09 | 2.38E-02 | 7.98E-03 | 2.79E-06 | 3.18E-02 | - |
| Zn | 5.00E-05 | 3.35E-06 | 5.88E-09 | 1.67E-04 | 5.58E-05 | 5.88E-07 | 2.23E-04 | - |

and Cr apart from regular cleaning of re-suspended dust in indoor areas.

The present study provides the latest geographical evidence of chemical and morphological characterization of indoor dust particles in households using biomass and LPG-based fuel and the associated health risk assessment in a sub-tropical rural site in Telangana, India. Nevertheless, further research is essential from various regions across the country for more heavy metal analysis and factors impacting their levels. One of the major limitations of the present study is the analysis of few metals and that it took place only during winter. Moreover, the measurements were carried out in living area locations; future studies can include soil and road dust as well as kitchens and bedrooms to provide more comprehensive analysis of dust compositions in varied environments.

Conclusion

Indoor dust samples in rural households relying on biomass and LPG fuels in Kishannagar village, Telangana state, India, were analyzed for Al, Fe, Mn, Zn, Cr, Pb, Ni, and Co levels, morphological characteristics, source contribution, and estimation of health risks associated with heavy metals for women and young children. It was observed that there is a significant difference in mean concentrations of Al, Co, Cr, Fe, Zn, and Ni based on fuel type. The EF values showed that in both types of households Pb was contaminated indoor dust. Pearson's correlation and PCA analysis revealed that the biomass households were dominated by Al, Fe, and Mn from crustal sources and laterite-rich soils, and Zn from biomass burning deposits. On the other hand, LPG households were dominated by Cr and Pb from paints, and Co and Ni from unpaved roads, stainless steel utensils, and metal objects. SEM-EDX morphological analysis helped in differentiating source contributions from biomass and LPG households. The health risks associated with these heavy metals to women and children were investigated

using the EPA health risk model (USEPA 1996). The values from the model indicated that both non-carcinogenic and carcinogenic risks were within the safe levels for both subjects. Thus, the present study provides the latest geographical evidence of chemical and morphological characteristics of indoor dust particles for varied fuels in a rural site in Telangana, India. The present study revealed that there is an urgent need for indoor air quality (IAQ) regulations; for instance, for leaded paints (Pb-Cr) and cooking chulhas. Further, there is a need to bring awareness of government schemes such as Pradhan Mantri Ujjwala Yojana (PMUY) sponsored by the government of India; these are significant as they help in the transition from biomass to LPG and thus reduce risk.

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