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# Polycyclic aromatic hydrocarbons in house dust and surface soil in major urban regions of Nepal: Implication on source apportionment and toxicological effect



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#### HIGHLIGHTS

- Concentration of  $\sum_{16}$  PAH in soil was 1.5 times higher than dust.
- HMW-PAHs were most abundant than LMW-PAH both in soil and dust.
- PAH chemicals both in soil and dust showed weak correlation with TOC/BC.
- Source apportionment study found mixed source from pyrogenic and petrogenic release.

### G R A P H I C A L A B S T R A C T



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#### ABSTRACT

#### Urban centers have turned to be the provincial store for resource consumptions and source releases of different types of semi-volatile organic compounds (SVOCs) including polycyclic aromatic hydrocarbons (PAHs), bringing about boundless environmental pollutions, among different issues. Human prosperity inside urban communities is unambiguously dependent on the status of urban soils and house dusts. However, environmental occurrence and sources of release of these SVOCs are challenging in Nepalese cities, as exceptionally very limited data are accessible. This motivated us to explore the environmental fate, their source/sink susceptibilities and health risk associated with PAHs. In this study, we investigated the contamination level, environmental fate and sources/sink of 16 EPA's priority pollutants in surface soil and house dusts from four major cities of Nepal. Additionally, the toxicological effect of individual PAH was studied to assess the health risk of PAHs. Generally, the concentrations of $\sum_{16}$ PAHs in surface soil were 1.5 times higher than house dust, and ranged 767-6770 ng/g dry weight (dw) (median 1810 ng/g dw), and 747-4910 dw (median 1320 ng/g dw), respectively. High molecular weight-PAHs both in soil and dust were more abundant than low molecular weight-PAHs, suggesting the dominance of pyrogenic source. Moderate to weak correlation of TOC and BC with PAHs in soil and dust suggested little or no role of soil organic carbon in sorption of PAHs. Source diagnostic ratio and principal component analysis indicated fossil fuel combustion, traffic/vehicular emissions and combustion of biomass are the principal sources of PAHs contamination in Nepalese urban environment. The high average TEQ value of PAHs in soil than dust suggested high risk of soil carcinogenicity compared to dust.

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#### 1. Introduction

Polycyclic aromatic hydrocarbons (PAHs) are semi-volatile organic compounds (SVOC). They are a class of persistent organic pollutants (POPs), and are omnipresent ecological contaminants. Some of them have been recognized as a cancer-causing and mutagenic substance (De La Torre Roche et al., 2009; IARC, 2010; Wang et al., 2011; Blokker et al., 2013; Shen et al., 2013; Lemieux et al., 2015). Likewise, they are an endocrine disrupting chemicals (EDCs) and can cause potential health hazards, including abnormalities in reproductive function, hormonal imbalance, testicular damages, development of cancer, neurological disorders, pre-mature birth, skin allergies, asthma, and several neuro developmental disorders (Bostrom et al., 2002; Deziel et al., 2013; Kim et al., 2013; Yang et al., 2015). Realizing the severe health risks and environmental threat concerned with PAHs, USEPA has categorized 16 different PAHs as priority pollutants that need further monitoring in different environmental matrices. These 16 EPA's priority pollutants are naphthalene (NAP), acenaphthene (ACE), acenaphthylene (ACY), fluorene (FLU), phenanthrene (PHE), anthracene (ANT), fluoranthene (FULA), pyrene (PYR), benzo[a]anthracene (BaA), chrysene (CHR), benzo[*b*]fluoranthene (BbF), benzo[*k*]fluoranthene(BkF), benzo[*a*]pyrene (BaP), dibenz[a,h]anthracene (DahA), indeno[1,2,3*cd*]pyrene (IcdP), and benzo[*g*,*h*,*i*]perylene (BghiP) (Kwon and Choi, 2014). Besides, the International Agency for Research on Cancer (IARC) has also recommended eight PAHs (BaP, BaA, BbF, BkF, CHR, IcdP, DahA, and NAP) as category-II pollutants in light of their cancer causing characteristics and mutagenic potential (IARC, 1987).

Despite PAHs enter into the environment by both natural processes (forest fires and volcanic activities), and the anthropogenic sources (traffic, fossil fuel combustion, and industrial processes)(Aydin et al., 2014; Wang et al., 2015), notwithstanding, the most widely recognized source of PAHs are anthropogenic origin. It results from partial combustion or pyrolysis of fossil fuels and biomass, together with the spillage of petroleum (Bamford and Baker, 2003; Dong and Lee, 2009; Wang et al., 2011). Anthropogenic sources of PAHs can be separated into two groups, i.e. petrogenic and pyrogenic. Burning of biomass generates pyrogenic PAHs, while petrogenic PAHs originates from oil-based commodities, including kerosene, gasoline, diesel fuel, lubricating oil, and asphalt (Baek et al., 1991; Yunker et al., 2002).

Atmospheric PAHs can also enter surface soil by wet and dry deposition (Kaya et al., 2012), and are promptly adsorbed by soil particles and soil organic matter (SOM). Following changes in environmental conditions, soil may re-emit the previously adsorbed PAHs into the air (Tao et al., 2008). A crop being the primary food source, an uptake of PAHs from polluted soil and their subsequent consumption leads to bioaccumulation of PAHs in human by means of food chain (Li et al., 2008). Therefore, profound understanding of the concentrations and dispersion of PAHs in soil may boost pollution control, reduce crop damage, and minimizes the human exposure. Soil contaminated with PAHs is a significant environmental problem worldwide. It is viewed as a steady indicator of the level of environmental pollution (Wild and Jones, 1995; Liu et al., 2001; Mueller et al., 2006; Wang et al., 2012) and has attracted considerable attention worldwide (Bourotte et al., 2005; Callen et al., 2011). On the other side, PAHs in indoor environments are released from different activities, for example, utilization of electric or gas stove for heating and cooking, coal and candle burning, smoking and parquet flooring (Huynh et al., 1991; Turner et al., 1992; Chao et al., 1997; Heudorf and Angerer, 2001; Huali et al., 2002; Guo et al., 2003; Ohura et al., 2004; Chen et al., 2005; He et al., 2005; Li et al., 2005). Incense burning is another essential source of PAHs in many South Asian countries. Moreover, the penetration of air and soil loaded with PAHs inside indoor environment from outdoor may also marginally influence indoor concentration (Chuang et al., 1995; Sanderson and Farant, 2004). Organic pollutants are known to build up in indoor environment due to the limited ventilation and lack of direct sunlight (Butte and Heinzow, 2002; Santillo et al., 2003). The house dust in such indoor environment may archive, hence, the analyses of organic pollutants in house dust can give a reliable indication of the extent of indoor contaminations (Butte and Heinzow, 2002).

Past investigation have discovered that high level of PAHs in surface soil and house dusts not only influence the physicochemical characteristics of soil, but also equally impact the environment and the human population living there (Blake et al., 2007; Haugland et al., 2008; Jiao et al., 2009; Mostert et al., 2012). Thus, knowledge about the dissemination of PAHs in surface soil and house dusts together with their likely sources of release are basic to limit the ecological dangers. Evidence suggests that anthropogenic activities identified with industrializations and urbanization intensely impact pollution levels in urban areas (Liu et al., 2010; Pataki et al., 2011; Luo et al., 2012; Vane et al., 2014; Gu et al., 2016a; Jiang et al., 2016; Marquez-Bravo et al., 2016). Urban centers have turned to be provincial store for asset utilization and sources of chemical emissions, bringing about boundless environmental pollutions among different issues (Chung et al., 2007; Cachada et al., 2016; Gu et al., 2016a). Human health inside urban communities is unequivocally reliant on the status of urban soils and house dusts (Imperato et al., 2003; Gu et al., 2016b). Therefore, understanding the behavior of organic pollutants, determining their spatial assortment and source/sink susceptibilities will help to shape an extremely conclusive approach in urban areas to avert contamination. Moreover, the knowledge about organic pollutants in soil will additionally help to evaluate health risks, to drive their environmental fate and to improve the air quality. The assurance of the levels and the spatial conveyance of these SVOCs are challenging in Nepalese cities, as exceptionally restricted data are accessible. Detailed about PAHs concentrations and environmental fate in the urban soil and dust of Nepal have not been studied so far. This absence of information poses troubles for pollution control and management. In this manner, this study investigates the environmental concentrations, spatial distribution pattern, and emission sources of PAHs in soil and house dusts from four major urban region of Nepal. The database generated from this investigation will be significantly useful in characterizing urban soil and dust with respect to PAHs emissions in Nepal. Knowledge about PAHs source comprehends the regional and global dynamics of their movement through various environmental compartments. Additionally, this study would be able to assess the quality of soil on regional scale which will be useful for making decision in land use planning for urban region.

#### 2. Materials and methods

Detailed descriptions of the materials and methods are described in supporting information.

#### 2.1. Soil and dust sampling

Details about study area, sampling locations and pretreatment methods are well discussed in our previous studies (Yadav et al., 2016, 2017a, 2017b). Briefly, four major cities of Nepal were selected for the collection of surface soil and house dusts. Both the surface soil and house dusts were collected during October 2014 using stainless steel shovel at depth of 0-15 cm (vegetation removed). Altogether 72 surface soil samples were acquired and mixed to make 24 representative samples. Hence, each sample was a combination of at least 3 sub samples obtained in different direction at distance of 5 m. Likewise, 24 house dusts were gathered from 24 different households in in all four cities (6 each) considering domestic, educational, residential, commercial, public places and office premises. Both the surface soil and dust samples were wrapped in aluminum foil and packed in zipper bag, and transported by air-mail to Organic Geochemistry Laboratory of Guangzhou Institute of Geochemistry, Guangzhou, China keeping in ice-box.

#### 2.2. Determination of total organic carbon (TOC) and black carbon (BC)

The total organic carbon (TOC) in soil and dust were determined as per method detailed by Ramu et al. (2010). Briefly, about 2–3 g of dried, sieved and homogenized soil/dust was mixed with 3 mL of HCI (10%) to eliminate inorganic carbon, rinsed thrice with Milli-Q water and dried overnight in oven at 45 °C. A portion of the soil/dust samples were taken to determine TOC. The TOC analysis was performed with Elemental Carbon-Hydrogen-Nitrogen (CHN) Analyzer (Elementar VARIO EL III). The analysis of BC in soil/dust was done following chemo-thermal oxidation (CTO-375) technique described elsewhere (Gustafsson et al., 2001; Elmquist et al., 2008). Briefly, 2–3 g of dried, sieved and homogenized soil/dust was burnt in a muffle furnace to thermal oxidation (375 °C, 18 h) under constant air flow. The samples were then mixed with 1 N HCl to remove inorganic carbon. Later, the remaining carbon content was determined by CHN Analyzer as BC.

#### 2.3. Sample extraction and analysis

Both surface soil and house dust were spiked with  $5 \,\mu\text{L}$  (200  $\mu\text{g/L}$ ) of the deuterated PAHs recovery standard (naphthalene-d8, phenanthrene-d10, acenaphthene-d10, perylene-d12, and chrysened12). Small chips of copper granules were added in extracting flask to remove the elemental Sulphur. The samples were then soxhlet extracted with dichloromethane (DCM) for 24 h. The extractant was then reduced to 0.5 mL volume by rotary evaporator (Heildolph 4000, Germany) and were solvent exchanged to hexane. Later, the aliquant was purified following alumina/silica column (8 mm i.d) closely packed with neutral alumina (3 cm, 3% deactivated), neutral silica gel (3 cm, 3% deactivated) and anhydrous sodium sulphate (1 cm). Final cleaned fraction was eluted with 15 mL mixture of DCM and hexane (1:1 by vol.) and eluted fraction was concentrated to 25 µL under gentle nitrogen stream. Before GC–MS analysis, 5 µL (200 µg/L) of hexamethylbenzene (Aldrich Chemical, Gillingham, Dorset, USA) was used as internal standard. Details about chemical extractions and GC-MS analysis are discussed in our previous work (Devi et al., 2016). Totally, 16PAHs were detected using GC-MS Agilent (6890 N/5975 MSD), attached with a 30 m  $\times$  0.25 mm i.d  $\times$  0.25 µm film thickness DB-5 capillary column coupled with HP-5972 mass selective detector operated in EI mode (70ev). The injector temperature was 270 °C, while detector temperature was fixed to 280 °C during analysis. The oven temperature was kept at 60 °C for 5 min and increased to 290 °C at a rate of 3 °C min<sup>-1</sup>. Helium was used as carrier gas at consistent flow of 1.5 mL min $^{-1}$ .

#### 2.4. Quality assurance and quality control (QA/QC)

Strict QA and QC protocol were followed both during sampling and experimental analysis. The performance of GC–MS was tested daily by injecting 6 sets of PAHs calibration standards. The instrument deviation was less than  $\pm$  10%. A total of 6 laboratory blanks were analyzed in the similar way as original samples. NAP was the only chemical regularly detected in blank samples. Method detection limits (MDLs) were calculated as the mean plus 3 times the standard deviations of all the blank samples. When the PAHs were not detected in blank samples, the MDLs were calculated as 3 times signal-to-noise ratio obtained from lowest spiked calibration standard. The MDL ranged from 20 to 200 pg/g for PAHs. The average recovery of deuterated-PAHs both in soil and dust were 79%  $\pm$  11% for acenapthene-d10, 82%  $\pm$  14% for phenanthrene-d10, 80%  $\pm$  10% for Chrysene-d12, and 89%  $\pm$  12% for perylene-d12. PAH concentrations in soil/dust were expressed on dry weight (dw) basis and were corrected for blank but not for recovery.

#### 2.5. Statistical analysis

Statistical analysis of GC–MS quantified data such as descriptive statistics, and graphical presentation such as box and whisker plots, profile and relative abundance were made with MS excel 2010/IBM SPSS statistics (v-21). Observed data with non-detection limit (nd) was changed to zero for statistical calculation. The spatial distribution map of PAHs was made using ESRI-Arc GIS geospatial (version 10.3) software.

#### 3. Results and discussion

#### 3.1. Overall comments on PAHs

The concentration of  $\sum_{16}$  PAHs,  $\sum_{8}$  Car-PAHs, together with individual PAH measured in soil and dust in this study has been presented in Table 1. All the 16 EPA's priority pollutants were detected at each site with 100% detection frequency (DF). Generally, the concentration of  $\sum_{16}$  PAHs were measured 1.5 times higher in soil than dust, and ranged 767-6770 ng/g dw (median 1810 ng/g dw), and 747-4910 ng/g dw (median 1320 ng/g dw), respectively. This suggests the soil in Nepalese urban region have been more influenced by deposition of PAHs from vehicular emission and industrial release. Contrary to this, the dust in indoor environment is relatively protected from vehicular pollution and industrial release. Eight carcinogenic PAHs (CHR, BaA, BbF, BkF, BaP, IcdP, DahA and BghiP) were also detected high in soil than dust. The concentration of  $\sum_8 Car$ -PAHs in soil and dust ranged from 430 to 4730 ng/g dw (median 1130 ng/g dw) and 415-2760 ng/g dw (median 651 ng/g dw), respectively (Table 1). The concentration of well-known carcinogen, BaP in the soil and dust ranged from 61.5-344 ng/g (median 77.5 ng/g dw) and 61.2-419 ng/g dw (median 93.7 ng/g dw), respectively. The sum of the concentrations of 8Car-PAHs was approximately >50% of the  $\sum_{16}$  PAH analyzed in soil and dust

High molecular weight PAHs (HMW-PAHs) were more abundant (70–75%) both in soil and dust than low molecular weight PAHs (LMW-PAHs) (24–30%). The concentration of HMW-PAHs ranged from 538 to 5370 ng/g dw (1290 ng/g dw) and 523–3820 ng/g dw (median 886 ng/g dw) in soil and dust, respectively. The high level of HMW-PAHs both in soil and dust indicates dominance of pyrogenic source over petrogenic source (Zakaria et al., 2002; Vinas et al., 2009). The elevated level of HMW-PAHs in soil could be because of higher persistency and tendency of HMW-PAHs to accumulate in soil (Chung et al., 2007). The concentrations of LMW-PAHs were measured low, and accounted for only 24–30% of  $\sum_{16}$ PAHs, which might be because of their degradation (by photolysis) and volatilization from soil (Morillo et al., 2007; Marquez-Bravo et al., 2016). Nepal being tropical region (annual

Table 1

Statistical summary of  $\sum_{16}$  PAHs,  $\sum_{8}$  Car-PAHs and individual PAH in soil and dust (ng/g).

Compounds	Surface soil				House dust			
	Min	Max	Median	Std. Dev.	Min	Max	Median	Std. Dev.
NAP	35.3	187	125	37.2	49.2	305	103	55.1
ACY	26.8	79.8	29.5	11.0	18.3	58.1	31.2	7.25
ACE	8.57	130	22.4	37.6	5.33	117	17.9	27.3
FLU	34.4	62.8	50.9	8.79	35.2	70.6	48.3	8.90
PHE	75.0	468	216	126	71.8	417	173	85.5
ANT	49.2	470	84.1	129	45.1	121	61.7	18.8
$\sum$ LMW-PAHs	229	1400	527	349	225	1090	434	203
FLUA	65.8	318	89.4	65.2	66.4	506	128	105
PYR	42.3	327	66.9	71.8	41.4	554	107	114
CHR	70.9	2950	713	919	125	298	157	44.5
BaA	31.6	254	51.9	80.8	31.3	497	78.4	106
BbF	9.64	276	16.4	93.9	9.67	490	18.4	122
BkF	38.0	321	47.7	92.9	38.2	446	74.8	100
BaP	61.5	344	77.5	87.7	61.2	419	93.7	93.6
IcdP	42.9	182	47.7	33.1	43.4	170	115	21.9
DahA	112	140	113	7.23	43.4	156	46.2	31.5
BghiP	62.9	253	63.8	42.3	62.9	282	68.1	48.5
$\sum$ HMW-PAHs	538	5370	1290	1490	523	3820	886	788
$\sum$ Car-PAHs	430	4730	1130	1360	415	2760	651	569
$\sum_{16}$ PAHs	767	6770	1810	1840	747	4910	1320	991

average temperature ranged from 10 to 40 °C), degradation by photolysis seems true. Chemical structure and molecular weight of PAHs compounds equally influence photolysis reactions (Korfmacher et al., 1980; Behymer and Hites, 1988; Finlayson-Pitts and Pitts, 1997). For example, linear 2-ring PAHs and some clustered PAHs can rapidly degrade under direct sunlight. Further, LMW-PAHs are more susceptible to photolysis, in light of the fact that these compounds are more bioavailable and will have longer exposure times to sunlight (Korfmacher et al., 1980). LMW-PAHs can also easily subject to weathering compared to HMW-PAHs (Giger and Blumer, 1974; Lee et al., 1977). Weathering mainly results from evaporation and dissolution which impact more volatile and soluble LMW-PAHs (Hostettler et al., 2007). The level of evaporation depends on the kind of petroleum included involved. Light products containing light PAHs evaporate readily, whereas heavier ones lose as little as 5-10% of their aggregate volume (Philp, 2007). Likewise, dissolution depends basically on the structure of the PAHs and declines with increase in ring number and alkylation level of the PAHs (Stout et al., 2001). The CHR was identified as the most abundant PAH measured in soil, and accounted for 39% of  $\sum_{16}$  PAHs, while PHE exceeded in dust, accounting for 13% of  $\sum_{16}$  PAHs. The PAH congeners follow the order from high to low: CHR > PHE > NAP > DahA > FLUA > ANT > BaP > BghiP > PYR > Flu > IcdP > BkF > BaA > ACY > ACE > BbF in soil, and PHE > CHR > FLUA >> DahA > PYR > NAP > BaP > BkF > BaA > ANT > BghiP > FLU > IcdP > ACY > ACE > BbF in dust.

#### 3.2. Concentration, profile and spatial distribution of PAHs in surface soil

In soil, PAHs results from both natural process as well as anthropogenic release (mainly released from fossil fuel combustions, vehicle pollutions, biomass combustions, industrial process and leakage of oils) (Peng et al., 2011). Results of the  $\sum_{16}$  PAHs along with individual PAH measured in soil in this study are given in Table S1. Relatively high concentrations of  $\sum_{16}$  PAHs were measured in Birgunj and Kathmandu than those in Pokhara and Biratnagar. The concentration of  $\sum_{16}$  PAHs ranged from 925 to 4840 ng/g dw (median 2460 ng/g dw), 880-6600 ng/g dw (median 1720 ng/g dw), 791-5350 ng/g dw (median 2630 ng/g dw) and 961-4070 ng/g dw (median 1940 ng/g dw) in Kathmandu, Pokhara, Birgunj, and Biratnagar, respectively (Fig. S1). Urban areas are more prone to PAH contaminations due to high density of automobiles and industrial activities (Stanković et al., 2008). Majority of PAHs received in such areas are believed to accumulate in surface soil (Agarwal et al., 2009). Kathmandu (part of Bagmati zone with population about 1.4 millions), the capital city of the country marginally has the largest numbers of registered vehicles (with 1,042,856 registered vehicles as on 2013), while Birguni is the gateway of Nepal as most of its trade with India through open border (CBS/NPCS, 2011; DoTM Nepal, 2017). Birgunj (part of Narayani zone with population over 0.2 millions) holds the second largest number of registered vehicles (with 625,089 registered vehicles as on 2013) after Kathmandu, which directly corresponds to elevated level of PAHs in soil (CBS/NPCS, 2011; DoTM Nepal, 2017). Brick manufacturing is another important industry in Kathmandu, which utilizes large amounts of coal imported from India (FAO, 1993). It is estimated that >100 brick kilns are located on the outskirt of Kathmandu city alone which indiscriminately usages low quality coal (CBS, 2006; ICIMOD, 2007; Kishida et al., 2009). Additionally, Nepal encounters intense power deficiency throughout the year with daily power cut for 10-18 h. This has prompted utilization of about 250,000 diesel-based small generators in Kathmandu city alone (Chen et al., 2015).

The concentration of  $\sum_{16}$ PAHs measured in soil in this study was compared with neighboring countries and other urban regions of the world and are given in Table S2. When compared with neighboring countries, the concentrations of  $\sum_{16}$ PAHs in this study were in the same range as compared to those reported in Delhi (India) (1910 ng/g), Shanghai (China) (1800 ng/g) (Wang et al., 2013) and Beijing (China) (1640 ng/g) (Li et al., 2006), however these concentrations

were relatively higher as compared to Yellow River Delta basin in China (118 ng/g) (Yuan et al., 2014) and hilly region in India (458) (Devi et al., 2016). Masih and Taneja (2006) reported 4-5 times high level of  $\sum$  PAHs (9370 ng/g) than this study in surface soil of Agra, India. The level of  $\sum_{16}$  PAHs in soil in this study was 3–4 times greater than those reported in urban soil from Torino in Italy (704 ng/g) (Cachada et al., 2009), Ljubljana in Slovenia (791 ng/g) (Cachada et al., 2009), Ulsan in South Korea (960 ng/g) (Kwon and Choi, 2014) and Terragona in Spain (438 ng/g) (Nadal et al., 2007). The sources of these PAHs were linked with road traffic pollutions. Besides, the level of  $\sum_{16}$  PAHs in soil in this study was also comparable with study reported in urban soil of Bergen in Norway (1470 ng/g) (Haugland et al., 2008). However, the  $\sum_{16}$  PAHs level measured in soil in this study was 2–3 times lower than those reported in urban soil from New York in USA (5400 ng/g) (Marquez-Bravo et al., 2016) and Glasgow in Scotland (8337 ng/g) (Cachada et al., 2009). Moreover, much higher level of  $\sum_{16}$  PAHs than this study was also reported in urban soil from Greater London in UK (18,000 ng/g) (Vane et al., 2014). This showed declining level of soil  $\sum$  PAHs in European countries and some developed countries in last decades. On contrary, the levels of  $\sum_{16}$  PAHs in soil in Nepal are still comparable to those of India and China indicating more use of fossil fuels and petroleum product in urban region in Nepal.

In order to classify the contamination level of PAHs in soil, the levels of  $\sum_{16}$ PAHs measured in this study were compared with contamination classification proposed by Maliszewska-Kordy-bach (1996) who categorized four different class of contamination level; uncontaminated-( $\sum_{16}$ PAH < 200 ng/g), weakly contaminated-( $\sum_{16}$ PAH = 200–600 ng/g), contaminated-( $\sum_{16}$ PAH = 600–1000 ng/g), and heavily contaminated soil ( $\sum_{16}$ PAH > 1000 ng/g). Based on this classification, we found the surface soil in Nepal is heavily contaminated with PAHs.

The composition profile of PAHs as depicted in Fig. 1 and Fig. S2 showed that 4-ring PAHs were most dominant followed by 3-ring and 5-ring PAHs, and accounted for 53%, 20% and 17% of  $\sum_{16}$ PAHs, respectively. This finding is consistent with previous studies of ring-PAHs in urban soil from Guangzhou (Ke et al., 2017) and Shanxi (Jiao et al., 2017) in China. HMW-PAHs (4–6 rings) are representative of pyrogenic source, while LMW-PAHs (with 2–3 rings) indicate petrogenic emissions (Ke et al., 2017). In this study, 4–6 ring PAHs contributed 76% of  $\sum_{16}$ PAHs, strongly implying dominance of pyrogenic sources (Vinas et al., 2009).

The profile of the individual PAH in soil is shown in Fig. 2 and Fig. S3. It is evident from Fig. 2 that the  $\sum_{16}$  PAHs level in soil in this study was mostly dominated by CHR, followed by PHE, NAP, DahA and FLUA, and accounted for 22–50%, 6–18%, 5–7%, 4–7% and 4–5% of  $\sum_{16}$  PAHs, respectively. CHR is an important indicator of biomass/coal combustions (Wang et al., 2013; Yang et al., 2013). This is true for many developing nation especially in case of Nepal where biomass burning or coal combustion is the principal and economically viable source of energy in many Nepalese houses for cooking and heating (Pokhrel et al., 2015). This finding is consistent with previous study by Wang et al. (2015), who also found high proportion of CHR (11.2%) in urban soil from Shanghai in China. Next to CHR, PHE was the second most abundant PAH detected in Kathmandu (386 ng/g dw), followed by Pokhara (317 ng/g dw), Birgunj (156 ng/g dw) and Biratnagar (141 ng/g dw), respectively. Similar pattern of individual PAH has been reported previously in urban soil around the world (Lors et al., 2012; Radzi et al., 2016; Liu et al., 2017). This suggests that those selected PAHs (CHR and PHE) may be relatively stable, thereby resulting abundance in soil. The concentration of BaP was less detected in soil, and accounted for 3-5% of  $\sum_{16}$  PAHs.

The spatial distributions map showing the distribution patterns of  $\sum$  HMW-PAHs and  $\sum$  LMW-PAHs in four Nepalese cities are shown in Fig. 3. It is evident from Fig. 3 that HMW-PAHs were more prevalent than LMW-PAHs at all sites. LMW-PAHs are relatively more volatile, and can more readily desorb from soil becoming more susceptible to



Fig. 1. Box and whisker-plots showing concentrations of ring-PAHs in soil (top) and dust (bottom) of Nepalese cities. The central box represents the concentration from 25 to 75 percentile. The middle bold line represents the median value. The asterisk/stars are extreme outlier that are >3-times beyond interquartile range. This graph showed the abundance of 4-ring PAHs both in soil and dust.

degradation (Pannu et al., 2003), while HMW-PAHs are persistent in the environment and are recalcitrant to degradation due to dense clouds of pi-electron surrounding the aromatic rings (Johnson et al., 2005). However, exceptionally high levels of PHE measured in soil in this study were due to fact that PHE is thermodynamically the most stable compound among 3-ring parent PAHs (Vane et al., 2014). Substantially high level of  $\sum_{16}$  PAHs was observed at BRS-6 (4880 ng/g dw) and BRS-1(3980 ng/g dw) in Birgunj, KTS-4 (4290 ng/g dw) in Kathmandu, and PKS-2 (3970 ng/g dw) in Pokhara (Fig. S4). Both sites BRS-6 and BRS-1 are located in suburban-residential area. As a result, emission from biomass combustion used for cooking and heating in residential houses would lead to high concentration of PAHs in surface soil. Further, a number of small and medium scale industries are located very close to site BRS-6, which could also impact level of PAHs in soil (Odabasi et al., 2016; Cetin et al., 2017). KTS-4 is urban commercial area, while PKS-2 is urban-residential area with close proximity to airport. High level of  $\sum_{16}$  PAHs was also measured at KTS-3(3520 ng/g dw) in Kathmandu, PKS-5 (3530 ng/g dw) in Pokhara, and BTS-5 (3770 ng/g dw) in Biratnagar. Likewise, relatively low level of  $\sum_{16}$  PAHs was detected at BRS-3(922 ng/g dw) in Birgunj and BTS-2 (1010 ng/g dw) in Biratnagar. Both KTS-3 and PKS-5 are located in industrial area, surrounded by small and medium scale industries. As a result, the industrial emission would lead to high level of PAH at KTS-3 and PKS-5. This finding is consistent with previous study by Radzi et al. (2016), who also measured high level of PAHs near industrial area in urban soil of Malaysia.

#### 3.3. Concentration, profile and spatial distribution of PAHs in house dust

The concentrations of  $\sum_{16}$  PAHs together with individual PAH analyzed in house dusts in this study are given in Table S3. Similar to soil samples, higher levels of  $\sum_{16}$  PAHs were also measured in Kathmandu and Birgunj than Pokhara and Biratnagar (Table S3). The concentration of  $\sum_{16}$  PAH ranged from 1010 to 4540 ng/g dw (median 1498 ng/g dw), 1060-2480 ng/g dw (median 1360 ng/g dw), 763-2570 ng/g dw (median 1410 ng/g dw) and 948-3310 ng/g dw (median 1260 ng/g dw) in Kathmandu, Birgunj, Pokhara and Biratnagar, respectively. The levels of  $\sum_{16}$  PAHs measured in dust in this study were compared with previous studies around the globe and are stated in Table S4. The levels of  $\sum_{16}$  PAHs observed in this study were 3–40 times higher than those reported in house dust from Brno in Czech Republic (39.1 ng/g) (Melymuk et al., 2016), Berlin in Germany (300 ng/g) (Fromme et al., 2004) and California in USA (990 ng/g) (Whitehead et al., 2013), but comparable with those from Kuwait (1675 ng/g) (Ali et al., 2016), Brittany in France (1029 ng/g) (Raffy et al., 2017), Jeddah in Saudi Arabia (2650 ng/g) (Ali et al., 2016), and Kocaeli in Turkey (2489 ng/g) (Civan and Kara, 2016). However, the  $\sum_{16}$  PAHs level in dust in this study was 3-10 times lower than those reported in Guizhou in China



Fig. 2. Profile of individual PAH showing % contribution to  $\sum_{16}$ PAHs in soil (top) and dust (bottom).The stacked plot showing dominance of CHR and PHE in soil and dust, respectively.

(6780 ng/g) (Yang et al., 2015), Ottawa in Canada (9530 ng/g) (Maertens et al., 2008), Texas in USA (28,800 ng/g) (Mahler et al., 2010), and several other Chinese cities (10,300 ng/g) (Qi et al., 2014).

The concentration of well-known carcinogenic BaP ranged from 61 to 180 ng/g dw (median 97 ng/g), 80–419 ng/g dw (median 114 ng/g dw), 62–183 ng/g dw (median 111 ng/g dw) and 76–302 ng/g dw (median 87 ng/g dw) in Kathmandu, Pokhara, Birgunj, and Biratnagar, respectively. The concentration of BaP in dust in this study was compared with German indoor air quality guideline for PAHs in house dust recommended by German Federal Environmental Agency (Heudorf and Angerer, 2001). According to this guideline, the concentration of BaP in dust shouldn't exceed 10,000 ng/g limits to cause adverse health effect. In this study, the measured BaP in all four cities were 1–2 orders of magnitude lower than prescribed limit, suggesting no adverse health effect.

Like soil, 4-ring PAHs were also most dominant PAHs in dust, followed by 5-ring PAHs and 3-ring PAHs and accounted for 36%, 26% and 22% of  $\sum_{16}$ PAHs, respectively (Fig. 1 and Fig. S2). Moreover, the 2–3 ring PAHs which are relatively more volatile, measured low. The LMW-PAHs are more liable to degradation and are usually less toxic, while HMW-PAHs are persistent and more toxic in the environment. This suggests ubiquitous pollution of HMW-PAHs and higher risk of PAHs exposure in Nepalese houses. High proportion of 4–6 ring PAHs was also reported previously in indoor dust from China (Qi et al., 2014). The profiles of the individual PAH measured in dust are illustrated in Fig. 3. It is clear from Fig. 3 that PHE was the most recognized PAH, followed by CHR, FLUA, DahA and PYR, and accounted for 10–13%, 11–12%, 8–11%, 8–9% and 6–10% of  $\sum_{16}$ PAHs, respectively. Similar patterns of PAHs have been reported previously in house dust from Kuwait (Gevao et al., 2007). The spatial distribution map showing the distributions of HMW-PAHs and LMW-PAHs in dust of Nepalese cities are shown in Fig. 4. It is evident from Fig. 4 that the HMW-PAHs were also dominant in dust, and accounted for 64–79% of  $\sum_{16}$  PAHs. Substantially higher levels of HMW-PAHs in house dust than LMW-PAHs have been reported earlier in China (Qi et al., 2014), Saudi Arabia and Kuwait (Ali et al., 2016). HMW-PAHs mainly originate from high-temperature combustion source, for instance, vehicular emission, while LMW-PAH mainly derived from moderate temperature combustion process, for instance, coal burning and petroleum source (Han et al., 2011; Yang et al., 2015). Under standard temperature of 25 °C in most houses, the HMW-PAHs get associated with dust, whereas LMW-PAHs still remain in vapor phase. Elevated level of  $\sum_{16}$  PAHs was observed at KTD-5 (4410 ng/g dw) in Kathmandu and BID-1 in Biratnagar (3220 ng/g dw). KTD-5 is urban-commercial area, while BID-1 is suburban residential area. Higher concentrations of  $\sum_{16}$  PAHs were also observed at BRD-5 (2200 ng/g dw) and BRD-4(1970 ng/g dw) in Birgunj, PKD-3(2040 ng/g dw) and PKD-5(1960 ng/g dw) in Pokhara, KTD-1 (1840 ng/g dw) and KTD-4 (1740 ng/g dw) in Kathmandu, and BID-3 in Biratnagar, respectively. Both BRD4 and BRD 5 are suburban residential areas; PKD-3 is urban area with intense traffic while PKD-5 is related to urban residential area. Likewise, KTD-1 and KTD-4 are closed to heavy traffic emission area, while BID-3 is close to airport. Relatively low level of  $\sum_{16}$  PAHs was measured at BRD-6 in Birgunj which is urban residential area.

#### 3.4. Spearman's rank correlation analysis

TOC and BC content in soils are known to influence the contamination level of PAHs in urban soil and dusts (Bucheli and Gustafsson, 2000; Agarwal and Bucheli, 2011; Duan et al., 2015; Cachada et al., 2016). In this study, both the TOC and BC contents were measured higher in house dust than surface soil. The % TOC contents in soil and dusts ranged from 0.51–10.6% (median 4.0%) and 0.10–2.47% (median 0.88%), respectively. BC in soil was quantified in the range of 0.07– 1.86% (median 0.32%) and 0.03–0.58% (median 0.10%) for dust and soil, respectively. BC/TOC ratio in soil ranged from 0.02–0.67% (median 0.16%) which is much lower than those reported in urban soil from India (6–22%) (Agarwal and Bucheli, 2011), Germany (18–70%) (Lorenz et al., 2006), and UK (12–33%) (Rawlins et al., 2008), but comparable to background soil from UK and Norway (<1) (Nam et al., 2008).

A spearman's rank correlation coefficient analysis was performed to investigate the influence of TOC and BC in dissemination of PAHs in soil/ dust (Table S5 and S6). Correlation analysis result showed that the individual PAH was weakly-moderately correlated with TOC, indicating non-equilibrium condition for adsorption between PAHs and TOC. PHE was the lonely PAH in soil which moderately linked with TOC (r =0.542, p < 0.05). Weak correlations between PAH with TOC are common in soils and have been widely reported (Bucheli et al., 2004; Wang et al., 2013; Vane et al., 2014; Bi et al., 2016). Slightly weaker correlation of PAHs with TOC suggests constant and continuous input of fresh PAHs in Nepalese soils until equilibrium reached between TOC and PAHs (Wang et al., 2010).

Although BC is considered as better adsorbent for PAHs than TOC in soil/dust due to co-emission of BC and PAHs in soil, the sorption ability of BC significantly depends on molecular weight and study area (urban or remote) of the PAH (Agarwal and Bucheli, 2011; Wang et al., 2017). In this study, BC was weakly-moderately related with LMW-PAHs, but lack with HMW-PAHs (Table S5). The lack of correlation between BC and HMW-PAHs could be due to either, i) BC determination is less accurate than PAHs determination, ii) the fact that CTO-375 method doesn't detect all types of BC (Hammes et al., 2007), iii) the low BC/TOC might inhibit sorption to BC because of natural attenuation (Cornelissen et al., 2005). This finding is contrasting to previous study reported in Delhi which showed significant correlation of BC in soil with individual



Fig. 3. Spatial map showing distribution of HMW- and LMW-PAHs in soil from Nepal. The concentrations of HMW-PAHs were more prevalent than LMW-PAHs.

PAH, except NAP (Agarwal and Bucheli, 2011). However, slightly different than this, BC in Swiss soil showed significant correlation with LMW-PAHs only (Brandli et al., 2008). Hence, our finding is more or less consistent with data presented by Brandli et al. (2008). PAHs in the atmosphere get associated with BC after their release from source materials. Consequently, the LMW-PAHs get partition more rapidly from BC to vapor phase, while HMW-PAHs tend to remain sorbed to particulate phase (Bucheli et al., 2004). After deposition to soil, the vapor phase LMW-PAHs is free to partition and equilibrate more readily with TOC than HMW-PAHs. This indicates diffusive source of PAHs pollutions (Bucheli et al., 2004). In this study, the HMW-PAHs such as BbF (r = -0.614, p < 0.05) and BkF(r = -0.531, p < 0.05) was significantly and negatively correlated with BC in dust. This finding is in contrast to the earlier study reported in Swedish soil, where BC was not correlated with HMW-PAHs (Brandli et al., 2008).

Additionally, Spearman's rank correlation analysis was also applied on individual PAH data to investigate the interrelationship. Strong and positive relation was observed among individual PAH both in soil and dusts (Table S5 and S6). Comparatively, HMW-PAHs were more strongly and positively inter-correlated than LMW-PAHs, suggesting dissimilar source. Additionally, strongly significant correlation among FLUA, BaA, BkF, BaP, PYR, ANT, and PHE (r = 0.9, p < 0.05) suggests their similar sources of origin.

#### 3.5. Source apportionment study

Source categorization of environmental pollutants plays a significant role in the assessment of health risk and environmental management, particularly in case of complex environment, for instance, urban areas where many point source and diffuse source are predominant rather than solitary sources. Therefore, multivariate approach or simple compound profile and isomeric ratios are regularly utilized for source characterization study (Cachada et al., 2009). In this study, isomeric ratio (also known as diagnostic ratio; DR) and principal component analysis (PCA) were applied to investigate the possible source of PAHs in soil and dust and are discussed below.

#### 3.5.1. Diagnostic ratio (DR)

Knowledge about the sources of PAHs in the multi-environmental matrices is an important step for comprehending the fate and transport process. DR of different PAHs compounds are used to differentiate their similar or dissimilar source (Devi et al., 2016). For instance, the DR of ANT/(ANT + PHE), BaA/(BaA + CHR), FLUA/(FLUA + PYR) and IcdP/ (IcdP + BghiP) are used to distinguish petrogenic source from and pyrolytic one (Jamhari et al., 2014). The DR ratio of ANT/(ANT + PHE) < 0.1 indicates petroleum emissions, whereas DR ratio > 0.1 reflects biomass combustion (Kanzari et al., 2014). Likewise, FLUA/(FLUA + PYR) ratio smaller than 0.4 specifies petroleum emissions, between 0.4 and 0.5 suggests combustion of natural gas, while DR ratio > 0.5 is representative of biomass and coal combustions (Saha et al., 2009). DR ratio of IcdP/(IcdP + BghiP) and BaA/(BaA + CHR) smaller than 0.2 are indicators of petrogenic emission, while DR ratio of BaA/(BaA + CHR) between 0.2 and 0.35 and IcdP/(IcdP + BghiP) ratio between 0.2 and 0.5, point toward mixed sources from fossil fuel combustion, crude oil, and vehicular emissions. DR ratio of IcdP/(IcdP + BghiP) and BaA/ (BaA + CHR) > 0.5, suggests contributions from biomass and coal combustion (Hien et al., 2007).

In this study, the DR cross plots of PAHs in soil are illustrated in Fig. 5. Considering the ratio of IcdP/(IcdP + BghiP), the DR value ranged between 0.2 and 0.5 except two sites in Kathmandu, strongly indicating input of PAHs derived from fossil fuel combustions (Yunker et al., 2002; Devi et al., 2016). The cross plots of IcdP/(IcdP + BghiP) and BaA/(BaA + CHR) represent the mixed sources of PAHs mostly from petrogenic emissions (Yin et al., 2008). For ANT/(ANT + PHE), the DR ratios at all study sites were >0.1, indicating dominance of fossil fuel combustion and biomass combustion. The cross plots between FLUA/ (FLUA + PYR) and ANT/(ANT + PHE) suggest combustion of biomass as the major sources of PAHs in soil. BaP/BghiP ratio ranged from 0.89–4.90, well above 0.6, strongly indicating PAHs contribution mainly from traffic emissions (Katsoyiannis et al., 2007) (see Fig. 5).

The estimated DR ratios of different PAHs in dusts also showed similar sources of PAHs emission except BaA/(BaA + CHR) (Fig. 6). The BaA/ (BaA + CHR) ratio in dust ranged from 0.14–0.69, which strongly indicates combustion of biomass, wood and coal. This finding is true in case of Nepal, as large amounts of solid biomass fuel such as wood, cow dung flakes, crop residues, coal lignite and charcoal are utilized as primary fuel for cooking and heating in Nepalese houses (Pokhrel et al., 2015). These biomass fuels are inexpensive and are more commonly available, making them attractive alternatives against cooking gas (Bonjour et al., 2013; Pokhrel et al., 2015). This finding is consistent with previous studies by Qi et al. (2014) and Bhargava et al. (2004), who also reported



Fig. 4. Spatial map showing distribution of HMW- and LMW-PAHs in dust from Nepal. The concentrations of HMW-PAHs were more prevalent than LMW-PAHs.



Fig. 5. Cross plots of different diagnostic ratio used for source identification of PAH in soil. The concentrations of HMW-PAHs were more prevalent than LMW-PAHs.

traffic emissions, cooking method and biomass burning as the major source of PAH in house dust in Asia.

#### 3.5.2. PCA analysis

Principal component analysis (PCA) is a well-known effective tool for studying the distinct source of environmental pollutants (Liu et al., 2010; Devi et al., 2016). PCA analysis was performed on whole soil PAHs data set. Loading value >0.50, was taken into account for interpreting the contribution of PAHs isomer. Five major PCs were extracted with Eigen value > 1, and contained 85.98% of the total variance (Table S7). PC 1 accounted for 44.55% of the total variance, and was highly loaded with HMW-PAHs (FLUA, PYR, BaA, BbF, BkF, BaP, DahA, IcdP and BghiP). BghiP, IcdP, BaP, and DahA are good indicator of vehicular emissions (Motelay-Massei et al., 2003; Liu et al., 2010; Zhang et al., 2012). Less volatile PAHs, for instance, IcdP, BghiP, DahA are mainly released from diesel exhausts (Callen et al., 2012; Dumanoglu et al., 2017). BaA, BbF and BkF are representative of fossil fuel combustions (Harrison et al., 1996). Previously, vehicular emission was identified as dominant source of PAHs in urban soil near Pearl River Delta (Bi et al., 2003). FLUA, BaP and PYR are taken as tracer of coal combustions (Zhang et al., 2012; Yuan et al., 2014). Hence, PC 1 is referred as mainly originated from vehicular emissions and industrial combustions of fossil fuels and coals. PC 2 explained 19.77% of the total variance with high loading on LMW-PAHs (FLU, PHE, ANT and NAP) and negative loading on CHR. PHE, ANT and CHR are fingerprints of wood combustions (Khalili et al., 1995), while CHR is marker for combustion of coke oven and biomass (Wang et al., 2013; Yang et al., 2013). NAP results from wood burning and biomass burning (Zhang et al., 2008). Therefore, this factor represents emission from residential combustion of biomass/wood. PC 3 represented 8.41% of the total variance, and was highly loaded with ACE (0.609), TOC (0.613) and BC (0.510). ACE is good predictor of coal combustions (Wang et al., 2009; Lin et al., 2013). Roughly, 8% variation in soil PAHs data was due to TOC and BC that suggest other than soil properties factor such as gas absorption and deposition of locally emitted PAHs significantly influence soil PAH concentration (Cetin et al., 2007). Therefore, PC 3 is identified as emission from coal combustion and influence of soil organic carbon. PC 4 accounted 6.99% of the total variance but didn't contain any significant loading. PC 5 explored 6.24% of the total variance and was loaded with ACE (0.811), which is indicator of coal combustions.

Multivariate PCA analysis was also applied to dust PAHs dataset to distinct the possible sources in indoor environment, and are presented in Table S8. For house dust, four major PC were extracted with Eigen value >1 following varimax rotation. PC 1 accounted 51.2% of total variance and was highly loaded with LMW-PAHs (NAP, ACY, ACE, FLU, PHE, and ANT) and HMW-PAHs (FLUA, BaA, BbF, BkF, DahA, IcdP and PYR). BaA, BbF, BkF, IcdP and DahA are indicators of fossil fuel combustions (Harrison et al., 1996; Yang et al., 1998). NAP, ACY, FLU, PHE and ANT represent emissions from petrogenic source (Fang et al., 2004; Qi et al., 2014) together with incomplete combustions of biomass or wood burning (Zhang et al., 2008). Therefore, PC1 is identified as emission from fossil fuel combustions and biomass burning. PC 2 contained 12.8% of the total variance, and was highly loaded with IcdP, BghiP and DahA. High loadings on IcdP, BghiP and DahA are good indicators of vehicular emissions (Fang et al., 2004; Jiang et al., 2009). Hence, this factor is taken as petrogenic source. PC 3 represented 8.31% of the



Fig. 6. Cross plots of different diagnostic ratio used for source identification of PAH in dust. The concentrations of HMW-PAHs were more prevalent than LMW-PAHs.

total variance and had moderate loading on IcdP and ACY. IcdP and ACY are good contributor of petrogenic emission (Jiang et al., 2009). Hence, PC 3 is also recognized as petrogenic origin. PC 4 explored 7.16% of the total variance and was moderately loaded with TOC (0.725) and BC (0.579).

#### 3.6. Estimation of toxicological effects of PAHs

BaP is well known carcinogen which is used to evaluate the carcinogenic potency of individual PAH based on toxic equivalence factor (TEF). TEF is considered as the carcinogenic strength of individual PAH with respect to BaP (Tsai et al., 2004; Lin et al., 2008; Zhu et al., 2014). The toxicity equivalency quantities (TEQs) of PAHs are estimated by multiplying the concentration of individual PAHs with their corresponding TEF values. TEF value of individual PAH used in this study was acquired from Tsai et al. (2004). The average TEQs for PAH in soil was estimated as  $294 \pm 121$  ng/g. As per Canadian soil quality guideline, the recommended TEQs limit in soil for the protection of Environment and health is 700 ng/g (CCME, 2010; Liu et al., 2010). All soil samples in this study irrespective of city showed TEQs value much below safe limit, suggesting little risk to Nepalese population due to soil PAH pollution. Moreover, the estimated TEQ value in soil in this study was 2-12 times higher than those reported in urban soil from Viseu in Portugal (24 ng/g) (Cachada et al., 2012), Kocaeli in Turkey  $(90 \pm 131 \text{ ng/g})$ (Cetin, 2016), Terragona in Spain (124 ng/g) (Nadal et al., 2004), and Beijing in China (181 ng/g) (Liu et al., 2010), but were comparable to those reported in Shanghai in China (236 ng/g) (Wang et al., 2013) and Lisbon in Portugal (229 ng/g)(Cachada et al., 2012). However, the estimated TEQs for soil was 2-3 times lower than previously reported in urban soil from Shanghai (892 ng/g) (Jiang et al., 2009) and Nanjing (445 ng/g) (Wang et al., 2015) in China and Agra in India (650 ng/g) (Masih and Taneja, 2006). The estimated TEQs for 16 different PAHs in soil were: NAP (0.117 ng/g), ACY (0.033 ng/g), ACE (0.037), FLU (0.050 ng/g), PHE (0.244 ng/g), ANT (1.52 ng/g), FLUA(0.126 ng/g), PYR(0.105 ng/g), BaA(9.91 ng/g), CHR(10.60 ng/g), BbF (8.38 ng/g), BkF (11.0 ng/g), BaP (129 ng/g), IcdP (6.47 ng/g), DahA(117 ng/g) and BghiP (0.085). BaP is well-known for its high cancer potency because of high carcinogenic contributor among different PAH followed by DahA and BbF (Pufulete et al., 2004). In this study, the TEQs for BaP were also estimated maximum (129 ng/g) suggesting high risk of soil carcinogenicity. Equally, other PAHs such as DahA (117 ng/g) and BkF (11 ng/g), and CHR (10.6 ng/g) also indicated higher TEQs value. Because of non-availability TEQs limit for PAHs in Nepalese soil, we compared our result of TEQs with Dutch guideline value (32.96 ng/g) (Crnković et al., 2007; Wang and Lang, 2010). When compared with Dutch limit, the estimated average TEQs (294 ng/g) was 8-9 times higher than Dutch target value of 32.9 ng/g. The TEQs of BaP (129 ng/g) was 3–4 times greater than Dutch target value.

For dust, the average TEQs for PAHs was estimated as  $243 \pm 126$  ng/g. This TEQs value is 1.5–2 times higher than those reported in house dust in Kuwait (162.2 ng/g) (Gevao et al., 2007), office floor dust (155 ng/g) and lecture room (151–165 ng/g) from Jordan (Maragkidou et al., 2017). However, the estimated TEQs in this study was 2 times lower than those reported in corridor dust in Jordon

(483 ng/g), but comparable with big corridor dust in Jordan (220 ng/g) (Maragkidou et al., 2017). The individual TEQ for 16 PAHs was NAP (0.11 ng/g), ACY (0.03 ng/g), ACE (0.03), FLU (0.05 ng/g), PHE (0.18 ng/g), ANT (0.67 ng/g), FLUA(0.16 ng/g), PYR(0.14 ng/g), BaA(11.6 ng/g), CHR(1.71 ng/g), BbF (7.47 ng/g), BkF (11.6 ng/g), BaP (136 ng/g), IcdP (62.0 ng/g), DahA(11.2 ng/g) and BghiP (0.10).

#### 4. Conclusions

In this study, concentration, profile, spatial distribution, sources and environmental fate of 16 PAHs was investigated in soil and dust matrices of Nepal. Overall the concentrations of  $\sum_{16}$  PAHs were about 1.5 times higher in soil than dust. The carcinogenic PAHs were also measured high both in soil and dust, and accounted for >50% of total PAHs. Generally, HMW-PAHs were more abundant than LMW-PAHs both in soil and dust, suggesting the dominance of pyrogenic source. In terms of compositional pattern, CHR was the most abundant chemical in in soil, while PHE exceeded in dust samples. The 4-ring PAH was identified as the most abundant PAH among different ring-PAHs, which is consistent with global studies. Relatively, Birgunj and Kathmandu showed higher level of  $\sum_{16}$  PAHs both in soil and dust than those in Pokhara and Biratnagar, and were related with large number of registered vehicles in the cities. The concentration of  $\sum_{16}$  PAHs in this study were comparable to PAH concentrations reported in some megacities around the world. TOC and BC were moderately to weakly related with PAHs both in soil and dust indicating little influence of soil organic carbon in sorption of PAHs. Source Diagnostic ratio and principal component analysis confirmed combustion of biomass, fossil fuel combustion and traffic/vehicular emissions as the principal sources of PAH in Nepalese urban environment. The high average TEQ value estimated for PAHs in soil than dust suggested relatively higher risk for soil carcinogenicity compared to dust carcinogenicity.

#### **Conflict of interest**

The authors declare no competing financial interest.

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#### Appendix A. Supplementary data

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