ABSTRACT  This study was conducted in the Central Himalayan middle hills to understand the nature of polycyclic aromatic hydrocarbons (PAHs) embedded in aerosol particles, their sources and human health risk assessments. The level of sum of 15 particle-phase PAHs was between 9 and 335 ng/m³, with an average concentration of 73±66 ng/m³. There were strong seasonal differences in total suspended particles (TSP) and particle-bound PAH concentrations with higher concentrations in winter, followed by premonsoon and lowest in monsoon. The main contributor to the suspended particles was 5-ring PAHs (32%), followed by 4-ring (29%), 6-ring (28%), and 3-ring PAHs (11%). Conversely, the gas-phase PAHs showed that 3-ring PAHs contributed utmost to the total particles. The molecular ratios and principal component analysis indicated that both petrogenic and pyrogenic sources, particularly fossil fuel combustion, biomass combustion, and car exhausts, were the major sources of PAHs. The overall average Benzo(a)pyrene equivalent concentration of particulate PAHs was 11.71 ng/m³, which substantially exceeded the WHO guideline (1 ng/m³), and indicated the potential health risks for local residents. The average lifetime inhalation cancer risk (ILCR) estimates associated with carcinogenic PAHs was 8.78×10⁻⁶ for adults, suggesting the possible cancer risk and 2.47×10⁻⁵ for children, signifying extreme carcinogenic effects of PAHs on children’s health. Therefore, strict measures should be taken to reduce PAHs emissions in the region.

KEY WORDS  Aerosol, Polycyclic aromatic hydrocarbons, Long-range transport, Himalayas, Nepal

1. INTRODUCTION

Polycyclic aromatic hydrocarbons (PAHs) are a class of organic contaminants with two or more fused aromatic rings that originate from incomplete burning of organic matter (Rajput et al., 2014; Singh et al., 2013; Zhang and Tao, 2009). They are considered to have remarkable mutagenic, carcinogenic, and teratogenic properties due to the presence of aromatic rings (Suvarapu et al., 2012; IARC, 2010). Moreover, PAHs are unaffected by biodegradation, and due to their hydrophobic
characteristics and low solubility, they can bio-accumulate in the ecosystems (Zhu et al., 2008; Conde et al., 2005). The origins of PAHs in the environment include anthropogenic and natural sources. Volcanoes, forest fires, earth’s thermal and geologic permutations include natural sources (Juhasz and Naidu, 2000), while human activities include the incomplete combustion of fossil fuels (petroleum, oil, coal, gas), domestic and commercial heating with biomass (wood and others), industrial exhausts, vehicular traffic emissions (motor vehicles exhaust), waste incineration, and tobacco smoke (Majrjan et al., 2022; Kamal et al., 2015; Chang et al., 2006). PAHs and their metabolites are of great concern because of their presence by widespread sources and persistence in air, water, and soil and, importantly, their toxicity and harm to human, animal, and plant health (Kim et al., 2013; Ravindra et al., 2008). On the basis of their carcinogenic potency, the United States Environmental Protection Agency (USEPA) has identified 16 priority PAHs: including Naphthalene (NaP), Acenaphthylene (AcPy), Acenaphthene (AcP), Fluorene (Flu), Phenanthrene (Phe), Anthracene (AnT), Fluoranthene (FluA), Pyrene (Pyr), Benzo(a)anthracene (BaA), Chrysene (Chr), Benzo(b)fluoranthene (BbF), Benzo(k)fluoranthene (BkF), Benzo(a)pyrene (BaP), Indeno(1,2,3-cd)pyrene (IndP), Dibenzo(a,h)anthracene (DbA), and Benzo(g,h,i)perylene (BghiP).

Several studies have been conducted on aerosol chemical characteristics (e.g., soluble ions, carbonaceous species, and particulate mercury) over the Indo-Gangetic Plains (IGP) and Himalayan areas (Guo et al., 2022, 2020, 2017; Tripathee et al., 2021, 2017, 2016; Chen et al., 2020, 2019). However, limited studies exist on the chemical composition of aerosol particles over the middle hills of the Himalayas, where there are regional and local impacts on aerosol chemistry. Besides, there have been sparse studies reported on the concentrations, deleterious nature of airborne PAHs and their adverse impacts on human health in the middle hills of the Central Himalayas, though much research has pointed out that the urban aerosols from polluted surrounding areas have posed a potential threat to lives and ecosystems (Chen et al., 2017, 2015). Meanwhile, PAH concentrations in soils were presented in a central Himalayan region (Bi et al., 2016), which suggested that the big cities had the highest PAH levels, while the high Himalayas had the lowest (Bi et al., 2016).

The PAHs of lower molecular weight are volatile and found chiefly in the gas phase, while the higher molecular weight PAHs are usually found to be attached to particulate matter (Yang et al., 2018; Lee et al., 2001). These compounds exhibit a wide range of gas-particle partitioning characteristics in the air, which plays a fundamental role in controlling their fate, transport, variation, and removal mechanisms through dry and wet deposition courses (Esen et al., 2019; Harner and Bidleman, 1998). Various factors, such as ambient temperature, relative humidity, liquid vapor pressure, the composition of chemicals, and the nature of the particles affect the partitioning behavior of PAHs (Gaga et al., 2012). Understanding the gas/particle-phase distribution of PAHs in the atmosphere is important for environmental as well as health risk evaluation, as some of the species are considered to be carcinogenic even at lower concentrations. Nevertheless, we are still unaware of this partitioning distribution of PAHs over the study region. Therefore, it is important to well understand the composition, characteristics, origins, and transport of atmospheric contaminants (e.g., PAHs) and their impacts on human health and the ecosystems in this region. Moreover, this study is also important as the contaminants (e.g., PAHs, mercury, black carbon etc.) could be easily transported to the glaciers and pristine ecosystems of the Himalayan Tibetan Plateau (HTP) as evidenced from prior studies (Chen et al., 2019; Kang et al., 2019; Tripathee et al., 2019). The HTP glaciers act as the archive of contaminants deposited through wet and dry deposition from the heavily polluted South Asian regions. The glacier melting from the HTP releases such deposited pollutants to glacier rivers on which a large population downstream relies for fresh water, thereafter impacting the health of the residents and the whole ecosystems (Zhang et al., 2017). Therefore, understanding is important of the fate and characteristics of atmospheric contaminants like PAHs over the South Asian regions, especially in the mid-hills of the Himalayas with limited local sources.

Under the Atmospheric Pollution and Cryospheric Changes (APCC) network (Kang et al., 2019), this research presents the results of 15 priority PAHs in the particulate phase from TSP samples in Dhulikhel, Nepal, collected from the period of January to July 2018. This study is intended to determine the concentrations, seasonality and to recognize the possible sources of PAHs in atmospheric particulate samples from characteristic molecular ratios and principal component analysis (PCA). Furthermore, the study aimed to evaluate the carcinogenic health risks from the atmospheric PAHs by inhalation in the study site. The results of this study are useful for the con-
control and management of atmospheric PAHs in the region.

2. MATERIALS AND METHODS

2.1 Study Area and Collection of Samples

The sampling was performed within the premises of Kathmandu University, located near Dhulikhel, a town in the central Himalayan middle hills, Nepal (Fig. 1). The sampling location (27.601°N, 85.538°E, 1,510 m) lies about 25 km southeast which is upwind of the major leading city of Kathmandu Valley, Nepal. The capital city, Kathmandu, is a heavily polluted urban location, with

Fig. 1. Location of the sampling site (Dhulikhel) in Nepal. (a) Sampling site, (b) Land use map showing the sampling location with black triangle, (c) Wind rose diagram during the sampling period.
nearly more than 2 million inhabitants. Banepa is another major town, which is located about 2.5 km northwest of the study area. Previous studies have ascribed that Dhulikhel is impacted by the extensive pollutant transport from the highly polluted IGP region as well as local sources from nearby urban cities (Tripathee et al., 2021; Shrestha et al., 2010). Temperature and relative humidity at the study location during the study phase is displayed in Fig. S1. The temperature was noted to be warm and humid during summer and cold and dry during the winter.

Aerosol sampling was carried out from January to July 2018. The total suspended particulate (TSP) sampler was established on the rooftop of the building (15 m above ground) of Kathmandu University bounded by agricultural fields, residential housing, and forests with limited local emissions. Overall, 48 TSP samples were collected with the Qingdao Laoying TSP Cyclone (T2034) at 100 L/min of flow rate for 24 hours (Tripathee et al., 2021; Chen et al., 2019). Briefly, the sample collection was done on pre-combusted quartz fiber filters (diameter 90 mm, Whatman, Maidstone, UK). The quartz fiber filters were pre-combusted (550°C, 6 h) to lessen any residual organic matter. During the pre-monsoon season, 2 to 3 samples were taken each week to capture the heavy pollution loadings during dry periods, and during other periods, one sample per week was taken. The collected samples were relocated to a clean disc and kept in a freezer prior to the laboratory analysis. The weight of all quartz filters was evaluated gravimetrically before and after the sampling to obtain the aerosol mass concentrations.

2.2 PAHs Extraction and Analysis

This study followed the procedure for extracting and analyzing PAHs according to the prior research (Maharjan et al., 2022; Chen et al., 2015). For the pre-treatment of PAHs, 5.08 cm² of each filter was cut and sited in a sample bottle and was submerged in a mixed solution (20 mL) of dichloromethane (DCM) and n-hexane (1:1). Sonication of the mixtures ultrasonically thrice each for 30 minutes was done. Spiking of each sample was done with deuterated PAH, Chrysene-d12 as recovery surrogates, and was left for about 30 minutes. With a rotary evaporator, the mixtures obtained were then concentrated to about 0.5 mL and were moved into a multi-layered column containing active silica gel, neutral alumina, and Na₂SO₄ (anhydrous). Then, n-hexane (10 mL) and mixed solution (1:1) of DCM/n-hexane (20 mL) was poured to elute the column. After that, the solvent was blown underneath the nitrogen gas stream to get 1 mL ultimate volume. The final solution was then moved to a sample vial to be measured. Samples were examined for the 15 priority PAHs (USEPA classified) by using Gas chromatography-mass spectrometry (GC-MS). Further details on full methods and quality control and assurance have been presented in the Supplementary text (Text S1).

2.3 Estimation of PAHs Gas/Particle Partitioning

The PAHs partitioning among the gas and the particle phase can be approximated from its octanol-air partition coefficient (K_{OA}). The K_{OA} values for PAHs can be determined from the method of gas chromatographic retention time as given in equation 1 (Harner and Bidleman, 1996):

\[ \log K_{OA} = A + \left(\frac{B}{T}\right) \]  

where the parameters of regression, dependent on temperature, are given as A and B (Odabasi et al., 2006; Harner and Bidleman, 1998). Table S1 lists different K_{OA} of PAHs in the sampled periods as estimated by modifying the equation to the ambient temperature (average) in the study site (winter: 10°C, pre-monsoon: 18°C, and monsoon: 23°C).

K_p, the gas-particle partition coefficient is expected using K_{OA} in equation 2, assuming that the principal distribution course is absorption (Ayyildiz and Esen, 2020; Wang et al., 2013; Harner and Bidleman, 1998):

\[ \log K_p = \log K_{OA} + \log f_{OM} - 11.91 \]  

where the fraction of organic matter on TSP is given by f_{OM}. The average organic carbon (OC) concentrations in aerosols in this study were 25.92, 21.62, and 7.65 μg/m³ for winter, pre-monsoon, and monsoon seasons, respectively. OM was calculated based on OC values using the equations in the previous study in Himalayas and Tibetan Plateau (Chen et al., 2018, 2015):

\[ OM = OC \times 1.6 \]

\[ f_{OM} = OM / TSP \]

\[ f_{OM} = (OC \times 1.6) / TSP \]

The transformation of OC values to organic matter (OM) contributed about 12.42%, 12.41%, and 8.54% to the total suspended particles in the respective sampled seasons. Using the K_p value (m³ μg⁻¹), atmospheric PAHs distribution between the particle and gas phases is
given as follows:

\[ K_p = \left( \frac{C_p}{C_{\text{TSP}}} \right) / C_g \]  

(3)

where and \( C_g \) and \( C_p \) (ng/m\(^3\)) are the concentration of PAHs in the gaseous and particle phases, respectively, and \( C_{\text{TSP}} \) (μg/m\(^3\)) is the total aerosol particles concentration in air (Verma et al., 2017; Harner and Bidleman, 1998).

2.4 Assessment of Health Risk

Human health risk/threat assessment can be done using human exposure to PAHs occurring through the routes of inhalation, ingestion, or dermal contact (USEPA, 1989). The inhalation of particulate PAHs in the air was taken into account for the carcinogenic risk assessment in this study. For estimating the health risks related to exposure of PAHs, the Benzo(a)pyrene equivalent concentration (BaPeq) is used and it is based on carcinogenic potencies of individual PAHs relative to BaP (Chen et al., 2016; Nisbet and Lagoy, 1992). The toxicity equivalency factor (TEF) of each PAH relative to BaP carcinogenic strength multiplied by its corresponding concentration determines the BaPeq of each PAH (ng m\(^{-3}\)) as given by the equation below:

\[ \text{Total BaPeq} = \sum_i (C_i \times \text{TEF}_i) \]  

(4)

The lifetime average daily dose (LADD) of PAHs is the intake quantity per kilogram of body weight per day of a pollutant which has the potential of causing adverse health impacts once the body absorbs it over a long time span (mg/kg/day). The incremental lifetime cancer risk (ILCR) is determined using the LADD of PAHs. The details on the estimation of LADD are given in the Supplementary text (Text S2). The equation for estimating ILCR (Bortey-Sam et al., 2015; Jamhari et al., 2014) is shown as follows:

\[ \text{ILCR} = \text{LADD} \times \text{CSF} \]  

(5)

where CSF denotes the cancer slope factor and its value of 3.14 (mg\(^{-1}\) kg day) is taken in this study (Chen and Liao, 2006). The standard values were acquired from USEPA (2011).

Also, the Lifetime Lung Cancer Risk (LLCR) through inhalation exposure to PAHs is estimated by the equation given below as used by Chen et al. (2017):

\[ \text{LLCR} = \sum \text{BaPeq} \times \text{UR}_{\text{BaP}} \]  

(6)

where \( \text{UR}_{\text{BaP}} \) denotes the cancer unit risk by BaP inhalation and the value is \( 8.7 \times 10^{-5} \) as specified by the World Health Organization (WHO).

3. RESULTS AND DISCUSSION

3.1 Overview and Seasonal Characteristics of TSP and PAHs

The overview of the mean \( \sum \) PAHs concentrations and ambient TSP in the study area, Dhulikhel, is presented in Table 1. The comparison of average concentrations of PAHs at Dhulikhel with other regions worldwide is shown in Fig. 2. The sum of PAHs and aerosol mass concentrations during the study period ranged from 9 to 335 ng/m\(^3\) and 62 to 442 μg/m\(^3\), respectively. The average concentration of total PAHs obtained in Dhulikhel (73 ± 66 ng/m\(^3\)) was similar to those reported in other places of Asia, such as Eerduosi, China (Wu et al., 2014), Nanjing, China (Wang et al., 2006), rural Jamshe dpur, India (Kumar et al., 2020), and Kolkata, India (Ray et al., 2017) but lower than the ones reported in Kathmandu, Nepal, a heavily polluted urban city (Chen et al., 2015), and a typical rural site in Nepal, Lumbini (Chen et al., 2016). The values were also lower than the reported levels in other cities of India such as Delhi, (Sarkar and Khillare, 2012), Amritsar, (Kaur et al., 2013), Agra (Rajput and Lakhani, 2012), and in China, Beijing, (Wang et al., 2008). The

| S.N | PAHs | Ring | Mean | SD | Minimum | Maximum |
|-----|------|------|------|----|---------|---------|
| 1   | AcPy | 3    | 0.38 | 0.33 | 0.00    | 0.92    |
| 2   | AcP  | 3    | 0.81 | 0.44 | 0.19    | 1.52    |
| 3   | Flu  | 3    | 2.29 | 0.56 | 1.24    | 4.04    |
| 4   | Phe  | 3    | 3.67 | 0.83 | 2.25    | 6.21    |
| 5   | AnT  | 3    | 0.57 | 0.34 | 0.18    | 1.14    |
| 6   | FluA | 4    | 5.30 | 4.35 | 1.08    | 17.93   |
| 7   | Pyr  | 4    | 5.91 | 5.59 | 0.77    | 24.10   |
| 8   | BaA  | 4    | 4.30 | 3.93 | 0.03    | 14.85   |
| 9   | Chr  | 4    | 5.96 | 5.68 | 0.61    | 23.83   |
| 10  | BbF  | 5    | 9.27 | 7.49 | 0.18    | 34.87   |
| 11  | BkF  | 5    | 7.63 | 7.14 | 0.02    | 27.07   |
| 12  | BaP  | 5    | 7.82 | 7.83 | 0.59    | 37.14   |
| 13  | IndP | 6    | 9.49 | 10.15| 0.88    | 56.19   |
| 14  | DbA  | 5    | 0.82 | 0.49 | 0.29    | 2.47    |
| 15  | BghiP| 6    | 10.84| 13.30| 0.34    | 82.75   |

| S.N | TSP  |
|-----|------|
| 1   | 263.58 |
| 2   | 108.04 |
| 3   | 61.54  |
| 4   | 442.45 |

Table 1. Summary of total concentration of particle-bound PAHs (ng/m\(^3\)) and TSP (μg/m\(^3\)) in the study site.
average TSP mass concentration during the study period was \((264 \pm 108 \mu g/m^3)\). TSP mass exhibited a reasonable correlation with PAH concentrations \((r = 0.46; p < 0.05)\) in the study site as presented in Fig. S2, suggesting that PAH concentrations were dependent on aerosol particles. Previous studies on the chemical composition of ambient aerosols have suggested that anthropogenic emissions considerably influence the atmospheric composition in Dhulikhel via short and long-range pollutants transported from the Kathmandu valley and the IGP region (Bhattarai et al., 2022; Tripathee et al., 2021; Gautam et al., 2011; Shrestha et al., 2010). Such transported polluted air mass can have a significant impact on the atmospheric chemistry of the middle hill sites (Guo et al., 2021).

Clear seasonality of the TSP and total PAHs concentrations was witnessed in the present study (Fig. 3). The higher concentrations of PAHs was found in the winter ranging from 42–170 with an average of 110 ng/m³ followed by pre-monsoon ranging from 13–311 with an average of 72 ng/m³ and lower concentration levels in the monsoon season ranging from 16–135 with an average of 42 ng/m³. In addition, the KOA-based approach was used to calculate the concentration of PAHs in the gas phase, which also exhibited clear variations seasonally with higher concentration levels in monsoon, and reduced to minimum concentrations in winter (Table 2). Generally, PAHs with two or three rings, having relatively lower log \(K_{OA}\) values, are mostly bound to the gaseous phase, PAHs with four rings are found in both the particle and gas phase, and PAHs with higher rings (five or more), with higher log \(K_{OA}\) values, are present in the particulate phase. However, the temperature rise possibly increases the gas partitioning of PAHs with four or more rings. The PAH species with lower molecular weight, such as Flu, AcP, AcPy and Phe were the predominant compounds in the gas phase, whereas the higher molecular weight ones such as BghiP, IndP, BaP, BaF, Chr, BaA were found to be attached to particles and at higher concentrations than those in the gas phase. In addition, seasonal comparisons of PAH were performed by the Mann-Whitney Rank Sum Test using U-statistics due to the skewed distribution of the data. Statistically, PAH concentrations were significantly higher during the winter \((U = 9, n1 = 9,\)
The composition of PAHs is important for determining their potential sources of emission (Li et al., 2017). 15 PAHs are classified into 4 groups on the aromatic ring number basis: including 3-ring PAHs (AcPy, AcP, Flu, PhA, and AnT), 4-ring PAHs (FluA, Pyr, BaA, and Chr), 5-ring PAHs (BbF, BkF, BaP, and DbA), and 6-ring PAHs (IndP and BghiP) (Kaur et al., 2013). Lower molecular weight, LMW (3-rings) and middle molecular weight, MMW (4-rings) are more volatile than PAHs with higher molecular weight, HMW (5, 6-rings) and are partitioned between gaseous phase and particulate phase, which can be transported far and wide, even to the remote atmosphere. Generally, due to their low vapor pressures, most of the HMW PAHs are existent in the particulate phase.

### Table 2. Calculated gas-phase PAH concentrations (Cg) in the sampled seasons in the study site.

| S.N | PAHs | Winter | Pre-monsoon | Monsoon |
|-----|------|--------|-------------|---------|
| 1   | AcPy | 2507.246 | 2270.5 | 3694.516 |
| 2   | AcP  | 2616.039 | 3264.183 | 11175.449 |
| 3   | Flu  | 1521.261 | 3864.065 | 22920.169 |
| 4   | Phe  | 452.748  | 919.286  | 4745.825  |
| 5   | AnT  | 97.814   | 125.471  | 355.263   |
| 6   | FluA | 76.207   | 92.532   | 189.353   |
| 7   | Pyr  | 71.074   | 93.617   | 159.543   |
| 8   | BaA  | 1.395    | 1.901    | 3.190     |
| 9   | Chr  | 1.774    | 2.529    | 5.728     |
| 10  | BbF  | 0.120    | 0.404    | 1.380     |
| 11  | BkF  | 0.059    | 0.240    | 0.844     |
| 12  | BaP  | 0.079    | 0.182    | 0.549     |
| 13  | IndP | 0.008    | 0.030    | 0.117     |
| 14  | DbA  | 0.001    | 0.001    | 0.007     |
| 15  | BghiP| 0.007    | 0.027    | 0.091     |
|     | Total | 7345.833 | 10634.968 | 42622.025 |

n2 = 9, p < 0.05) and pre-monsoon (U = 67, n1 = 9, n2 = 30, p < 0.05) compared to the monsoon season. In addition, winter-time PAH concentration was also significantly higher than that of the pre-monsoon season (U = 56, n1 = 9, n2 = 30, p < 0.05). It can be seen that the TSP mass concentration was higher to more than twice in winter than that in the monsoon period. Many previous studies have shown a similar seasonal trend of PAH levels (Kumar et al., 2020; Mehmood et al., 2020; Morakinyo et al., 2019).

In the winter and pre-monsoon periods, the TSP mass and PAH concentrations were higher, suggesting high aerosol loads during the cold and dry seasons due to high emissions and less precipitation. Nevertheless, such weather conditions that promote the accumulation of air pollutants and atmospheric boundary layer heights are lower with stable atmospheric conditions and less dispersion of pollutants (Ram and Sarin, 2010). Further, the reduced photo-degradation and reduced volatility during lower temperature periods play an essential role in enhancing atmospheric pollutants (e.g., PAHs) during this period (Kaushal et al., 2021; Elzein et al., 2020). During the monsoon, low concentrations of particulate loading could be attributed to fewer pollution sources (e.g., brick kilns are not active and less biomass burning activities). In addition, the washing out of emitted contaminants by high precipitation during monsoon also dilutes the concentrations as shown in Supplementary figure (Fig. S3). The site is under the influence of large-scale pollutant transport from the IGP region as well as local sources around the Kathmandu valley (Shrestha et al., 2010). Strong westerly winds during dry periods carry pollutants from the IGP region that can be transported over the Himalayan region. Intense biomass burning (BB) activities occur in the IGP region during dry periods causing intense haze over the region, which could have transported and deposited over the Himalayan region via long-range transport of atmospheric aerosols (Tripathee et al., 2021; Rajput et al., 2011; Ram and Sarin, 2010). The back trajectories of air-masses with satellite observations have been checked in a former study (Tripathee et al., 2021), in the sampling site over different seasons. The results revealed that during the dry period (winter, pre-monsoon), the air masses from IGP and surrounding polluted regions reached the study site Dhulikhel, through atmospheric transport and that higher aerosol loads in those regions during dry periods were particularly caused by BB and other anthropogenic emissions. Such transported pollutants could have given rise to the high loadings of aerosol and particulate bound PAHs in our site during those periods. To further understand the influence of BB on PAH concentrations, we checked the correlations with BB tracer (nss-K+) and shown in Supplementary figure (Fig. S4). The PAHs concentration was also positively correlated with nss-K+ in winter (r = 0.74; p < 0.05) and pre-monsoon (r = 0.57; p < 0.01), (Fig. S4), supporting our assumption that BB could be a potentially crucial source of particulate PAHs in the dry periods over the study site.
and are adsorbed into fine particles. It has been found that 4-ring PAHs are released through coal combustion and BB, whereas 5- and 6-ring PAHs are mostly discharged from fuel processes with burning in high temperatures such as motor vehicular exhausts (Maharjan et al., 2022; Chen et al., 2018).

The composition of PAHs in Dhulikhel, Nepal, was predominated by 5-ring PAHs (32%), followed by 4-ring (29%) and 6-ring PAHs (28%). The 3-ring LMW PAHs constituted only 11% of the total PAHs in Dhulikhel (Fig. 4). This indicates that particle-bound PAHs result from petroleum combustion, such as vehicular emissions, followed by emissions from coal combustion origin and BB. The dominance of HMW PAHs shows that the sources have resulted from high-temperature courses, such as engine fuel burning (Tobiszewski and Namieśnik, 2012). A higher contribution of 4-ring PAHs suggests that there is a dominant influence of emission from fossil fuels (motor vehicle exhausts and coal combustion) (Chen et al., 2015). The results are reasonable as our site is upwind of the polluted valley with numerous brick kiln emissions using low-grade coal and near to the Arniko highway with the frequent flow of heavy vehicles. Therefore, it indicates that most of the PAHs in our study site have originated from the local combustion of fossil fuel, including coal and BB, along with automobile exhausts.

### 3.3 PAHs Source Apportionment

To classify potential sources of atmospheric PAHs, characteristic molecular ratios and PCA approach were used.

#### 3.3.1 Diagnostic Ratio Analysis

Anthropogenic activities, such as pyrogenic sources from partial combustion of organic materials like biomass, fossil fuels and emissions from petroleum products are the primary sources of atmospheric PAHs (Boonyatumanond et al., 2006). Here, the PAH sources at Dhulikhel were recognized by comparing the ratios of selected PAHs. The ratios of Ant/(Ant + Phe), FluA/(FluA + Pyr), BaA/(BaA + Chr), IndP/(IndP + BghiP) and BaP/BghiP were calculated on a seasonal basis.

The Ant/(Ant + Phe) ratio value > 0.1 usually indicates the pyrogenic (combustion) origin, and < 0.1 indicates the petrogenic sources (Kamal et al., 2014; Kong et al., 2011; Budzinski et al., 1997). In this study, most of the values exceeded 0.1, implying input from the pyrogenic origin in all winter, pre-monsoon and monsoon seasons. FluA/(FluA + Pyr) ratios are indicative of petrogenic source input, liquid fossil fuel combustion, or BB (Fang et al., 2010; Mandalakis et al., 2002; Rogge et al., 1993). In the present study, most of the emissions were from petrogenic sources during the pre-monsoon and monsoon season, while emissions from liquid fossil fuel (petroleum) combustion, biomass, and coal-burning were prevalent in the winter season. Also, IndP/(IndP + BghiP) and BaA/(BaA + Chr) ratios are indicative of petrogenic and pyrogenic source input (Akyüz and Çabuk, 2010; Yunker et al., 2002). In the current study, the ratio values of IndP/(IndP + BghiP) indicated emission from petroleum combustion and burning of coal, grass, and wood burning. Further, the ratio value of BaA/(BaA + Chr) was mostly greater than 0.2, indicating the existence of mixed sources (petroleum, biomass, and coal combustion). The BaP/BghiP ratio is considered an indicator of traffic and non-traffic sources, the values of which indicated the influence of traffic sources too in all the sampled seasons. The cross-plot analyses of diagnostic ratios are presented in Fig. 5. The analysis implies that the emissions of PAHs in the study site are from petrogenic as well as pyrogenic sources, particularly from petroleum combustion, coal and biomass burning, and automobile exhausts.

#### 3.3.2 Principal Component Analysis (PCA)

The PCA was performed for investigation of the major sources of emissions of atmospheric PAHs in Dhulikhel, Nepal. A varimax rotation along with Kaiser Normalization was executed to identify the factors (Eigen value > 1). The results obtained are given in Table 3. Three prin-

![Fig. 4. Percentages of different ring PAHs in TSP in the study site.](image-url)
Principal components (PCs) were obtained, accounting for 91% of the total variance.

PC-1 described 52.95% of the total variance and was found with FluA, Pyr, BaA, Chr, BaP, BbF, BkF, BghiP and IndP. Among these, FluA and Pyr are principally emitted from incomplete burning of coal, pyrolysis of fuel, and oil burning (Ravindra et al., 2008; Harrison et al., 1996). BaA and Chr are emitted from diesel and natural gas combustion (Khalili et al., 1995). BaP, BbF, BkF, IndP and BghiP are reported as markers of vehicular emission by various studies (Fang et al., 2010; Chang et al., 2006; Park et al., 2002; Simcik et al., 1999; Harrison et al., 1996). Therefore, PC-1 represented the contribution of combustion of fossil fuel sources. PC-2 accounted for about 29% of total variability and loaded highly on LMW PAHs like AcPy, AcP, Phe and AnT, which could be identified as combustion origin (coal) (Wang et al., 2008; Mastral et al., 1996). PC-3 showed a variance of 9% and was loaded with Flu and Phe. Therefore, the PCA results showed that the study area was affected by diverse sources of fossil fuel combustion and vehicular emissions.

3.4 Health Risk Assessment

Carcinogenic human health risks from the atmospheric PAHs through inhalation of air particles and possible health risk implications were assessed. The health risk estimation was done by determining the values of total BaPeq, LLCR, and ILCR in Dhulikhel over the sampling
periods. The total BaPeq of 15 PAHs during the sampling period was found to vary from 1.26 to 51.49 ng/m³, having a mean value of 11.71 ng/m³, indicating the likely adverse health risks for local inhabitants (Table 4). This value exceeded the standard (1 ng/m³) as given by the WHO guideline and European Commission (EC) (WHO, 2000; Chen et al., 2017b). BaP alone contributed to about 67% carcinogenicity of the total PAHs in the study site.

The average LLCR was calculated for Dhulikhel as $1.01 \times 10^{-3}$, which was above the standard cancer risk level, as the upper limit of LLCR values specified by USEPA and the EC is $<10^{-6}$ to $10^{-4}$ per year (Jahmhari et al., 2014). Furthermore, the health risk assessment was performed by calculating the LADD values of PAHs for the age-specific group (adults and children) and then estimating their associated values of ILCR. According to most of the regulatory standard programs, the ILCR value between $10^{-6}$ to $10^{-4}$ specifies potential cancer risks, representing the rise in cancer chance by one in a million people, whereas an ILCR value exceeding $10^{-4}$ is considered as significant cancer risk and an ILCR value of $<10^{-4}$ represents negligible risks of cancer (Chen and Liao, 2006; USEPA, 1989). The average ILCR associated with carcinogenic PAHs in Dhulikhel was estimated to be $8.78 \times 10^{-6}$ for adults and $2.47 \times 10^{-5}$ for children, respectively. The ILCR value obtained in this study for adults suggests that it falls under the potential risk of cancer. This value is comparable to the average ILCR value of Lumbini, Nepal ($7.10 \times 10^{-6}$), and Jamshedpur, India ($15.78 \times 10^{-6}$) (Kumar et al., 2020), but lower than that obtained in the urban cities, Kathmandu, Nepal ($1.04 \times 10^{-5}$) (Neupane et al., 2018) and Amritsar, India $7.2 \times 10^{-5}$ (Kaur et al., 2013). On the contrary, the average ILCR value obtained in the study site for children suggests that the cancer risk level in this site is very high, and the adverse consequences of carcinogenic PAHs on children's health are more serious. These results indicate the need for attention to mitigate the emissions from contributors of total carcinogenic potential to reduce the adverse human health risks from inhalation of airborne PAHs.

### 4. CONCLUSIONS

This study offers the measurements of concentrations, seasonality and evaluates the sources and health risk assessment of particle-bound PAHs in ambient aerosols from January to July 2018 in Dhulikhel, a middle hill region of the Central Himalayas. The average levels of TSP ($264 \pm 108 \mu g/m³$) and PAHs ($73 \pm 66 ng/m³$) obtained exhibited similarity with those of some South-Asian cities. Evident seasonal variation of total PAHs was observed with higher levels during the dry season (pre-monsoon and winter) than those in the wet period (mon-
soil), attributed to higher emission sources and less dispersion during dry periods. The dominant species was 5-ring PAHs (32%), followed by 4-ring (29%) and 6-ring PAHs (28%).

Source apportionment using diagnostic ratios and PCA revealed mixed sources of emission like petrogenic as well as pyrogenic sources, particularly from fossil fuel combustion, biomass burning, and automobile exhausts to be the possible pollution sources in the site. Anthropogenic emissions from surrounding polluted areas and the IGP region could be responsible for particulate loading in the Himalayas. The mean total BaPeq level of PAHs in Dhulikhel was 11.71 ng/m³, signifying the potential adverse health risks for local inhabitants. The average ILCR associated with carcinogenic PAHs was estimated to be 8.78 ×10⁻⁶ for adults, indicating the potential risk of cancer and 2.47 ×10⁻⁵ for children, indicating serious carcinogenic effects of PAHs on children's health. This study delivers the first dataset of PAHs in the Himalayan middle hills, which is essential to formulate control strategies to reduce atmospheric emissions and health threats owing to PAHs over the region.

ACKNOWLEDGEMENT

The Pan-Third Pole Environment Study for a Green Silk Road (Pan-TPE) (XDA20040501), National Natural Science Foundation of China (41705132, 41630754), CAS “Light of West China” program and the State Key Science and Technical Support Program of China (2016YFC0203101) supported this study. Dr. Lekhendra Tripathee is thankful to the Asia-Pacific Network for Global Change Research (APN) for the grant (Grant reference: CRECS 2020-07MYTripathee) and Chinese Academy of Sciences for providing President’s International Fellowship Initiative (PIFI) as Young Staff (2020FYC0001). This study is part of the research initiative: Atmospheric Pollution and Cryospheric Change (APCC). The authors would like to express their deep gratitude towards Bhaskar Shrestha for the help and support.

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**Table S1.** Parameters of equation (1) and octanol-air partition coefficients (K_{OA}) of PAHs at 10°C, 18°C, and 23°C in winter, pre-monsoon and monsoon season, respectively.

| S.N | PAHs | A   | B   | log K_{OA} (10°C) | log K_{OA} (18°C) | log K_{OA} (23°C) |
|-----|------|-----|-----|-------------------|-------------------|-------------------|
| 1   | AcPy⁰ | -1.97 | 2476 | 6.77              | 6.53              | 6.39              |
| 2   | AcP⁰  | -2.20 | 2597 | 6.97              | 6.72              | 6.57              |
| 3   | Flu⁰ | -2.61 | 2833 | 7.40              | 7.12              | 6.96              |
| 4   | Phe⁰ | -3.37 | 3293 | 8.26              | 7.94              | 7.75              |
| 5   | AnT⁰ | -3.41 | 3316 | 8.30              | 7.98              | 7.79              |
| 6   | FluA² | -4.34 | 3904 | 9.45              | 9.07              | 8.84              |
| 7   | Pyr² | -4.56 | 3985 | 9.51              | 9.13              | 8.90              |
| 8   | BaA⁰ | -5.64 | 4746 | 11.12             | 10.66             | 10.39             |
| 9   | Chr² | -5.65 | 4754 | 11.14             | 10.68             | 10.40             |
| 10  | Bb⁰  | -6.40 | 5285 | 12.27             | 11.75             | 11.45             |
| 11  | BkF⁰ | -6.42 | 5301 | 12.30             | 11.79             | 11.48             |
| 12  | BaP⁰ | -6.50 | 5382 | 12.51             | 11.99             | 11.67             |
| 13  | IndP⁰| -7.00 | 5791 | 13.45             | 12.89             | 12.55             |
| 14  | DbA⁰ | -7.17 | 5887 | 13.62             | 13.05             | 12.71             |
| 15  | BghiP⁰ | -7.03 | 5834 | 13.57             | 13.01             | 12.67             |

\(^{a}(\text{Harner and Bidleman, 1998}); \(^{b}(\text{Odabasi et al., 2006})

**Fig. S1.** Temperature (°C) and relative humidity (%) in Dhulikhel during the sampling period.

**Fig. S2.** Correlation between concentrations of PAHs and TSP mass in the study site.
Fig. S3. Seasonal variations of TSP mass (μg/m³) and PAH (ng/m³) concentrations with precipitation in the study site during the sampling period.

Fig. S4. Correlations coefficient between concentrations of Σ PAHs and nss-K⁺ during (a) Winter, (b) Pre-monsoon and (c) Monsoon season.
**Text S1. Details on Methods (Extraction and Analysis of PAHs) and Quality Control/Quality Assurance**

Analyses of PAHs were done at the State Key Laboratory of Cryospheric Sciences, Northwest Institute of Eco-Environment and Resources, Lanzhou, China by using Gas chromatography–Mass spectrometry (GC-MS). An ITQ 1100 GC-MS system (Thermo Electron Corporation, USA) was used equipped with a (30 m × 0.25 mm × 0.25 μm) TG-SMS capillary column. The carrier gas (high-purity helium) was used at a constant flow rate of 1.0 mL/min. The mass spectrometer was functioned at a full scan in 70 eV electron impact mode at a mass scanning range of m/z = 50–650. The temperature of the injector and the transfer line was 280°C and, the ion source temperature was 230°C. The GC oven temperature gradient was carried out: the initial temperature was 70°C, which was retained for 3 minutes. The temperature was then increased to 150°C at 25°C/min, increased to 280°C at 3°C/min and finally increased to 300°C and held for 10 minutes. Samples were examined for the fifteen PAHs listed by the USEPA as priority PAHs: including Acenaphthylene (AcPy), Acenaphthene (AcP), Fluorene (Flu), Phenanthrene (Phe), Anthracene (AnT), Fluoranthene (FluA), Pyrene (Pyr), Benzo(a)anthracene (BaA), Chrysene (Chr), Benzo(b)fluoranthene (BbF), Benzo(k)fluoranthene (BkF), Benzo(a)pyrene (BaP), Indeno(1,2,3-cd)pyrene (IndP), Dibenzo(a,h)anthracene (DbA), and Benzo(g,h,i)perylene (BghiP). As Naphthalene (NaP) was detected with high concentrations in the laboratory and field blanks, it was not analyzed in this study.

Strict quality assurance and control procedures were followed for the analysis. The extraction and analysis of field blanks for the air samples were done using the same method as the samples. Method detection limits (MDLs) were calculated as 3 times the standard deviation of the mean blank concentrations. The detection limits were 0.15 (AcPy), 0.32 (AcP), 0.74 (Phe), 0.18 (AnT), 0.45 (Flu), 0.13 (FluA), 0.05 (Pyr), 0.04 (BaA), 0.01 (Chr), 0.07 (BbF), 0.06 (BkF), 0.03 (BaP), 0.16 (IndP), 0.06 (DbA), and 0.08 pg/m^3 (BghiP). Chrysene-d12 was added as the PAH surrogate standard to the samples before the extraction to check the extraction, cleanup, and analysis processes. The recovery efficiency of Chrysene-d12 was 69–109% for the TSP samples.

**Text S2. Details on the lifetime average daily dose (LADD) estimation of PAHs**

Benzo(a)pyrene (BaP) is mostly used as a marker of toxicity for all PAHs due to its high carcinogenic potency and its presence in the environment (Petry et al., 1996). International Agency for Research on Cancer (IARC) has classified total carcinogenic PAH considered as probably carcinogenic to humans (BaA, BaP, and DbA) and as possibly carcinogenic to humans (BbF, BkF, and IndP) (IARC, 2010).

The equation for estimating the lifetime average daily dose (LADD) of PAHs is shown as follows:

\[
LADD = \frac{Cs \times IR \times ET \times EF \times ED}{BW \times AT} \times cf
\]

where, LADD is the amount of intake per kilogram of body weight per day of a chemical suspected of having adverse health effects when absorbed into the body over a long period of time (mg/kg/d); Cs is the concentration of BaPeq for total carcinogenic PAH in the air (ng m⁻³); IR is the air inhalation rate (0.83 m³/h for adults), (0.5 m³/h for children up to six years) and ET is the exposure time (21 h/d); EF is the exposure frequency (350 d/year); ED is the lifetime exposure duration (years), 70 years for adults and 6 years for children; BW is the body weight (kg), it was assumed that BW = 70 kg for adults and 15 kg for children; AT is the averaging time (days) for carcinogens which is ED × 365 d. So, AT for adults = 25,550 and children = 2190 days, cf is the unit conversion factor, (10⁻⁶) (Bortey-Sam et al., 2015; Bozek et al., 2009).

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