# CHARACTERIZATION AND SOURCE IDENTIFICATION OF POLY CYCLIC AROMATIC HYDROCARBONS (PAHS) FOR COASTAL INDUSTRIAL CITY MANGALORE, INDIA

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## Abstract

*Polycyclic aromatic hydrocarbons (PAHs) are ubiquitous environmental pollutants generated primarily during the incomplete combustion of organic materials. These compounds are contributed to the atmosphere due to various anthropogenic activities in the form of particulate matter. In this study Particulate matter, $PM_{10}$ samples were collected from a Traffic site (Town hall) and Industrial site (KSPCB) of a coastal city Mangalore in India during post the monsoon period between October to December 2014. The samples were analysed for PAHs namely seven Fluorene (Flu), Acenaphthene (Ace), Chrysene (Chr),*

*Benz(a)anthracene (B(a)A), Benzo(a)pyrene (B(a)P), Benzo(b)fluoranthene (B(b)F), Indeno (1,2,3-c,d) and pyerene (Ind) using fluorescence spectrophotometer. The quarterly average of TPAHs concentration of the industrial site varied from 12 ng/m$^3$ to 109 ng/m$^3$ with an average of about 70.2 ng/m$^3$ whereas TPAHs concentration of traffic site varied from 39 ng/m$^3$ to 252 ng/m$^3$ with an average of 109 ng/m$^3$. Further it was observed that the TPAH concentrations showed increasing trend TPAH$_{oct}$ < TPAH$_{Nov}$ < TPAH$_{Dec}$ due to meteorological factors. Concurrently TPAH concentrations at traffic site was 1.8 times higher than that of the industrial site. The source apportionment study carried out using Principal Component Analysis (PCA) assisted by varimax rotation revealed that there were only two types of principal components PC1 and PC2. Both the PCs were observed to have variances of 66.21% and 14.38% respectively and classified to originate from fossil fuel burning predominantly diesel/petrol combustion in vehicles for traffic site and the rest from other type of fuels for the industrial site.*

## 1. Introduction

Ambient air quality has been worsened over the past decades due to the release of suspended particulate matter from numerous anthropogenic and natural activities. The release of these particulate matters in the atmosphere has become a grave environmental concern since it unswervingly affects the human health through the respiratory system leading to chronic and pulmonary diseases to the population susceptible to long-term exposures. The hazards caused by fine (≤2.5µm) and coarse particles (≥2.5µm to ≤10µm) are distinct. The impact of fine particles (PM$_{2.5}$) are higher compared to coarse particles (PM$_{2.5-10}$) since these particles goes deep into the respiratory tract and gets trapped and affects the lungs pertaining to respiratory disorders. Various studies concluded that PM$_{10}$ concentration is a strong indicator for several health effects both in India (Gupta, Karar, & Srivastava, 2007) and in other parts of the world (Braga et al., 2001). Numerous Indian researchers' works on optimizing the receptor model technique to find a holistic approach for source apportionment of particulate matter (Anu, Rangabhashiyam, Rahul, & Selvaraju, 2015; Selvaraju, Pushpavanam, & Anu, 2013). Therefore studies on these particulates is necessary for obtaining better knowledge and exposure on their behaviour and composition for improvising the monitoring and control of particulate matter. Polycyclic Aromatic Hydrocarbons (PAHs)

are a group of organic compounds consisting of two or more fused aromatic rings made up of carbon and hydrogen atoms. These are prevalent components of particulate matter ($PM_{10}$ and $PM_{2.5}$) and are suspected to be carcinogenic, Whereas some PAHs are associated with acute and chronic health complications (Guo, 2003). The United States Environmental Protection Agency (EPA) monitors 16 priority PAHs in air and found to be carcinogenic in experimental animals after inhalation or intra tracheal ingestion, increasing concern about the levels of these carcinogens in ambient air. PAHs are instigated from incomplete combustion of either natural (volcanic eruptions and forest fires) or anthropogenic processes like industrial emissions, motor vehicular emissions, refuse incineration, and residential heating (Abdel-Shafy & Mansour, 2016; Erik Dybing, 2013; Vu, 2010).

Among these automobile emissions were found to be the major contributor of PAHs in urban areas (Velasco, Siegmann, & Siegmann, 2004). Furthermore PAHs may be found both in the gas and particulate phase of ambient air or in mixtures of both phases since they range from semi volatile molecules to molecules with high boiling points (Erik Dybing, 2013). Generally urban atmosphere were found to be the major contributor of PAHs compounds and the dispersion of PAHs occurs through atmospheric transport and exists almost everywhere leading to its exposure to human beings (Vu, 2010). Hence studies on PAHs in ambient air grasps attention towards greater focus of research in recent years. Numerous studies have been carried out in the Indian scenario for PAHs emissions (Alankar Sharma, 2014; Masih, Saini, Singhvi, & Taneja, 2010) having TPAH concentrations 52.96 $ng/m^3$ and 97.93 $ng/m^3$ respectively. The present study emphasizes the prevalence of PAHs in the collected particulate matter samples by identifying PAHs compounds present in those samples and their concentration in the atmosphere during the post monsoon period, Likewise the study estimates the sources contributing to these PAHs compounds using a receptor modelling technique called Principal Component Analysis.

## 2. Materials and Methods

### 2.1 Study Area

Mangalore is located at 12.87°N 74.88°E in the Dakshina Kannada district of Karnataka. It has an average elevation of 22 metres (72 Ft.) above mean sea level. Mangalore is situated on the west coast of India, and is bounded by the Arabian Sea to its west and the Western Ghats to its east. The city, as a municipal entity, spans an area of 184.45 $km^2$ (71.22 sq mi). Mangalore experiences a tropical monsoon climate and receives about 95 per cent of its total annual rainfall within a period of about six months from end of

May to October, and a post monsoon period prevails from November to February, hot and humid from March to May.

## 2.2 Sampling Sites

Particulate Matter samples were collected from Town hall and Baikampady Industrial Area for Traffic and Industrial Sites respectively (Figure 1). The selection of sampling sites has been done according to the land use pattern in those areas.

Air samples for Baikampady Industrial area has been collected from the roof top of Karnataka State Pollution Control Board (KSPCB) building. The site is located close to various types of small-scale industries (Iron and Steel, Smelting, Fertilizers etc.). In the same way for traffic site, samples were collected from roof top of Town Hall building which is located 400 meters away from city bus stand. The site is located near to a major city junction with heavy vehicular traffic and also surrounded by various commercial complexes, hotels and hospitals. Thus the site characteristics pose to be suitable for Traffic Site.
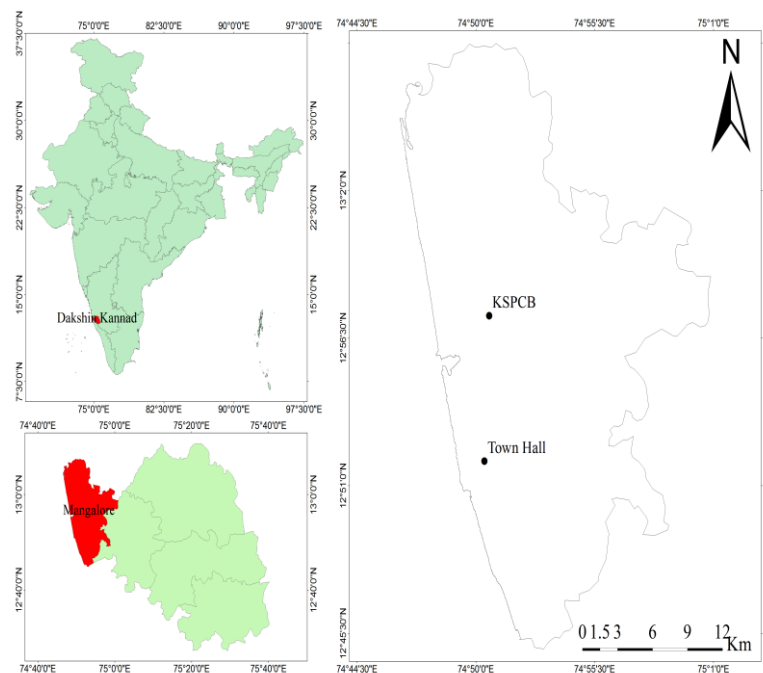


**Figure 1:** *Location of Sampling Sites at Mangalore*

## 2.3 Sampling Methodology

Sampling was carried out as per Guidelines prescribed for the measurement of ambient air pollutants (CPCB, 2011) for a period of 24 hours ($8 \times 8 \times 8$) at a flow rate of $1m^3$/min. The samples were collected every 8 hours (06:00 to 14:00, 14:00 to 22:00, and 22:00 to 06:00) on Glass Fibre Filters (Whatman GF/A $20.3 \times 25.4$ cm) using High Volume

RSPM Samplers (Envirotech APM460 BL) (Figure 2). The sampling time was distributed between the sample stations for two weeks alternatively in a month among the sampling stations during the study period. The mass concentration of $PM_{10}$ particles were measured by gravimetric technique using weighing balance (Oahu pioneer with accuracy 0.0001g). The filter papers were kept in a desiccator before and after sampling for 24 hours at a temperature of $27\pm30^{0}C$ and at a RH of $55 \pm 2\%$ to remove the moisture present in them.
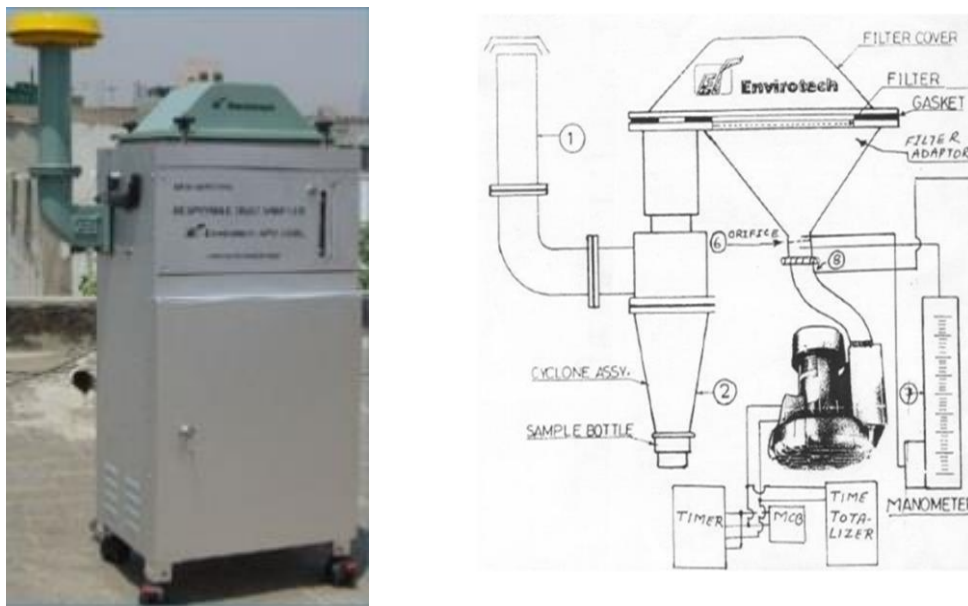


**Figure 2:** *RSPM Samplers used for Particulate Matter Sampling*

## 2.4 PAHs Extraction

Each sampled filter paper containing particulate matter was cleaned by ultrasonication in 5 ml DCM for 15 min. For MIP Extraction of PAHs, decanted solution of sample (leachate) (5 ml) was mixed with 10 mg MIP and agitated in water bath at 100 rpm for 3 hours. The MIP was then separated from the particulate leachates by centrifugation. The PAHs selectively adsorbed on the MIP were re-extracted with 1 ml methanol. This extract was spiked with 1 $\mu gL^{-1}$ of a mixed standard of PAHs (10 $\mu L$) and then analysed for PAHs by fluorescence spectrophotometer. Extraction procedure was followed according to (R J . Krupadam et al., 2009) with necessary modifications.

## 2.5 Concentration of PAHs in Air

The concentrations of all fluorescent PAHs were determined for particulate samples of ambient air as given below.

$$V = Q \times t \quad \longrightarrow \quad (1)$$

Where,

Q = Average flow rate of sampling $m^3$/min

T = sampling time, in min.

V = total sample volume at ambient condition in $m^3$

The concentration of PAH compound in ng/$m^3$ in the air sampled is given by

C (ng/$m^3$) = $C_s \times V_e$/ $V_{i \times} V_s$

Where,

$C_s$: Concentration of PAH compound in (ng/mL) sample extracted

$V_e$: Final Volume of the extract in mL

$V_i$: Initial volume (mL)

$V_s$: Volume of the air sampled $m^3$

## 2.6 Quality Control and Data Analysis

In order to estimate the PAHs present in low concentration, spiking technique was involved. Blank filter papers were spiked with known concentrations of PAHs standard mixture of 0.1 ppb, 0.5 ppb and 1 ppb respectively. All the Sample filter papers were spiked with 10 µl of 1ppb PAH mix and analysed. Spiked concentrations were deducted while analysing the concentrations of the sample.

Wavelength accuracy checks were made 3 times for different emission/excitation slit width. Selective excitation maxima for the individual target analyse was taken for the analysis.

## 2.7 Principal Component Analysis

Principal component analysis (PCA) is a factor analysis method by reducing dimensions of large set of data into factors or components. In the present study, PCA was used to explore the major possible sources of the PAHs. PCA transforms large set of multivariate data set into smaller set of independent components with minimum data loss (Singh, Malik, Kumar, Saxena, & Sinha, 2008). The PCA method is performed using software SPSS 16.0. Significant numbers of components were retained by using the scree plot. In general, scree plot displays the eigenvalues associated with a component or factor in descending order versus the number of the components or factors. These scree plots can be used in PCA to visually assess which components or factors explain most of the variability in the data. In this

study the scree plot (Figure 3) shows a steep curve, in which first two components were selected. Remaining factors were excluded due to their flat trend.
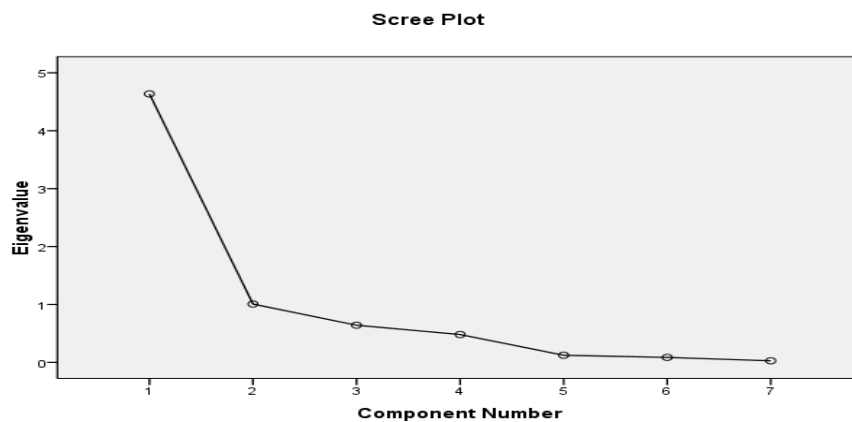


**Figure 3:** *Eigen Values of the Various Components*

## 3. Results and Discussion

### 3.1 PM$_{10}$ Analysis and Characterization

The PM$_{10}$ concentration during the study period ranges from 97 µg/m$^3$ to 73 µg/m$^3$. The concentration was found to have not exceeded the National Ambient Air Quality Standards (NAAQS). The monthly PM$_{10}$ concentrations at both sites were illustrated in Figure 2. It was evident that the PM$_{10}$ Concentrations increases from October to December due to various meteorological conditions.
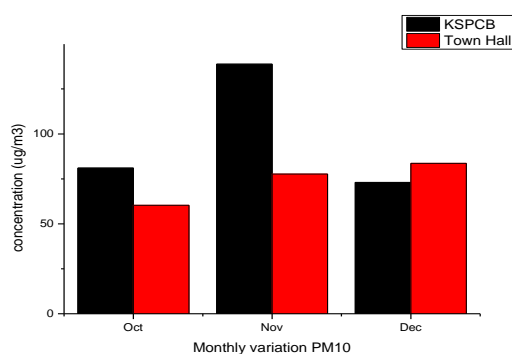


**Figure 4:** *PM$_{10}$ Concentration during the Study Period*

### 3.2 PAHs Concentration Profiles

Quarterly average of Total Polycyclic Aromatic Hydrocarbons (TPAH) concentrations of KSPCB Industrial Site varied from $12ng/m^3$ to $109ng/m^3$ with an average of $70.2ng/m^3$. While TPAH concentrations of Town hall Traffic Site varied from $39ng/m^3$ to $252ng/m^3$ with an average of $109ng/m^3$. It is observed that PAHs concentrations fluctuated continuously during the study period due to the various meteorological factors, such as wind direction and also affected by the stability of PAHs molecule.

The PAHs compounds Ace, B(b)F, Ind estimated from samples collected at KSPCB site were found to possess higher concentration ranging from $10ng/m^3$ to $29ng/m^3$. Whereas Chr, B(a)A possess a significant concentration. B(a)P a more stable PAH, possess concentration ranging from $1.31-12.41ng/m^3$ with an average of $4.36 ng/m^3$.

Similarly concentrations of Ace, Chr, B(b)F estimated from Town hall Traffic site were found to be significant with Ind and Carcinogen B(a)P concentration ranging from 10 to $48ng/m^3$ and 0.2 to $13ng/m^3$ respectively.

Among the estimated PAHs, Acenaphthene and Ind concentrations were comparatively higher than the other PAHs at both the sites, since both the sites were observed to have many likely markers of the above mentioned PAHs. In Town hall site heavy traffic, grilled meat cooking, gas stoves for cooking in restaurants and many activities like smoking, open burning wastes and road dust from construction work exhibit the presence of PAHs. Likewise in KSPCB industrial site PAHs were contributed through emissions from small scale industries like electroplating industries, metal welding industries, smelting industries and so on. Also emissions from Petrochemical and Fertilizers industries located in the vicinity of the sampling location.

**Table 1:** *Quarterly Average concentrations (Avg), Minimum (Min), Maximum (Max) of individual PAH ($ng/m^3$) in $PM_{10}$ at KSPCB and Town hall*

| PAHs | KSPCB | | | Town Hall | | |
|------|-------|-----|-----|-----------|-----|-----|
|      | **Min** | **Max** | **Avg** | **Min** | **Max** | **Avg** |
| Flu | 0.07 | 2.952 | 0.5 | 0.230 | 6.98 | 2.79 |
| Ace | 8.16 | 64.8 | 22.46 | 16.34 | 112.11 | 48.56 |
| Chr | 0.29 | 31.05 | 8.53 | 0.885 | 28.48 | 10.28 |
| BaA | 0.16 | 20.21 | 8.01 | 0.127 | 24.36 | 8.80 |
| BaP | 1.31 | 12.41 | 4.36 | 0.240 | 13.85 | 4.94 |

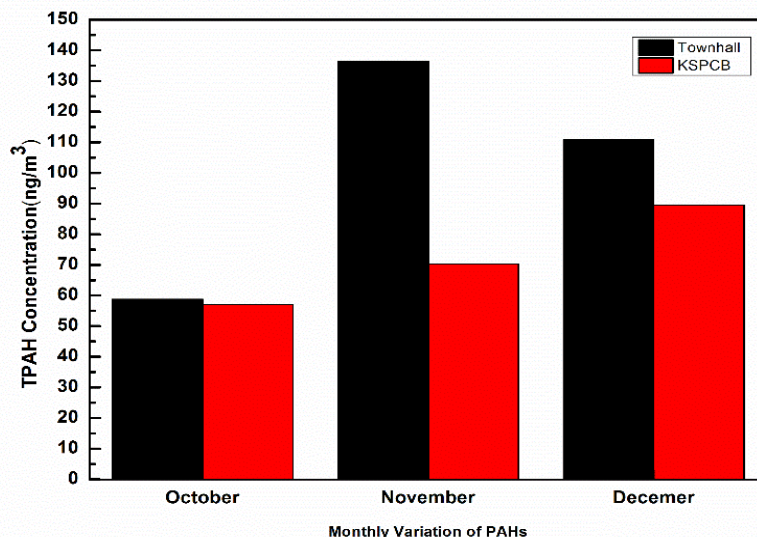| BbF | 0.23 | 29.08 | 10.48 | 2.17 | 29.80 | 10.67 |
|-----|------|-------|-------|------|-------|-------|
| Ind | 6.656 | 46.36 | 18.74 | 6.37 | 44.47 | 22.73 |



**Figure 5:** *Monthly variation of TPAHs Concentration at KSPCB and Town hall Sites*

### 3.3 Spatial Variation

A significant variation with respect to spatial conditions was observed at KSPCB and Town hall sampling site (Figure 4 and Figure 5). Total polycyclic aromatic hydrocarbons (TPAH) concentration at Town hall was at higher levels compared to the TPAH concentration at KSPCB site.

TPAH concentrations of KSPCB Industrial site were observed to be 53.71ng/m$^3$ in October, 73ng/m$^3$ in November and 88.46ng/m$^3$ in December. This phenomenon shows an increasing trend of TPAH$_{oct}$<TPAH$_{Nov}$<TPAH$_{Dec}$ concentrations during the study period. This rise in TPAH concentrations was due to the influence of meteorological factors such as temperature decrease, low wind speeds at the sampling sites. Further it was found that TPAH concentration at town hall was 1.8 times higher than KSPCB site in November and successively 1.4 times higher during October and December. Uncertain rain in the month of October has also decreased the PAHs concentration in Mangalore and similar kind of results were reported by (Kulkarni K.S., 2014) for Mumbai region which lies on the same coast of western India.

Minimum TPAH concentration of 73.75ng/m$^3$ was observed in October 2014 and maximum, 135ng/m$^3$ was observed in November 2014. This rapid increase in the TPAH concentrations would have been caused by the post monsoon trading during months of

November and December at New Mangalore Port Trust which in turn has increased the traffic on the highway NH66 leading to higher contribution of TPAHs from the incomplete combustion of heavy duty vehicles.
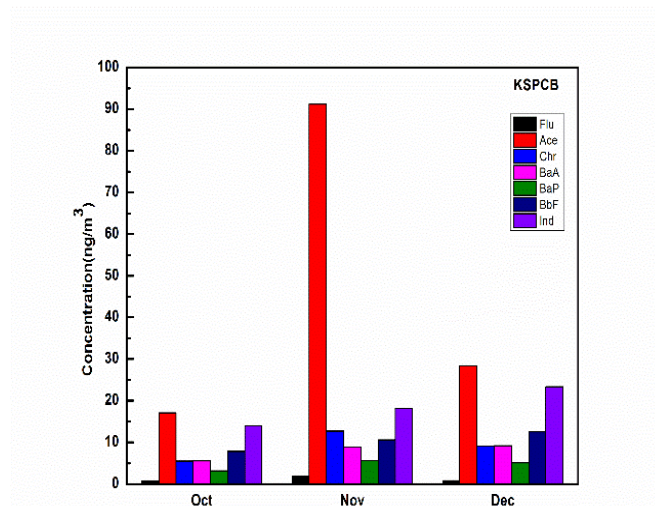


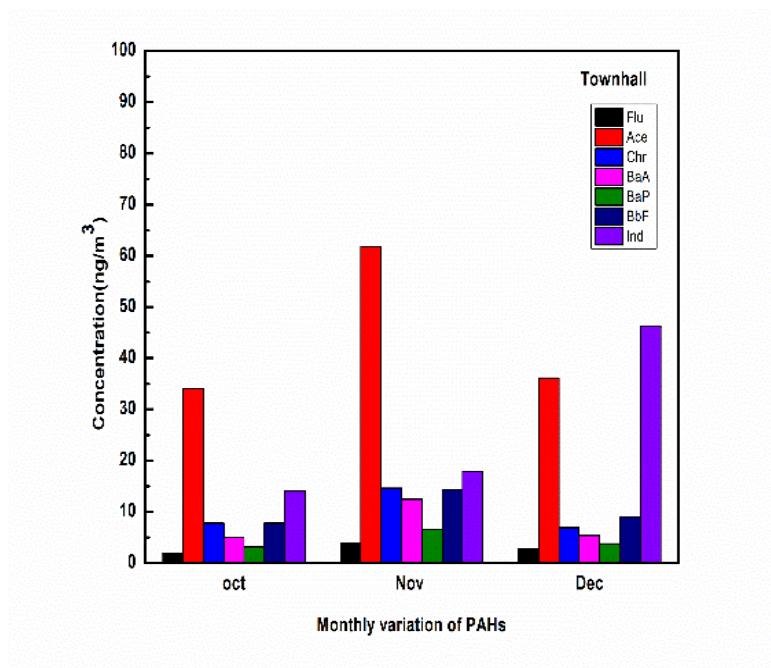**Figure 6:** *Monthly variation of PAHs at KSPCB Industrial Site*



**Figure 7:** *Monthly variation of PAHs at Town hall Traffic Site*

## 3.4 Source Apportionment of PAHs Compounds

The Principal Component Analysis (PCA) using varimax rotation for the seven estimated PAHs compounds yielded two principal components PC1 and PC2 with a variance of 66.21% and 14.38% respectively. Table 2 PCA results reveal that the $PM_{10}$ emissions from these sources have a very high cumulative variance of 80%.

The PC1 exhibits 66.21% of variance and it indicates that the sources of PAHs compounds prevailing in the $PM_{10}$ samples have been contributed from combustion of coal, wood, gas emissions and natural gas. PC1 possess a higher factor loading (>0.70) for Ace, Chr, BaA, BaP, BbF loadings. Whereas moderate factor (>0.50) loadings for Flu. The result of higher loadings for low molecular weight (LMW) PAHs indicates the sources of these PAHs from burning of coal, wood and cow dung cakes (Nasrin R. Khalili, 1995). PAHs compound like Fluorene possessing a moderate factor loading (0.673) was found to be a dominant marker for gas emission. Likewise BaP and BbF are emitted from natural gas combustion, Chrysene and Benz (a) anthracene are reported as molecular markers for wood combustion(Nasrin R. Khalili, 1995; Ravindra, Sokhi, & Vangrieken, 2008). Thus PC1 can be categorized as emissions from coal, wood, gas emissions and natural gas i.e., emissions from fuels other than diesel/petrol combustion.

Similarly PC2 with a variance of 14.38% shows strong factor loadings for Ind. (Matt F. Simcik, 1999) reported that Ind can be used as finger print to identify diesel emissions. Also the PAHs having moderate loadings are associated with the vehicular emission (Nasrin R. Khalili, 1995; Randolph K . Larsen & Joele.Baker, 2003) and confirms the emissions of these PAHs from vehicular source emissions in the vicinity of the sampling location. Thus PC2 indicates the emissions at the traffic site were contributed by fossil fuel burning predominantly from gasoline/diesel combustion from vehicles.

Thus the two principal components PC1 and PC2 can be labelled as emissions from combustion sources emissions and diesel/gasoline (Figure 7) combustion emissions respectively.

**Table 2:** *PCA factor loadings using varimax rotation for samples from both Traffic and Industrial Sites*

| PAHs | PC1 | PC2 |
|---|---|---|
| Fluorene | 0.673 | 0.235 |
| Acenaphthene | 0.874 | 0.013 |
| Chrysene | 0.959 | 0.002 |
| Benz(a)anthracene | 0.953 | 0.075 |
| Benzo(a)pyrene | 0.893 | 0.282 |

| | | |
|---|---|---|
| Benzo(b)fluoranthene | 0.824 | 0.051 |
| Indeno(1,2,3-c,d) pyerene | 0.83 | 0.986 |
| % variance explained | 66.21 | 14.38 |
| Eigen values | 4.635 | 1.007 |
| Predicted Sources | Combustion (Coal, wood, natural gas) | Diesel/Gasoline |

Values (>0.70) are loadings

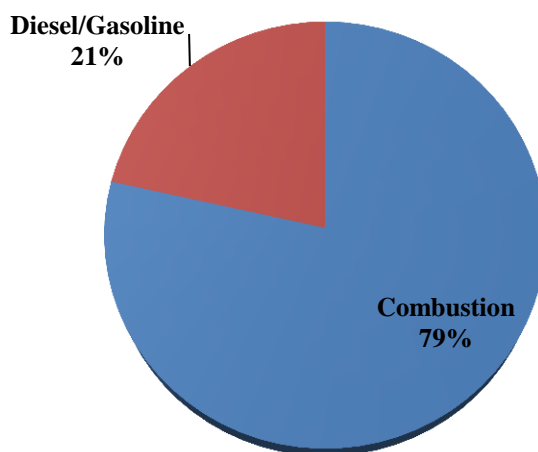Values (>0.50) are moderate loadings



**Figure 8:** *Source apportionment of PAHs from Traffic and Industrial Sites*

## 4. Conclusion

Air samples of particulate matter ($PM_{10}$) with aerodynamic diameter $\leq 10\mu m$ were collected and analysed for PAHs present in those samples. Seven PAHs Fluorene (Flu), Acenaphthene (Ace), Chrysene (Chr), Benz(a)anthracene (B(a)A), Benzo(a)pyrene (B(a)P), Benzo(b)fluoranthene (B(b)F), Indeno (1,2,3-c,d) and pyerene (Ind) compounds have been detected using Fluorescence Spectrometer. The average TPAHs Concentration at KSPCB Industrial Site and Town hall Traffic Site were found to be 70.2ng/m$^3$ and 109ng/m$^3$respectively. The spatial variation of TPAHs concentration during the study period showed an increasing trend due to the impact of meterological conditions prevailing in the sampling locations. Source apportionment of the PAHs using principal component analysis

using varimax rotation yielded two principal compound PC1 and PC2. PCA analysis revealed that the sources of emissions at KSPCB industrial site has been contributed from the emissions of combustion of fuels other than gasoline/diesel, whereas the sources of emissions at Town hall traffic site has been contributed from the gasoline/diesel vehicle combustion. Thus the study concluded that the sources of PAHs in the study area were mainly from vehicular emissions and combustion emissions from fuels other than gasoline/diesel.

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