Evaluation of seasonal variations in abundance of BTXE hydrocarbons and their ozone forming potential in ambient urban atmosphere of Dehradun (India)

Neetu Bauri · Pradip Bauri · Krishan Kumar · V. K. Jain

Abstract In Dehradun, ambient volatile organic compounds (VOC) samples were collected for three seasons viz. summer, winter and monsoon (during period 2012–2013) to investigate seasonal variations at five different sampling sites. The samples were quantified for aromatic VOCs by gas chromatography (GC-FID) to monitor benzene, toluene, m, p-xylene, o-xylene and ethylbenzene. BTXE comprise an important group of VOCs mostly prevalent in a typical urban environment. They were monitored because they are known to cause impacts on climate, health and on vegetation. Toluene was found to be the most abundant VOC among the measured ones in the atmosphere of Dehradun. The maximum mean concentration of VOCs was observed in winters and lowest during summers for BE species. Toluene ambient concentration was rather found to register highest during winters and lowest in monsoons. Kruskal–Wallis test showed statistically significant differences seasonally ($p < 0.05$). High toluene to benzene T/B (>1) observed ratio indicates vehicular emission as their major source. BTEX were also evaluated for their ozone-forming potential (OFP). Toluene and xylenes were found as the highest contributing hydrocarbons towards ozone forming potential among BTXE.

Keywords VOC · Urban · Kruskal–Wallis test · OFP · Interspecies ratio · Dehradun

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Introduction

In today's time, urban air pollution has now become synonymous with a wide variety of pollutants among which volatile organic compounds (VOCs) are considered as an important one. Generally, the acronym VOC implies non-methane volatile organic compounds (NMVOC) (Demeestere et al 2007). They have both anthropogenic as well as biogenic sources. Anthropogenic emissions in urban atmosphere consist of vehicular exhaust, gasoline evaporation, solvents usage, leakage from natural gas (CNG) and liquefied petroleum gas (LPG), air fresheners, tobacco smoke, dry cleaning chemicals, landfill sites, municipal solid waste, etc. (Zou et al. 2003; Caselli et al. 2010; CPCB 2010; Majumdar and Srivastava 2012; Suvarapu et al 2013). Biogenic VOCs (BVOCs) emission mainly comes from vegetation (forests), termites, wetlands and tundras (CPCB 2010). They include isoprene (C_5H_8), monoterpenes ($C_{10}H_{16}$), and sesquiterpenes and several oxygenated species (Ciccioli et al 1993; Koknig et al 1995; Kesselmeier et al 1996; Komenda et al 2001.). BVOCs are known to have minimal contribution as compared to anthropogenic emissions in urban areas (Wang et al 2012).

VOC's are precursors of a number of secondary air pollutants such as PAN, tropospheric ozone and secondary organic aerosols (Atkinson 2000; Elbir et al 2007; Lau et al 2010). Wang and Zhao (2008) reported that toluene, ethylbenzene, xylene and trimethylbenzenes readily undergo reaction with OH radical to produce ozone, photochemical oxidants and smog. VOC's have serious health implications. WHO 2000 report mentioned that people residing in urban areas are at risk of developing 10 leukaemia cases per million of inhabitants of lifetime exposure at 1.7 µg/m³ (Buczynska et al 2009). VOCs are known to play an important role in plant physiology and ecology (Seco et al 2007). Plants growing in polluted urban environments are more prone to experience significant detrimental changes in timing of key activities, for example,

flowering, leaf senescence, leaf surface characteristics along with their growth (Honour et al 2009)

Mono aromatic compounds like benzene, toluene, xylenes and ethylbenzene are collectively named as BTXE form an important group found in urban regions (Lee et al 2002). In ambient atmosphere, BTXE tend to remain in gaseous phase (Rad et al 2014) and constitute 60 % of NMVOCs (Lee et al 2002).

Several studies pertaining to the measurement of BTXE have been carried out by researchers all over the world. For example, Lee et al (2002) studied that similar to other Asian cities, the VOC levels in urban areas in Hong Kong were affected both by automobile exhaust and industrial emissions. Fernàndez-Villarrenga et al. (2004) reported that the BTEX along with chlorinated hydrocarbons dominated the urban air of A Coruña, Spain. Their work acknowledged traffic and evaporative losses as the major source of VOCs by applying principal component analysis (PCA) and correlation studies. In Nigeria, emissions from industries and traffic were identified as dominant sources of VOC arising out of anthropogenic origin (Ojiodu et al 2013). In Malaysia, the research work of Al Madhoun et al. (2011) reported a negative correlation between temperature and concentration levels of benzene and identified that major source of benzene comes from gasoline-based vehicular emissions. In India, Srivastava et al. 2005a; Srivastava et al. 2005b; Srivastava et al. 2005c and Srivastava 2005 carried out source apportionment study of VOCs in ambient air of Delhi and Mumbai. Their study found diesel combustion engines to be a major contributor to ambient VOCs in Delhi. Evaporative emissions and sewage sludge was also found to contribute significantly. However, in Mumbai, evaporative emissions were found to contribute maximum to total (TVOC) concentration to ambient air. Kumar and Tyagi (2006) investigated the benzene and toluene profiles in ambient air of Delhi. They found higher concentration of benzene than the limit values prescribed in the UK (16 $\mu g \ m^{-3}$, annual average) and European Commission (5 $\mu g \ m^{-3}$, annual average). Hoque et al (2008) studied the variation of BTEX spatially and temporally in the urban atmosphere of Delhi. They found vehicular exhaust to be a major contributor of BTEX in urban air of Delhi. Majumdar et al 2011 conducted a study in ambient air of Kolkata. A multimedia mass balance model, TaPL3 was applied to estimate the environmental distribution and load of BTEX in different environmental departments (air, water and soil). BTEX were found to be resided in air compartment and were followed by soil and water with total environmental load as high as 9.7×104 Kg. Pandit et al (2011) studied the distribution of atmospheric non-methane hydrocarbons (NMHCs) and carried out their source apportionment at urban sites in Mumbai, India. Their study identified vehicular exhaust, refineries, petrochemical production facilities, paint solvent and polymer-manufacturing industries as their sources. Chaudhary

and Kumar (2012) studied the mixing ratio of BTEX in ambient air of Firozabad, Uttar Pradesh. Their finding showed a significant difference between daytime and night-time concentration levels of BTEX species. Singla et al (2012) assessed the concentration of benzene, toluene and xylene (BTX) along the roads and petrol pump in Agra. Interspecies ratios and interspecies correlation showed that BTX concentration levels were influenced by road traffic.

Though most of the literature available on VOCs is reported by the international scientific community, studies by the Indian researchers are rather limited. Those documented are basically aimed at the monitoring and characterization of VOCs in the megacities with very little attention given to the high-altitude areas. The aims of present work produced in this paper are to infer the variations in ambient concentration levels of BTXE hydrocarbons in Dehradun on seasonal basis and to study their interspecies ratio and evaluation of their ozone-forming potential. Study pertaining to characterization of BTXE species in Dehradun has not been reported so far. Therefore, results of study presented in this research paper would be useful for researchers, policymakers and other stakeholders in formulating effective pollution control strategies for abating the rising pollution levels in Dehradun.

Material and methods

Dehradun is the capital city of Uttrakhand and nestled in the Doon valley surrounded by hills of the Himalayas. It lies between 29° 58′–31° 2′ 30″ N latitudes and 77° 34′ 45″–78° 18′ 30″ E longitudes at an altitude of 2100 ft above the mean sea level (http://dehradun.nic.in/). It has a total area of 3088 km^2 (http://dehradun.nic.in/). The area receives an annual rainfall of about 2073.3 mm with maximum rainfall occurring during the months of monsoon season (http://dehradun.nic.in/). The rainfall and temperature variations within the city are quite considerable due to altitudinal variation and local orography with presence of Mussoorie hills. Since it is a valley, the condition here is conducive for the formation of inversion during most of the period of the year (Banerjee et al 2002; Rana et al 2009).

In recent years, Dehradun city has evolved as an important business, educational and cultural destination in northern India. According to an estimate of Uttaranchal Urban Development Project (Uttaranchal Urban Development Project 2007), one million Indian and foreign tourists visit this city every year in the form of floating population. Enhanced vehicular traffic density has been observed in the city which has been contributed both by the vehicles registered here and others registered elsewhere but moving on the roads of Dehradun (Kishore et al 2014). They normally cause traffic jams on almost all intersections of the city. Three wheelers known as Vikram are normally found running in

the city. They are both diesel and petrol-driven vehicle and are poorly maintained and often carry more passengers than required. Chauhan et al 2010 estimated that concentrations of annual NO_X and SO_2 varied between 23.66 and 30.37 μg/m^3 and 21.72–28.17 μg/m^3 at Clock Tower (Ghantaghar), respectively. Suspended particulate matter (SPM) concentration in the city is found to exceed the National Ambient Air Quality Standards (NAAQS). Uttaranchal Urban Development Project (Uttaranchal Urban Development Project 2007) registered the increased levels of suspended particulate matter (SPM) from 250 to 400 μg/m^3 during 2003–2005 in Dehradun. Dehradun has been experiencing growth in human number and their associated activities in the form of number of vehicles, narrow roads, parking facility problems, ineffective implementation of laws, urbanization, industrialization and several unplanned developmental projects which has resulted in severe ecological degradation and profoundly found its impacts on urban climate alarmingly (Singh et al 2013).

Five representative sites from Dehradun were chosen for ambient air sampling on the basis of land use and land cover pattern, traffic count, availability of the electricity and security of the sampling equipment. These are represented in Table 1 and Fig 1. (S1) ISBT is the interstate bus terminal which is located near the intersection of Saharanpur road and Haridwar bypass road. (S2) Ghantaghar is located in the central part of the city with heavy traffic density. (S3) Panditwari is a medium density traffic area. (S4) Malsi: Sampling here has been done at a petrol pump in this area and is the gateway to Mussoorie and (S5) Doon University is a premier institute of learning located on the southeastern part of Dehradun city.

The data of meteorological parameters for Dehradun like temperature (T), relative humidity (RH), wind speed, wind direction and rainfall data were obtained from meteorological station of Dehradun (IMD, Dehradun). The wind rose plots were drawn using Lake Environmental software using wind speed and wind directions.

Average wind speed was found to be 0.84, 0.47 and 0.56 m/s for summer, winter and monsoon, respectively with frequent calm among seasons. The wind roses for Dehradun are provided in Figs 2a–c. In summer, winds mainly prevailed from west (with 1.0–1.4 m/s), while winters were dominated by winds from west (1.0–1.4 m/s), northwest (with 1–1.4 m/s to 1.4–1.9 m/s) and northeast (varied from 1.0 to 1.4 m/s to 5.5 to 7.0 m/s). Moreover, monsoon was found to experience a mixed pattern of winds from southwest (1.0–1.4 m/s) and northeast (ranging from 1.0–1.4 m/s to 7.0–8.5 m/s). In summers, the average minimum and average maximum temperatures recorded were 21.07 and 36.41 °C with mean relative humidity of 39.33 %. Whereas winter experienced 21.55 and 9.84 °C as average maximum and average minimum temperatures, respectively with average relative humidity of 76.51 %. Average relative humidity was

observed in monsoon month as 80.95 % with 31.48 °C as average maximum and 21.17 °C as average minimum temperature. Average monthly rainfall measured to be 0.05, 6.80 and 7.20 mm in summer, winter and monsoon seasons, respectively.

The monitoring in different seasons was carried out in May 2012 (summer), February 2013 (winter) and September 2013 (monsoon). The sampling of VOCs in the ambient atmosphere was carried out according to the standard method developed by NIOSH 1501 using large activated coconut shell charcoal (specially treated) tubes (ORBO™ tubes, Supelco). The sampling was carried out during daytime from 8:00 AM to 5:00 PM for 1 day at each individual site. Seven to eight samples were collected at each site. Also, the samples were procured alternately due to a single low volume sampler. Air was drawn through the activated charcoal tubes using a portable constant flow low volume sampler (Envirotech) with a flow rate of 200 ml/min. The sampling duration was 60 min. Breakthrough test was performed prior to sampling, and therefore, flow rate and sampling duration were adjusted accordingly. One blank from each respective site was collected. Thirty-six samples for summer season, 32 for winter and 30 for monsoons were analysed for BTXE hydrocarbons.

Analysis was done by GC-FID (Shimadzu, Auto sampler, Model AOC- 20i, GC-2010) fitted with RTX-VGC column (75 m, 0.45 i.d, 2.55-μm film thickness) and GC solution software. VOCs were quantified using the multipoint external standard calibration procedure using VOC Standard (HC BTEX/MTBE Mix, 2000 μg/ml each in methanol) were supplied by Supelco (Sigma Aldrich).

The quality assurance and quality control

To verify the validity and reproducibility of the data procured, some samples were chosen and their duplicate measurements were carried out. For laboratory, blank unexposed ORBO™ tube was analyzed for BTXE similar to the exposed ones. Field blanks were not attached to sampler and undergo the same storage and transportation conditions. Breakthrough was checked prior to sampling by adjusting the flow rate with the duration of sampling. Standards used for calibration were analyzed weekly to check the GC performance.

Ozone-forming potential

In this study, for calculation of Ozone-forming potential maximum incremental reactivity (MIR) scale was used. This scale has been widely used by many researchers worldwide like Hoque et al 2008 (Delhi, India), Tan et al. 2012 (Foshan, China), Zheng et al. 2009 (China) to rank hydrocarbons on the basis of their contribution to ozone formation. Carter 1994 proposed the ozone-forming

Table 1 Description of sampling sites

| S. No.	Location ID	Site	Site type	Geological coordinates	
				Latitude	Longitude
1	S1	ISBT	Traffic	30.2888° N	77.9968° E
2	S2	Ghantaghar	Commercial cum traffic	30.3243° N	78.0416° E
3	S3	Panditwari	Residential cum commercial	30.3327° N	77.9953° E
4	S4	Malsi	Petrol pump	30.3783° N	78.06751° E
5	S5	Doon University	Residential cum institutional	30.2643° N	78.0390° E

potential (MIR scale) which measures the contribution of particular VOC to ozone formation. It is calculated as product of VOC concentration by its MIR coefficients (dimensionless) (Carter 1994). MIR is defined as the amount of ozone formed per gram of VOC added to a VOC-NO_X mixture, reflecting how much a VOC would contribute to the ozone formation in the air mass (Carter 1994; Hoque et al 2008). In high NO_X conditions, usually the concentration of radicals formed by reaction with VOCs decides the amount of ozone formation, whereas NO_X levels tend to limit the ozone formation process during low NO_X scenario (Carter 1994; Tan et al. 2012). MIR coefficients were given by Carter 1994. Highly reactive VOC would yield higher MIR (Czader et al. 2008). Many factors affect the photochemical O_3 formation like NO_X concentration,

solar radiation intensity, meteorological conditions along with VOC reactivity's (Tan et al. 2012).

The following equation was used to determine the contribution of VOCs to ozone formation, given by Carter 1994:

$$\mathrm{OFP(k)} = \mathrm{Concentration\ (k)} \times \mathrm{MIR\ coefficient} \qquad (1)$$

where, ozone-forming potential (OFP) (k) is the ozone-forming potential of hydrocarbon (k), MIR coefficient is the maximum incremental reactivity coefficients of the VOC concerned.
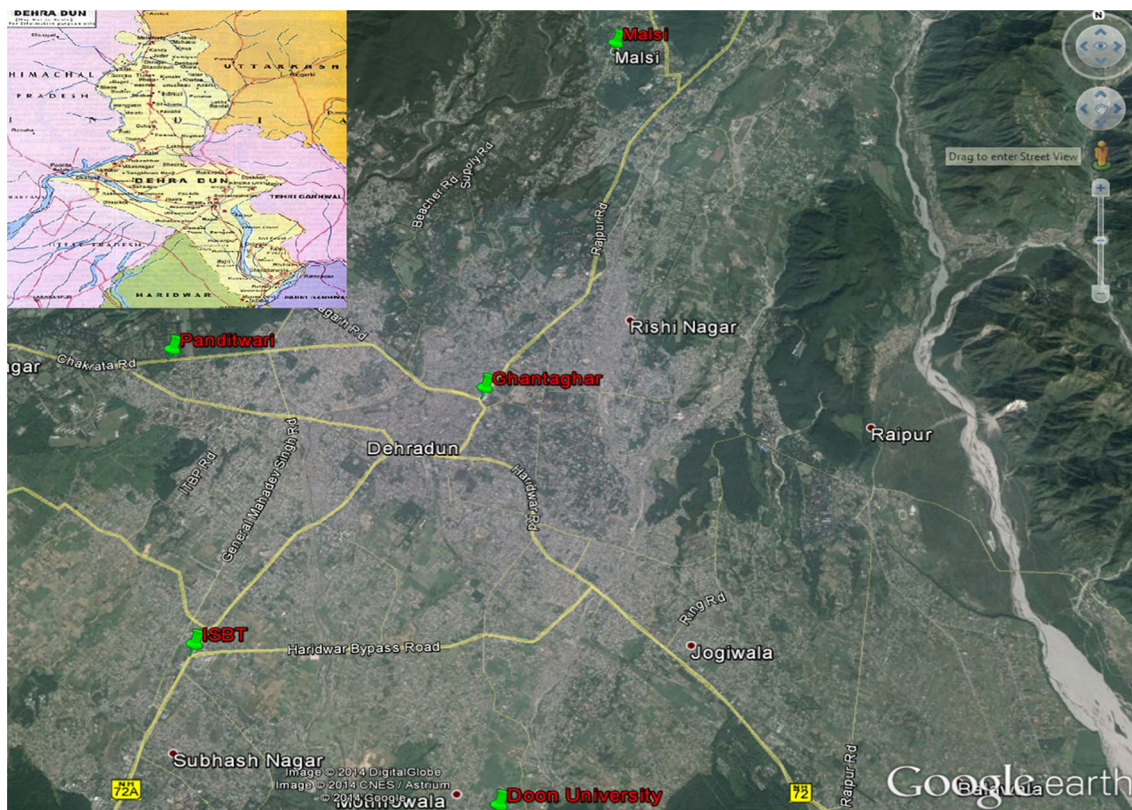
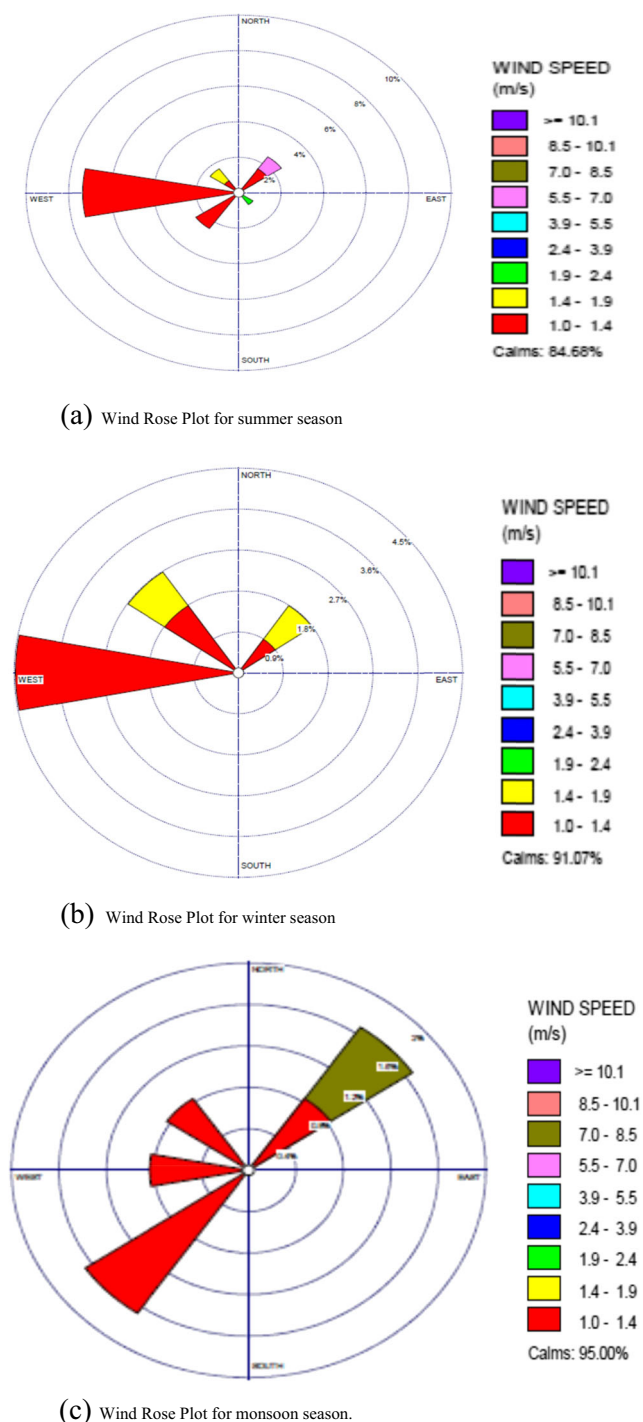**Fig. 1** Sampling sites in Dehradun (Map)

(a) Wind Rose Plot for summer season

(b) Wind Rose Plot for winter season

(c) Wind Rose Plot for monsoon season.

Fig. 2 **a** Wind rose plot for summer season. **b** Wind rose plot for winter season. **c** Wind rose plot for monsoon season

Results and discussion

Seasonal variability of BTXE hydrocarbons in Dehradun

The seasonal mean concentration of five monitored aromatic VOCs (BTXE) in Dehradun at five respective sites is shown in Fig 3. All the sites have been found to exhibit seasonal

variability. Among BTXE, toluene was found to be the most abundant monoaromatic species in the ambient atmosphere of Dehradun. Similar trend has also been observed by researchers in other urban areas by Muezzinoglu et al 2001 in Turkey; Kerbachi et al 2006 in Algiers; Martins et al 2007 in Rio de Janerio; Hoque et al 2008 in Delhi; Majumdar et al 2011 in Kolkata; Pandit et al 2011 in Mumbai; Mohan and Ethirajan 2012 in Chennai; Rad et al 2014. Among BTXE, the mean concentration of toluene was followed by benzene, m, p-xylene, o-xylene and ethylbenzene successively in three seasons for all the sites. Similar abundance pattern of BTXE species have also been reported by Hoque et al 2008. Mean concentration of all the VOCs were found to be maximum at Malsi and showed minimum concentration levels at Doon University in all specified seasons. At Malsi, sampling was carried out at petrol pump and hence, vehicular emissions, refuelling, degreasing activities and evaporative emissions would have enhanced the observed VOC levels. Similar sources for aromatic VOCs have been reported by Kountouriotis et al 2014; Majumdar et al 2008 at petrol pumps. On the other hand, Doon University being a residential cum institutional area is quite far off from the main city activities. The activities here are basically concerned with domestic, agriculture and a bit of traffic for the purpose of transportation. The mode of transportation also varied here as people mostly used scootys, bikes and cars. The influence of heavy duty vehicles is found to be very less here. VOCs get diluted through atmospheric processes (during transportation to this site) therefore; people residing at some distance from main streets would tend to experience reduced exposure of the concerned VOCs. Toluene was found to vary at Malsi as 118.38 ± 39.32 μg/m^3, 193.83 ± 54.68 μg/m^3 and 134.65 ± 36.30 μg/m^3 in summer, winter and monsoon, respectively. A maximum value of 260 μg/m^3 is recommended by WHO for toluene averaged over a week as a limiting level from the human health perspective (Parra et al 2009; Mohan and Ethirajan 2012). Seasonal mean concentration of toluene has been found to have comparable values at both ISBT and Ghantaghar.

ISBT, Ghantaghar and Panditwari experienced the following sequence of mean concentrations of VOCs: winter > monsoon > summer for benzene and ethylbenzene hydrocarbons. However, Malsi followed the order being highest in winters and lowest during summers for BTXE species. Similar results have also been observed by Yoshikado 1994 in Kanto Plain, Hoque et al 2008 in Delhi; Hoshi et al 2008 in Tokyo; McNabola et al 2008 in Dublin, Ireland; Mohan and Ethirajan 2012 in Chennai, India. Throughout the year, Dehradun city, situated in a valley, experiences frequent inversions which are quite prevalent in winters most of the time (Rana et al 2009). Dissipation of VOCs in inversion conditions is difficult due to atmospheric stability. Therefore, inversion conditions during winters could be the possible reason for high-observed VOCs level. Borbon et al (2002) concluded that NMHC distribution in winters is dominated by motor vehicle exhaust while in summers; emissions
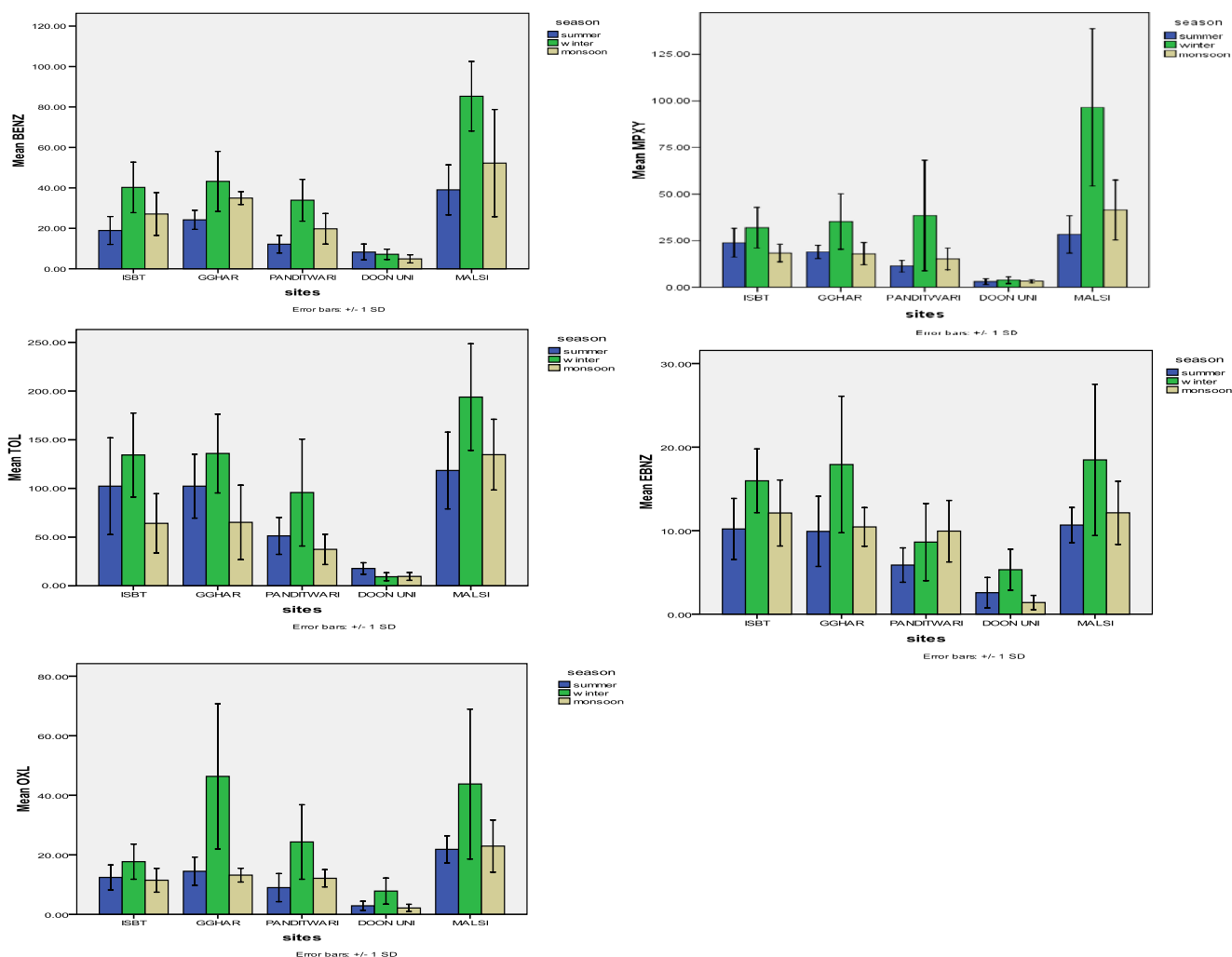
Fig 3 Seasonal variations of BTXE ($\mu g/m^3$) in urban air of Dehradun

come from evaporative mechanism from fuel and solvents. Vehicular emissions comprise direct emissions from exhaust along with other contributing components which consist of hot soak emissions during engine cooling (Na et al. 2004; Rubin et al 2006; Langford et al 2010), evaporative emissions comprising solvent usage and gasoline evaporation during spillage and filling of fuel tanks (Srivastava et al 2005c; Langford et al 2010), emissions from brakes and tyre wear (Caselli et al 2010). Moreover, in summers, the rate of photochemical reactions of hydrocarbons with OH radical's increases which accounts for less concentrations being observed (Na et al 2005; Mohan and Ethirajan 2012). Relatively higher OH radical strength is observed in summer season (Hakola et al 2003; Hoque et al 2008). In monsoons, benzene levels are observed to be comparatively

lower than other respective seasons as pollutant levels (BTXE) tend to get diluted because of heavy rains. It can be further supported as in Dehradun that the average monthly relative humidity was observed to be 80.95 % and average monthly rainfall 7.20 mm for monsoon season. However, the average relative humidity (monthly) during summers and winters was observed to be 39.33 % and 76.51 %, respectively. The average rainfall (monthly) was recorded to be varied as 0.05 and 6.80 mm in summers and winters, respectively. This phenomenon in monsoons has also been observed by Lee et al 2002 in Hong Kong; Srivastava et al 2006 in Mumbai; Ojiodu et al 2013 in Southwestern Nigeria. Similar trends for VOCs' seasonal variations have also been observed in Ahmdabad, Firozabad, Delhi, Agra and Mumbai by Sahu and Lal 2006; Chaudhary and Kumar 2012; Singh et al. 2012a, 2012b; Singla et al 2012 and Pandit et al 2011 in India. Similar trends for seasonal variations being highest in winters and lowest during summers have also been observed in several other studies carried out in different parts of the world, e.g. by Lee et al 2002 (Hong Kong); Mohamed et al 2002 (United States); Zalel and Yuval 2008

Table 2 Kruskal–Wallis statistics for Dehradun

VOCs	Benzene	Toluene	m, p-Xylene	o-Xylene	Ethylbenzene
p value	0.002	0.017	0.004	0.001	0.011

Table 3 Intercity variations in reported BTXE levels ($\mu g/m^3$)

City under study	Benzene	Toluene	Xylenes	Ethylbenzene	Period of study
Dehradun[a]	29.76	83.54	42.87	9.93	2012–2013
Delhi[b,1]	48–110	85–204	45–131	7–27	October 2001–September 2002
Delhi[c]	9.07–20.36	20.55–57.61	10.80–16.77	–	November 2010–February 2011
Mumbai[d]	45.31–539.95	29.16–303.27	0.21–1.66	0.20–3.01	May 2001–April 2002
Kolkata[e]	13.8–72.0	21.0–83.2	29.9–78.5	7.6–21.6	November 2003–February 2005
Carmen[f] (Mexico)	5.42	11.23	8.32	3.97	June 2012–December 2012
Michigan[g] (USA)	1.13	2.61	2.61	0.52	2004–2005
Hong Kong[h] (China)	2.75–15.11	4.58–137.15	4.13–33.08	2.20–13.70	September 1997–September 1999
Egypt[i]					
Ramsis	87.20	213.80	214.57	43.40	June 2004–August 2004
Haram	46.23	111.80	110.85	22.77	June 2004–August 2004
Kaohsiung[j] (Taiwan)					
Rush hours	22.38	212.39	49.11	41.84	July 2003–October 2003
Non-rush hours	5.25	45.55	11.80	10.73	July 2003–October 2003
Algeria[k]	9.6	15.2	3.2	0.9	November 1996, November–December 2007, January–March 1998, May–August 2008

Xylenes represent sum of ortho, meta and para isomers of xylene

[a] (this study)

[b] (Hoque et al 2008)

[c] (Singh et al 2012b)

[d] (Srivastava et al 2006)

[e] (Majumdar et al 2011)

[f] (Cerón et al 2013)

[g] (Jia et al 2008)

[h] (Lee et al 2002)

[i] (Khoder 2007)

[j] (Liu et al 2008)

[k] (Kerbachi et al 2006)

[1] Represents passive sampling

Table 4 Interspecies ratios among different monitoring sites in ambient air of Dehradun

Sites	Statistics	T/B	m, p-X/B	o-X/B	EBENZ/B
ISBT	Mean±SD	3.88±1.88	1.04±0.58	0.58±0.32	0.53±0.33
Ghantaghar	Mean±SD	3.26±1.42	0.73±0.31	0.67±0.37	0.38±0.16
Panditwari	Mean±SD	3.29±1.84	1.09±1.00	0.79±0.57	0.46±0.26
Doon University	Mean±SD	2.02±1.02	0.57±0.28	0.65±0.48	0.46±0.34
Malsi	Mean±SD	2.90±1.42	0.97±0.48	0.54±0.28	0.26±0.12

(Ahuja, Israel); Rad et al 2014 (Ahvaz, Iran). The seasonal mean concentrations followed the trend being highest in winters for BTXE species. Minimum ambient concentration levels were observed during summers for benzene and ethylbenzene species, while for toluene, least concentrations were recorded in monsoons. Toluene is also used as solvents in coatings, paints, adhesives and cleaning agents in repair workshops (Alghamdi et al 2014). Hence, during summers, due to high vapour pressure, toluene emissions would tend to arise from evaporation from solvents along with vehicular emissions. m, p-Xylene and o-xylene rather showed a mixed pattern being lowest in summers for Panditwari, Doon University and Malsi only. At ISBT and Ghantaghar xylenes (m,p-xylene and o-xylene) showed near about similar fluctuations in summer and monsoon season.

The mean ambient concentration of benzene at almost all sites is a matter of serious concern as it exceeded the limits set by CPCB in its revised NAAQS (2009) which is 5 μgm^{-3} (annual) for industrial, residential as well as for ecologically sensitive area. Moreover, it is also found to be present beyond the levels of air quality standards given by WHO (5–20 μgm^{-3}, annual average) and European Union (5 μgm^{-3}, annual average). Benzene is very well known for its genotoxicity and carcinogenicity (International Agency for Research on Cancer, IARC 1995; Massolo et al 2009) and is believed to mainly come from emissions from cars (Al Madhoun et al. 2011). In a study carried out by Villeneuve et al 2013 in Toronto, association between ambient VOC concentrations and cancer mortality were observed.

Results of Kruskal–Wallis test (nonparametric one-way ANOVA) was carried out on BTXE data sets and it ascertained that the differences in the mean concentrations of VOCs in summer, monsoon and winter were statistically significant ($p < 0.05$). It is one-way analysis of variance (ANOVA) by ranks which is used to test whether samples originate from same distribution or not. For Kruskal–Wallis test, the null hypothesis is assumed that there is no significant difference in the VOCs concentration in Dehradun. Alternative hypothesis states that there is significant statistical difference among the VOCs. The result of test in Table 2 shows that significant statistical differences ($p < 0.05$) were obtained among the concentrations of all measured VOCs in Dehradun. Therefore, the null hypothesis is rejected and the alternative hypothesis is accepted. Post hoc test was then carried out to ascertain whether there is significant difference in means of any two seasons. Based on the results, it

can be concluded that the sources of aromatic VOCs might have originated from different sources seasonally.

Intercity comparison of aromatic species against observed levels in Dehradun

A comparison of mean concentrations of monoaromatic hydrocarbon viz. BTXE in Dehradun with other cities of the world is presented in Table 3. It may be inferred that benzene, toluene and xylene mean concentrations are comparatively found to be on the lower side than those reported in some parts of India like in Delhi, Mumbai and Kolkata. Whereas ethylbenzene mean concentration in Dehradun was found to be three times higher than that observed in Mumbai but comparatively low as found in Kolkata and Delhi. Rather, a common trend was observed as far as the abundance of BTXE hydrocarbons is concerned: toluene > xylenes > benzene > ethylbenzene for the mentioned reported research work in Table 3 except for Mumbai. In Mumbai, Srivastava et al 2006 estimated the abundance sequence as: benzene > toluene > ethylbenzene > xylenes. A different pattern was also observed by Liu et al (2008) in Taiwan during non-rush hours as benzene as the lowest abundant species among the BTXE hydrocarbons.

BTXE concentrations in our study are found to be slightly higher than those reported by researchers in other countries, e.g. Kerbachi et al 2006 in Algeria; Jia et al 2008 in Michigan; Cerón et al 2013 in Mexico. However, Khoder 2007 in Greater Cairo (Ramsis and Haram) reported higher mean concentrations than our respective study. Liu et al 2008 carried out their study in

Table 5 MIR coefficients and mean seasonal concentration of BTXE hydrocarbons

VOCs	MIR coefficient[a]	Mean VOC ($\mu g/m^3$)		
		Summer	Winter	Monsoon
Benzene	0.42	19.77	42.22	27.29
Toluene	2.70	76.28	113.08	61.27
Ethylbenzene	2.70	7.68	13.18	8.92
m, p-Xylene	8.20	16.48	41.78	18.85
o-Xylene	6.50	11.65	27.85	12.03

[a] (source: Carter 1994)

Taiwan and concluded that concentrations during rush hours are found to be four to five times higher than non-rush hours. The respective concentrations of toluene, ethylbenzene and xylenes in our study are found to be lower than those reported by Liu et al 2008. Mean concentration of benzene in our study is found to be higher than non-rush hours but are comparable to rush hours in Taiwan by Liu et al 2008.

Interspecies ratios of BTXE species in Dehradun

Interspecies ratios among BTXE have been widely used as indicator to identify emission sources (Kerbachi et al 2006; Guo et al 2007; Khoder 2007). Each VOC has its own distinct photochemical reactivity towards OH radicals. The abundance of highly reactive species usually decrease during daylight as they tend to undergo photochemical reactions, whereas the concentration of relatively less reactive species increases due to accumulation (Liu et al 2008). Benzene and toluene have been found to be relatively more stable than xylenes having lifetime of 12.5 days, 2.0 days and 7.8 h, respectively (Prinn et al 1987; Liu et al 2008). Xylenes (m-xylene) with comparably shorter lifetime do not remain much longer in the atmosphere (Prinn et al 1987; Liu et al 2008).

The mean interspecies ratios among BTXE are provided in Table 4. Toluene to benzene (T/B) ratio is commonly used to indicate traffic-related emissions (Hoque et al 2008) in urban areas. Benzene and toluene are constituents of gasoline and are released into the atmosphere by motor vehicle exhaust (Cerón et al 2013). Generally T/B ratio approaching 1.0 refers traffic originated emission sources while this ratio would tend to increase as the proximity to pollution sources reaches (Gelencsér et al 1997). Highest T/B ratio observed was at ISBT (3.88) whereas relatively lower but comparable values at Ghantaghar (3.26) and Panditwari (3.29) suggesting a similar source, i.e. vehicular emissions. Doon University and

Malsi experienced lowest values of T/B with values 2.02 and 2.90, respectively in comparison to other sites. ISBT, Ghantaghar and Panditwari showed almost similar values for all ratios inferring that the nature of source of BTXE could be similar. ISBT is a traffic influenced area where buses, trucks, bikes, scootys and cars from in and around the Dehradun moving on the roads would justify the high-observed ratio. Ghantaghar is a commercially hit area, experiences high traffic throughout the day from the adjoining Rajpur road, Chakrata road, Gandhi road and Darshanlal chowk. Several painting, auto repair, degreasing, dry cleaning, photo film developers and other commercially viable shops are present nearby which might use solvents; therefore, these would also enhance the toluene emissions. Toluene and m, p-xylene may be released from both solvent usage as well as vehicular emissions (Na et al 2003). On the contrary, Doon University campus is an area which is far from intense traffic. Here, T/B ratio (2.02) comparably low than other sites could come from the use of solvents (toluene) in nearby area being used in auto repair shops, cleaning shops along with vehicle either parked or moving inside the campus could attempt to justify the observed toluene to benzene ratio.

T/B ratios found in the present study were similar to those reported in other cities, e.g. Delhi (1.80–2.54), Rome (2.80), Izmir (1.87–2), Santiago (2.01), Ramsis (2.45), La Plata (2.8) and Carmen (2.45–2.8) (Brocco et al 1997; Hartmann et al 1997; Muezzinoglu et al 2001; Khoder 2007; Hoque et al 2008; Massolo et al 2009; Cerón et al 2013). On the other hand, our observed ratios are lower than those estimated in Bangkok (10.22), Hong Kong (7.74), Osaka (7.19), Japan (4.1–6.5) and Sydney (4.04) (Tsujino and Kuwata 1993; Gee and Sollars 1998; Lee et al 2002; Tiwari et al 2010), but found to be higher than Agra (0.74) (Singla et al 2012). One could try to infer a difference in type of vehicle, fuel, their composition, local and industrial activities from the varying magnitude of T/B ratios.

Table 6 Comparison of ozone formation potential of Dehradun with other reported studies

| Sites | Ozone formation potential (OFP) | | | | |
	Benzene	Toluene	m, p-Xylene	o-Xylene	Ethylbenzene
Dehradun[a]					
Summer	8.30	205.96	121.95	75.72	21.43
Winter	17.73	305.30	309.20	181.00	36.77
Monsoon	11.46	165.43	139.47	78.19	24.89
Delhi[b]	37	452	550	226	47
Porto Alegre[c]	9	100	295	–	54
Yokohama[d]	6.7	20.1	9.1	2.9	12.6
Beijing[e]	3.52	30.80	71.58	23.89	13.87
Nigeria[f]	1.54–1.63	7.88–7.99	25.7–36.81	3.87–10.71	6.48–11.02

Superscript denotes the work carried out by researchers in theirrespective cities

Where [a] (this study), [b] (Hoque et al 2008), [c] (Grosjean et al 1998), [d] (Tiwari et al 2010), [e] (Duan et al 2008) and [f] (Olumayede and Okuo 2013)

The mean values of m, p-xylene/benzene and o-xylene/benzene and ethylbenzene/benzene were found to be relatively less which suggests that there is more benzene concentration and less observed ambient concentration levels of more reactive species like xylenes and ethylbenzene. It also implies that xylenes and ethylbenzene might have been involved in photochemical reactions with hydroxyl radicals (Hoque et al 2008). In addition, it could also be inferred that xylene could have been transported from the surrounding area to the sampled site. Therefore, small X/B ratios suggested that photochemical reactions were active and aged air mass could have been transported to the area under consideration. X/B ratio (0.28–1.09) in the present study is found to be comparable to Hoque et al 2008 (0.34–0.87) in Delhi.

Ozone-forming potential estimation for BTXE species in Dehradun

It is usually calculated by the product of maximum incremental reactivity coefficient (MIR) with their mean concentration of respective VOC (Carter 1994). The MIR coefficients and mean VOCs concentration are provided in Table 5. In this study, xylenes (m, p-xylene plus o-xylene) were found to contribute greatly towards ozone formation potential as compared to others among BTXE hydrocarbons except for summers. Summer followed the trend having toluene as the most important VOC in terms of OFP then the rest. Similar trend for toluene in terms of OFP has also been observed by (So and Wang 2004) in Hong Kong. While in winter, a different pattern was rather observed which follows the sequence m, p-xylene > o-xylene > toluene > ethylbenzene > benzene. Our winter results are truly in agreement with those reported by Grosjean et al 1998 at Porto Alegre; Hoque et al 2008 in Delhi; Tiwari et al 2010 in Yokohama. Benzene showed lowest OFP among BTXE in all seasons despite being regarded as the most hazardous species in C_6-C_9 aromatic VOCs (Na et al 2005) and toxic in nature as it affects the central nervous system in humans and considered as group 1 human carcinogen (International Agency for Research on Cancer and IARC 2004; Han and Naeher 2006). In Seoul, both toluene and xylene were together accounted for 94 % of the total OFP and are usually emitted through evaporative activities in atmosphere by solvents usage and gasoline spillage in summers (Na et al 2005). Hoque et al 2008 and Na et al 2005 found that xylenes contribute majorly to the ozone formation on the basis of MIR scale among BTEX group of VOCs.

Table 6 shows the ozone formation potential (OFP) of BTXE species measured in Dehradun in comparison with other cities. Our observed concentrations are lower than those observed by Hoque et al 2008 in Delhi but greater than the data of Grosjean et al 1998 at Porto Alegre; Duan et al 2008 in Beijing, China; Tiwari et al 2010 in Yokohama, Japan; Olumayede and Okuo 2013 in Nigeria. In general, based on MIR scale contribution of VOCs to ozone formation potential, the trend follows as: xylenes (m, p-xylene and o-xylene) > toluene > ethylbenzene > benzene except in Yokohama.

Conclusion

Monoaromatic VOCs viz. BTXE were sampled at five sites in ambient urban atmosphere of Dehradun. Toluene was estimated as the most abundant species among BTXE in Dehradun. For benzene and ethylbenzene ISBT, Ghantaghar and Panditwari experienced the highest mean concentrations of VOCs in winters followed by monsoon and summer. BTXE hydrocarbons recorded highest mean concentration in winters. Dehradun being lying in a valley, the condition here is conducive for the formation inversion during most of the period of the year. This could be the reason for high-observed ambient concentration levels during winters. Toluene registered a summer minima. Highest T/B ratio observed was at ISBT (3.88), whereas relatively lower but comparable values at Ghantaghar (3.26) and Panditwari (3.29) suggesting vehicular emissions could be the dominant source along with others prevalent in ambient air levels of Dehradun. The small X/B ratios suggested that photochemical reactions were active and aged air mass could have been transported to the area. Xylenes (m, p-xylene and o-xylene) and toluene were found to contribute greatly towards ozone formation assumed on the basis of ozone formation potential. Particularly, xylenes (sum of all the isomers of xylene) showed maximum OFP during winters and monsoons. While in summers, OFP was found to be dominated by toluene.

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