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Volatile organic compounds over Eastern Himalaya, India: temporal variation and source characterization using Positive Matrix Factorization

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Abstract

A first ever study on the characterization of volatile organic compounds (VOCs) has been made over a Himalayan high altitude station in India. A total of 18 VOCs (mono aromatics-BTEX (benzene, toluene, ethylbenzene, xylene), non-BTEX substituted aromatics and halocarbon) have been measured over Darjeeling (27.01° N, 88.15° E, 2200 m a.s.l.) in the eastern Himalaya in India during the period of July 2011–June 2012. The annual average concentration of the sum of 18 target VOCs (TVOC) was $376.3 \pm 857.2\ \mu g\ m^{-3}$. Monoaromatics had the highest contribution (72 %) followed by other substituted aromatics (22 %) and halocarbon (6 %) compounds. Toluene was the most abundant VOC in the atmosphere of Darjeeling with the contribution of ~37 % to TVOC followed by benzene (~21 %), ethylbenzene (~9 %) and xylenes (~6 %). TVOC concentrations were highest during the postmonsoon season with minimum solar radiation and lowest during the premonsoon season with maximum solar radiation. Anthropogenic activities related mainly to tourists like diesel and gasoline emissions, biomass and coal burning, use of solvent and solid waste emissions were almost equal in both the seasons. Seasonal variation in TVOCs over Darjeeling was mainly governed by the incoming solar radiation rather than the emission sources. Source apportionment study using Positive Matrix Factorization (PMF) model indicated that major fraction of (~60 %) TVOC were contributed by diesel and gasoline exhausts followed by solvent evaporation (18 %) and other sources. Diesel exhaust was also found to have the maximum potential in tropospheric ozone formation. The atmospheric loading of BTEX over Darjeeling was found to be comparable with several Indian metro cities and much higher than other cities around the world.

1 Introduction

The studies on volatile organic compounds (VOCs) have gained much attention because of their ability in modifying oxidizing capacity of the atmosphere as well as

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health implications to humans. VOCs play an important role in the formation of photochemical smog and tropospheric ozone by reacting with hydroxyl radicals (OH) in the presence of NO_x (Atkinson, 2000). They also have the potential towards stratospheric ozone depletion and enhancement of the global greenhouse effect (Guo et al., 2004a). VOCs comprise a wide range of compounds including aliphatic and aromatic hydrocarbons, alcohols, aldehydes, ketones, esters, and halogenated compounds. Many VOCs react with hydroxyl radicals (OH) and/or nitrate (NO_3) radicals to form secondary organic aerosol (SOA) by nucleation and condensation with a significant aerosol yield and thus they influence gas phase pollutants directly and particle-phase pollutants indirectly (Atkinson, 2000).

There is as such no general source for VOCs as there are numerous compounds in this group, which can be emitted from very different sources (Yurdakul et al., 2013). In addition to the biogenic sources of VOCs (Williams and Koppmann, 2007), some well documented anthropogenic sources are gasoline powered and diesel powered motor vehicles (Demir et al., 2011), fuel storage (Lanz et al., 2008), biomass burning (Yokelson et al., 2008), natural gas (Latella et al., 2005), LPG (Lai et al., 2005), industrial processes and solvents (Lanz et al., 2008) etc.

High levels of VOCs have been observed in Asian countries and these have been considered to be originating from vehicular emissions (Srivastava et al., 2005a). Among the Asian countries, India is the second largest contributor to the emission of non-methane VOCs (Kurokawa et al., 2013). In spite of growing population and associated increase in vehicular and industrial activities, the studies on VOCs in India are limited. Some of those important studies have been conducted in the recent past mostly in metro cities such as in Delhi, the capital city of India (Hoque et al., 2008; Khillare et al., 2008; Srivastava, 2005; Srivastava and Singh, 2005; Srivastava et al., 2005b, c; Gurjar et al., 2004; Padhay and Varshney, 2000), in Mumbai, a metro city and financial capital of India situated in western India (Srivastava and Som, 2007; Srivastava et al., 2004a, 2006a, b; Srivastava, 2004b), in Kolkata, a metro city in eastern India (Dutta et al., 2009; Mujumdar et al., 2008; Som et al., 2007; Mukherjee et al., 2003), in Hyderabad,

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a metro city in southern India (Rekhadevi et al., 2010), in Agra in northern India, (Singla et al., 2012), in Firozabad in northern India (Chaudhury and Kumar, 2012) etc. In India there is no legislation of VOC as a whole except national ambient air quality standard for Benzene by Central Pollution Control Board of India. Globally US Occupational Safety and Health Administration (OSHA) and World Health Organization (WHO) have proposed some guidelines and recommendations for VOCs and not compulsory for governments to follow (Han and Naeher, 2006).

Where almost all the studies were conducted over several cities in India, no such study on VOCs have been ever made over Himalaya in India. Such studies over Himalayan region are of paramount interest as the ecology of the Himalaya is under serious threat from various forms of pollutants (Bostrom, 2002). The increase in the loading of atmospheric pollutants over the Himalaya is a matter of concern, since most of the glaciers in the region have been retreating since 1850 (Mayewski et al., 1979) with increasing melting rates. The rising anthropogenic interferences for rapid urbanization and development in the Himalaya not only affect the immediate landscape environment, but also the atmospheric environment which is becoming an increasing concern (Momin et al., 1999). The anthropogenic activities such as increasing vehicular traffic due to increased tourism-related activities, biomass burning and fuel wood burning for cooking and heating are the causes of concern for most of the Himalayan high altitude hill stations in India which apparently look like pollution-free regions as situated far away from the Indian mega-cities.

The present study on the characterization of VOCs has been made over a high altitude (2200 m a.s.l.) hill station, Darjeeling (27.01° N, 88.15° E) at eastern Himalaya and the first ever study conducted over Indian Himalaya to the best of our knowledge. Our earlier studies (Chatterjee et al., 2010, 2012; Adak et al., 2014; Sarkar et al., 2014) over the same region showed high aerosol loading during premonsoon (March–May) due to vehicular emissions related to tourist activities and during winter (December–February) due to massive biomass burning. In addition to the local sources, pollutants were also found to be accumulated over this region transported from long distant regions like

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Indo–Gangetic Plain (IGP) and other Asian sub continents. Sarkar et al. (2014) found enhancement of Black Carbon aerosols over Darjeeling during postmonsoon (October–November) due to transported plumes of biomass burning from northern India. The seasonal variation of aerosols associated to the variation in emission sources (local and transported) as observed from earlier studies have prompted us to make a year-long study on VOCs over Darjeeling as major aerosol sources over this region are generally the major sources of VOCs too.

The present study is thus mainly focused on (1) the identification of the major factors governing seasonal variation of VOCs, (2) contribution of long distant source regions, (3) source apportionment of VOCs using Positive Matrix Factorization (PMF) and their potential in tropospheric ozone formation.

2 Study site and synoptic meteorology

The study has been carried out at a high altitude hill station Darjeeling (27°01′ N, 88°15′ E, 2200 m a.s.l.) at eastern Himalaya in India. The map showing geographical location of the measurement site and adjacent regions in Darjeeling has been given in detail in our earlier study (Adak et al., 2014).

The seasonal average along with minimum and maximum of surface meteorological parameters; temperature (T) in °C, wind speed (WS) in $m\,s^{-1}$, relative humidity (RH) in % and rainfall (mm) are given in Fig. 1. The entire study period is divided into four seasons; winter (December–February), premonsoon (March–May), monsoon (June–September) and postmonsoon (October–November). Figure 1 shows that the temperature was highest during monsoon and lowest in winter whereas relative humidity shows monsoon maximum and premonsoon minimum. Wind speed was found to be maximum in premonsoon which was ∼ 2 times than that in other seasons. We did not observe much variation between daytime and nighttime wind speed except in premonsoon when daytime wind speed was much higher (∼ 1.8 times) than night-time wind speed. The surface reaching solar radiation was maximum during premonsoon

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and was much higher than postmonsoon, monsoon and winter. The total amount of rainfall over the entire sampling days was 421.4 mm. However, ∼ 95 % rain occurred during monsoon (397 mm) only.

3 Methodology

3.1 Sampling and analysis of VOCs

The study was carried out in the campus of National Facility on Astroparticle Physics and Space Science, Bose Institute, Darjeeling. Samples were collected on a roof top of the building of Bose Institute at a height of about 20 m from the ground level at day (7 a.m. to 7 p.m.) and night (7 p.m. to 7 a.m.) basis for a year long period from 7 July 2011 to 25 June 2012. The samples were collected once a week. A total of 90 samples were collected during the study period, using a custom made glass sampling tube containing charcoal and chromosorb. The tubes were pre-conditioned by heating over night at 200 °C temperature. The tubes were connected with a low flow air suction pump (SKC, USA). The flow rate was maintained at ∼ 100 mL min^{-1}. The flow was measured before and after each sampling event using a flow meter. After sampling the ends of the tubes were sealed well with the Teflon tape and cap and kept at 4 °C for analysis.

The analysis was done by thermal desorption followed by detection on GC-MS in accordance with USEPA TO-17 compendium method for the determination of target VOCs and described in details in the authors' previous publications (CPCB, 2007, 2010; Srivastava and Som, 2007; Majumdar et al., 2014). In short the thermal desorption of sorbent tube was done by heating at 180 °C for 25 min. 100 µL of desorbed gas was injected into Varian Make GC-MS (Now Agilent; GC-MS model: (Model 450GC-240MS)). Target VOCs were separated using DB 624 capillary column of 30 m length and 0.32 mm internal diameter. Helium gas with flow rate of 1 mL min^{-1} was used as carrier gas with split ratio 1 : 20, GC oven was programmed for 35 °C hold for 4 min

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and ramped to 210 °C. For estimation of the target compounds external five point calibration curve was prepared in triplicate using VOC mix 20 by Dr. Ehrenstorfer GmbH, Germany.

4 Results and discussion

4.1 General characteristics of VOCs over Darjeeling

All the 18 VOCs measured in this study denoted as TVOC (Total VOCs) have been classified in to three groups; mono-aromatics BTEX, non-BTEX substituted aromatics (iso-propylbenzene, *n*-propylbenzene, 1,3,5 trimethylbenzene, 1,2,4 trimethylbenzene, sec-butylbenzene, 4-isopropyltoluene, 2-chlorotoluene, 1,4 dichlorobenzene, *n*-butylbenzene, naphthalene), and halocarbons (1,1 dichloroethane, 1,2 dichloroethane, chloroform and carbon tetra chloride). The annual average concentrations of each VOC for each group along with their minimum and maximum concentrations over the entire period of study have been given in Table 1. The concentration of TVOC over Darjeeling was found to vary widely from as low as 6.6 $\mu g\,m^{-3}$ to as high as 4707.5 $\mu g\,m^{-3}$ over the entire period of study (July 2011–June 2012). The annual average concentration of TVOC was $376.3 \pm 857.2\,\mu g\,m^{-3}$. BTEX was found to have the highest contribution (72 %) followed by non-BTEX substituted aromatics (22 %) and halocarbon (6 %) compounds. BTEX varied over a wide range between 1.5 and 3975.6 $\mu g\,m^{-3}$ with an average of $275.1 \pm 685.7\,\mu g\,m^{-3}$. Toluene was found to be the most abundant VOC in the atmosphere of Darjeeling with the contribution of ~ 37 % to TVOC followed by benzene (~ 21 %), ethylbenzene (~ 9 %) and xylenes (~ 6 %). The concentration of non-BTEX aromatics, too, varied widely from a very low (0.3 $\mu g\,m^{-3}$) to a very high (912.1 $\mu g\,m^{-3}$) value with an average of $88.6 \pm 220.1\,\mu g\,m^{-3}$. On the other hand, halocarbon compounds, unlike other VOCs, did not show such large variability during the study period. The concentration of halocarbons varied from 1.5 to 73.3 $\mu g\,m^{-3}$ with an average of $21.5 \pm 15.4\,\mu g\,m^{-3}$. TVOC and most of its components showed their mini-

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mum concentrations during premonsoon (15 May 2012) and maximum concentrations during postmonsoon (21 November 2012).

4.2 Factors affecting seasonal variations of VOCs

Figure 2 shows the seasonal variations of VOCs over Darjeeling. The concentration of TVOC was maximum in postmonsoon ($1649.9 \pm 875.4\,\mu g\,m^{-3}$) followed by monsoon ($117.1 \pm 88.3\,\mu g\,m^{-3}$), winter ($60.4 \pm 28.2\,\mu g\,m^{-3}$) and minimum during premonsoon ($35.9 \pm 9.7\,\mu g\,m^{-3}$) BTEX and non-BTEX substituted aromatics showed similar seasonal patterns. The high postmonsoon concentrations were found to be $1228.2 \pm 534.1\,\mu g\,m^{-3}$ and $404.0 \pm 336.1\,\mu g\,m^{-3}$ and the low premonsoon concentrations were found to be $12.9 \pm 3.3\,\mu g\,m^{-3}$ and $3.5 \pm 1.5\,\mu g\,m^{-3}$ for BTEX and non-BTEX substituted aromatics respectively. Unlike BTEX and non-BTEX, halocarbons showed highest abundance in winter ($33.5 \pm 10.4\,\mu g\,m^{-3}$) with small variabilities between premonsoon ($19.1 \pm 4.2\,\mu g\,m^{-3}$), postmonsoon ($17.6 \pm 4.5\,\mu g\,m^{-3}$) and monsoon ($14.5 \pm 5.2\,\mu g\,m^{-3}$).

Postmonsoon and premonsoon are the tourist seasons over Darjeeling. Darjeeling experiences huge emissions of fossil fuel burning from large numbers of tourist vehicles during these two seasons compared to other seasons. We had made rough measurements on vehicle counts and consumption of fossil fuel over Darjeeling earlier in the year of 2005 (Adak et al., 2010). We observed that the number of light and medium duty vehicles was 6000–6700 day^{-1} during premonsoon and postmonsoon whereas 3000–3600 day^{-1} during winter and monsoon. The total consumption of fossil fuel (petrol and diesel) was 6500–7500 $L\,day^{-1}$ during premonsoon and postmonsoon whereas it was 3500–4500 $L\,day^{-1}$ during winter and monsoon. In addition to the vehicular emissions, various other anthropogenic activities get increased in premonsoon and postmonsoon seasons. The tourist activities remained almost same in these two seasons but VOCs showed significant variations with high level in postmonsoon and low level in premonsoon. The other factors related to the sinks of VOCs played major roles dominating the emission sources of VOCs, leading to the significant variation between postmonsoon and premonsoon. Observed seasonal trends can thus be addressed by

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the characteristics of the prevailing meteorology, and most importantly, the availability of solar insolation in these two seasons. Darjeeling recorded maximum solar insolation in premonsoon ($360 \pm 140\,W\,m^{-2}$; Fig. 1) which could help in the photolysis of ozone, carbonyls, water vapour etc leading to the formation of OH radicals in the atmosphere (Ho et al., 2004). This plays key role in atmospheric clean-up and degradation of VOCs during premonsoon. Another important meteorological factor is wind speed which was observed to be maximum during premonsoon months ($1.4 \pm 0.5\,m\,s^{-1}$; Fig. 1). This could favour the ventilation and dispersion of VOCs from the study site. On the other hand, the solar insolation ($220 \pm 100\,W\,m^{-2}$) and wind speed ($0.65 \pm 0.2\,m\,s^{-1}$) during postmonsoon were much lower than premonsoon. Thus, although the VOC emissions remained comparable, VOC degradation was maximum in premonsoon than postmonsoon leading to premonsoon low and postmonsoon high VOC concentrations. In addition to the local emissions, transported carbonaceous compounds could also contribute significantly in enhancing carbonaceous compounds over eastern part of Himalaya during postmonsoon. Bonasoni et al. (2010), Marinoni et al. (2010), Dumka et al. (2010) and Kaskaoutis et al. (2014) have shown the influence of carbonaceous compounds (mainly Black Carbon) over Himalayas due to transported plumes associated to crop residue burning over Punjab and adjacent Indo Gangetic Plain regions during postmonsoon seasons. Our recent study (Sarkar et al., 2014) showed the impact of this transported biomass burning plumes on Black Carbon aerosols over Darjeeling in the same study period. These biomass burning plumes could also bring significant amount of VOCs over Darjeeling enhancing their concentrations during postmonsoon.

The tourist activities remained low both during monsoon and winter months over Darjeeling. The solar insolation during monsoon and winter was comparable in magnitude ($180 \pm 80\,W\,m^{-2}$). Darjeeling recorded maximum temperature during monsoon ($15.8 \pm 0.9\,^{\circ}C$) which may lead to increased evaporative emissions for certain VOC species with higher vapour pressure from vehicular service stations, and also from waste decomposition in the hotter months (Talapatra and Srivastava, 2011). VOC emis-

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sion from solvent evaporation is less significant at very low temperature (5.3 ± 2.1 °C) during winter.

In general, variation in VOC concentrations between hotter and colder months over plain land cities is addressed with the help of vertical advection through boundary layer dynamics in addition to other meteorological factors. The low VOC concentration during summer is generally associated to favourable vertical mixing due to high boundary layer/mixing height whereas comparatively higher VOC concentration during winter is associated to calm and stable atmospheric condition with low boundary layer/mixing height restricting vertical dissipation. The boundary layer dynamics has been used for addressing seasonal variation of VOCs for most of the studies conducted over several Indian cities (Talapatra and Srivastava, 2011 and several references therein). But the case of Darjeeling is unique, unlike plain land cities, the seasonal variation in VOC concentration could not be addressed through boundary layer dynamics as the station itself is situated at a height of 2.2 km, well above the boundary layer. But there is a probability that boundary layer could reach the altitude of Darjeeling during premonsoon under high convective activities. Thus, VOCs emitted from plain land regions could reach Darjeeling after their vertical advection and could contribute and enhance VOC concentrations over Darjeeling. But, photochemical degradation under high solar insolation over Darjeeling could have hindered the development in VOC concentrations during premonsoon.

4.3 Day and night time VOCs: role of anthropogenic and meteorological factors

VOC concentrations over Darjeeling were compared between day and night time for different seasons in order to investigate the potential impact of the variability in emission sources and/or meteorological factors between day and night time. The night to day ratio was greater than 1.0 in each season. We infer that although the emissions were high, the photochemical degradation could decrease the day-time VOC concentrations. Thus, night-time VOCs could be attributed to the VOCs generated during night (which could not degrade by photolysis) plus residual VOCs generated during day-time.

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Another important factor is higher wind speed during daytime which could favour the dispersion of VOCs more than night. The ratio for TVOC was highest during premonsoon (1.9) followed by postmonsoon (1.4), monsoon (1.2) and minimum during winter (1.1). The highest ratio in premonsoon could be due to the removal of VOCs by efficient and faster photo-degradation by very high solar insolation favoured by much higher dispersion due to higher wind speed during day time in premonsoon leading to high level of VOCs compared to the other seasons. However, the minimum value of the ratio in winter could be due to massive biomass burning during winter nights which could enhance night-time VOC concentrations.

4.4 Contribution of long distant source regions to VOCs over Darjeeling

The transport of air masses from distant sources could affect the pollutant concentrations at the study site in conjunction with the local sources. In order to investigate the transport of VOCs from long distances, we have computed 36 h air-mass back trajectories, arriving at an altitude of 500 m a.g.l. over Darjeeling for all the days on which VOCs were measured, using Hybrid Single Particle Lagrangian Integrated Trajectory (HYSPLIT) model (http://www.arl.noaa.gov/ready/hysplit4.html). Over the entire period of study, we have identified three major source regions for long range transport as shown in Fig. 3. The frequency of transport from each of the source regions has also been shown in the figure. Region 1 corresponds to the transport from SE directions and the air masses originated from southern part of West Bengal, India and Bangladesh with the frequency of 32 %. The average TVOC concentration associated to Region 1 was found to be $117.2 \pm 86.1\,\mu g\,m^{-3}$. Region 2 corresponds to the transport from W/NW directions and the air masses originated from eastern and central part of Nepal with the frequency of 42 %. The associated TVOC concentration was found to be $831.5 \pm 955.2\,\mu g\,m^{-3}$. Region 3 corresponds to local/regional sources and the air masses originated mainly from the E/SE directions with the frequency of 26 %. The major regions were northern part of West Bengal and the average TVOC concentration was found to be $620.1 \pm 535.4\,\mu g\,m^{-3}$. Thus, the contri-

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bution from Nepal (Region 2) was found to be ~7 and 1.5 times higher than West Bengal/Bangladesh (Region 1) and local/regional sources (Region 3) respectively. The contributions from each source regions were also investigated for different seasons. The average TVOC concentrations associated to respective source regions along with their frequencies have been given in Table 2 season-wise. It was observed that during monsoon, all the air masses originated from Region 1 with 100 % frequency with the average TVOC concentration of $117.2 \pm 86.1\ \mu g\,m^{-3}$. Similarly, during winter, 100 % air masses originated from Nepal with the TVOC concentration of $60.9 \pm 28.0\ \mu g\,m^{-3}$. During postmonsoon, 60 % air masses originated from local/regional sources (Region 3) and 40 % originated from Nepal (Region 2) with the average TVOC concentrations of $1206.8 \pm 628.3\ \mu g\,m^{-3}$ and $2978.1 \pm 1538.1\ \mu g\,m^{-3}$ respectively. It was observed that 50 % air masses originated from local/regional and 50 % originated from Nepal during premonsoon and the TVOC concentration associated to Nepal was found to be slightly higher ($46.2 \pm 12.5\ \mu g\,m^{-3}$) than local/regional sources ($34.2 \pm 11.3\ \mu g\,m^{-3}$). This result indicates that the air masses coming from Nepal carried more VOCs and thus more polluted compared to other source regions. It is important to mention over here that the altitudes of the air masses were below 1000 m a.s.l. throughout their trajectories/pathways originating from their source regions. Thus the air masses could pick up the boundary layer pollutants of the regions they passed over before reaching our observational site.

As Nepal was found to be most polluted source regions, an attempt was made to roughly estimate the contribution of TVOC from Nepal in postmonsoon and premonsoon seasons. During these two seasons, air masses originated both from Nepal and local/regional source regions and thus contribution from Nepal was estimated in terms of the relative concentrations associated to these two regions. The estimation has been made by the following equation:

% contribution from Nepal = $(EC_{Nepal}/MC_{Total}) \times 100 = ((MC_{Total} - MC_{Local})/MC_{Total}) \times 100$, where EC_{Nepal} is the estimated concentrations of TVOC coming only from Nepal i.e. additional amount of TVOC coming from Nepal. MC_{Total} is the measured concentra-

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tion of TVOC on respective days when air masses generated from Nepal i.e. with the total contribution of both Nepal and local air masses. MC_{Local} is the average measured concentration of TVOC on all the days when air masses originated from local sources i.e. contribution from local sources only. It was observed that VOCs from Nepal contributed to the TVOC concentration over Darjeeling by 38–54 % with the average of ∼ 53 % during postmonsoon and 32–65 % with the average of ∼ 50 % during premonsoon.

4.5 Effect of local and long distant sources on the variability–lifetime relationship for VOCs

The relationship between the variability in concentrations and the life time of VOCs can be used to estimate the distance of their source regions regardless the influence of the regional transport. The following empirical equation was first proposed by Jobson et al. (1998).

$$S_{\ln x} = A\tau^{-b}$$

Where $S_{\ln x}$ is the SD of the natural logarithm of the concentration X of VOC, τ is the atmospheric lifetime of VOC, A and b are the fit parameters.

The value of exponent b lies between 0 and 1 and describes the influence of the source contribution. The value of b will approach zero when sampling site is closed to a source and the variability–lifetime relation will be "weaker". In the extreme case, when $b = 0$, the variability will not depend on the atmospheric lifetime but will depend on the variability of the emission sources. In remote areas b will approach 1 (Jobson et al., 1998; Ehhalt et al., 1998; Wang et al., 2005) where the distance of sampling site is longer from the potential sources. The variability concept is based on the assumption that the chosen compounds have more or less the same source distribution. The compounds reported in this paper, are mostly of anthropogenic origin (aromatic hydrocarbons and halocarbons). We have used their concentrations for premonsoon and postmonsoon as the sources of VOCs are same in both the seasons. We have

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calculated the back trajectory analysis for each sampling day and based on the trajectories we have separated the transported air masses from the local emissions (as discussed above). Figure 4 shows the relationship of variability with lifetime for different VOC species separately for long range transport and local emissions. It can be seen that the value of b was higher for long range transport ($b = 0.19$, $R^2 = 0.79$) than for local emission ($b = 0.09$, $R^2 = 0.64$). The value of b for long distant sources (0.19) as obtained in the present study was found to be slightly lower than 0.22 as observed over Mount Tai, China (Ting et al., 2009) and 0.23 as obtained in the Mediterranean Intensive Oxidant Study (MINOS) in August 2001 on Crete (Gros et al., 2003). But, it was much lower than 0.44 as observed over the remote NARE locations (Jobson et al., 1999) and 0.41 on a cruise through the western Indian Ocean during the INDOEX field study (Karl et al., 2001). Thus, Darjeeling does not represent a remote site where the variability is strongly dependent on the lifetime of VOC but represent a typical urban site in the vicinity of sources where the sources dictate the variability and not the chemistry. The longest source regions (Central part of Nepal or southern part of West Bengal/Bangladesh) for VOCs over Darjeeling as estimated from HYSPLIT trajectory models were within 200 km from Darjeeling.

4.6 Characterization of sources of VOCs by Positive Matrix Factorization receptor model

In recent years, an advanced receptor model, Positive Matrix Factorization (PMF), has been applied extensively in identifying VOC contributing sources at different locations in the world (e.g., Jorquera and Rappengluck, 2004; Latella et al., 2005; Xie and Berkowitz, 2006; Brown et al., 2007; Song et al., 2007; Yuan et al., 2009). PMF does not require any priori knowledge on the exact VOC emission profiles, and it can be used to apportion source contributions solely based on observations at the receptor site, thus avoiding VOC decay adjustment problem. More details about the PMF method were described by several studies (Paatero and Tapper, 1994; Paatero, 1997; Reff et al., 2007).

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In this study, the PMF method has been applied to identify the possible sources of VOCs over Darjeeling. Table 3 shows the source profiles derived by the PMF model. Eight factors were selected according to the resulted stable *Q* values. Figure 5 shows the percentage contribution of each VOC associated to each of eight sources.

Table 3 shows that Factor 1 is dominated by high values of BTEX with much higher concentrations of benzene and toluene followed by ethylbenzene and xylene. Toluene to benzene ratio was found to be 0.9 in this factor. Thus factor 1 could be associated to the gasoline-related emissions. VOC emissions from gasoline may occur along many pathways like, evaporative emission from gas stations and bulk terminals and exhaust released from the gasoline-powered vehicles during gasoline combustion (Watson et al., 2001; Choi and Ehrman, 2004). BTEX are the major components of vehicular exhaust, as shown by many studies (Watson et al., 2001; Guo et al., 2006, 2007; Som et al., 2007). High VOC emissions from tourist vehicles during premonsoon and postmonsoon seasons and the gasoline vapours from the frequent use of the gas stations are the most important contributors to this source over Darjeeling.

Factor 2 is also dominated by BTEX. Toluene was found to have the maximum contribution followed by benzene, ethylbenzene and xylene. The toluene to benzene ratio is 2.5 in this factor. Previous study of the authors (Som et al., 2007) reported the same ratio in a study made over Kolkata, India for the VOCs emitted from diesel-driven vehicles. This factor is associated to diesel exhaust. It is interesting to observe that the number of petrol and diesel driven vehicles are nearly same over Darjeeling and PMF result indicates the percentage contribution of TVOCs from Diesel and gasoline sources are also comparable (discussed later in details).

Factor 3 is characterized by the high values of TEX. TEX being the primary constituents of solvents (Guo et al., 2004b; Choi et al., 2011), often used as a solvent in paints, coatings, synthetic fragrances, adhesives, inks and cleaning agents, in addition to its use in fossil fuel (Borbon et al., 2002; Chan et al., 2006). This factor can therefore be assigned to the solvent usage and related emission. The rapid growth in tourism

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related infrastructure like hotels, resorts, restaurants etc over Darjeeling could be the reason for high VOC emission from solvent usage.

Factor 4 is characterized by the high values of *n*-propyl benzene, 2-chloro toluene and BTEX and could be assigned to solid waste disposal. Majumdar et al. (2014) reported the high values of these compounds in municipal waste dumping stations in Kolkata, India. With the dramatic increase in tourists and changing consumption patterns, Darjeeling is facing immense problems of waste management. The existing systems of waste management are technically unscientific and the infrastructure is insufficient to manage the waste.

Factor 5 is dominated mainly by chloroform and carbon tetrachloride and thus the factor could be associated to chlorine bleach containing house hold products. Odabasi et al. (2008) showed that house hold cleaning agents and fresheners produce these two VOCs significantly. Chloroform and carbon tetrachloride are the major compounds along with several halogenated compounds in chlorinated bleach products.

Factor 6 is dominated by m-xylene and ethylbenzene followed by *n*-butylbenzene and toluene and could be assigned to industrial sources (Yuan et al., 2010). Although there is no industry in Darjeeling, but the VOCs could be transported from low land townships and cities. The m-xylene to ethylbenzene ratio in this factor was found to be 1.8. The ratio of m,p-xylene to ethylbenzene (X/E ratio) is used as indicator for the age of the VOCs in the atmosphere (Elbir et al., 2007; Guo et al., 2004b, c). The ratio becomes smaller as the VOCs get older in the atmosphere, because m,p-xylene is more reactive than ethylbenzene. Kuntasal (2005) found X/E ratio to be varied between 3.8–4.4 in fresh emissions at various environments. The low ratio in this study (1.8) suggests that the species were not emitted in situ but aged/transported.

Factor 7 is dominated by chloroalkanes, benzene and toluene and could be associated to coal and biomass burning (Fernandez-Martinez et al., 2001; Barletta et al., 2009). Coal burning is a significant anthropogenic source in Darjeeling as it is used for the domestic cooking purpose and it is also used in a large scale for coal engines in the toy trains. In addition to that, massive biomass burning during winter to get warmth

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against cold and probable transportation of biomass burning species from northern Indian states (as discussed earlier) during postmonsoon could enhance those VOCs in the atmosphere of Darjeeling.

Factor 8 is characterized by high values of aromatics with high molecular weight like 1,2,3-tri methyl benzene, 1,2,4-trimethyl benzene, o-xylene. Liu et al. (2005) reported high emissions of these VOCs from asphalt related road construction works. Road construction works were in progress in and around Darjeeling during few sampling events. Thus the factor 8 could be assigned to the asphalt related emission.

Figure 6 shows the percentage contributions of each source to the total VOC loading over Darjeeling during the entire study period. It can be seen that the major sources are diesel exhaust (32 %) and gasoline exhaust (29 %) followed by solvent evaporation (18 %). Chlorine bleach containing house hold products and solid wastes contributed equally (6 %) whereas industrial sources situated at the regions far from Darjeeling, coal/biomass burning and asphalt related constructional works contributed nominally by 4, 3 and 2 % respectively. Thus it can be concluded that the major source of VOCs over Darjeeling is gasoline and diesel driven vehicular activities which contributed by more than 60 %.

4.7 Ozone formation potential of VOC sources

In order to investigate the potential of various VOC sources (as derived from PMF model) to the tropospheric ozone formation over the study area we have computed the ozone formation potential (OFP) of each source using the Maximum Incremental Reactivity (MIR) values derived by Carter (2008). To do this, we have used the equation derived by Na and Kim, (2007).

$$\mathrm{OFP}_i = S_i \times \sum_{j=1}^{n} (\alpha_{ji} \times \mathrm{MIR}_j)$$

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Where, OFP_i is the estimated contribution of ith source to OFP, S_i is the total mass contribution of the source i, α_{ji} is the mass fraction of species j in source i and MIR_j is MIR value of species j. Figure 7 shows the relative contribution of each source to OFP. It can be seen from the figure that diesel exhaust has the maximum potential (45 %) followed by solvent (24 %) and gasoline exhaust (18 %). Although, Gasoline exhaust contributes more (29 %) towards TVOC concentration than solvent usage (18 %), the later source is contributing more towards tropospheric ozone generation. The MIR values of the individual species are also responsible for the total OFP of a source along with the corresponding source strength. Thus, vehicular emissions and solvents could play the key role in the formation of tropospheric ozone and have the potentials to modify the tropospheric ozone budget.

4.8 Comparison with other studies

The concentration of BTEX over Darjeeling (present study) has been compared with that over several metro cities in India and also with other cities in Asian, European, African, Arabian and American countries (Table 4). We have taken the sum of BTEX (not TVOC) for comparison as the data of BTEX is more available in the literature.

Table 4 shows that BTEX over Darjeeling is lower than the commercial, industrial and the areas with high traffic density (traffic intersection) over Delhi, the capital city of India; traffic intersection and petrol pumps over Mumbai, a metro city in western India and Hyderabad, a metro city in south-eastern India. This is quite expected as the vehicular and industrial activities over those metro cities are much higher than Darjeeling. But the most interesting fact is that BTEX over Darjeeling shows ~ 3, ~ 2 and ~ 5 times higher concentrations than residential areas over Kolkata (a mega city in eastern India), Delhi and Mumbai respectively. Even, Darjeeling shows higher BTEX concentrations than commercial areas of Mumbai and much higher than roadside (~ 10 times) and petrol pump (~ 7 times) areas in Agra, a city in northern India with much less vehicular activities compared to other Indian metro cities.

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BTEX over Darjeeling was found to be much higher (10–25 times) than the residential/industrial/commercial areas of Turkey, Houston, Rome and Paris; 2–6 times higher than residential/commercial areas of Bangkok, Yokohama, Kuwait and Hongkong; 1.5–2 times higher than roadside/industrial/commercial areas of Kaohsiung, Sanghai and Beijing. Darjeeling shows much higher (~ 8 times) BTEX concentration than Gongga Mountain, a high altitude (1640 m a.s.l.) remote station in southwestern China. However, BTEX over a commercial area with heavy traffic density in Cairo, Egypt shows 1.7 times higher concentration than that over Darjeeling.

In our earlier study (Sarkar et al., 2014), we also reported much higher concentration of black carbon aerosols over Darjeeling compared to other high altitude Himalayan stations in India and Nepal and some of the metro cities in India like Ahmedabad, Bangalore, Trivandrum and Chandigarh. The present study corroborate with that findings. The major source for black carbon aerosol and VOCs over Darjeeling is same, vehicular emissions. Thus, Darjeeling represents a typical urban atmosphere at eastern Himalaya with high loading of carbonaceous pollutants. This could be due to high anthropogenic emissions related to tourist activities, high population density and moreover it's unique orography and land use pattern with narrow roads, unplanned township, poor administrative control on solid waste disposal and burning of these wastes, unplanned constructions of buildings/hotels/resorts which reducing open space/area which in turn prevents ventilation and dispersion of pollutants.

5 Conclusion

The major findings of the study on VOCs conducted over Darjeeling, a high altitude hill station over eastern Himalaya in India are as follows:

1. The annual average concentrations of TVOC, BTEX, non-BTEX aromatics and halocarbons were 376.3±857.2, 275.1±685.7, 88.6±220.1 and 21.5±15.4 $\mu g\,m^{-3}$ respectively with the maximum contribution from BTEX (72 %), non-BTEX aromat-

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ics (22 %) and halocarbons (6 %). Toluene was found to be the most abundant VOC over Darjeeling which contributed 37 % to the TVOC.

2. Concentration of TVOC showed well defined seasonal variations with maximum in postmonsoon ($1649.9 \pm 875.4\ \mu g\,m^{-3}$) followed by monsoon ($117.1 \pm 88.3\ \mu g\,m^{-3}$), winter ($60.4 \pm 28.2\ \mu g\,m^{-3}$) and minimum during premonsoon ($35.9 \pm 9.7\ \mu g\,m^{-3}$). The seasonal variation in VOC concentration was mainly governed by the photochemical degradation process rather than the emission source strength. Although, the anthropogenic activities related to massive tourist influxes during premonsoon and postmonsoon were comparable, the solar radiation made the difference between premonsoon and postmonsoon VOC concentrations.

3. Other than local sources, two major regions were identified for VOCs over Darjeeling; Nepal and southern part of West Bengal, India/Bangladesh. It was observed that VOC concentration over Darjeeling was higher when air masses arrived from Nepal than West Bengal, India/Bangladesh and local/regional source regions. The relationship between variability and lifetime of VOC was discussed and it was observed that Darjeeling represents the site in the vicinity of sources as compared with other studies.

4. Positive matrix facorization receptor model was used to characterize the sources of VOCs over Darjeeling. It was observed that the major source of VOC over Darjeeling was emission from petrol and diesel driven vehicles which contributed by more than 60 % followed by solvent evaporation (18 %) and other sources.

5. Diesel exhaust was found to have the maximum potential (45 %) in the formation of tropospheric ozone followed by solvent evaporation (24 %) and gasoline exhaust (18 %).

6. The atmospheric loading of BTEX over Darjeeling was comparable with Indian metro cities and much higher than other Asian, American, African, Arabian and European countries.

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Thus we found that Darjeeling represents a typical urban atmosphere over eastern Himalaya in India from the point of view of VOC pollution. The high VOC pollution over Darjeeling draws a serious attention as it could significantly affect human health as well as the sensitive ecosystem over this part of Indian Himalaya. Study result emphasis the need for better pollution control system for the vehicles plying on the road of Darjeeling. Imposing regulations on uncontrolled solvent usage is also necessary. Better Solid waste management system is also called for. This year long data set of VOC can be used to make further studies on the modification of the budget of tropospheric ozone, NO_x and other gaseous and particulate pollutants. This would, in turn, help us to make studies on the implications of VOCs for regional atmospheric chemistry over eastern Himalaya.

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Table 1. Statistical summary of the concentration of each VOC component over the entire period of study (all the concentrations are in $\mu g\,m^{-3}$).

	Mean	SD	Max	Min
1,1-Dichloroethane	2.80	4.58	19.75	BDL
1,2-Dichloroethane	0.44	0.79	6.82	BDL
Chloroform	17.89	17.03	86.81	1.62
Carbon Tetrachloride	0.18	0.27	1.65	BDL
Benzene	81.19	212.16	1166.20	2.01
Toluene	140.67	430.03	2304.38	2.67
Ethylbenzene	32.69	93.35	563.50	1.06
m-Xylene	19.93	38.59	216.68	0.91
o-Xylene	0.87	1.47	7.60	BDL
Isopropylbenzene	12.07	44.88	267.06	0.66
n-Propylbenzene	4.82	9.66	48.48	BDL
2-Chlorotoluene	5.79	13.33	75.42	BDL
1,3,5-Trimethylbenzene	24.56	99.44	647.09	0.87
1,2,4-Trimethylbenzene	3.21	6.14	45.16	BDL
sec-Butylbenzene	3.72	12.20	104.61	0.05
4-Isopropyltoluene	28.45	125.05	752.62	0.72
1,4-Dichlorobenzene	0.38	0.98	6.87	BDL
n-Butylbenzene	3.85	7.86	58.23	0.04
Naphthalene	1.12	3.84	36.46	BDL

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Table 2. TVOC concentrations for various source regions over different seasons and entire study period.

Period	Regions	Direction	Source region	Frequency	[TVOC] $\mu g\,m^{-3}$
Annual	Region 1	S/SE	Bangladesh and West Bengal	32	117.2 ± 86.1
	Region 2	W/NW	Nepal	42	831.5 ± 955.2
	Region 3	E/SE	Local/Regional	26	620.1 ± 535.4
Monsoon	Region 1	S/SE	Bangladesh and West Bengal	100	117.3 ± 86.5
Postmonsoon	Region 3	E/SE	Local/Regional	60	1206.8 ± 628.3
	Region 2	W/NW	Nepal	40	2978.1 ± 1538.1
Winter	Region 2	W/NW	Nepal	100	60.9 ± 28.0
Premonsoon	Region 2	W/NW	Nepal	50	46.2 ± 12.5
	Region 3	E/SE	Local/Regional	50	34.1 ± 11.3

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Table 3. Source profiles of several factors estimated from PMF model.

	Gasoline Exhaust	Diesel Exhaust	Solvent, Paint	Solid Waste Disposal	Chlorine Bleach Products	Industrial Source	Coal Burning	Asphalt Related Emission
	Factor 1	Factor 2	Factor 3	Factor 4	Factor 5	Factor 6	Factor 7	Factor 8
1,1-Dichloroethane	0.00	0.01	0.00	0.03	0.16	0.00	2.14	0.00
1,2-Dichloroethane	0.01	0.04	0.01	0.00	0.01	0.01	0.16	0.03
Chloroform	0.69	0.21	0.00	0.45	13.03	0.55	2.62	0.26
Carbon Tetrachloride	0.00	0.00	0.00	0.00	0.08	0.00	0.06	0.00
Benzene	43.35	22.49	0.00	2.43	1.90	0.00	2.49	0.00
Toluene	37.82	55.35	18.36	2.70	1.23	0.99	1.59	1.25
Ethylbenzene	3.15	15.09	0.36	3.09	0.00	2.97	0.19	0.24
m-Xylene	2.29	5.15	1.50	3.36	0.13	5.43	0.00	0.00
o-Xylene	0.00	0.00	0.00	0.00	0.02	0.14	0.00	0.48
Isopropylbenzene	0.42	0.50	8.71	0.37	0.34	0.10	0.00	0.18
n-Propylbenzene	0.00	0.41	0.09	3.09	0.06	0.25	0.07	0.26
2-Chlorotoluene	0.41	0.00	0.00	3.89	0.07	0.09	0.11	0.46
1,3,5-Trimethylbenzene	0.00	0.11	0.14	0.01	0.00	0.00	0.09	0.34
1,2,4-Trimethylbenzene	0.00	1.15	0.00	0.00	0.03	0.02	0.21	1.41
sec-Butylbenzene	0.00	0.00	1.17	0.03	0.00	0.06	0.36	0.00
4-Isopropyltoluene	0.00	0.00	26.18	0.75	1.01	0.32	0.00	0.53
1,4-Dichlorobenzene	0.13	0.01	0.02	0.00	0.01	0.00	0.06	0.01
n-Butylbenzene	0.95	0.00	0.00	0.00	0.00	1.99	0.00	0.00
Naphthalene	0.14	0.00	0.03	0.00	0.01	0.00	0.06	0.17

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Table 4. Comparison of BTEX concentration over Darjeeling with other cities in India and other countries

Location	Nature of Site	Sum of BTEX ($\mu g\,m^{-3}$)	Study Period	Reference
Darjeeling	High altitude tourist station	331.0	Jun 2011–Jul 2012	Present Study
Indian Metro Cities				
Delhi	Residential area	186.0	Oct 2001–Sep 2002	Hoquea et al. (2008)
	Commercial area	421.0		
	Industrial area	411.0		
	Traffic intersection	456.0		
Kolkata	Commercial-cum-residential area	132.5	Dec 2003–Feb 2005	Majumdar et al. (2011)
Mumbai	Residential	75.8	May 2001–Apr 2002	Srivastava et al. (2006)
	Commercial	256.6		
	Industrial	281.7		
	Traffic intersection	655.6		
	Petrol pump	587.6		
Hyderabad	Road side	370.2	NA	Rekhadevi et al. (2010)
	petrol pump	2978.8		
Agra	Roadside	30.0	Apr 2010–Mar 2011	Singla et al. (2011)
	Petrol pump	47.1		
Other cities in Asian, European, African, Arabian and American countries				
Beijing, China	Road Side, High traffic density	173.7	Aug 2005	Song et al. (2007)
Gongga Mountain, China	High altitude remote station	40.3	Jan 2008–Dec 2011	Zhang et al. (2013)
Hong Kong, China	Residential area	91.7	Sep–Nov 2010	Lam et al. (2013)
Sanghai, China	Commercial	191.7	Jan 2007–Mar 2010	Cai et al. (2010)
Yokohama, Japan	Residential-cum-commercial-cum-industrial	115.9	Jun 2007–Nov 2008	Tiwari et al. (2010)
Ulsan, Korea	Residential	23.8	Mar 2010–Feb 2011	Lee et al. (2012)
Kaohsiung, Taiwan	High traffic density	202.8	Jul and Oct 2003	Liu et al. (2008)
Bangkok, Thailand	Commercial	61.6	Jan–Dec 2009	Ongwandee et al. (2011)
Paris, France	Residential-cum-Industrial-	17.9	Jan–Feb 2010	Ait-Helal et al. (2014)
Rome	High traffic density	15.9	Dec 2010–Dec 2011	Fanizza et al. (2014)
Cairo, Egypt	Commercial	558.9	Jun–Aug 2004	Khoder et al. (2007)
Ankara, Turkey	Residential area	13.5	Jan–Jun 2008	Yurdakul et al. (2013)
Kuwait, UAE	Residential-cum-commercial	127	Aug 2010–Nov 2011	Al Khulaifi et al. (2014)
Houston, USA	Highly industrialized	14.7	Aug–Sep 2006	Leuchner et al. (2010)

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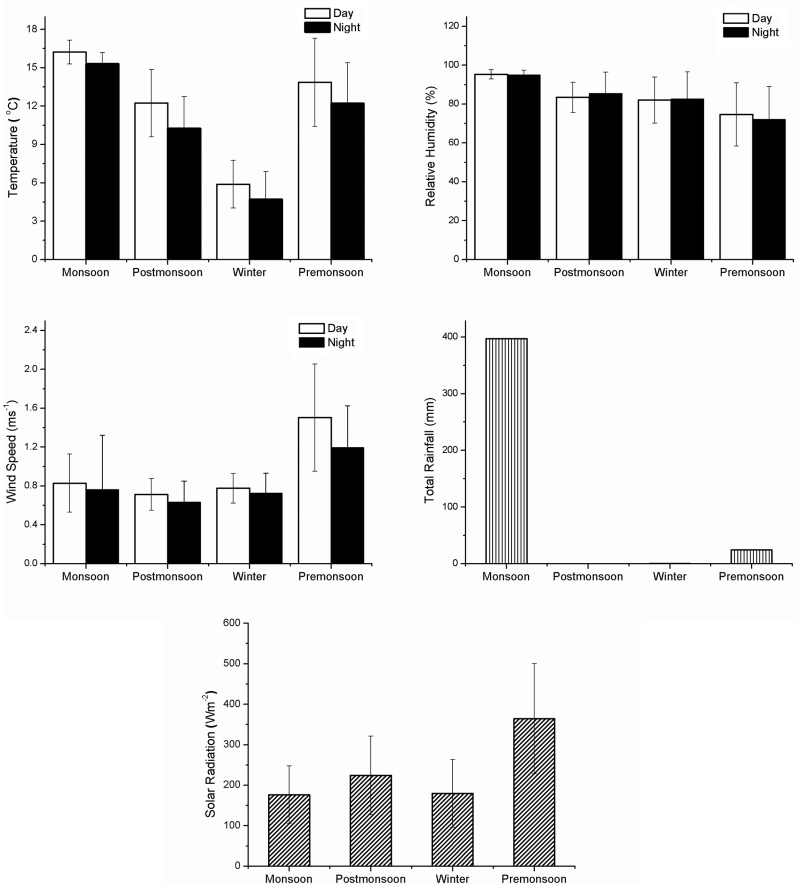

Figure 1. Seasonal variation of micro-meteorological parameters over Darjeeling during the study period.

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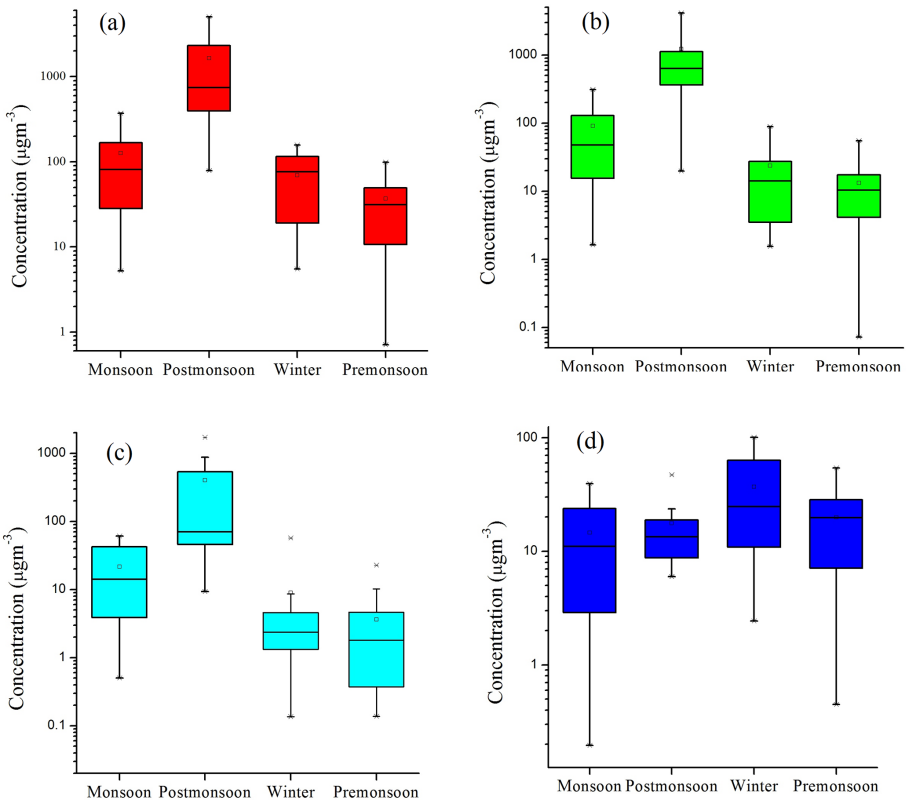

Figure 2. Seasonal variation of **(a)** TVOC, **(b)** mono aromatics-BTEX, **(c)** non-BTEX substituted aromatics and **(d)** halocarbons shown in box-whisker plot. The lower boundary of the box, the horizontal line inside the box and upper boundary of the box represent 25th percentile, median and 75th percentile respectively. The whiskers below and above represent minimum and maximum respectively.

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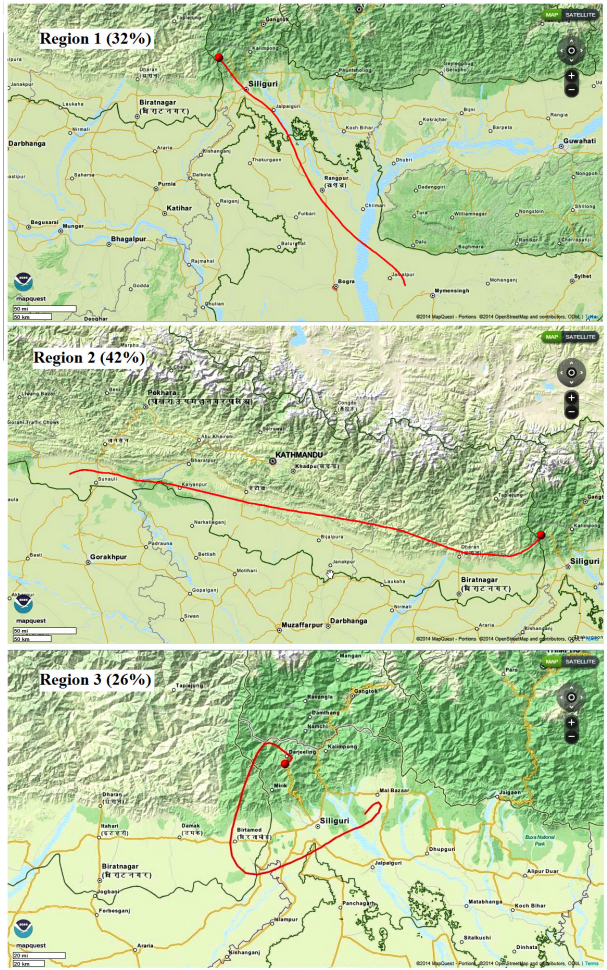

Figure 3. Source regions of VOCs obtained from air mass trajectories of HYSPLIT model.

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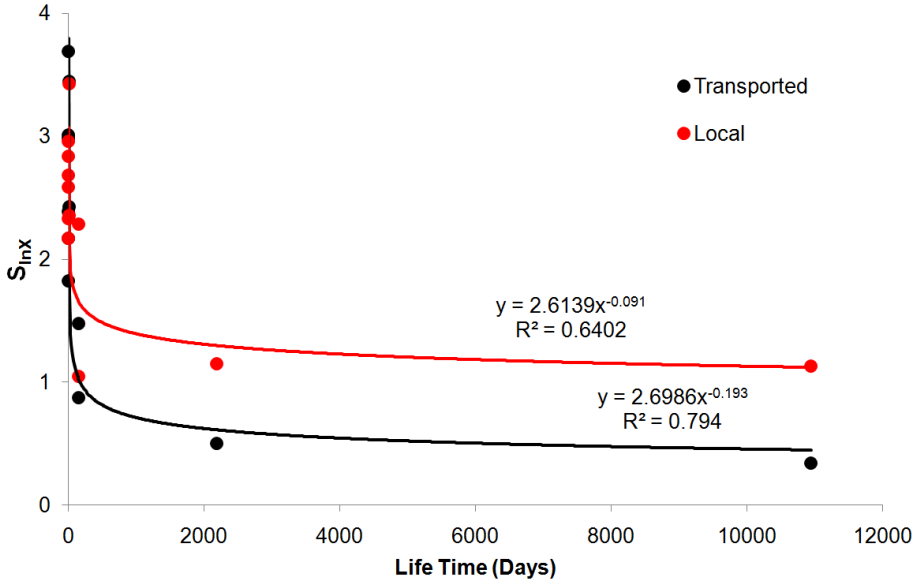

Figure 4. Variability–lifetime relationship of different VOCs for local/regional and long distant source regions.

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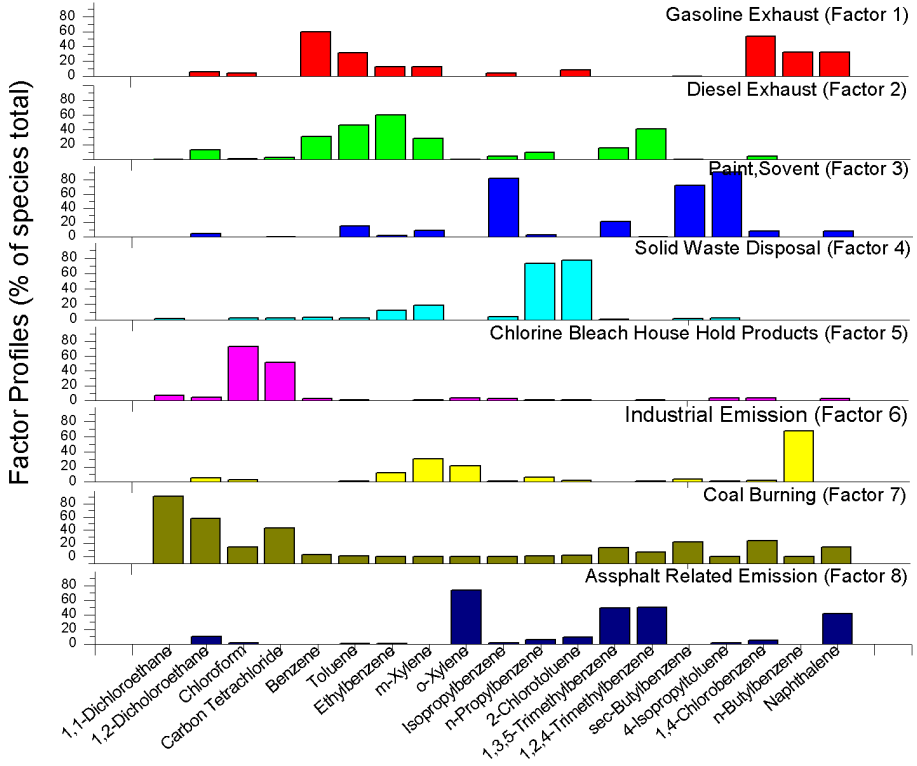

Figure 5. VOC source profiles estimated from PMF model.

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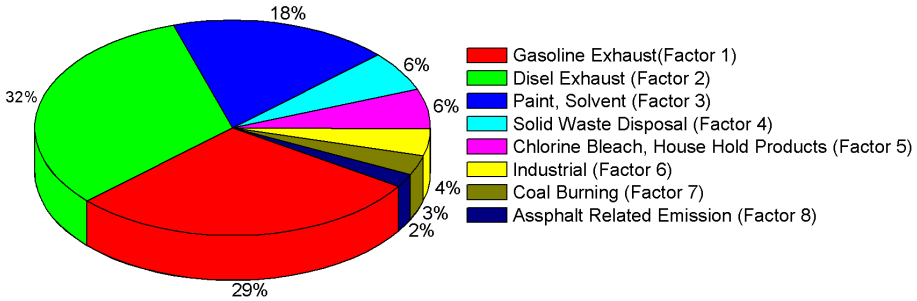

Figure 6. Percentage contribution of various sources for VOCs over Darjeeling.

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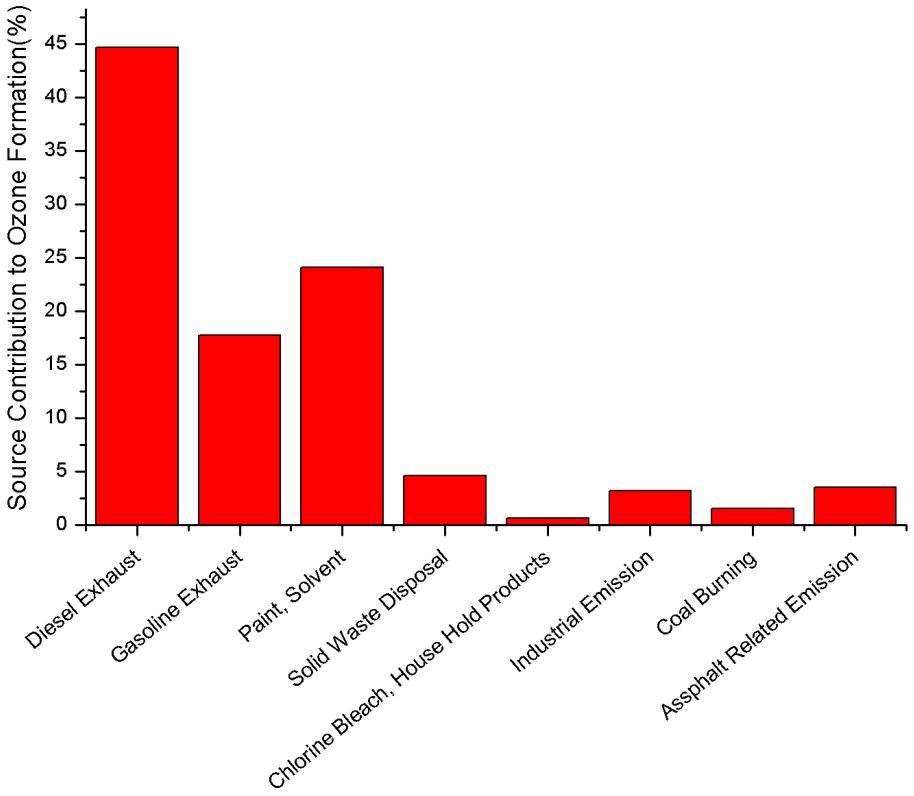

Figure 7. Ozone formation potential of each source of VOCs.

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