



Research article

Emissions of non-methane volatile organic compounds from a landfill site in a major city of India: impact on local air quality

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ARTICLE INFO

Keywords:

NMVOCs
Landfill
India
TD-GC-FID
Urban
Pollution
Air quality
Environmental chemistry
Environmental hazard
Environmental impact assessment
Environmental pollution
Fossil fuel
Environmental science

ABSTRACT

Emissions from landfills are a significant source of non-methane volatile organic compounds (NMVOCs) in urban environments. NMVOCs play an important role in atmospheric chemistry, and elevated concentrations of some compounds are responsible for air quality deterioration. This study is based on the measurements of a suite of 20 C₂–C₈ NMVOCs at 21 upwind and downwind sites of the largest landfill in western India. Ethane, ethylene and aromatics were the dominant compounds; the concentrations of BTEX in the downwind regions were up to three times higher than their concentrations at upwind sites. The emission ratios of BTEX and other NMVOCs were different from those for residential, commercial, and industrial sources characterizing the emissions from burning and decomposition of organic material. The slope of Δ Toluene/ Δ Benzene of 0.64 is about three times higher than that determined at the main road junctions of the city. Ranking by Prop-Equiv, the top NMVOCs were isoprene, cis-2-Butene, m + p-xylenes, propylene, ethylene and trans-2-Butene account for 72–75% of the total Prop-Equiv concentrations. Alkenes played the dominant role in ozone formation, followed by aromatic and alkane groups. In addition to landfill emissions, contributions from traffic-related emissions to ambient concentrations of aromatic VOCs were also significant at some sites. Although the experiment was not designed to characterize the emissions from a specific source, the analysis suggests the substantial contributions from both decomposition and burning of landfill materials. The main difficulty in characterizing VOC emissions from landfills is the spatial and temporal variability of emissions from a large area.

1. Introduction

Poor air quality is a serious problem in many cities of the world due to the large emissions of both gaseous and particulate pollutants from a variety of anthropogenic activities (Carmichael et al., 2009; Baudic et al., 2016; Wang et al., 2017; Dias and Tchepel, 2018; Shah et al., 2019). The fast urbanization, industrial-economic growth, transportation rate, and use of fossil fuels have resulted in the increasing concentrations of key air pollutants in the South Asia region (Sahu et al., 2016a, 2016b, 2020a; Srivastava, 2005; Yadav et al., 2019, 2017). Volatile organic compounds (VOCs) are important components of urban air pollution. On a global scale, VOCs are precursors for the formation of tropospheric ozone (O₃). Ozone is produced from the oxidation of VOCs

by hydroxyl (OH) radicals followed by a chain of reactions in the presence of NO_x (=NO + NO₂) and sunlight (Liu et al., 2008; Monks, 2005). Oxidation of some VOCs with high photochemical reactivity contributes to formation of secondary organic aerosols (SOA) (Seinfeld and Pandis, 2016). The higher concentrations of O₃ in the lower atmosphere are known to have adverse effects on human health, terrestrial plants, and outdoor materials (Jerrett et al., 2009; Ainsworth et al., 2012; Lelieveld et al., 2015; Yadav et al., 2016). Aromatic VOCs such as benzene, toluene, and 1–3 butadienes are potentially toxic and exposure to elevated concentrations can be harmful to human health (Na et al., 2003; Fang et al., 2019). The International Agency for Research on Cancer (IARC) and the US Department of Health and Human Services (DHHS) have determined that benzene is carcinogenic to humans (e.g.,

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Received 18 February 2020; Received in revised form 10 April 2020; Accepted 20 July 2020

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Band, 2012). Therefore, monitoring based studies of VOCs in urban air are essential to frame the regulation policies.

In major cities, aromatic VOCs are released mainly from the combustion of fossil fuels in gasoline and diesel engines, gasoline evaporation, solvent usage and bio-decomposition of wastes in landfill areas (Chan et al., 2002; Filella and Penuelas, 2006; Ortiz et al., 2002; Sahu et al., 2016a). Typically, congested traffic and stagnant wind conditions are the major factors favoring accumulations of pollutants, resulting in their concentrations (Ho et al., 2004). The emissions from landfill sites include greenhouse gases (GHGs), VOCs, trace gases containing nitrogen and sulfur, and particulate matters (PM) (Randazzo et al., 2020). VOCs represent less than 1% (by volume) of the total gas emitted from the landfill (Schuetz et al., 2003; Tassi et al., 2009). Among the VOCs, the concentrations of aromatic compounds are particularly high near the landfill (Kim et al., 2005; De la Rosa et al., 2006). Most previous studies of VOCs have widely used the gas chromatography-mass spectrometry (GC-MS) methods (e.g., Conte et al., 2017; Chiriac et al., 2009; Corréa et al., 2012).

However, mostly focused on traffic-related emissions, benzene and toluene concentrations are reported for different urban regions of the world (e.g., Barletta et al., 2005; Chaudhary and Kumar, 2012; Civan et al., 2011; Keymeulen et al., 2001; Miller et al., 2011; Parra et al., 2006; Shaw et al., 2015; Tiwari et al., 2010; Velasco et al., 2007). The measurement-based studies of aromatic VOCs are limited only to a few

cities of India (e.g., Hoque et al., 2008; Lal et al., 2012; Majumdar et al., 2011; Sahu et al., 2016a, 2016b; Sahu et al., 2010; Sahu and Lal, 2006a, 2006b; Srivastava, 2005). However, the measurements of VOCs near specific emission sources such as landfill, major road junction, etc. are scarce in India. This is the first comprehensive measurement-based study of a large suite of ambient VOCs near a major landfill site in Ahmedabad city. The main objective of this study is to determine the composition and characterize the emission ratios (ERs) of VOCs. We have also investigated the change in the concentrations of different VOCs with the distance, and the contributions of other sources in the downwind regions.

2. Materials and method

2.1. Study site and local meteorology

The Pirana landfill (22.98°N, 72.56°E) is located in Ahmedabad city in the Gujarat state (province) of India. It is the 5th largest city in India with a population of about 6.5 million (according to the census year 2011) and covers an area of 464 km². The city is located along the banks of the Sabarmati river as shown in Figure 1. Further, details about Ahmedabad city and major emission sources are reported in our earlier study (Sahu et al., 2016a). The city has the only landfill site at Pirana, where municipal solid waste (MSW) has been dumped for ~30 years. The

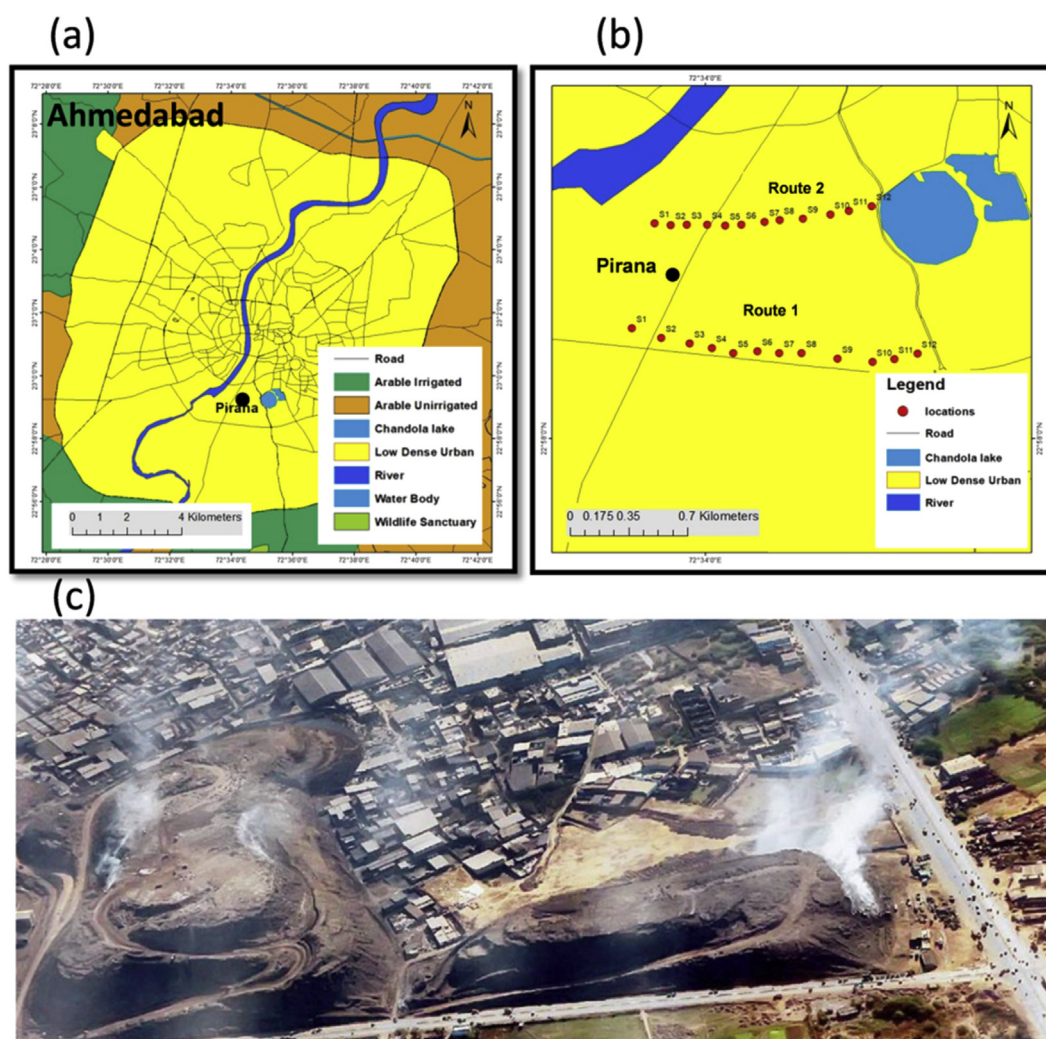


Figure 1. (a) The road map of Ahmedabad city and a black circle indicate the location of the Pirana Landfill site, (b) sampling sites along the Route-1 and Route-2, and (c) a picture of smoke emanating from some parts of the Pirana landfill.

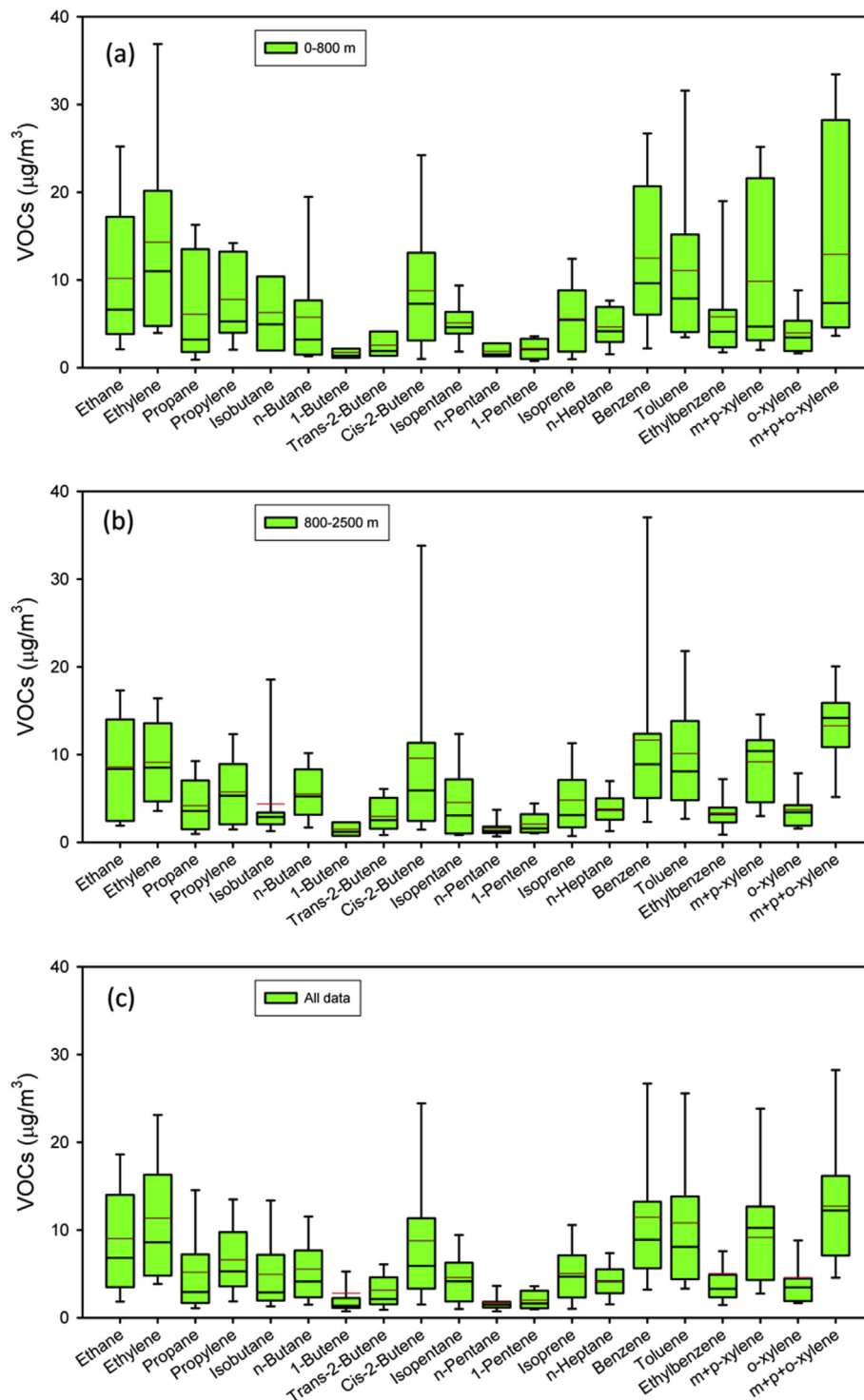


Figure 2. Box-whisker plots of VOCs concentrations measured at different sites within the distances of (a) 0–800 m, (b) 800–2500 m, and (c) 0–2500 m from the Pirana landfill site in Ahmedabad city, India during May 2017.

Pirana landfill site can be located in the southern part of the city along the east bank of the Sabarmati river. The total allocated area for the Pirana landfill is ~84 ha, of which ~65 ha of the land has been used for the MSW disposal since 1980. An average of ~2100 tons/day of waste is dumped on the site. At present, the average landfill height and depth are ~52 m and 22 m, respectively. The wastes dumped at landfill sites are mainly residuals of domestic and commercial consumptions. The site is a non-engineered open pit, and the wastes collected from different parts of the city are dumped without proper segregation. The MSW dumped at

Pirana has been broadly classified as organic matters (72%), earth material (13.5%), plastic (6.8%), paper (5.7%), and metal/glass (2%) (Singh et al., 2008). Among a mixture of gases, the burning and decomposition of landfill organic materials leads to the release mainly of CH₄ (45–60%) and CO₂ (40–55%) (e.g., Abushammala et al., 2016). Also, trace amounts of various VOCs are emitted from the decomposition and burning of organic materials. The concentrations of VOCs in surrounding areas of the landfill depend upon the waste composition and stages of the decomposition and fire.

Table 1. Summary of NMVOC concentrations ($\mu\text{g}/\text{m}^3$) measured at upwind and downwind sites near the Pirana landfill area.

VOCs	Upwind			Downwind					
	0–500 m			0–800 m			800–2500 m		
	Min	Max	Average	Min	Max	Average	Min	Max	Average*
Ethane	1.6	11	7.0 ± 5	1.4	54	10 ± 9	1.8	77	8.6 ± 6.2
Ethylene	8.5	9.2	8.8 ± 0.4	3.9	100	14 ± 11	3.5	189	9.1 ± 5.1
Propane	1.1	18	5.9 ± 8	0.7	17	6.1 ± 5.8	0.8	47	4.2 ± 3.0
Propylene	4.4	10	6.5 ± 3	2.1	26	7.8 ± 4.7	1.3	48	5.7 ± 3.8
Isobutane	1.3	8	3.7 ± 4	1.6	28	6.3 ± 4.6	1.2	29	4.4 ± 5.6
n-Butane	2.0	8	4.8 ± 3	1.3	21	5.8 ± 6.2	1.6	11	5.5 ± 2.9
1-Butene	1.1	23.6	8.7 ± 13	1.1	11	1.7 ± 0.8	0.7	6.4	1.5 ± 0.9
Trans-2-Butene	1.5	12	5.2 ± 6	1.0	17	2.6 ± 1.7	0.8	28	3.0 ± 1.9
Cis-2-Butene	3.2	12	5.6 ± 4	0.8	27	8.8 ± 7.6	1.2	37	9.1 ± 10
Isopentane	1.3	4.9	3.3 ± 2	1.8	20	5.1 ± 2.5	0.8	15	4.6 ± 4.1
n-Pentane	1.2	2.5	1.7 ± 1	0.7	3.7	1.8 ± 1.0	0.7	3.9	1.6 ± 0.8
1-Pentene	1.0	1.7	1.3 ± 1	0.7	7.2	2.2 ± 1.1	1.0	5.2	2.1 ± 1.3
Isoprene	2.5	6.3	4.4 ± 2	0.8	28	5.5 ± 3.8	0.4	32	4.6 ± 3.7
n-Heptane	1.7	5.5	3.4 ± 2	1.5	22	4.7 ± 2.2	1.1	11	3.7 ± 1.9
Benzene	3.8	12	7.3 ± 3	1.3	85	13 ± 8.2	1.3	101	11 ± 11
Toluene	2.9	11	8.3 ± 4	3.4	45	13 ± 11	1.7	255	10 ± 6.5
Ethylbenzene	2.2	3	2.5 ± 0.4	1.6	32	7.8 ± 9.3	0.5	62	3.4 ± 1.8
m + p-xylenes	1.0	11	7.4 ± 5	2.0	68	13 ± 14	2.0	183	8.7 ± 4.4
o-xylene	1.8	4.5	3 ± 1.3	1.6	32	6.1 ± 8.2	1.3	52	3.6 ± 2.3
m + p + o-xylenes	4.6	16	10 ± 5	3.5	77	18 ± 21	4.0	235	13 ± 5.2

* Major outliers are not considered.

2.2. Instrumentation

As shown in Figure 1, the air samples were collected at different sites along two different routes henceforth referred to as Route-1 and Route-2 within 2.5 km from the landfill center. The sampling along the Route-1 and Route-2 were conducted on 05, 06, and 18 May 2017 (see Table S1). For further details, see Supplementary material-Heliyon-20-07-2020. The air samples were collected in pre-evacuated glass flasks (~800 mL) at a pressure up to ~2 bar using an oil-free 'Metal Bellow' compressor (High speed Appliances, India, mini diaphragm type, Model No. IS 4722). We collected 48 samples during the campaign and replicas for 10% of the samples. These samples were analyzed using a gas chromatography-flame ionization detector (GC-FID) (7890A-G3440A, Agilent, Palo Alto, CA, USA) coupled with thermal desorption (TD) system (UNITY2, Air Server 3 channel module, Markes International, UK). An "Ozone Precursor" cryo-trap was used for the pre-concentration of NMVOCs present in air samples. An ultra-pure helium gas (Research Grade 99.9999%, Linde, USA) was used as a carrier gas. The zero-air produced by the Parker HPZA-3500-220 and hydrogen using the Parker H2PD-300-220 gas generators were used as fuel gases in the FID. The nitrogen gas (999.9999%) generated using the Parker UHPN2-1100-220 generator was used to purge the TD and also as a make-up gas in the FID.

The chromatogram signals of different NMVOCs were calibrated using dynamic dilutions of a standard mixture containing $\sim 1.0 \pm 5\%$ ppm of each compound (LE732C 4/15, Linde, USA). A gas calibration unit (GCU-advanced v2.0, Ionicon Analytik GmbH, Austria) was used for the dilution and hence to get known concentrations. In GCU, the standard mixture flow rate was adjusted using a mass flow controller (MFC-std) to provide variable amounts of the calibration gas. The dilution gas (zero-air) flow of 500 sccm/min maintained using another mass flow controller (MFC-dil). The zero-air gas is generated by passing ambient air through a VOC-scrubber catalyst (heated at 350 °C). However, a very stable zero-air is required to determine the background and hence quantify TD-GC-FID performances, including the sensitivity and limit of detection (LOD). Therefore, instead of using ambient air

directly (with variable VOCs) as a direct input to GCU, the ultra-purity zero-air generator (Parker HPZA-3500-220) was connected in tandem to GCU. The LOD of each VOC was determined by the following equation (Rovelli et al., 2019).

$$\text{LOD} = 3 \times \frac{\sigma_{\text{blank}}}{\text{Sensitivity}} \quad (1)$$

where σ_{blank} represents the standard deviation of blank (zero-air) signals. The sensitivity of each VOC was determined using the multipoint calibration curve. The blank level (zero) is important for determination of the LOD. For each VOC, the sensitivity depends on the relation between the set volume mixing ratio (ppbv) and normalized peak area which showed excellent linear fits with correlation coefficients (r^2) > 0.97 at $p < 0.05$. The precision was calculated as the relative standard deviation (RSD) of the measurements at different set values of 2, 4, 8 ppbv with 5 replicates for each. The overall precisions for different NMVOCs presented in this study were in the range of 2–15%. The overall accuracies of all NMVOCs were determined to be better than 10%, which includes the uncertainties of calibration mixture (~5%) and flow rates (~2%) of both the standard and dilution gases. The key operating parameters of TD-GC-FID used for the analysis of NMVOCs in air samples are presented in Table S2. Additional details of TD-GC-FID system including temperature programming and key operational parameters are reported in our previous papers (Sahu et al., 2016a; Tripathi et al., 2020). The ranges of ambient temperature, relative humidity (RH), and wind speed recorded during the sampling periods were 27–35 °C, 40–70%, and 0.1–3.1 m/s, respectively. The wind flows were from either northwest (NW) or southeast (SW) directions.

We have estimated the Propylene-Equivalent (Propy-Equiv) concentration (PEC) to assess the roles of different NMVOCs in the formation of ozone. The Propy-Equiv concentration of a given NMVOC (i) can be defined as provided in Chameides et al. (1992).

$$\text{Propy-Equiv}(i) = \frac{C_i \times k_{\text{OH}}(i)}{k_{\text{OH}}(\text{C}_3\text{H}_6)} \quad (2)$$

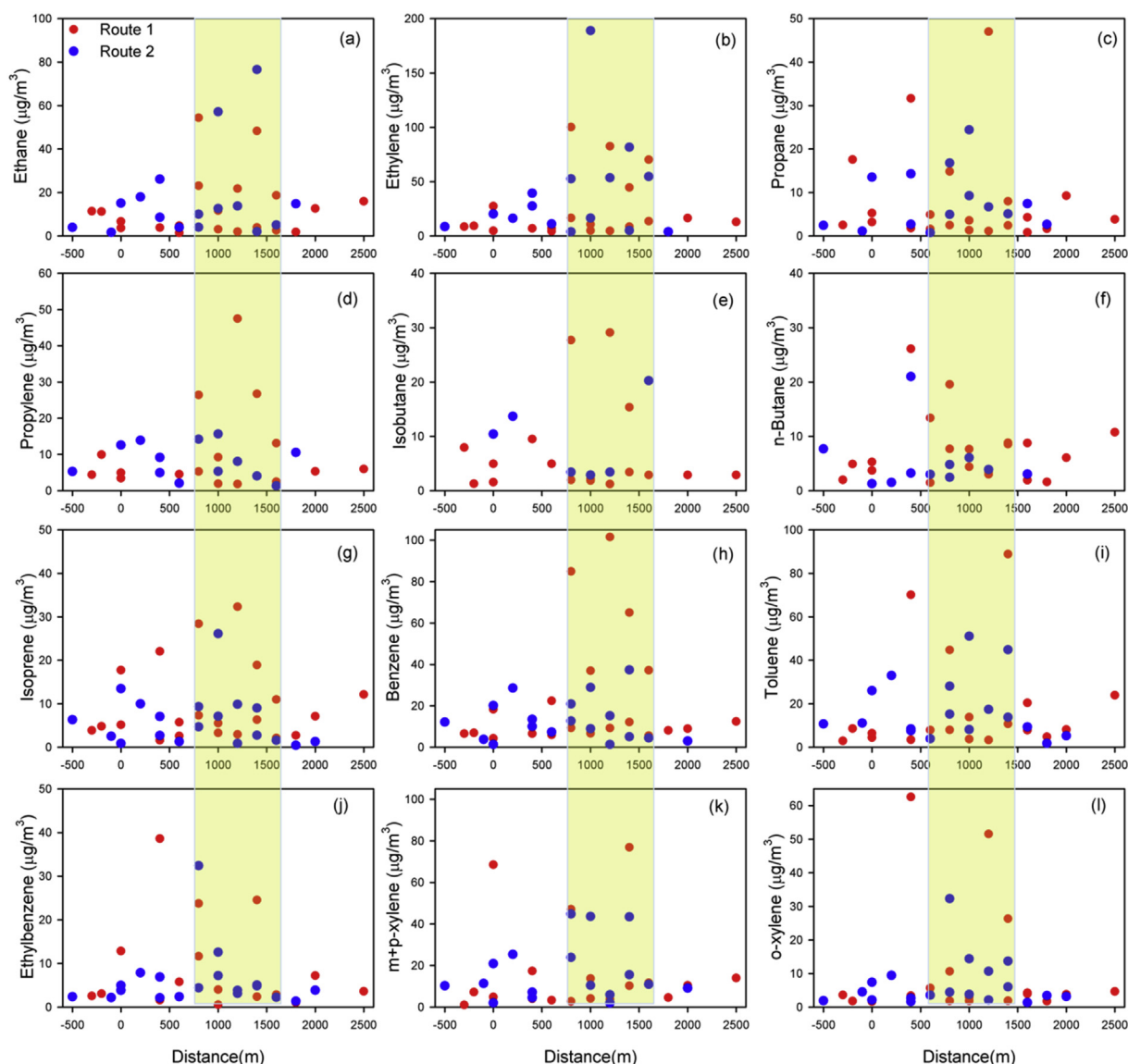


Figure 3. The concentrations of NMVOCs measured at different distances surrounding the Pirana landfill. The shaded region represents receptor sites with the major enhancements of NMVOCs, while negative distances refer to the upwind sites.

The Propy-Equiv (i) is a measure of the concentration of a compound (i) on an OH reactivity-scale normalized with respect to the reactivity of propylene (C_3H_6). Whereas, C_i is expressed in 'ppbc' which can be obtained by multiplying the volume mixing ratio (ppb) with the number of carbons present in a given compound. The ozone formation potential (OFP) defined by the following expression has also been used to assess the role of different NMVOCs in the formation of ozone (Carter, 1994).

$$OFP_i = [VOC_i] \times MIR_i \quad (3)$$

where OFP_i represent the ozone formation potential of a given VOC(i). $[VOC_i]$ and MIR_i are the concentration and maximum incremental reactivity (MIR) of VOC_i , respectively. In the present study, the values of MIR are taken from Carter (1994).

3. Results and discussion

3.1. Concentrations of NMVOCs

The statistics of the concentrations of different NMVOCs measured at different sites within the distances of 0–800 m, 800–2500 m from the

Pirana landfill site are shown in the box plots (Figure 2). Although the concentrations of NMVOCs show substantial variations at each site, the change in their concentrations with distance can be noticed very clearly. As expected, the concentrations of almost all NMVOCs measured at nearby sites (within 0–800 m) were significantly higher than their concentrations at greater distances (800–2500 m). The major outliers (a few) measured at both nearby and far distances were not included in the box plots. Therefore, the comparative discussions are based on the dataset which have successfully passed the statistical t-test ($p < 0.05$). To ensure this, we have removed the outliers from each dataset. The change in the concentrations of aromatic compounds and particular of xylenes with the distance was particularly significant. The average concentrations of m + p-xylenes and o-xylene were 13.0 ± 13.9 and $6.1 \pm 8.2 \mu\text{g}/\text{m}^3$ between 0–800 m which decreased to 8.7 ± 4.4 and $3.6 \pm 2.3 \mu\text{g}/\text{m}^3$ between 800–2500 m of distances, respectively. While the average concentrations of ethane decreased slightly from $10.2 \pm 8.4 \mu\text{g}/\text{m}^3$ at 0–800 m to $8.6 \pm 6.2 \mu\text{g}/\text{m}^3$ at 800–2500 m of distances. Among different NMVOCs measured in the present study, alkane and aromatic groups make substantial contributions to ambient concentrations. Overall, ethane ($9.0 \pm 6 \mu\text{g}/\text{m}^3$), ethylene ($11.4 \pm 8 \mu\text{g}/\text{m}^3$), benzene ($11.5 \pm 9 \mu\text{g}/\text{m}^3$) and toluene ($10.8 \pm 8 \mu\text{g}/\text{m}^3$) were the dominant species at all distances. In addition to the

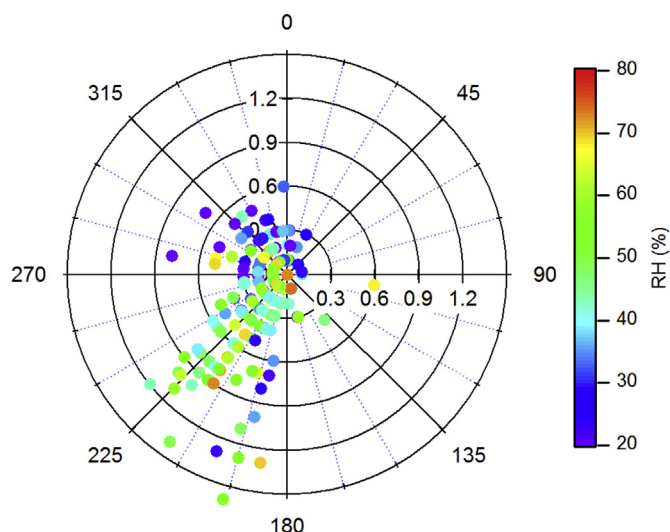


Figure 4. Wind polar plot color-coded with the level of relative humidity (RH) and the magnitude of wind speed is in the unit of m/s.

dominance of alkane and aromatic compounds, the concentration of *cis*-2-Butene ($8.8 \pm 8 \mu\text{g}/\text{m}^3$) was also high. As inferred from the percentile data, the concentration of each NMVOC at nearby sites show larger variations compared to those at greater distances. At nearby sites, the concentrations of benzene, toluene, ethylbenzene, and xylenes (BTEX) showed larger ranges of variations compared to those of other NMVOCs. The higher concentrations at nearby sites are expected due to the proximity to emission sources. However, on average, except for 1-Butene and *trans*-2-Butene, the concentrations of NMVOCs at both nearby and far downwind sites were about 1–3 and 1–1.6 times higher than those measured at upwind sites, respectively (see Table 1). These differences with reference to upwind sites are small as the upwind locations sites were very close to the emissions sources. On the other hand, the samples collected at far upwind sites (>800 m) were predominantly influenced by the traffic-related emissions. As a result, the emissions of NMVOCs from the landfills are challenging to distinguish from background-concentrations. Furthermore, this becomes worse when other emission sources are also present in the surrounding areas. Therefore, the absolute enhancements in ambient concentrations of NMVOCs due to landfill emissions are difficult to estimate at different downwind sites.

A modeling tool can help to resolve contributions from individual sources is critical for air-toxics exposure assessments (Stein et al., 2007). Several studies have reported the dispersion modeling of pollutants using different models such as AERMOD, CALPUFF, LAPMOD, WRF-CALPUFF and HYSPLIT (Stein et al., 2007, 2015). In this study, the Hybrid Single-Particle Lagrangian Integrated Trajectory model (HYSPLIT) has been used to compute the dispersion of air masses from the Pirana landfill site. According to the Lagrangian approach, the dispersion particles are computed by taking into account the number of different possible trajectories, including the stochastic nature of turbulence that acts on particles. The HYSPLIT dispersion results computed for three different days (05, 06, and 18 May) are presented in Figure S1. The dispersion of plumes covers both the routes in the downwind of the Pirana landfill site for each day. The significant reductions of \sim ten times in the particle concentrations can be noticed in the downwind between 800–2500 m distances. However, the measurements show smaller reductions partly due to the emissions from various other sources (i.e., traffic, industries, residential, etc.), which were not considered in the simulations. It is consistent with the explanation that other sources also contribute to ambient concentrations in addition to landfill emissions.

Although samples were collected up to maximum distances of \sim 2.5 km impact of landfill emissions could be widespread. As simulated using the dispersion model, downwind regions covering distances of 10–12 km

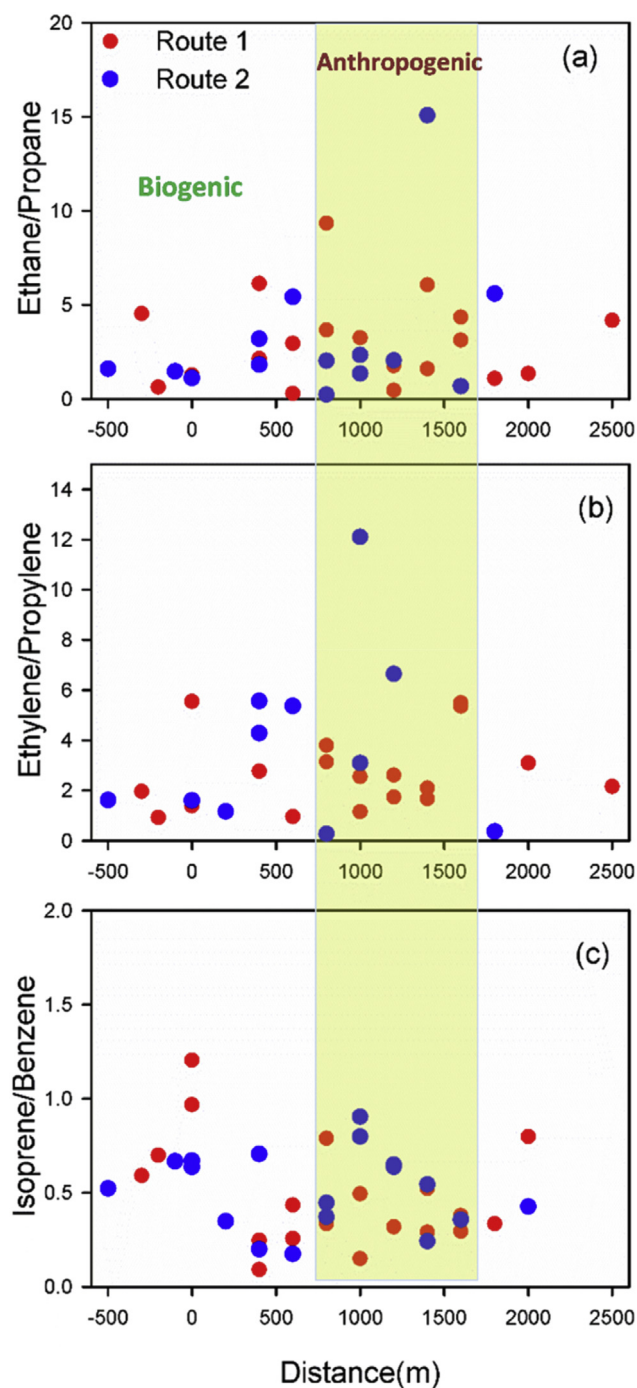


Figure 5. The ratios of pairs of NMVOCs measured at different distances from the Pirana landfill along the Route-1 and Route-2. The shaded region represents the downwind sites with the major enhancements of NMVOC concentrations.

along the Route-1 and 8–10 km along the Route-2 were substantially affected by the landfill emissions. However, depending on prevailing meteorological conditions, the spatial extent impacted by landfill emissions could vary significantly.

3.2. Variations of NMVOC and their ratios with distance

The variations in the concentration of different NMVOCs with distance from the landfill site are shown in Figure 3. The negative and positive distances refer to the sites located in the upwind and downwind regions, respectively. The concentrations of almost all NMVOCs at the

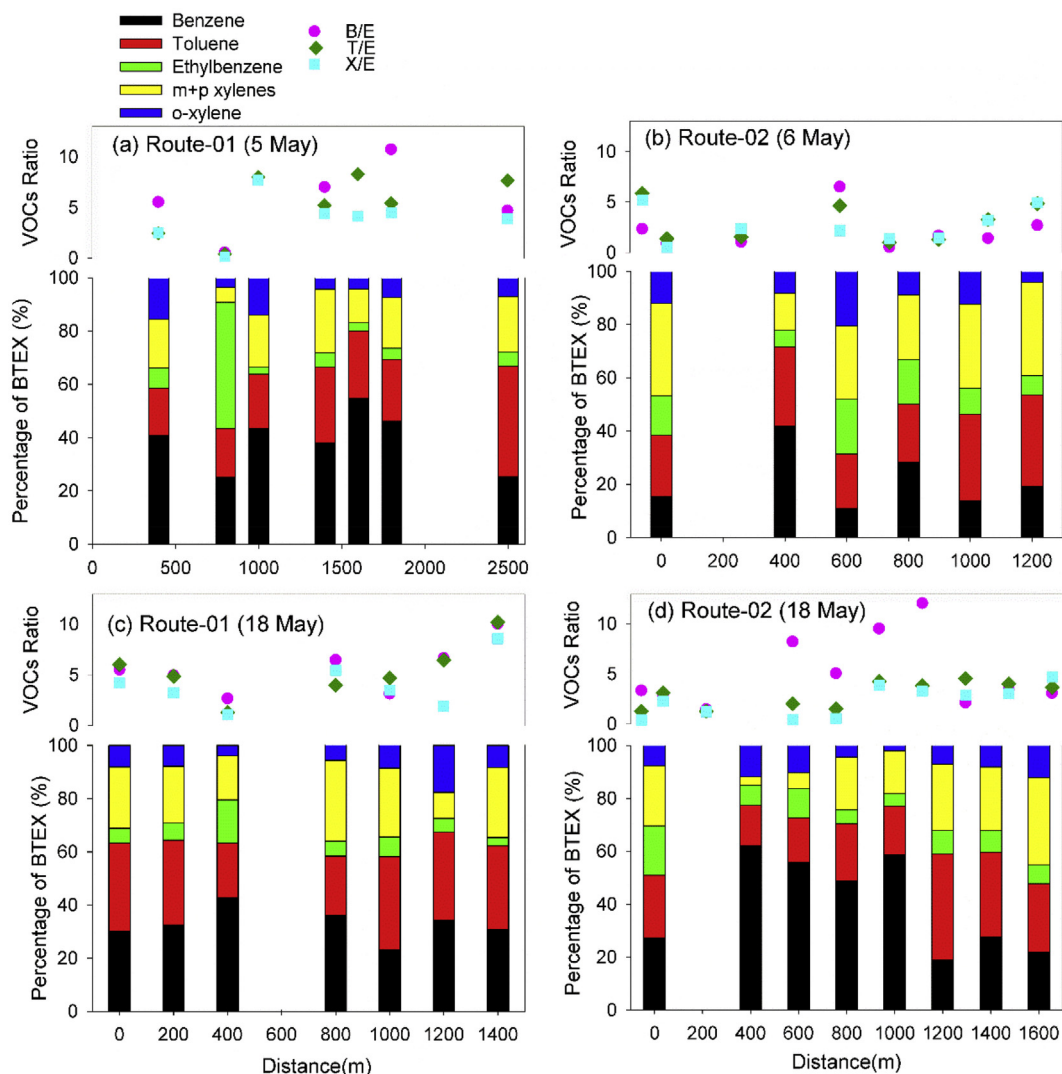


Figure 6. The composition of and ratios (B/E, T/E, X/E, B/T) measured at different distances from the Pirana landfill along Route-1 and Route-2.

upwind sites are lower than their respective concentrations measured at downwind sites located within 1600 m. The major enhancements of NMVOCs were measured at receptor sites located between 800–1600 m of distances along both the routes. This is consistent with the prevailing NW and SW winds recorded during the sampling periods (Figure 4). The sites along the Route-1 and Route-2 were in the downwind of the landfill during the prevailing NW and SW wind flows, respectively. Therefore, the variability is mainly due to the changes in emission and wind parameters at a given distance. The enhancements of almost all NMVOCs at receptor sites along the Route-1 were higher than those measured along the Route-2. As described in Table S1, in addition to emissions from the landfill, the vehicular traffic and industrial activity could significantly contribute to the concentrations of NMVOCs measured along the Route-1. In particular, BTEX concentrations at the receptor sites along Route-1 were much higher than their concentrations measured at about the same distances along Route-2. Between 800–1600 m, average concentrations of benzene and toluene were 35 and 29 $\mu\text{g}/\text{m}^3$ along the Route-1, while were 13 and 17 $\mu\text{g}/\text{m}^3$ along the Route-2, respectively. The average concentrations of ethane and ethylene were 21 and 32 $\mu\text{g}/\text{m}^3$ along the Route-1, and 17 and 39 $\mu\text{g}/\text{m}^3$ along the Route-2, respectively. The similar concentrations of ethane and ethylene along both the routes indicate their predominant biogenic emissions from the biodecomposition of organic materials.

3.3. Spatial fingerprinting of anthropogenic and biogenic sources

The molar emission ratios (ERs) of several pairs of NMVOCs were used to approximately characterize and differentiate the contributions of biogenic and anthropogenic sources (Sahu et al., 2017; Tang et al., 2009). However, with definite uncertainties, the ratios of ambient air concentrations of NMVOCs can also account for the variations caused by the boundary layer and meteorological processes and are useful to characterize the major emission sources. On the other hand, the ratios do not necessarily take account of the chemical loss mainly by the reactions with hydroxyl (OH) radicals, as the pairs of NMVOCs can have different rate constants for the reactions with OH. In the present study, the air samples were collected in the morning hours within a time span of 2–3 h. Therefore, the variations in the ratios of NMVOCs caused due to the different reaction rates of oxidation by OH radicals can be assumed to be small.

The ratios of ethane/propane, ethylene/propylene, and isoprene/benzene measured at different distances along both the routes are plotted in Figure 5. Several NMVOC compounds from the alkene group are dominant species that were released from the natural biological decomposition of organic materials present in the landfill. Therefore, relatively higher and lower ratios suggest predominance of biogenic and anthropogenic emissions, respectively. Except for a few samples, the

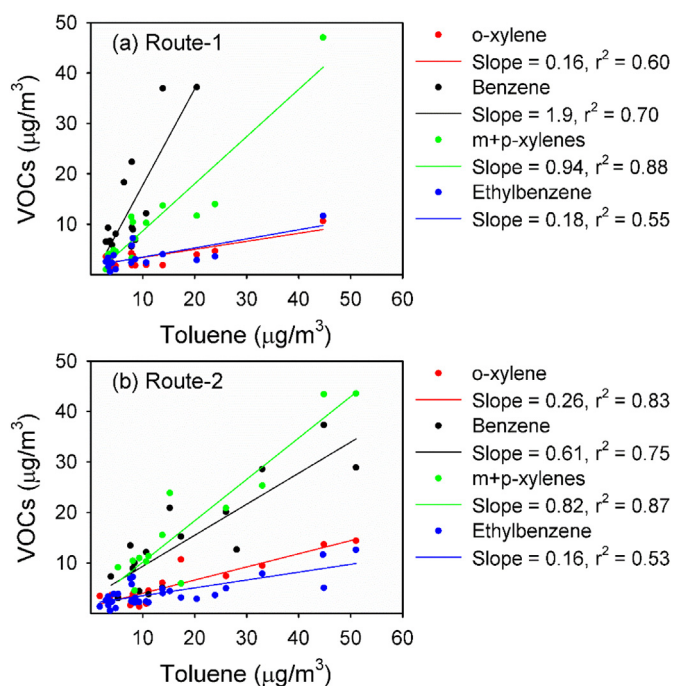


Figure 7. Correlations between the concentrations of different aromatic NMVOCs with toluene measured near the landfill sites along the Route-1 and Route-2.

ratios of these NMVOCs at the sites very near to the landfill (<500 m) were slightly higher than those measured at major receptor sites. Although the emissions from various sources affect the measurements, contributions from biogenic (decomposition of organic materials) near the landfill sites seem to be higher than those measured at far downwind sites. In particular, the higher isoprene/benzene ratios (>0.5 µg/µg) near the landfill site indicate the dominance of biogenic emissions. The ethylene/propene ratios measured near the landfill sites are similar to those reported at biogenic sites (e.g., Goldstein et al., 1996; Lamanna and Goldstein, 1999; Rhew et al., 2017). On the other hand, the higher ethane/propane ratios (1.5–5) in the downwind of fossil fuel combustion and biomass burning emissions are reported in the literature (Hough, 1991; Rudolph, 1995).

3.4. Aromatic NMVOCs and BTEX ratios

Diagnostic BTEX ratios with ethylbenzene-based normalization have been used to identify the major emission sources (Buczynska et al., 2009; Sahu et al., 2020b). The emission rates of BTEX have been investigated using their ratios to ethylbenzene and also for comparison with other data from the different landfills (Durmusoglu et al., 2010). The ratios of B/E, T/E, X/E, and B/T measured at different distances from the Pirana landfill are shown in Figure 6. The ratios of B/E, T/E, and X/E show similar dependence to those of BTEX concentrations with the distance. The ratio of B/T has been widely used for the source apportionment studies to assess the emission characteristics of NMVOCs from vehicular and industrial sources. On the other hand, the B/T ratio shows a different variation, which increases with distance up to 500 m and then decreases gradually at greater distances from the landfill. The ratios of B/E, T/E, X/E and B/T varied in the ranges of 0.39–13, 0.33–7, 0.40–3.5, and 0.12–8 along the Route-1, and 0.35–7.4, 0.87–9, 0.34–2 and 0.51–8.6 along the Route-2, respectively. The median ratios of B:T:E:X were 3.1:2.8:1:3 and 2.1:4:1:3.2 along the Route-1 and Route-2, respectively. At the Datianshan landfill of Guangzhou in China, the BTEX ratios of 4:6.5:1:2 and 3:5:1:2 were measured in winter and summer, respectively (Zou et al., 2003). The B/T ratios of ~0.015, 0.11, 0.5, and 0.27–0.5 have been reported for the emissions from the landfill, vehicle exhaust, and urban environment (mix), respectively (Dincer et al., 2006). However, a wide range of B/T ratios between 0.12 and 5.3 was reported for the emissions from five different landfill sites in Korea (Kim et al., 2006). A very high B/T ratio of ~8 was measured near a landfill site in Tokyo, Japan (Urase et al., 2008). In the present study, the large variations of BTEX ratios were partly due to the sampling at both upwind and downwind sites. Another cause of variations could be the disproportionate mixing of NMVOCs released from other emission sources such as industrial plume, vehicle exhaust, etc. Nonetheless, the higher B/T ratios (>1.0) measured up to ~1000 m of distances in the downwind region suggest the predominance of landfill emissions. However, both B/T and B/E ratios at the downwind sites along the Route-1 were particularly higher than those measured along the Route-2. In tunnels experiments, the B/E ratios <5 and T/E ratios <6 were measured which characterize the vehicle emissions, while their higher ratios suggest the contributions from other sources.

The ambient air concentrations of benzene and toluene were measured at 12 different road junctions of Ahmedabad city during the pre-monsoon months of the year 2015 (Sahu et al., 2016a). The average

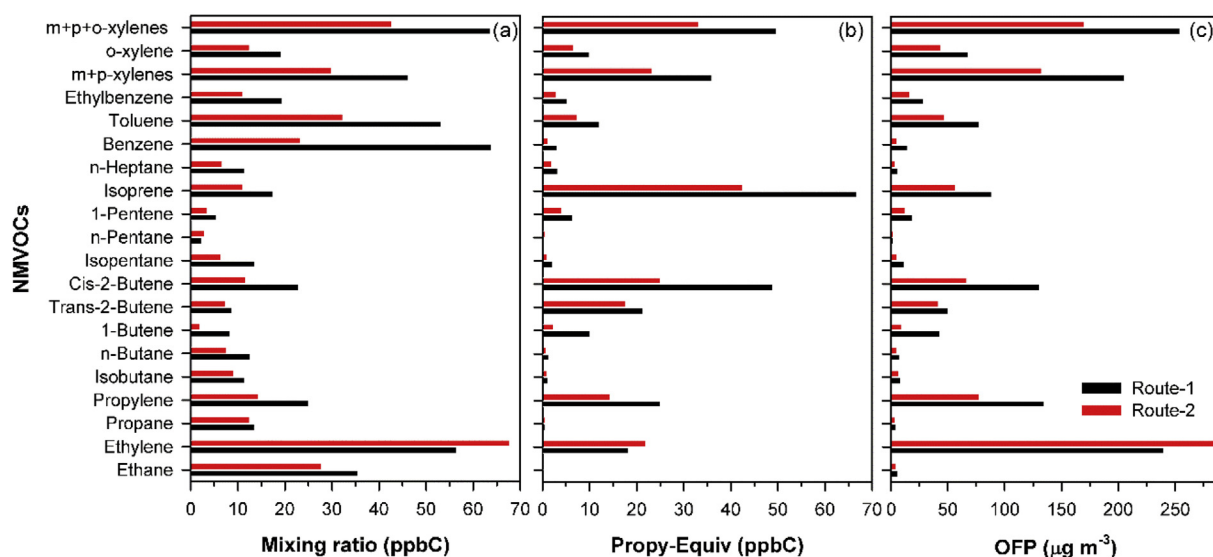


Figure 8. The average mixing ratios (ppbC), Propy-Equiv concentrations (ppbC), and the ozone formation potentials (OFPs) of different NMVOCs measured along the Route-1 and Route-2 near the Pirana landfill.

B/T ratios of 0.33–0.5 were estimated from the data measured at different road junctions of Ahmedabad city. In the present study, however, except for a few sites, B/T ratios near the landfill sites are significantly higher than those measured at major road junctions. The comparison suggests characteristically different emission profiles of NMVOC for vehicle exhaust and landfill sources.

3.5. Correlation between BTEX concentrations

Among the aromatic compounds (BTEX) are mainly emitted from fossil fuel combustion and evaporative losses (Buczynska et al., 2009). Scatter plots between the concentrations of benzene, ethylbenzene, m + p-xylenes, and o-xylene with the concentration of toluene measured along Route-1 and Route-2 are shown in Figure 7. The concentrations of benzene, m + p-xylenes and o-xylene show strong correlations with toluene with $r^2 = 0.60$ – 0.88 for the Route-1, and $r^2 = 0.75$ – 0.87 for the Route-2. The slope of $\Delta B/\Delta T$ showed significantly different values of 1.9 and 0.64 along the Route-1 and Route-2, respectively. The slopes of $\Delta E/\Delta T$, Δm , p-xylenes/ ΔT and Δo -xylene/ ΔT were 0.18, 0.94 and 0.16 for the Route-1, while were 0.16, 0.82 and 0.26 for the Route-2, respectively. Schroder et al. (2018) reported the slope of Δo -xylene/ $\Delta T \sim 0.17$ in an urban plume over the northeast United States.

The correlations between the concentrations of benzene and toluene measured in the present are compared with our previous measurements at different road junctions of the city (Sahu et al., 2016a). The concentrations of NMVOCs measured at road junctions were predominantly influenced by vehicle exhaust emissions. The concentrations of benzene and toluene measured under both types (landfill and traffic) of emissions show strong correlations ($r^2 = 0.70$ – 0.87), indicating their co-emissions (see Figure S2). However, the overall slope ($\Delta T/\Delta B$) of 0.64 determined using all the data measured at landfill sites is ~ 3 times higher than those determined using the data measured at road junctions.

4. Propylene-Equivalent concentration of NMVOCs

The photochemical production of O_3 is influenced by several factors including the reactivity of NMVOCs, NO_x concentration, intensity of solar radiation, and meteorological conditions (Tan et al., 2012). The contribution of particular NMVOC to the production of O_3 is related to its reaction rate with OH radicals and level of NO_x . Under high NO_x regimes, the O_3 production rate is determined by the concentrations of radicals produced from the oxidations of NMVOCs. In the lower NO_x regimes, the concentrations of NO_x that limit the production of O_3 (Carter, 1994). Therefore, the importance of different NMVOCs in atmospheric chemistry is ranked in terms of their concentrations and assessed based on the number of C-atoms and reaction rates with OH radicals. The concentration-based representation has the advantage of being simple and, therefore, easy to interpret. However, it has a disadvantage to investigate the sensitivities leading to the formation of secondary oxidants such as O_3 and SOA as it does not account for the reaction rates of NMVOCs. Therefore, several reactivity-based scales have been proposed to assess the role of different compounds in the formation of O_3 .

As shown in Figure 8, ranking by Prop-Equiv concentration, the top 5 NMVOCs along the Route-1 were isoprene (67 ppbC), cis-2-Butene (49 ppbC), m + p-xylenes (36 ppbC), propylene (25 ppbC) and trans-2-Butene (21 ppbC). While the top 5 NMVOCs along the Route-2 were isoprene (42 ppbC), cis-2-Butene (25 ppbC), m + p-xylenes (23 ppbC), ethylene (22 ppbC) and trans-2-Butene (18 ppbC). These top NMVOCs along the Route-1 and Route-2 accounted for $\sim 72\%$ and 75% of the total Prop-Equiv concentrations of 271 and 174 ppbC, respectively. This result indicates that the alkene and aromatic series compounds play the most important role in the production of O_3 near the landfill. The higher Prop-Equiv concentrations of aromatic NMVOCs along the Route-1 could be due to additional contributions from traffic and industrial related emissions beside those from the landfill sources. The contributions of different

NMVOCs to the Prop-Equiv concentrations are consistent with a recent study reporting the dominance of alkene and aromatic compounds in the Foshan area, Pearl River Delta (PRD), China (Tan et al., 2012). The higher Prop-Equiv concentrations of NMVOCs near the landfill site in Ahmedabad could lead to the higher formation rates of O_3 . Earlier studies (Lal et al., 2000; Sahu and Lal, 2006a) reported the pronounced diurnal variations of O_3 in the pre-monsoon season, suggesting the high photochemical production efficiency of O_3 at Ahmedabad.

The OFP has also been used to estimate the contributions of different VOCs in the formation of O_3 (e.g. Tan et al., 2012). The OFP values determined for different VOCs are plotted in Figure 8(c). Except for ethylene and isoprene, the OFPs of VOCs show similar relative contributions to those of Prop-Equiv concentrations. Overall, the Prop-Equiv concentration and OFP scales show good agreements ($r^2 > 0.65$) along both the routes. As shown in Figure 8(c), ranking by OFP in decreasing order, the top 5 NMVOCs were ethylene, m + p-xylenes, propylene, cis-2-Butene, and isoprene along both the routes. These top NMVOCs accounted for $\sim 70\%$ and 75% of the total OFP along the Route-1 and Route-2, respectively. In reality, both the Prop-Equiv and OFP based methods have limitations as the production efficiency of O_3 also depends on the level of NO_x and intensity of sunlight.

5. Conclusions

The concentrations of C_2 – C_8 NMVOCs were measured at 21 different sites in the downwind of the largest landfill (Pirana) in Ahmedabad city during a pre-monsoon month of year 2017. The residential areas were the major receptors because of predominant NW and SW wind flows during the study period. The concentrations of almost all NMVOCs measured at nearby sites within 800 m from the landfill were higher than their concentrations at the greater distances. The concentrations of all NMVOCs show substantial variations with the distance from the landfill site. The values of total BTEX ($\sum BTEX$) in the upwind regions were 18–84% higher than the mean value measured at the upwind sites. The meteorological parameters and inferences of other sources played an important role in varying influences of the landfill emissions. The relative abundances of NMVOCs did not change much with the distance. Alkane and aromatic compounds made the major contributions to ambient concentrations. The contributions of biogenic emission from the decomposition of the landfill organic materials at nearby sites were higher than those at far downwind sites. Although air sampling covered the distances of up to ~ 2.5 km, the impact of landfill emissions in the downwind regions covering greater distances were simulated using the dispersion model. Among NMVOCs, cis-2-Butene, m + p-xylenes, propylene/ethylene, and trans-2-Butene explain $\sim 75\%$ of the total Prop-Equiv concentrations. In the OFP scale, except for ethylene and isoprene, VOCs show similar relative contributions to those of Prop-Equiv concentrations. In reality, in addition to the values of Prop-Equiv concentration/OFP of ambient VOCs, the concentrations of NO_x are important in controlling the ozone level. In the past few decades, application of proton transfer reaction-mass spectrometry (PTR-MS) technique has gained popularity for the real-time measurements of many VOCs at low concentrations (pptv/ppbv) (e.g., Byliński et al., 2019; Tripathi and Sahu, 2020). Our future studies will involve a comprehensive measurement of NMVOCs using PTR-ToF-MS in different seasons.

Declarations

Author contribution statement

Lokesh Sahu: Conceived and designed the experiments; Wrote the paper.

Pragnesh N. Dave, Ravi Yadav: Analyzed and interpreted the data; Wrote the paper.

Nidhi Tripathi: Contributed reagents, materials, analysis tools or data; Wrote the paper.

Samiksha Bajaj, Kashyap Patel: Performed the experiments; Wrote the paper.

Funding statement

This research did not receive any specific grant from funding agencies in the public, commercial, or not-for-profit sectors.

Competing interest statement

The authors declare no conflict of interest.

Additional information

Supplementary content related to this article has been published online at <https://doi.org/10.1016/j.heliyon.2020.e04537>.

Acknowledgements

The financial support for the research was provided by Physical Research Laboratory (PRL). The authors gratefully acknowledge the NOAA Air Resources Laboratory (ARL) for the provision of the HYSPLIT transport and dispersion model and/or READY website (<https://www.ready.noaa.gov>) used in this publication. Pragmesh N. Dave acknowledges PRL for offering the Teachers Training Program with supports. Samiksha Bajaj acknowledges PRL for offering the Summer Internship Programme (SIP) with supports.

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