Impact of COVID-19 Pandemic Lockdown in Ambient Concentrations of Aromatic Volatile Organic Compounds in a Metropolitan City of Western India

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Abstract The real-time Benzene, Toluene, Ethylbenzene, and Xylenes (BTEX) concentrations were measured in a metropolitan city of India during January to May of 2020 and 2014-2015-2018 to assess the impact of emission reduction during the COVID-19 lockdown. The total BTEX ($\Sigma$BTEX) concentrations were 11.5 ± 9.0, 15.7 ± 16, 5.3 ± 5.0, 2.9 ± 2.0, and 0.93 ± 1.2 ppbv in January–May 2020, respectively. The evening rush hour peaks of BTEX during lockdown decreased by 4–5 times from the same period of years 2014-2015-2018. A significant decline in background concentrations suggests a regional-scale reduction in anthropogenic emissions. The contributions of $\Sigma$TEX compounds to $\Sigma$BTEX increased from 42% to 59% in winter to 64%–75% during the lockdown under hot summer conditions. While emission reductions dominated during the lockdown period, the meteorological and photochemical factors may also have contributed. Meteorological influence on actual observed BTEX data was removed by normalizing with ventilation coefficient (VC). The actual ambient air reductions of 85%–90% and VC-normalized reductions of 54%–88% of the BTEX concentrations during lockdown were estimated compared to those during the same period of 2014-2015-2018. The estimated changes using nighttime data, which take into account BTEX photooxidation removal, are ~8% lower than the VC-normalized estimates using all data. These significant reductions in BTEX concentrations are consistent with the change in people’s movement as inferred from mobility data during the lockdown. Although enforced, the significant decline in ambient BTEX levels during lockdown was a good change for the air quality. The study suggests a need for more effective science-based policies that consider local and regional factors.

Plain Language Summary Outbreaks of the COVID-19 pandemic necessitated the implementation of strict lockdown in India, which drastically decreased anthropogenic emissions. The elevated levels of a group of aromatic volatile organic compounds known as benzene, toluene, ethylbenzene, and xylene (BTEX) can adversely impact human health. The real-time continuous measurements of ambient air BTEX concentrations were conducted in a major city of India during January–May 2020. The concentrations of all BTEX compounds declined drastically during the COVID-19 lockdown period. We incorporated the VC in the ambient air BTEX concentrations to reduce the meteorological influences. During the lockdown, different BTEX compounds were reduced by 54%–88% compared to the same period during the normal years. In addition to reduced anthropogenic activities, the balance between photochemical processes and evaporative emissions seems to control BTEX concentration and composition during the lockdown period. Although enforced, the decrease of ambient BTEX concentrations was a good change for air quality as these compounds are primary pollutants and precursors for secondary pollutants. This is the most comprehensive study, investigating the impact of the lockdown on ambient BTEX concentrations in India.

1. Introduction

The deterioration of air quality is a serious issue in many urban regions due to the high loadings of various gaseous and particulate pollutants in ambient air (Baudic et al., 2016; Carmichael et al., 2009). The rapid and uncontrolled urbanization of South Asian cities has resulted in the increase of air pollutants, adversely affecting the health and life expectancy of the large population. In recent years, the frequent occurrence of poor air quality episodes in megacities (Singh et al., 2021) and several urban regions of India is mainly due to the ever-increasing number of vehicles (Gurjar et al., 2010). Among the particulate and gaseous pollutants, the emission of volatile...
organic compounds (VOCs) from anthropogenic sources is one of the factors determining air quality. Volatile organic compounds refer to a group of organic chemicals that have high vapor pressures (>0.01 kPa at 20°C) and can easily evaporate into the atmosphere (Herrmann, 2010). Volatile organic compounds are released from various natural and anthropogenic sources (Goldstein & Galbally, 2007; Tripathi et al., 2020). The primary anthropogenic sources include the emissions from fossil fuel combustion, solvent usage, biomass/biofuel burning, and power plants (Sahu, 2012). In the atmosphere, VOCs are short-lived due to their rapid oxidation by hydroxyl radical (OH), nitrate radical (NO3), and ozone (O3; Atkinson & Arey, 2003; Heard & Pilling, 2003). The photooxidation of VOCs and reaction cycles involving oxides of nitrogen (NOx = NO + NO2) can lead to the formation of secondary pollutants, such as ozone (O3) and secondary organic aerosols (SOA; Ahlberg et al., 2017; Liu et al., 2008; Majdi et al., 2019). The elevated O3 levels in the lower atmosphere are known to have adverse impact on human health, plants, and outdoor materials (e.g., Lelieveld et al., 2015). Several studies have reported relatively high SOA formation yields from the oxidation of aromatic VOCs (e.g., Liu et al., 2019; Peng et al., 2017; Platt et al., 2014). Vehicle exhaust emission is a dominant source of aromatic VOCs in megacities of developed and developing countries (Brito et al., 2015). The health impacts of vehicle emissions are strongly related to emission composition (Drozd et al., 2019).

The ambient concentrations of pollutants in megacities can have widespread impacts on human health and ecosystem degradation (Baklanov et al., 2016). In recent decades, the air quality monitoring programs have also included other chemicals, such as VOCs, in addition to the criteria pollutants (e.g., CO, NOx, O3, and PMs; Lee et al., 2002). Volatile organic compounds are key air pollutants in urban and industrial regions known to negatively affect air quality and human health through myriad pathways (Mellouki et al., 2015). Environmental exposure studies have mainly focused on aromatic compounds among many VOCs. The United States Environmental Protection Agency (USEPA) has identified several aromatic VOCs as toxic or carcinogens (Sanchez et al., 2008). The International Agency for Research on Cancer (IARC) and the Department of Health and Human Services (DHHS), US, have identified benzene (C6H6) as a human carcinogen that can cause leukemia (e.g., Band, 2012). Exposure to non-permissible levels of toluene (C7H8) can cause eye, nose, and throat (ENT) irritation. Therefore, long-term exposure to elevated aromatic VOCs concentrations above the permissible limits can cause acute and chronic health issues. Thus, the accurate representation of aromatic compounds is important in the atmospheric modeling of secondary pollutants (O3, SOA, etc.) and human exposure. The BTEX (Benzene, Toluene, Ethylbenzene, and Xylenes) compounds are generally associated with traffic-related emissions (combustion and evaporative loss of fossil fuels) in the urban regions, while TEX compounds are also released from the solvents, paint production, and printing materials (Baudic et al., 2016; Fan et al., 2014; Liu et al., 2008; Salameh et al., 2014; Zhang et al., 2015). Benzene and toluene are mainly emitted from automobile exhaust, while toluene has other significant sources, such as fuel evaporation and industrial/solvent emissions. Regardless of the source, BTEX species contribute substantially up to 47% to the ambient air concentrations of non-methane hydrocarbons (NMHCs) in urban regions (Barletta et al., 2008; Cai et al., 2010; Gros et al., 2011). The major anthropogenic emissions of VOCs in India are from residential combustion (41%), transportation (37%), and industrial non-combustion (13%; Li et al., 2014). The BTEX concentration in ambient air exhibits strong seasonal and diurnal variations associated with the change in sources and atmospheric processes.

The lockdown due to the COVID-19 pandemic led to reductions in emissions of greenhouse gases (GHGs) and air pollutants, including particulate matters (PM2.5 and PM10) and gaseous pollutants (e.g., NO2, O3, CO, SO2, etc.) in many countries (e.g., Forster et al., 2020; Shi & Brasseur, 2020; Singh et al., 2020; Sokhi et al., 2021). The associated decrease in air pollution has been observed from satellite and ground-based observations (Bauwens et al., 2020; Biswal et al., 2021; Shi & Brasseur, 2020). The observations over China and India showed larger reductions in NOx emissions than predicted 28% and 48%, respectively (Forster et al., 2020). However, the changes in the ambient concentrations of VOCs during the lockdown have been rarely reported, mainly due to the lack of measurements (Fu et al., 2020; Resmi et al., 2020). In India, the measurement-based studies of aromatic VOCs are limited only to a few cities (e.g., Dave et al., 2020; Hoque et al., 2008; Majumdar et al., 2011; Sahu, Pal, et al., 2016; Sahu, Yadav, & Pal, 2016; Sahu et al., 2020; Sahu & Saxena, 2015). Nonetheless, in-situ ground-based VOC measurements are important to constrain and reduce the uncertainties in the emission inventories of air pollutants (Borbón et al., 2013; Hu et al., 2015). There is a lack of understanding of the emission, concentration, composition, and spatiotemporal variations of BTEX in India.
The transport, construction, and industrial activities in India were almost completely shut down during the COVID-19 lockdown period. The change in the fuel consumption data during the lockdown suggests 50%–60%, 90%, 40%, and 70% reductions in transport, aviation, industrial, and construction activities, respectively (PPAC, 2020). The Google mobility data show declines of up to 70% in transport movement activities (Biswal et al., 2021) across India during the lockdown. The studies conducted in several cities of India reveal a considerable improvement in air quality during the lockdown period (Singh et al., 2020, and references therein). However, changes in ambient VOC levels due to COVID-19 lockdown are rarely reported, mainly due to the lack of ongoing measurement programs over the Indian regions. The present study is based on time-resolved continuous measurements of aromatic VOCs (BTEX) in Ahmedabad metropolitan city in western India during January–May 2020 and 2014, 2015, and 2018. The primary objective is to assess the impact of a nationwide lockdown against the COVID-19 pandemic on ambient BTEX concentration and composition. We have estimated the changes in the ventilation coefficient (VC) normalized BTEX concentrations during the lockdown with reference to the pre-lockdown and equivalent lockdown periods of 2014-2015-2018, representing business as usual (BAU) conditions. The effects of meteorological and photochemical processes are investigated in detail.

2. Materials and Methods

2.1. Description of Study Site

The measurement site (23.036°N, 72.54°E; 55 m AMSL) in the campus of Physical Research Laboratory (PRL) is in the western part of Ahmedabad city. According to the census 2011, it is the 5th largest city of India with over 6.5 million population (Joshi, 2014). A layout map describing the measurement site and major roads of the city is presented in Figure S1 in Supporting Information S1. The city, with an area of ~464 km², is located along the banks of the Sabarmati river. Many large- and small-scale industries (textile, pharmaceutical, etc.) are located in the eastern and northern outskirts. The landfill site at Pirana, where municipal solid waste (MSW) materials have been dumped for over 30 years, can be located in the southern part of Ahmedabad along the east bank of the Sabarmati river. The study site is located in a highly developed and urbanized western part of the city, where emission from the transportation sector is the major local source of air pollutants (Chandra et al., 2016). The study site is surrounded by trees on all sides (within ~1 km) and is about 15–20 km from the industrial areas. In the fiscal year 2015-2016, the city had a total of ~3.6 million registered vehicles comprising of two wheelers (72%), four wheelers (15%), three wheelers (5%), and bus/truck (2%), which are increasing at the annual rate of ~10% (Source: RTO, Ahmedabad). Gasoline-fueled two-wheeled vehicles are known as large emitters of aromatic VOCs compared to other VOCs (Saxer et al., 2006; Platt et al., 2014). The emission standards (4-wheel vehicles) of India 2000, Bharat Stage II (Euro 2), Bharat Stage III (Euro 3), Bharat Stage IV (Euro 4), and Bharat Stage VI (Euro 6), were implemented during the years 2000, 2003, 2005, 2010, and 2020, respectively (https://dieselnet.com/standards/in/). It has been made mandatory for all vehicles to have the Pollution Under Control (PUC) certificate. Unlike other Indian cities, the contributions of biomass/biofuel burning emissions to ambient pollutants in Ahmedabad are significantly small compared to fossil-fuel combustion and industrial emissions (Sahu et al., 2017). However, particularly in the winter season, sporadic activities of local biomass burning can be noticed in the surrounding regions (Tripathi & Sahu, 2020). The Indo-Gangetic Plain (IGP), the Thar Desert, and the Arabian Sea are situated to the northeast, northwest, and southwest of Ahmedabad, respectively. We have also used the data from a continuous monitoring station of Maninagar, Ahmedabad, as a reference for the BAU conditions (2015 and 2018).

2.2. Instrumentation

An online C₂-C₆ VOC analyzer (AirmoVOC, Model: A22022, Chromatotec®, Saint-Antoine, France) is a thermal desorption-gas chromatography-flame ionization detector (TD-GC-FID) system used for the measurement of aromatic VOCs in ambient air. NMHCs including BTEX were pre-concentrated by pumping several liters of ambient air per minute through a 5-m-long Teflon tube (1/2” id) to a manifold and subsampling through a 1-m-long stainless steel (SS) line (1/4” id) to a Peltier-cooled adsorbent trap. Ambient air samples pre-concentrated on the adsorbent trap are desorbed into a metallic column (MXT30CE, 30 m × 0.28 mm, 1 μm film thickness, Restek Corp., USA). The GC column temperature was programmed with an initial temperature of 40°C, increased to 50°C (2°C min⁻¹), to 80°C (10°C min⁻¹), to 220°C (15°C min⁻¹), to 230°C (2°C min⁻¹), and finally to 260°C (9°C min⁻¹). An ultrahigh purity Hydrogen (H₂) gas (generated using Hydroxychrom USB...
4U, Model No: XXX916) was used as a carrier gas. Zero-air (produced using airmoPURE D 45 PSI, Model No: XXX031-D) and H₂ were used as fuel gases for the FID. The H₂ (FID and carrier gas) and zero-air flow rates were set at 30 mL min⁻¹ and 180 mL min⁻¹, respectively. A piezo valve controlled the H₂ carrier gas pressure at the heated column head. Systematic quality assurance (QA) and quality control (QC) procedures were performed for very accurate concentration measurements. The procedure includes regular evaluation of VOC response factors using a certified calibration mixture, chromatographic retention times, missing values, and measurements below the detection limit. The VISTACHROM® software developed by Chromatotec® was used for the calculation of retention time and peak area. Further details of the NMHC sampling and GC-FID separation, analysis, and calibration using a similar system are given elsewhere (e.g., Gros et al., 2011). The chromatographic peaks of different BTEX compounds were calibrated using dynamic dilutions of a standard calibration mixture containing \( \sim 1.0 \text{ ppmv (±5%) of each compound (LE732C 4/15, Linde, USA). The Dynamic Dilution Calibrator (DDCZ094, AM-5800) was used for multipoint calibration of the VOC analyzer. The DDC consists of two mass flow controllers (MFCs) to set the standard gas flow (0–100 sccm) and dilution gas flow (0–20 SLPM). The zero-air supply from the Zero Gas Generator (ZGGZ095, AM 5900) was used as a dilution gas in the DDC. The zero-air gas generator system consists of an oil-free compressor, pressure flow controller, water removal system, SO₃, NO, O₃, H₂S, CO, and hydrocarbon removal. Further, high-purity zero-air gas was generated by connecting the ultra-purity zero-air generator (Parker HPZA-3500-220) in tandem with the GCUZGGZ095 instead of using direct ambient air.

The response factors of BTEX compounds were quantified using multipoint calibration. As shown in Figure S2 in Supporting Information S1, the relations between the set (known) mixing ratio (ppbv) and normalized peak area of BTEX compounds show excellent linear response \( r^2 = 0.97–0.99, p < 0.05 \). The measurement precisions of BTEX analysis in five replicates of the standard mixture were in the range of 2%–7%. The limit of detection (LOD) was determined for each compound by the following equation (Dave et al., 2020):

\[
\text{LOD} = 3 \times \frac{\sigma_{\text{blank}}}{\text{Sensitivity}}
\]

where \( \sigma_{\text{blank}} \) represents the standard deviation of blank signals obtained using the analysis of zero-air. The sensitivity or response factor of a BTEX compound was determined using the linear fit of multipoint calibration. The LOD of benzene was 20 pptv, while those of toluene, ethylbenzene, and xylenes were 2–4 pptv. The relative standard deviations (RSD) or precisions of the measurements at different set values of BTEX were in the range of 2%–5%. The 1σ measurement uncertainties were calculated as the root sum of squares of the precision and all experimental uncertainties, such as those associated with flow rates and certified standard concentrations. The overall measurement uncertainties of BTEX were in the range of 8%–11%, which take account of the uncertainties of standard/calibration mixture (±5%) and flow rates (±2%) of standard and dilution (zero-air) gases. An additional consistency test was performed by investigating the correlation between isomers of xylene (m,p-xylenes and o-xylene), which have similar emission sources and lifetimes in the atmosphere. The mixing ratios of m,p-xylenes and o-xylene showed a strong correlation \( r^2 > 0.95, p < 0.05 \) during the study period. The BTEX concentrations during 2014 were measured using a high-resolution proton transfer reaction-time of flight-mass spectrometer (PTR-TOF-MS) instrument (Ionicon Analytik GmbH Innsbruck, Austria).

2.3. Meteorology, Long-Range Transport, and Lockdown

In the South Asia region, the long-range transport of air masses from different source regions is driven primarily by the progression of the Inter Tropical Convergence Zone (ITCZ; Asnani, 2005). The ITCZ shifts northward during northern hemisphere summer and southward during northern hemisphere winter. The circulations over the South Asia region in the summer and winter seasons are characterized by the southwesterly and northeasterly winds, respectively. The back trajectory model has been used to track the transport of different air masses during the study period. The 7-day isentropic backward air mass trajectories at 500 m AMSL were calculated using the Meteorological Data Explorer (METEX) of the Centre for Global Environmental Research (CGER), National Institute for Environmental Studies (NIES), Japan (http://db.cger.nies.go.jp/metex/trajectory.html). As shown in Figure S3 in Supporting Information S1, the monthly pattern of trajectories suggests the transport of air masses from the distinct regions associated with the change in meteorological conditions. In this study, we have presented
a detailed analysis of the dependence of BTEX mixing ratios on meteorological parameters, particularly on wind speed. The meteorological data were taken from the wunderground archive (www.wunderground.com).

The wind rose plots in Figure S4 in Supporting Information show the monthly contributions of wind from the different sectors. In January, the daily means of temperature, WS, RH, and pressure were 15.5–22.3°C, 1.8–4.2 m s\(^{-1}\), 45%–75%, and 1011–1018 mb, respectively. Most air mass trajectories indicate the long-range transport from the NE originated over the IGP during the January-February period. In March, the mean values of temperature, WS, RH, and pressure were 21.8–31.1°C, 1.9–4.1 m s\(^{-1}\), 24%–72% and 1008–1015 mb, respectively. In April, representing the summer conditions, the daily means of temperature, WS, RH, and pressure were 28.3–38.5°C, 1.8–4.2 m s\(^{-1}\), 22%–45%, and 1006–1011 mb, respectively. The back trajectories for April and May indicate the dominant transports from the NW and west directions, respectively. In May, the daily mean values of temperature, WS, RH, and pressure were 34.1–37.3°C, 2.3–4.9 m s\(^{-1}\), 23%–52%, and 1002–1007 mb, respectively. However, the transport of BTEX compounds from distant source regions will have a minimal contribution due to their short atmospheric lifetimes. On the other hand, the mixing with the air masses originated over distinct regions can change the background concentrations of BTEX, particularly during the intrusion of air masses from the free troposphere. The planetary boundary layer height (PBLH) was deeper in the summer (April-May) but shallower during the winter period (January-February).

The lockdown in India was imposed in four phases starting from 25 March–31 May 2020. Prior to the full lockdown, on 22 March 2020, a voluntary Janta (public) Curfew, for 14 hr, was ordered in India. The full lockdown was imposed on 25 March 2020 for 21 days (Phase I) and then extended until the end of May in 3 more phases, namely 15 April to 03 May (Phase II), 04–17 May (Phase III), and 18–31 May (Phase IV).

### 2.4. Dispersion Normalization

The atmospheric abundance of BTEX compounds is determined through the interplay of emissions, meteorology, transport, and loss processes. Thus, to quantify the impact of changes in any of these, an analysis must isolate the influence of the remaining important parameters. As the transition from a pre-lockdown condition to a lockdown condition occurred during the winter-to-summer transition, it is essential to account for the meteorological impact when estimating the effect of emission changes. We used the dispersion normalization method to reduce the influence of meteorological-driven dispersion on the pollutants. Previous studies (e.g., Dai et al., 2020; Gu et al., 2022; Mishra et al., 2021) have used the VC-normalization method to reduce the influence of local meteorology (dispersion) on the observed concentrations of pollutants for the estimation of the changes due to COVID-19 lockdown restrictions. In this method, the observed daily mean BTEX concentrations are normalized by the average VC estimated using the following equation.

\[
C_{\text{norm},i} = C_i \times \frac{V C_i}{V C_{\text{mean}}}
\]

where \(C_{\text{norm},i}\) is the dispersion normalized concentration and \(C_i\) is the measured concentration during period \(i\). \(V C_{\text{mean}}\) is the average value of VC over the entire sampling period. The VC has been calculated by multiplying the PBLH and WS obtained from ERA5 hourly data (https://cds.climate.copernicus.eu/) over Ahmedabad (e.g., Tiwari et al., 2016). In brief, VC is an indicator of the ability of the atmosphere to dilute and disperse the concentrations of ambient air pollutants over a particular region. The winter-to-summer transition can change the photochemical process, and its effect is discussed in this paper (Sections 3.2 and 3.7).

### 3. Results and Discussion

#### 3.1. Times Series of BTEX and Meteorological Parameters

As shown in Figure 1, the daily (0–23 hr), afternoon (14–17 hr), and evening-nighttime (18–24 hr) mean mixing ratios of actual ambient BTEX show large variations. During January-February, the daily means of benzene, toluene, ethylbenzene, m,p-xylene, o-xylene, and their sum (\(\sum\) BTEX) were in the ranges of 1.9–8.5, 1.2–29.6, 0.17–2.6, 0.31–5.5, 0.17–2.7, and 4.9–36.4 ppbv, respectively. All BTEX species showed significant decreasing trends in March during the transition from pre-lockdown to lockdown. During the lockdown period (25 March–31
May), the daily mean mixing ratios of benzene, toluene, ethylbenzene, and xylenes compounds in ambient air at the urban site of Ahmedabad, India, from 01 January to 31 May 2020.

The daily mean time series of $\Sigma$BTEX, $\Sigma$TEX, meteorological parameters, and PBLH exhibit substantial day-to-day variations (Figure 2). Despite large daily variability, the episodes of higher and lower BTEX concentrations coincided with the periods of weak and stronger winds, respectively. Therefore, the daily mixing ratios of BTEX compounds and WS seem to show anti-correlated variations. For example, the lower BTEX concentrations...
(∑BTEX <0.5 ppbv) during 23–26 April and 2–5 May coincided with relatively high wind speeds (>3 m s⁻¹), while higher levels (>2.0 ppbv) during 28–30 April under calm low-wind conditions. In the normal years, the measurements of benzene and other primary VOCs show strong day-to-day and episodic variations during the winter to summer transition period at the same site (Sahu et al., 2017). The seasonal variability of BTEX in the urban regions strongly depends on sources' strength, meteorological conditions, and concentrations of OH radicals and other oxidants (Filella & Peñuelas, 2006; Jiang et al., 2017). It is well reported that ambient concentrations of air pollutants are strongly affected by meteorology with low wind speeds causing a buildup of air pollutants (Lee et al., 2020). But the levels of BTEX measured under the strong wind conditions during the lockdown period are lower than their pre-lockdown values. However, the study period from pre-lockdown to lockdown also coincides with the transition from winter to summer. Hence, we have examined the impact of the winter to summer transition on meteorological and photochemical processes.

Figure 2. The time series variations of the daily mean (±1σ) (all data), afternoon, and nighttime ambient air mixing ratios of ∑BTEX, ∑TEX, PBL, and meteorological parameters at the urban site of Ahmedabad, India, from 01 January to 31 May 2020.
Typically, the higher emissions combined with the accumulation due to the slower chemical removal and lower PBLH lead to elevated levels of BTEX and other pollutants in the winter season. In contrast, the faster chemical removal and efficient dispersion due to higher PBLH are the key factors, leading to decreased ambient air BTEX concentrations during the summer season. The higher evaporative emissions in summer than those in winter could increase the levels of certain BTEX compounds. These counter-effects can reduce seasonal variation of the BTEX concentration but not necessarily the seasonal change in BTEX composition. However, mainly to account for the seasonal variation, we have also estimated the changes from the VC-normalized BTEX concentrations measured during the same period of normal/BAU years of 2014-2015-2018 at PRL and Maninagar stations in Ahmedabad. The data for the Maninagar station are taken from the Continuous Ambient Air Quality Monitoring Station (CAAQMS) of the Central Pollution Control Board (https://cpcb.nic.in). The BTEX data for intermittent years of 2016, 2017, and 2019 are not available. The year-to-year change in factors controlling BTEX levels remains a cause of some uncertainty in quantifying the impact of the lockdown.

### Table 1

| Sampling period | N | Benzene ± 1σ | Toluene ± 1σ | Ethylbenzene ± 1σ | Xylenes ± 1σ | ∑BTEX ± 1σ | ∑TEX ± 1σ | VC-normalized monthly reduction (%) with reference to the mean of years 2014-2015-2018 |
|-----------------|---|--------------|--------------|-------------------|-------------|----------|---------|-----------------------------|
| January 2020    | 1085 | 5.25 ± 1.1  | 3.85 ± 2.5  | 0.58 ± 0.3        | 1.80 ± 1.3  | 11.50 ± 9.9 | 10.90 ± 8.1 | 85 | 59 | 1 | −1 | 19 | 57 |
| February 2020   | 674  | 4.28 ± 1.9  | 7.84 ± 6.9  | 0.76 ± 0.7        | 2.48 ± 2.2  | 15.37 ± 16.6 | 14.61 ± 14.8 | −83 | −11 | −14 | −6 | −24 | 7 |
| March 2020      | 971  | 1.88 ± 0.8  | 2.69 ± 2.2  | 0.31 ± 0.2        | 0.91 ± 0.6  | 5.80 ± 5.5  | 5.49 ± 4.6  | 24 | 68 | 48 | 39 | 58 | 72 |
| April 2020      | 898  | 0.40 ± 0.2  | 1.43 ± 0.9  | 0.09 ± 0.06       | 0.33 ± 0.3  | 2.26 ± 2.9  | 2.16 ± 2.1  | 72 | 72 | 70 | 75 | 72 | 76 |
| May 2020        | 870  | 0.19 ± 0.05 | 0.54 ± 0.5  | 0.05 ± 0.04       | 0.11 ± 0.03 | 0.89 ± 1.2  | 0.84 ± 1.0  | 86 | 88 | 84 | 85 | 87 | 86 |

*N indicates the number of samples. *Negative values indicate enhancement.

### 3.2. Change in BTEX Concentration During the Lockdown

The monthly mean ambient mixing ratios of BTEX compounds during January–May 2020 and changes due to the COVID-19 lockdown with reference to the mean of years 2014-2015-2018 are presented in Table 1. During April-May, the estimated mean reductions in the actual ambient mixing ratios of benzene, toluene, ethylbenzene, and xylenes were 81%–90%, 78%–90%, 87%–93%, and 75%–89%, respectively. The ∑BTEX and ∑TEX concentration reductions were 78%–91%. During this period, the mean reductions in the VC-normalized mixing ratios of benzene, toluene, ethylbenzene, xylenes, ∑BTEX, and ∑TEX were 72%–86%, 72%–88%, 70%–84%, 75%–85%, 72%–87%, and 76%–86%, respectively. In May, the highest reductions of both ambient concentrations (≥90%) and VC-normalized concentrations (≥85%) of each BTEX compound were due to the stringent lockdown measures. In March, the moderate decreases are due to the measurements under transition from pre-lockdown (1–24 March) to lockdown conditions.

The actual and VC-normalized BTEX concentrations and values of meteorological parameters measured during January, February, pre-lockdown, and different phases of lockdown are plotted in Figure 3. The actual concentration of each BTEX compound shows a substantial decline from pre-lockdown (∑BTEX = 5.95 ± 3.6 ppbv) until the end of phase II or 03 May (∑BTEX = 1.41 ± 1.1 ppbv). This decline in BTEX concentrations was accompanied by a rapid increase in air temperature from 25.8 ± 2.7 to 34.5 ± 1.4°C, while no significant changes were observed in other meteorological parameters. The concentrations of BTEX compounds during phase III (∑BTEX = 0.92 ± 0.3 ppbv) and phase IV (∑BTEX = 0.78 ± 0.4 ppbv) were much lower than their values measured during the pre-lockdown period. During each phase of the lockdown, the reduction in the nighttime concentration was higher than the daytime concentration for each BTEX compound. In phase I, with respect to...
the means of the respective period of years 2014-2015-2018, the mixing ratios of benzene, toluene, ethylbenzene, and xylenes declined by ∼67%, 75%, 80%, and 63%, respectively (Table S1 in Supporting Information S1). The concentrations during phases II–IV show significantly higher reductions of 85%–91%, 84%–92%, 85%–94%, and 86%–89%, respectively. In phase I, the VC-normalized mixing ratios of benzene, toluene, ethylbenzene, and xylenes decreased by 64%, 73%, 54%, and 76%, respectively (Table 2). In comparison, there were higher reductions in the VC-normalized concentrations of these compounds by 74%–85%, 77%–87%, 82%–86%, and 79%–87%, respectively, during phases II–IV. These VC-normalized changes estimated for different phases could be primarily due to the lockdown-related emission drops. However, the prevailing meteorological factors account for about 10% reductions during the lockdown period.
The daily mean (±1σ) time series of the actual ambient air and VC-normalized mixing ratios of benzene, toluene, ethylbenzene, and xylenes measured during January–May 2020 are compared with the available data for January–May 2014-2015-2018 in Figure 4. During the January–March period, the actual and VC-normalized concentrations of aromatic VOCs show nearly similar trends for 2020 and 2014-2015-2018. In the summer season (April-May) of 2014-2015-2018, the BTEX mixing ratios show a slight decline compared to their values in winter. In contrast, both the actual and VC-normalized concentrations of all BTEX compounds during April-May 2020 showed significant reductions. Therefore, it is reasonable to attribute the reduction of anthropogenic emissions as the primary factor, which led to drastic declines in ambient concentrations of aromatic VOCs during the COVID-19 lockdown period.

The effect of photochemistry/radiation in the nighttime trends of BTEX can be assumed to be negligible due to negligible removal caused by the photooxidation due to OH radicals. As shown in Figures 1 and 2, the changes in the nighttime BTEX concentrations from pre-lockdown to lockdown remain substantial. However, the estimated changes using the VC-normalized nighttime data are slightly lower (∼8%) than VC-normalized estimates using all data. The differences between the nighttime and afternoon trends could be attributed to the photooxidation loss of BTEX by the OH (indirect effect of radiation). Further, we have estimated the photochemical age and used it to analyze the change in BTEX compositions in Section 3.7.

### 3.3. Mobility and Emissions Change

The google mobility change data set has been used as a proxy for the anthropogenic activity change during the lockdown (Archer et al., 2020; Biswal et al., 2021; Forster et al., 2020; Gama et al., 2020; Guevara et al., 2021). Here, we have analyzed the mobility change for Ahmedabad city from 01 March–31 May 2020. The estimated daily and phase-wise reductions for retail and recreation, grocery and pharmacy, parks, transit stations, work places, and residential locations are shown in Figures 5a and 5b, respectively. By considering the workplace and transit stations as proxies for traffic emission as used by Forster et al. (2020), a decline of up to 76% in traffic emissions can be attributed to the lockdown restrictions. The decline further reduced gradually to 58% in the fourth phase of the lockdown. However, the travel restrictions increased the number of residents living at residential locations by 39%. As shown in Table S2 in Supporting Information S1, the level of mobility in Ahmedabad changed significantly during the pre-lockdown and different phases of the lockdown.

### 3.4. Diurnal Variation of BTEX

The significant periodic variations of BTEX in short time scales (<1 day) represent their strong diurnal cycle. The average diurnal plots of actual ambient air BTEX concentrations for different months are shown in Figure 6. The weekend (Sunday) data have been analyzed separately to investigate the impact of the weekend effect. The concentrations of all BTEX compounds exhibit significant diurnal variability with two peaks, a primary peak between 20 and 22 hr and a secondary peak between 8 and 10 hr. The levels were high from the evening to

| Sampling period | Benzene   | Toluene   | Ethylbenzene | Xylenes   | ∑BTEX    | ∑TEX    |
|-----------------|-----------|-----------|--------------|-----------|----------|---------|
| Phase I (25 March–14 April 2020) | 0.88 ± 0.5 | 1.70 ± 0.7 | 0.13 ± 0.1 | 0.47 ± 0.2 | 3.18 ± 1.5 | 2.24 ± 1.0 |
| Phase II (15 April–3 May 2020)   | 0.26 ± 0.1 | 0.96 ± 0.9 | 0.05 ± 0.03 | 0.16 ± 0.09 | 1.43 ± 1.1 | 1.17 ± 0.9 |
| Phase III (4–17 May 2020)        | 0.21 ± 0.1 | 0.58 ± 0.2 | 0.05 ± 0.01 | 0.12 ± 0.04 | 0.97 ± 0.3 | 0.76 ± 0.3 |
| Phase IV (18–31 May 2020)        | 0.18 ± 0.1 | 0.48 ± 0.3 | 0.05 ± 0.01 | 0.12 ± 0.02 | 0.86 ± 0.4 | 0.64 ± 0.3 |

| VC-normalized phase-wise reduction(%) with reference to the mean of years 2014-2015-2018 |
|-----------------------------------------------|
| Phase I | 64 | 73 | 54 | 76 | 69 | 71 |
| Phase II | 74 | 77 | 84 | 83 | 75 | 75 |
| Phase III | 85 | 87 | 84 | 88 | 87 | 88 |
| Phase IV | 83 | 86 | 82 | 79 | 84 | 87 |
Figure 4. Comparison of time series variations of the daily (a) average (±1σ) of actual ambient air and (b) average of VC-normalized mixing ratios of different benzene, toluene, ethylbenzene, and xylenes compounds measured in Ahmedabad, India, during 01 January–31 May of the years 2014, 2015, 2018, and 2020.

Figure 5. The 7-day moving mean changes in the mobility for retail and recreation, grocery and pharmacy, parks, transit stations, workplaces, and residential locations in Ahmedabad city. (a) Daily change in mobility and (b) phase-wise change in mobility.
morning hours, while low concentrations were measured in the afternoon (14–17 hr). Overall, there are several similarities and differences between the monthly diurnal variations of BTEX compounds. Although the pattern remains the same, the levels and particularly the peak values of each BTEX compound decreased from January–May 2020. The mean evening (afternoon) $\sum_{BTEX}$ concentrations of 16.07 (6.2), 21.5 (7.3), 5.7 (2.3), 3.3 (0.53), and 2.2 (0.43) ppbv were measured in January–May, respectively. The declines of both afternoon and evening concentrations during the April-May period were particularly significant. The dependencies of anthropogenic emission strength and meteorological parameters on local time can be important factors, controlling the

Figure 6. The monthly diurnal variations of ambient air mixing ratios of benzene, toluene, ethylbenzene, and xylenes during January–May 2020 at the urban site of Ahmedabad, India. Gray circles for all the data points measured at ~40 min of time resolution, black circles for the mean ±1σ values, and red circles for Sunday. The monthly mean data of benzene, toluene, and xylenes measured from 2014-2015-2018 are plotted (open blue circle) for the comparison.
diurnal variation of primary pollutants in urban areas (Sahu et al., 2011). The primary and secondary peaks of BTEX concentrations coincide with evening and morning rush-traffic hours, respectively. For each month, relatively small and large BTEX amplitudes measured during the morning and evening rush-traffic hours coincide with the periods of strong and weak winds, respectively (Figure S5 in Supporting Information S1). The low concentrations in the afternoon can be attributed mainly to the dilution associated with the increased PBLH, stronger winds, and reductions in vehicular emissions. The oxidation reactions with OH radicals can also result in gradually decreasing mixing ratios of BTEX in the afternoon hours. From evening till early morning, the formation of the shallower nocturnal boundary layer (NBL) under the calm winds favored the accumulation of pollutants causing higher BTEX concentrations. Overall, the diurnal cycles of the PBLH and BTEX concentrations show anticorrelated variation for each month.

In the winter season, the benzene concentration does not show clear weekday-weekend differences, indicating significant contributions from non-vehicular sources. However, the concentrations of TEX compounds during the weekend were 15%–20% lower than the values measured during the weekdays. In the lockdown period, the concentrations of all BTEX compounds decreased by 15%–25% during the weekend. On the diurnal scale, the weekday-weekend differences were highest during the evening rush hours and lowest during the afternoon hours. For each month, except for benzene in the winter season, the lower levels of BTEX measured during the weekend were consistent with the reduction of vehicular traffic. The weekend-weekday differences in the concentrations can be attributed mainly to the change in emission, but variation in meteorological conditions could also be a factor.

The monthly mean diurnal variations of BTEX compounds measured for 2020 and 2014-2015-2018 are compared in Figure 6. Although the levels were different, the diurnal patterns of BTEX remain similar for each month of 2020 and 2014-2015-2018. During the January-March period, the BTEX concentrations during 2014-2015-2018 were slightly higher than their respective levels in the year 2020. In contrast, the average diurnal patterns of BTEX in April and May show large differences between 2020 and 2014-2015–2018. For each BTEX compound, the differences were largest during 18–24 hr. During the lockdown period, the peak concentrations of BTEX were 4–5 times lower than their peak values measured during the 2014-2015-2018 period.

3.5. Relative Change in BTEX Composition Due To the Lockdown

The daily time series percentage contributions of different compounds to $\sum$BTEX show large day-to-day variations (Figure 7). Benzene and toluene were the two most abundant species throughout the study period. In the winter and particularly in January, the contributions of benzene were higher than that of toluene. The contribution of toluene increased gradually during the winter-summer transition period and it became the most abundant BTEX compound in summer during the April-May period. The overall daily mean contributions of benzene, toluene, ethylbenzene, m,p-xylenes, and o-xylene were 9%–65%, 22%–86%, 1%–14%, 2%–16%, and 1%–9%, respectively. During the evening-night period, the mean contributions of benzene, toluene, ethylbenzene, m,p-xylenes, and o-xylene were in the ranges of 8%–66%, 23%–84%, 2%–23%, 3%–21%, and 2%–10%, respectively. However, in the afternoon, the abundances of more reactive compounds (i.e., ethylbenzene and xylenes) show decline compared to their fractions measured in fresh emissions during the evening-night period. Several factors related to the emission and atmospheric processes can cause day-to-day changes in the BTEX composition. The large day-to-day variations of BTEX composition, particularly in January, characterize the emissions from both vehicular and non-vehicular sources. The TEX compounds constitute ~42% (January), 59% (February), 58% (March), 64% (April), and 73% (May) to the total BTEX loads. This trend in the contribution of TEX compounds
associated with the increasing trend of ambient temperature suggests the increasing evaporation and solvent emissions during the winter-to-summer transition period. The TEX compounds are predominantly emitted from fuel evaporation and industrial sources, such as painting, architectural coating, manufacturing, printing, degreasing solvents, etc (e.g., Monod et al., 2001). Although there is a significant increase of TEX, still it reflects the lower contributions from the evaporation and solvent emissions as the reaction rates of TEX compounds with OH radicals are higher than that of the reaction of benzene with OH.

The compositions measured during evening-night and afternoon hours are compared to assess the effect of fresh emissions and photochemical processing on the observed BTEX proportions. The trend of BTEX composition noticed in the evening-night data indicates the varying contributions of major sources, such as vehicular traffic and industrial emissions and the change in meteorological conditions. However, the variation of BTEX composition in the afternoon reflects significant photochemical processes and sensitivity of emissions to meteorological variability. As presented in Figure S6 in Supporting Information S1, the monthly mean diurnal percentage contributions reflect the impact of major factors controlling the diurnal dependence of BTEX composition.

The mean compositions of BTEX measured during the night and afternoon hours of the pre-lockdown and different phases of the lockdown are shown in Figure 8. In the nighttime, the composition of BTEX does not show significant variation between different phases of the lockdown. During phases I–IV, the nighttime percentage contributions of benzene, toluene, ethylbenzene, m,p-xylenes, and o-xylene varied in narrow ranges of 23.8%–26.8%, 53%–54.4%, 2.1%–9.8%, 4.7%–6.3%, and 4.6%–5.9%, respectively. While their contributions in the afternoon show large variations of 23.9–38.5, 52.8–59.6, 2.3–12.4, 4.9–9.7, and 2.9–12.8, respectively. The composition of BTEX in the nighttime is governed mainly by the emission source, which does not seem to show significant changes between the different phases. The differences in the daytime BTEX composition between the different phases suggest the role of meteorological and photochemical processes. The highest contribution of benzene in January could be due to the enhanced biomass/biofuel burning emissions. In the above discussion, particularly the changes in relative BTEX composition during different phases, we have assumed that the impact of meteorological conditions is negligible. Further, the diagnostic ratio, photochemical, and meteorological parameters are analyzed to understand the composition change.

### 3.6. Diagnostic Ratios and Emission Sources

The ratios of BTEX in ambient air provide useful information to identify the emission sources, photochemical processes, as well as the transport and dilution processes (Han et al., 2020; Sahu et al., 2020; Dave et al., 2020). The relative BTEX concentration (B:T:E:X) data have been used to obtain the fingerprints of different emission sources, such as biomass/biofuel burning, vehicle exhaust, and industrial sources (e.g., Zhang et al., 2016). The monthly mean diurnal variations of the toluene/benzene (T/B), m,p-xylenes/ethylbenzene (m,p-X/E), and o-xylene/ethylbenzene (o-X/E) ratios are shown in Figures 9a–9c. Each of these ratios shows the lowest and highest enhancements in the afternoon and evening hours, respectively. The T/B ratio exhibited a strong peak during the evening rush-hour traffic due to the buildup of fresh vehicular emissions under a nocturnal inversion. From evening to morning, the T/B ratios in January–May varied in the ranges of 0.43–1.06, 0.77–2.04, 1.1–1.77, 1.82–4.04, and 1.72–4.03 ppb ppb⁻¹, respectively. The gradual decline of T/B from midnight to early morning reflects the change in major emissions from petrol-to diesel-fueled vehicles. This is consistent as the total mass emission of BTEX for diesel vehicles is mainly composed of benzene (60%), whereas xylene and toluene are dominant compounds in gasoline vehicles (Louis et al., 2016). In January, the lower T/B ratios reflect the significant influence of biomass burning emissions in addition to automobile exhaust. Very low T/B ratios of ~0.5 (Karl et al., 2007) and 0.37 (Simpson et al., 2011) have been reported for biomass burning emissions. The higher T/B ratios during the April-May lockdown period indicate significantly higher contributions from fuel evaporation and industrial processes than those in the winter season. In the tropical urban regions, evaporative emissions represent a substantial fraction of atmospheric VOCs at higher temperatures marked by the higher T/B ratios during April-May (Liu et al., 2015). Tunnel and roadside measurements indicate that the ambient T/B ratios of 1–2 characterize the impact of fresh vehicular emissions (Huang et al., 2015; Warneke et al., 2013). The lower ratios (<0.6) and higher ratios (>3) are attributed mainly to biomass burning and industrial sources, respectively (Akagi et al., 2011; Zhang et al., 2015).

The xylene/benzene ratio has also been used as a surrogate for traffic-related emissions when vehicle exhaust and evaporative losses during storage and refueling are dominant sources. Typically, the exhaust has distinctly higher
benzene proportions and lower xylene proportions compared to unburned fuel (Drozd et al., 2019). Therefore, the xylene/benzene ratio has been used to estimate the photochemical activity in urban regions (Seguel et al., 2013). In the daytime, the lower xylene/benzene ratios are due to a higher reaction rate of xylene with OH (m-xylene: $k_{\text{OH}} = 23.6 \times 10^{-12}$ cm$^3$ molecule$^{-1}$ s$^{-1}$; p-xylene: $k_{\text{OH}} = 14.3 \times 10^{-12}$ cm$^3$ molecule$^{-1}$ s$^{-1}$) compared with that of benzene ($k_{\text{OH}} = 1.2 \times 10^{-12}$ cm$^3$ molecule$^{-1}$ s$^{-1}$; Atkinson, 1990).

Diagnostic BTEX ratios with ethylbenzene-based normalization have been used to identify the dominant sources (Buczynska et al., 2009). Ethylbenzene and the xylene isomers are co-emitted and have source fingerprints that do not differ significantly (Nelson & Quigley, 1983). The diurnal profile of m,p-X/E ratio shows two sharp peaks, coinciding with the morning and evening rush-hour traffic. In the daytime, the declines of m,p-X/E ratio indicate the impact of photochemical aging as the reaction rate of m,p-xylenes with OH radicals is about three times faster than that of ethylbenzene (Huang et al., 2015). As shown in Figure 9d, the scatterplot between m,p-xylenes and ethylbenzene mixing ratios shows a strong correlation ($r^2 = 0.97$), confirming their common emission.

Figure 8. The compositions of the benzene, toluene, ethylbenzene, and xylenes species in ambient air at (a) night and (b) afternoon hours during the different periods and phases of COVID-19 lockdown at the urban site of Ahmedabad, India, from 01 January to 31 May 2020.
sources (Jiang et al., 2017). Most of the data measured during the evening to early morning hours fall along the ∆m,p-X/∆E slope line of 2.47, while the daytime measurements fall well below the slope line. Similar to the present study, Monod et al. (2001) have reported the ∆m,p-X/E ratios of ∼2.35 in urban air mainly influenced by traffic-related emissions. The m,p-X/E ratio in the present study is comparable to those reported for several urban areas, such as ∼3.4 in Munich (Rappenglück & Fabian, 1999), 3.0 in Sydney (Nelson et al., 1983), and 2.9 in different cities of UK (Derwent et al., 2000). During the April-May period, a slightly higher m,p-X/E ratio could be due to the higher contributions from the evaporative and industrial sources than those in the winter. Monod et al. (2001) have reported the higher m,p-X/E ratios of 3.8–4.5 for evaporative emissions than vehicular exhaust alone.

Toluene and ethylbenzene are emitted from both combustion and evaporation processes and have similar OH reaction rate constants. As shown in Figure 9f, the toluene and ethylbenzene mixing ratios exhibit a good correlation

Figure 9. (a–c) Diurnal variations of toluene/benzene (T/B), m,p-xylene/ethylbenzene (m,p-X/E) and o-xylene/ethylbenzene (o-X/E) ratios (ppb ppb$^{-1}$) and (d–f) scatterplots of m,p-xylene versus ethylbenzene, o-xylene versus ethylbenzene, and toluene versus ethylbenzene at the urban site of Ahmedabad, India, during January–May 2020.
but only for the higher values of xylenes (>1.75 ppbv). At lower concentrations of xylenes and ethylbenzene, the elevated concentrations of toluene suggest contributions from non-vehicular sources. In the winter season, the significantly higher $\Delta T/\Delta E$ slopes at lower values of xylenes could be due to additional contributions from biomass burning sources. However, at higher concentrations of xylenes, a good correlation ($r^2 > 0.50$) and the $\Delta T/\Delta E$ slope of $\sim 6.5$ indicate their predominant emissions from combustion processes. The $\Delta T/\Delta E$ slope agrees with the vehicular and urban emission ratios reported in previous studies (e.g., Monod et al., 2001; Parrish et al., 1998). Monod et al. (2001) have reported $\Delta T/\Delta E$ slopes of $\sim 8.3$ in urban air and $\sim 3.6$ in fuel (gasoline) composition.

The relative B:T:E proportions of 0.69:0.27:0.04 for biomass/biofuel burning, 0.31:0.59:0.10 for traffic emissions, and 0.06:0.59:0.35 for industrial emissions have been reported in a previous study (Zhang et al., 2016). A ternary plot (Figure 10a) reveals that the relative proportions of B:T:E were scattered mostly in the regimes of biomass/biofuel burning and traffic-related emissions in the winter season. During the summer/lockdown period, the B:T:E proportions represent the dominant emissions from traffic/evaporative sources. However, to a lesser extent, the industrial and solvent emissions also contribute to the BTEX concentrations during the entire study period. The scatterplot between the sums of TEX and BTEX shows a strong correlation ($r^2 = 0.91$) with an estimated $\Delta$TEX/$\Delta$BTEX slope of $\sim 0.73$ (Figure 10b). The data points measured at higher ambient temperatures ($>35^\circ$C) fall around the 1:1 line (TEX:BTEX). But data at lower temperatures ($<20^\circ$C) show significant departures from the 1:1 line. Therefore, consistent with the above discussion, the higher TEX:BTEX ratios indicate the enhanced evaporative emissions at higher summer temperatures.

### 3.7. Impact of Photochemical Process

The photochemical age, defined as the time-integrated exposure of an air mass to OH radicals, can be calculated using molar ratios of two different VOCs (Kleinman et al., 2003). The concentration ratios of VOC compounds with different OH-reaction rates and well-characterized sources are frequently used to estimate the photochemical age of air masses (e.g., Jobson et al., 1994; Parrish et al., 1998). The aromatic VOCs have different photochemical reaction rates in the atmosphere; hence, their concentration and composition change during transport from the source to the receptor site (Borbon et al., 2013; de Gouw et al., 2005). The differences in atmospheric ratios of primary VOCs from those at the emission sources could be mainly attributed to photochemical processes. The T/B ratio has been widely used to calculate the photochemical age of air in urban environments (Warneke et al., 2007). As shown in Figure S7 in Supporting Information S1, the T/B ratio shows higher values in summer due to additional emissions from the evaporation sources compared to winter in urban regions (Legreid et al., 2007; Schnitzhofer et al., 2008). Therefore, relying on the T/B ratio can be subject to significant uncertainties in estimating the photochemical age. Alternatively, the m,p-xylenes/ethylbenzene (X/E) ratio can also be used

Figure 10. (a) Ternary diagram representing relative proportions of benzene (b), toluene (T), and ethylbenzene (e); (b) scatterplot between the sums of TEX and benzene, toluene, ethylbenzene, and xylenes compounds color-coded by the ambient temperature at the urban site of Ahmedabad, India, during January–March and April–May 2020.
to estimate the photochemical age of air masses influenced by traffic-related emissions (Roberts et al., 1984; Xue et al., 2017). The oxidation rate of a VOC molecule \(X\) due to reaction with OH radicals can be given as follows:

\[
\frac{dX}{dt} = -k_x[OH][X]
\]  

where \(k_x\) is the reaction rate constant of a VOC \(X\) with OH radicals. Equation 3 can be integrated for an exposure period \((0-t)\) as follows:

\[
\ln \left( \frac{[X]_{t=0}}{[X]_{t=t}} \right) = -k_x[OH]t
\]

\[
[X]_t = [X]_{t=0} e^{-k_x[OH]t}
\]

A suitable pair of VOCs (\(VOC_1\) and \(VOC_2\)) emitted from the same source but have different reaction rates with OH radicals can be used to estimate the photochemical age.

\[
[VOC_1]_t = [VOC_1]_{t=0} e^{-k_{VOC_1}[OH]t}
\]

\[
[VOC_2]_t = [VOC_2]_{t=0} e^{-k_{VOC_2}[OH]t}
\]

We have used the molar ratio of \(X/E\) to assess the photochemical age of air masses as the OH-reaction rates of xylenes are higher than that for ethylbenzene \((k_{OH} = 7.0 \times 10^{-12} \text{ cm}^3 \text{ molecule}^{-1} \text{ s}^{-1} \); Miller et al., 2011; Xue et al., 2017). The photochemical age \((\Delta t)\) of an air mass was calculated from the measured concentration ratios of m,p-xylene to ethylbenzene by solving Equations 6 and 7.

\[
\Delta t = \frac{1}{[OH](k_E - k_X) \ln \left( \frac{X}{E} \right)_{t=0} - \ln \left( \frac{X}{E} \right)_{t=t}}
\]

where \(k_E\) and \(k_X\) are the OH-reaction rate constants of ethylbenzene and m,p-xylenes, respectively (Atkinson et al., 2005; Yassaa et al., 2006). We have considered a mean OH concentration \([OH]\) of \(1 \times 10^6\) molecule cm\(^{-3}\), which has been used as a standard value for the Asian air masses measured during the Intercontinental Chemical Transport Experiment (INTEX) campaign (Mao et al., 2009). The molar emission ratio of \((X/E)_{t=0}\) characterizes the fresh emissions from primary sources. In the present study, a \((X/E)_{t=0}\) value of 2.47 ppb ppb\(^{-1}\) was derived using the linear fit \((r^2 = 0.97)\) of ethylbenzene and m,p-xylene mixing ratios measured from the evening to early morning hours \((18–06 \text{ hr})\). Air masses measured during this period are expected to have a negligible photochemical reactivity and hence represent fresh emissions. The value of \((X/E)_{t=0}\) in the present study is comparable to the emission ratios reported for various source profiles of China (Liu et al., 2008) and in particular of \(\sim 2.0\) in Beijing urban air (Shao et al., 2011). Xue et al. (2017) have reported m,p-X/E ratios of \(\sim 1.84\) at Weibin urban site and \(2.03\) at Chencang suburban site, attributing to the fresh emissions in Baoji city of China. Shao et al. (2009) have reported the ratios of \(\sim 1.81\) in Beijing, China, using the measurements during 00–04 hr.

The dependencies of BTEX composition on the photochemical age during the winter and lockdown periods are shown in Figures 11a and 11b. In the winter season, the percentage contribution of benzene increased gradually with photochemical age up to \(\sim 10 \text{ hr}\) in moderately aged air masses. At higher photochemical ages \((>12 \text{ hr})\), the composition of BTEX shows very little change. The average relative contributions of benzene, toluene, ethylbenzene, m,p-xylenes, and o-xylene were 29%, 43%, 5%, 15%, and 7% in fresh emission plumes, respectively, while these were 52%, 35%, 4%, 5%, and 3% in aged air masses, respectively. The increase of benzene contribution

Figure 11. Dependencies of benzene, toluene, ethylbenzene, and xylenes (BTEX) composition (% of volume mixing ratio) on the photochemical age during (a) winter and (b) lockdown periods. The bottom plot (c) shows the change in \(\sum\)BTEX loading with the photochemical age during the entire study, winter (blue line exponential decay), and lockdown (red line exponential decay) periods.
is mainly due to the decrease of more reactive aromatic VOCs with photochemical aging. During the lockdown period, the percentage contribution of benzene shows a gradual increase with photochemical age up to ~5 hr. At higher ages, the contribution of benzene changed a little, but that of toluene shows an increasing trend. The decline in the contributions of ethylbenzene and xylenes was particularly significant. This result is consistent, given the faster oxidation of C8 aromatics than benzene and toluene. This is particularly true for the variation of xylenes, which have the highest reactivity toward OH radicals among the BTEX compounds (Atkinson & Arey, 2003). During the lockdown period, the contributions of benzene, toluene, ethylbenzene, m,p-xylenes, and o-xylene were 24%, 51%, 5%, 14%, and 6% in fresh emission plumes, respectively, while these were 35%, 55%, 3%, 5%, and 2% in aged air masses, respectively. Variation of BTEX composition in fresh emission plumes could be due to differences in source emission strength and meteorological parameters. The higher contributions of toluene indicate significant contributions from industrial and evaporative emissions. During the lockdown period, particularly in the daytime, photochemical aging is accompanied by higher ambient temperatures due to prevailing summer conditions. Therefore, in aged air masses, the higher abundances of toluene during the lockdown period than those during the winter season suggest significant contributions from evaporative or solvent sources.

As shown in Figure 11c, the BTEX concentrations during both winter and lockdown periods show a strong dependence on photochemical age. During both periods, the ∑BTEX levels decreased rapidly in moderately aged (5–10 hr) air masses. The following exponential-decay functions describe the relations of ∑BTEX concentration with the photochemical age during the winter and lockdown periods.

\[
[\sum BTEX]_{Winter} = 5.17 + 19.1 \times e^{-0.13\Delta t}
\]

\[
[\sum BTEX]_{Lockdown} = 1.66 + 7.15 \times e^{-0.23\Delta t}
\]

In relatively fresh air masses (Δt < 4 hr), the average ∑BTEX concentration of 7.0 ± 0.7 ppbv during the lockdown period was ~3 times lower compared to 21.3 ± 3.0 ppbv measured in the winter. In more aged air masses (Δt > 10 hr), the ∑BTEX concentration of 2.1 ± 0.6 ppbv during the lockdown period was ~4 times lower than that of 8.2 ± 0.8 ppbv in the winter season. The comparison of exponential-decay parameters suggests that the rate of decline of ∑BTEX concentration with photochemical age during the lockdown period was much greater than in the winter season. Therefore, in addition to emission reductions, the faster photochemical removal also contributed to the large decrease of BTEX in ambient air during the lockdown period. The distinct photochemical dependencies of BTEX composition during winter and lockdown can be linked to differences in the emission source type and strength, meteorological, and dilution conditions. By assuming similar seasonality in the OH concentration during the lockdown and normal years, the changes estimated using VC-normalized data account for the photochemical factor into account. A specific photochemical box model study, however beyond the scope of this study, could have better captured the changes caused by photochemical processes.

3.8. Dependence of BTEX Concentration on Wind Speed

The meteorological parameters play an important role in the dispersion, transformation, and removal of pollutants in the urban atmosphere (David & Nair, 2011). As discussed earlier, the time series of actual ambient air BTEX concentrations and WS showed anticorrelated variations throughout the study period but more clearly noticed during weak and stronger wind episodes. The dependencies of concentrations of different aromatic VOCs on WS for the winter and lockdown periods are shown in Figure 12. The BTEX concentrations decreased rapidly with increasing WS in lower WS regimes but showed a gradual decline in higher WS regimes. The following exponential functions describe the relations between the BTEX concentrations and WS.

(a) For the winter season

\[
[Benzene]_{Winter} = 1.9 + 4.1 \times e^{-0.36WS}
\]

\[
[Toluene]_{Winter} = 1.1 + 18 \times e^{-0.92WS}
\]

\[
[Ethylbenzene]_{Winter} = 0.19 + 3.7 \times e^{-1.4WS}
\]

\[
[Xylenes]_{Winter} = 0.45 + 13.2 \times e^{-1.4WS}
\]
In the above equations, the aromatic VOCs and WS values are in the units of “ppbv” and “m s$^{-1}$,” respectively. The decrease rate of each compound during the lockdown period is lower than that during the winter season. Except for benzene, the concentration and variability (percentiles) of VOCs decreased significantly with the increase of WS in the lower WS regime (<3 m s$^{-1}$), while they showed weaker dependencies in the higher wind regime (>3 m s$^{-1}$). The benzene concentration appears to be less sensitive to the change of WS than those of more reactive VOCs (TEX). In the low WS regime, the average concentrations of ∑BTEX and ∑TEX were 10.7 and 6.6 ppbv during the winter and 2.3 and 1.5 ppbv during the lockdown, respectively. At stronger winds, the average ∑BTEX and ∑TEX concentrations were 5.5 and 2.4 ppbv during the winter and 0.90 and 0.58 ppbv during the lockdown, respectively. The distinct relations of different VOCs with the WS reflect the different

\[
\sum_{BTEX}^{Winter} = 4.2 + 34 \times e^{-0.92 \times WS} \tag{15}
\]

\[
\sum_{TEX}^{Winter} = 1.8 + 34 \times e^{-1.1 \times WS} \tag{16}
\]

(b) For the lockdown period

\[
[Benzene]^{Lockdown} = 0.16 + 3.6 \times e^{-0.84 \times WS} \tag{17}
\]

\[
[Toluene]^{Lockdown} = 0.38 + 13 \times e^{-1.53 \times WS} \tag{18}
\]

\[
[Ethylbenzene]^{Lockdown} = 0.04 + 1.2 \times e^{-1.9 \times WS} \tag{19}
\]

\[
[Xylenes]^{Lockdown} = 0.08 + 6.7 \times e^{-2.2 \times WS} \tag{20}
\]

\[
\sum_{BTEX}^{Lockdown} = 0.69 + 22 \times e^{-1.4 \times WS} \tag{21}
\]

\[
\sum_{TEX}^{Lockdown} = 0.5 + 20 \times e^{-1.7 \times WS} \tag{22}
\]
background concentrations and OH reactivity. The higher background concentrations and slower chemical removal could cause a weaker WS dependence of benzene compared to other aromatic compounds. The stronger winds (>3 m s\(^{-1}\)) were recorded mostly in the daytime, coinciding with the deeper PBLH and higher chemical (OH) loss rates of VOCs. Therefore, the decrease of BTEX concentrations at stronger wind speeds could be due to dilution/ventilation and loss by OH radicals. Sahu, Yadav and Pal (2016) have reported a rapid decrease in the mixing ratios of both primary and secondary VOCs with the increasing WS for the same measurement site during the winter season of the year 2014.

4. Source Apportionment of BTEX Compounds

The positive matrix factorization (PMF) is a multivariate factor analysis tool recommended by the USEPA (US EPA) to quantify the contributions from different emission sources to various gaseous and particulate pollutants at the receptor site (Ling et al., 2011; Sahu et al., 2020). The source apportionment of BTEX at the receptor site was conducted using PMF (version 5.0) model (Norris et al., 2014) with the data measured in the present study. The PMF decomposes a matrix of speciated data into the matrices of factor contributions (\(G\)) and factor profiles (\(F\)). The PMF analysis is performed to determine the contribution of each factor, the total number of source factors, and the residual for each species as in the following equation.

\[
X_{ij} = \sum_{k=1}^{p} g_{ik} f_{kj} + e_{ij}
\]  

(23)

where \(X_{ij}\) is the concentration of a species \((j)\) in a sample \((i)\), \(g_{ik}\) is the contribution of a factor \((k)\) to a sample \((i)\), \(f_{kj}\) is the fraction of a factor \((k)\) of species \((j)\), and \(e_{ij}\) is the residual for a species \((j)\) in a sample \((i)\). The \(g_{ik}\) and \(f_{kj}\) can be determined by minimizing the following objective function \((Q)\).

\[
Q = \sum_{i=1}^{n} \sum_{j=1}^{m} \left( X_{ij} - \sum_{k=1}^{p} g_{ik} f_{kj} \right)^2
\]  

(24)

where \(u_{ij}\) is the uncertainty associated with \(X_{ij}\).

The four factor profiles of BTEX determined as traffic, residential bio-fuel/biomass burning, gasoline/solvent evaporation, and mixed industrial emissions at receptor site are shown in Figure 13. Traffic emission (Factor 1) was found to be the dominant contributor to ambient concentrations of all BTEX with toluene (35%) followed by \(\alpha\)-xylene (30%), benzene (29%), \(m/p\)-xylenes (29%), and ethylbenzene (28%) during the study period. This factor signifies the traffic-related emissions due to the incomplete combustion of fossil fuel (Yuan et al., 2009).

The contributions of residential bio-fuel/biomass burning emissions (Factor 2) to the mixing ratios of benzene, toluene, ethylbenzene, \(m/p\)-xylenes, and \(\alpha\)-xylene were 27%, 22%, 24%, 23%, and 21%, respectively. The significant contributions from biomass, wood, and crop residue burnings during the winter to early summer are related to post-harvesting practices in this region. In the winter season, the local activities of residential bio-fuel/shrub burnings for heating purposes in the morning and evening hours also contribute to ambient BTEX (Begum et al., 2011; Venkataraman et al., 2006; Yadav et al., 2019). Gasoline/solvent evaporation (Factor 3) is responsible for ~25% of toluene, indicating emissions from evaporative sources in the summer season (Monod et al., 2001; Sahu et al., 2020). The contributions of mixed industrial emissions (Factor 4) were about 22%, 18%, 26%, 28%, and 27% to the mixing ratios of benzene, toluene, ethylbenzene, \(m/p\)-xylenes, and \(\alpha\)-xylene, respectively. Factor 4 consists of the emissions from industrial sources in the surroundings of the study site. The contributions of each factor to the total BTEX during winter, pre-lockdown, and COVID-19 lockdown periods are shown in Figure 13. Factor 1 showed a significant contribution (~30%) to total BTEX in the winter period. The primary anthropogenic source of BTEX in urban air is the incomplete combustion of fossil fuels in vehicles (Sahu, Pal, et al., 2016; Sahu et al., 2020). However, the contribution of traffic-related emissions reduces to ~21% during the COVID-19 lockdown period.

It demonstrates the impact of lockdown due to the absence of major anthropogenic activities, which emit the BTEX compounds in higher amounts (Fan et al., 2014; Liu et al., 2008). The significant reductions in the total BTEX concentrations can be attributed to the restrictions in combustion-related activities, such as vehicular traffic and industrial production. The contributions from biomass burning were 30%, 34%, and 29% during the
winter, pre-lockdown, and COVID-19 lockdown periods, respectively. Despite the major restriction in industrial and transport sectors, bio-fuel emissions were not reduced significantly during the COVID-19 lockdown period due use of wood and kerosene for cooking activities with poor combustion technology in the slums regions. The results are consistent with a reported increase of ∼12% in biofuel consumption during the lockdown period (PPAC, 2020).

5. Conclusion

We have investigated the response of ambient BTEX concentrations due to the COVID-19 lockdown in a major urban region of Ahmedabad in western India. The changes in BTEX concentrations during the different lockdown phases were estimated with respect to the data measured during 2014-2015-2018. The BTEX concentrations showed large day-to-day variations mainly controlled by the local emissions and meteorological parameters with a strong transition from pre-lockdown to lockdown conditions in response to the COVID-19 restrictions. In addition to the emission reductions, the changes in ambient air BTEX concentrations during the lockdown could also be due to the differences in meteorological and photochemical processes to a certain extent. Therefore, to account for the variability associated with meteorological conditions, we compared the actual and VC-normalized concentrations during the same period from 2014-2015-2018. In comparison to normal years, the reductions of the actual ambient (70%–90%) and VC-normalized (54%–88%) BTEX concentrations during the lockdown periods are in line with the mobility reductions. Although the VC-based approach covers meteorological influences reasonably, uncertainty could come from other processes that affect the abundance of BTEX. The estimated changes using the nighttime data, which take account of photooxidation removal of BTEX, show slightly lesser (by ∼8%) reductions than VC-normalized estimates using all data. Therefore, the reductions in BTEX levels during the lockdown period were primarily due to reductions in emissions, despite the influences of meteorological and photochemical factors. The photochemical age, estimated using the m,p-xylene/ethylbenzene ratio, was used to investigate the

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**Figure 13.** The explained contributions of four identified sources to ambient air benzene, toluene, ethylbenzene, and xylenes at Ahmedabad using the PMF 5.0 analysis of the United States Environmental Protection Agency.
change in BTEX concentration and composition of fresh emissions and aged air masses. During the lockdown period, benzene and toluene accounted for ~24% and 51% of the total BTEX in fresh emission plumes, while 35% and 55% in photochemically aged air masses, respectively. At higher ambient temperatures, the TEX/BTEX ratios were close to 1.0, indicating the enhanced evaporative emissions in summer. While at lower temperatures, the TEX/BTEX ratios were significantly lower than 1.0, indicating significant contributions from biomass/biofuel burning sources in winter. All BTEX compounds showed elevated concentrations from the evening to early morning and the lowest in the afternoon. A prominent peak during the evening rush hours also coincided with stagnant winds. The BTEX concentrations decreased with the increase in WS and were particularly sensitive in the lower WS regime. However, varied from compound to compound, the BTEX concentrations decreased exponentially with the increasing WS. The results presented in this study will help implement the control strategies of VOCs emissions and hence the ambient levels of related secondary pollutants such as O$_3$ and PM$_{2.5}$ in the urban regions of India. The findings of this study suggest the implementation of effective science-based policies that consider local and regional emission sources of aromatic VOCs. The long-term measurement programs of BTEX and other VOCs need greater attention to understand the regional importance of different emission and photochemical processes in South Asia.

Data Availability Statement

The data used in this study are archived at a public data repository at figshare (https://figshare.com/s/2faa920ac695213a38ff).

Acknowledgments

The authors gratefully acknowledge the ECMWF (https://cds.climate.copernicus.eu). The weather data were taken from the wunderground archive (www.wunderground.com). The BTEX data for the Maninagar station were taken from the CA-AQMS of the Central Pollution Control Board (https://cpch.nic.in). We acknowledge the researchers involved in the ISRO-Bhuvan project (https://bhuvan.nrsc.gov.in/). We thank the USEPA (US-EPA) for providing the PMF model. We are grateful to Prof. Anil Bhardwaj (Director PRL), ISRO Chairman, and the ISRO-GBP team for their interest, encouragement, and support to conduct this study.

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