Source apportionment of BTEX compounds in Tehran-Iran using UNMIX receptor model

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Abstract

Understanding the distribution levels and sources of volatile organic compounds (VOC), mainly benzene, toluene, ethylbenzene and xylenes (BTEX), in the ambient atmosphere is important for efficiently managing and implementing the associated control strategies. We measured BTEX compounds at an industrial location in the west Tehran city (Iran), which is highly influenced by industrial activities and traffic during the winter and spring seasons during 2014-2015. A multivariate receptor model, UNMIX, was applied on the measured data for the identification of the sources and their contributions to BTEX compounds in a highly industrialised and trafficked atmospheric environment of Tehran city. Three main groups of sources were identified. These included solvent and painting sources (e.g. vehicle manufacturing), motorised road vehicles and mixed origin sources. While the solvent and painting sources and vehicle exhaust emissions contributed to about 5 and 29% of total BTEX mass, respectively, the mixed origin source contributed to about two-third (~66%) of the remaining mass. These mixed origin sources included rubber and plastic manufacturing (39%), leather industries (28%) and the unknown sources (33%). The mean concentrations of benzene, toluene, ethylbenzene and average xylene(o, p,m) compounds were measured as $28.96 \pm 9.12 \mu\text{g m}^{-3}$, $29.55 \pm 9.73 \mu\text{g m}^{-3}$, $28.61 \pm 12.2 \mu\text{g m}^{-3}$ and $25.68 \pm 10.58 \mu\text{g m}^{-3}$, respectively. A high correlation coefficient ($R^2 > 0.94$) was also found between predicted (modelled) and measured concentrations for each sample. Further analyses from UNMIX receptor model showed that the average weekday contributions of BTEX compounds were significantly higher during winter compared with those during spring. This higher concentration during winter may be attributed to calm wind conditions and high stability of

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the atmosphere, along with the after effect of government policies on the use of cleaner fuel in refineries that became operational in winter 2014.

Keywords: Source apportionment; BTEX compounds; UNMIX Receptor model; Urban environment; Tehran air quality

1. Introduction

The air quality of an urban region depends on a number of factors such as pollutant emissions, meteorological, geographical, solar radiation, deposition and atmospheric dispersion conditions (Kumar et al. 2014, 2015). Volatile organic compounds (VOCs) play an important role in deteriorating the quality of ambient air since these contain carbon atoms that have high vapour pressure and hence evaporate readily to the atmosphere (Hester and Harrison 1995). Long-term human exposure to VOCs has shown to increase adverse health effects (e.g. eye and nose irritation, allergy, liver and kidney dysfunction, neurological impairment and cancer) in the human population (Berglund et al. 1992; Kim and Bernstein 2009; Wallace 2001). Among the VOCs, emissions of benzene, toluene, ethylbenzene and xylene (hereafter referred as BTEX) belong to the group of VOCs. BTEX, which is the focus of this article, have found to show even more serious effects compared with the other VOCs on air quality and human health (Hoque et al. 2008). For example, BTEX is known to be toxic, mutagenic and carcinogenic (Crump 1994; Dutta et al. 2009). However, toluene, ethylbenzene and xylenes carcinogenicity and mutagenicity has not been proven although could be possible (Helmes et al. 1982; Mansha et al. 2011) and also, they are the precursors of toxic radical intermediates in the atmosphere (Possanzini et al. 2004). Exposure to high levels of BTEX components has been reported to trigger skin and sensory irritation, central nervous system depression and negative effects on the respiratory system

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(Cockerham and Shane 1993). Specific attention has been given to BTEX species, especially to benzene, due to their carcinogenic effects on human health (Park et al. 2002). These enter the atmospheric environment from a variety of anthropogenic sources. Some of which includes fossil fuels, incomplete combustion from gasoline, motor vehicles, an industrial production involving petrochemical process, paint, solvent, synthetic resin and synthetic rubber (Hoque et al. 2008).

United States Environmental Protection Agency (US EPA) estimates that, if an individual were to continuously breathe the air containing average benzene concentration of 0.13 to $0.45 \mu\text{g m}^{-3}$ over his or her entire lifetime, that person would theoretically have no more than a one-in-a-million increased chance of developing cancer as a direct result of continuously breathing air containing this chemical (Cleland et al. 2002). Despite the commitment of the Iranian cities to comply with the Iranian Department of Environment air quality limit values for BTEX, some Iranian cities, including Tehran have so far failed to effectively tackle the issue of excess BTEX emissions (Sarkhosh et al. 2013). Therefore, it is necessary to carry out a detailed characterization of the main sources of BTEX compounds to control excessive emissions.

In general, the management of ambient air quality involves the identification of the sources of pollutant substance emitted into the air. The understanding of the transport of the substance from the sources to downwind locations and the knowledge of the physical and chemical transformation processes that can occur during that transport are also important. Numerous mathematical models are used to estimate the change in observable airborne concentrations that might be expected to occur as a result of the chemical transformation in the atmosphere (Kumar et al. 2011; Shraim et al. 2016). Receptor models, which use multivariate statistical approach, are often applied to infer probable sources of air pollution in atmospheric environments (Kumar et al. 2013). Source

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apportionment methods usually includes receptor models such as principal component analysis, PCA (Al-Dabbous and Kumar 2014; Mouzourides et al. 2015), multiple linear regression, MLR (Larsen and Baker 2003), positive matrix factorization, PMF (Al-Dabbous and Kumar 2015; Paatero and Tapper 1994; Yang et al. 2013), chemical mass balance, CMB (Lu et al. 2005) and UNMIX model (Henry 2000).

UNMIX is one of the US EPA's recommended receptor models that has been used widely in the atmospheric environment studies (Larsen and Baker 2003). The UNMIX has undergone intense development over the years and is used in source apportionment of VOCs and particulate matter (Hopke 2003; Samek et al. 2016). The UNMIX had been successfully applied to identify the sources of VOC compounds in many cities worldwide such as Tehran, Iran , Beijing, China , Alexandria, Egypt and Lucknow, India .

A summary of various recent UNMIX applications is outlined in Table 1. The aim of this work is to apportion the sources of BTEX using the UNIMIX receptor model in a highly industrialised and trafficked atmospheric environment in the west Tehran city. There are two distinct parts (field measurements and receptor modelling) of this work. Firstly, we collected a total of about 100 samples on charcoal sorbent tubes were collected for laboratory investigations using gas chromatography from the west of Tehran during November 2014 to March 2015. Sampling location was carefully selected to represent possible emission sources of BTEX including residential, traffic and industrial activities. Secondly, the results were used for the source identification of mobile and industrial emissions using the UNMIX receptor model.

2. Materials and methods

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2.1. Sampling location

Tehran, the capital of Iran, is the largest urban area in Iran with a population of 8,791,378 (Iran 2015). This has heterogeneous emission sources (e.g. Iran Khodro company, motorised road vehicles and mixed origin sources) in and around the city (Figure 1). Growth in traffic volume, residents population and industrial sectors have resulted in increased emissions of air pollutants, including the BTEX (Chan et al. 2007; Jia et al. 2008). The sampling location (35.719275° longitude, 51.191613° Latitude) was situated close to a busy highway (Figure 1). As shown in Figure 1, the study area is characterised by surrounding one of the largest automotive industry, industrial plants and all types of vehicles (petrol, diesel and gasoline driven) plying on the highway throughout the year. In addition, massive demolition and construction projects are taking place all over the city.

2.2. Sample collection

A total of 100 BTEX samples were collected, twice in a day, over a 5 months period between November 2014 and March 2015. The sampling was performed during the morning peak hours from 08.00 h to 09.00 h (local time) and evening peak hours from 17.00 h to 18.00 h. The sampling site is close (~25 m) to a heavy traffic motorway and a number of industrial emission sources such as vehicle manufacturer (i.e. Iran Khodro company, IKCO, which is a leading Iranian vehicle manufacturer) and several solvents, synthetic resin, and synthetic rubber production facilities. Sampling was carried out at a height of 1.5 m above the ground level to represent a typical breathing height (Kumar et al. 2008) using the hand operated pumps (SKC 224-44MTX). Table 2 shows the meteorological parameters which were measured using Vane Anemometer (Testo 417) for wind speed during the study period at the site sampling. Mean

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temperature and relative humidity were measured at the site using Humidity/Temperature Pen 44550. The general climate of Tehran city is moderate and cold in winter, with high temperature and high solar radiation in the summer season.

2.3 Extraction and analysis

The collected samples were transported to the laboratory and analysed using the GC-flame Ionization detection (FID), which was optimised to achieve the best selectivity and sensitivity. The gas chromatograph (Clarus 600 model), which was used for BTEX determination, was equipped with an autosampler, two FIDs and two columns. The BTEX were sampled by drawing air through a charcoal sorbent tube (SKC.Inc. 224-44 EX model), which was 7 cm long and 4 mm internal diameter (ID) with flame-sealed ends and contained two sections of activated coconut shell charcoal (front 100 mg and back 50 mg) at a flow rate of 50 ml min⁻¹ for 20 min sampling frequencies were set up to run continuously at sampling site in a heavy traffic and industrial zone in Tehran city. The samples were then analysed with a gas chromatograph that was equipped with a GC-FID. At the end of each sampling period, the sorbent tube was capped with plastic samplers (not rubber) caps and pack securely for shipment. The sorbent tube containing BTEX sample were transferred into 2 ml vials and extracted by adding 1.0 ml of carbon disulphide (CS₂) with an agitation of 30-45 min in the ultrasonic bath. After extraction, it was transported to the laboratory for separation and detection was done in a GC-FID, using a capillary column of 30 m × 0.32 mm (ID). The temperature of the column, injector and detector were 40, 250 and 300 °C, respectively. The carrier gas was high in pure helium gas at a flow rate of 2 ml min⁻¹ and a volume of 1 µl and a 5:1 split, which was injected at 250 °C.

2.4. UNMIX receptor model

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UNMIX is a multivariate receptor model, which is used for identifying the contribution of various sources (Henry 1997). In some cases, there are more unknowns than equations and thus there may be many wildly different solutions that are all equally good in the least squares sense. Few modelling approaches allow the user know clearly when a reliable solution is not possible (Chow et al. 1990; Chow and Spengler 1986).

The basic assumption in the UNMIX was stated, which included the data and the compositions and contributions of the sources are all strictly positive (zero data values are not allowed). For each source, there are some samples that contain little or no contribution from that source. Two key algorithms were used in the UNMIX model. The first was NUMFACT algorithm, which estimates the number of sources (N) that can be resolved relative to the particular ambient data set, based on signal-to-noise considerations. The signal-to-noise ratio is greater than 1.5 for species that contribute significantly to a source. The second algorithm is for data edges to find the source points with one source missing. The sets of points in N-dimensional space uses geometric features in the data called "edges" to constrain both source profiles and contributions. These "good edge" will have some dates during which the contribution is zero for one factor and non-zero for the other factor when a particular source contribution falls to zero at the receptor. In the UNMIX reports, the species that have a correlation ≥ 0.8 were associated with the edge. To establish a relationship between each species and the BTEX concentration scatter plots were drawn. Also, PCA tool (Blifford and Meeker 1967) was used to identify the major source of atmospheric BTEX and analyse structure in multivariate data sets. PCA describes the total variance of all the observable variables (e.g., BTEX concentrations) as much as possible.

3. Results and Discussion

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3.1 *Ambient concentrations of BTEX*

Table 3 shows the measured compounds and univariate statistics pertaining to BTEX compounds and VOCs at the receptor site during the sampling period. The results clearly indicate that all compounds present concentrations of the same order of magnitude of $29.54 \mu\text{g m}^{-3}$ for Toluene, followed by benzene ($28.95 \mu\text{g m}^{-3}$), ethylbenzene ($28.60 \mu\text{g m}^{-3}$) and xylene (o; $28.40 \mu\text{g m}^{-3}$), respectively. The average concentrations of BTEX compounds in the present study in comparison with those reported from other cities in the world are shown in Table 4. Also, in the present study, the highest m,p,o-xylene concentrations are comparable to those observed in Cairo, Egypt (Khoder 2007) and Izmir, Turkey (Muezzinoglu et al. 2001). Benzene is a marker of vehicle exhaust (Hong et al. 2006) and evaporative emissions, whereas toluene, ethylbenzene and xylene that is also emitted from motor vehicles has other sources as well such as evaporation of solvents used in paint, plastic and leather manufacturing (Yuan et al. 2010). These differences can be attributed to the particular characteristics such as studied cities, sampling points, sampling periods, and main city activities as well as differences associated with vehicles, such as vehicle production quality and fuel composition (Gee and Sollars 1998; Marc et al. 2014).

3.2. Source apportionment

A total of 100 number of samples and 6 number of BTEX species were introduced into the UNIMIX model. Three sources, or factor, profiles of BTEX concentrations were extracted from the applied model (Table 5). Figure 2 shows the details of the sources and their contributions extracted by the UNMIX.

3.2.1 *Solvent and painting sources*

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The solvent and painting sources are recognised as a source 1 since this factor was mainly rich in toluene (41%), xylene (m) (30%) and xylene (o) (27%). These elements are often considered as good markers of industrial solvent and vehicle manufacturer sources (Chan et al. 2006). Characteristics of nonmethane hydrocarbons (NMHCs) in industrial, industrial-urban, and industrial-suburban atmospheres of the Pearl River Delta (PRD) region of south China (Chan et al. 2006). This profile mainly from of vehicle manufacturer (Iran Khodro Company (IKCO), which is a leading Iranian vehicle manufacturer. This factor has a fraction of about 8% of their mass contained in this factor. Although this factor had negative values. Its contribution uncertainty was three-times of its contribution estimation.

3.2.2 Vehicle exhaust and fuel evaporation

Vehicle exhaust and fuel evaporation is recognised as source 2 since this factor was dominated by benzene (40%) and ethylbenzene (31%). Most of these BTEX are predominant in the automobile exhaust emission and the evaporation of motor Vehicle. This factor accounted for about 33% of the total BTEX mass concentrations. The vehicular emissions may be from the traffic on the highway, gasoline and diesel engine vehicles. UNMIX is unable to separate the gasoline engine source profile from the diesel engine profile.

3.2.3 The mixed source

The mixed sources were recognised as the third factor since this was highly related to all species of BTEX. Thus, this contribution was interpreted as mixed emissions from rubber and plastics manufacturing (42%), leather industries (25%) and the unknown other sources (33%). This factor has a fraction of more than 59% of the total BTEX mass concentration (Figure 2).

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3.2.4 Overall summary: mass contributions

Figure 3 shows the average source mass contributions for the BTEX compounds at the sampling site. Three key sources of BTEX compounds were distinguished and their quantitative contributions made. The mixed sources made the highest contribution (~59%) to the total BTEX mass. These contributions came from the rubber and plastics manufacturing (42%), leather industries (25%) and the unknown sources (33%) that we could not explain. The second source was vehicle exhaust and fuel evaporation that made a contribution of 33%, followed by the solvent and painting sources contributing another ~8% of the total BTEX mass.

3.3. Diurnal variation of different source categories

In the present study, the daily temporal trends of BTEX compounds obtained from the UNMIX model at the receptor site is shown in Figure 4. The average weekday concentrations of BTEX were found to be significantly higher between November 2014 and early February 2015 during winter (average temperature 7 °C; average humidity 60%) than those during the middle of February to March 2015 during spring (average temperature 21°C, average humidity 47%). This is different from the west Tehran, where higher BTEX concentrations were observed between December 2014 and early February 2015. These higher concentrations could be attributed to two reasons. Firstly, calm conditions and high stability of the atmosphere prevails during the winter months when traffic rush hour and industrial emissions accumulate in the shallow boundary layer. Secondly, government policy on the use of cleaner fuel, which is imported from other countries to replace dirty fuels from refineries in several big cities including Tehran, Iran from February to March 2015, thereby reducing the concentration of BTEX compounds during these months.

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Figure 5 shows edge plots edge of the masses attributed to three sources by UNMIX plot. Edge plots allow the user to examine the edges in the data that define the additional constraints used by UNMIX. These edges in the data are the source of the additional constraints used by UNMIX to find a unique multivariate receptor model that fits the data (Henry 1997). The edges are the x and y-axes of the plots. In both plots, the x-axis is the edge associated with source 1, the industrial activities source. Points that are near the y-axis have very small contributions from the industrial activities. Data points near the x-axis in Figure 5a are associated with small contributions of source 2 that including vehicle exhaust and fuel evaporation. Finally, the x-axis in Figure 5b is associated with small values of the mixed contributions (i.e. source 3). All the edges in these plots are typical of “moderate” to “poor” edges. Since UNMIX uses edges to find the source compositions, poor edges will lead to increased variability in the source compositions (Henry 1997).

4. Summary and conclusions

We collected the samples of BTEX compounds over a period of 5 months at Iran khodro intersection sampling location in Tehran, Iran. The collected samples were chemically analysed in the laboratory and the data were used in the UNMIX receptor model (version 6) to identify their probable sources. The UNMIX results showed that the solvent and painting sources, motor vehicle exhaust and the mixed contribution, each contributing 8, 33 and 59%, respectively, were the major sources. The mixed contribution came from the rubber and plastics manufacturing (39%), leather industries (28%) and the unknown sources (33%).

The following conclusions are drawn:

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- The BTEX compounds of Tehran was apportioned by UNMIX receptor model during November 2014 to March 2015. The total concentrations of 4 BTEX compounds in the urban atmosphere ranged from $23.3 \mu\text{g m}^{-3}$ to $29.55 \mu\text{g m}^{-3}$, with an average value of $27.36 \mu\text{g m}^{-3}$.
- The source apportionment results obtained with the UNMIX model indicated three significant factors as solvent and painting sources, vehicular emissions and fuel evaporation and the miscellaneous sources as major sources of BTEX compounds. The temporal variations and source apportionment of BTEX compounds indicated the mixed sources, especially from rubber and plastics manufacturing and leather industries are the major source of BTEX compounds in Tehran, Iran.
- The modelled BTEX concentrations ($\mu\text{g m}^{-3}$) were compared with the measured BTEX concentrations ($\mu\text{g m}^{-3}$) as illustrated to assess the accuracy of the model results. A high correlation coefficient ($R^2 = 0.98$) was found between the Fitted and measured BTEX concentrations.
- The mixed sources (rubber and plastics manufacturing, leather industries and the unknown other sources) shows the highest contribution to local BTEX with an average contribution of approximately 59%.

While the study presents a unique insight into the BTEX sources and contribution in Tehran, there are some explainable limitations of the work. For example, the UNMIX has physical assumptions and constraints like the other receptor models. Such limitations can be obviated using the other receptor models such as the PMF and CMB. Using the comparison of these results or the measurement of specific molecular markers may contribute to confirming the contribution of

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BTEX compounds. However, the results could assist the regulatory authorities to improve air quality and reduce the potential effects of the environment on human health by taking appropriate actions to control sources of BTEX compounds. Furthermore, urban cities are large sources of atmospheric pollution. Understanding the chemical composition and contribution of the sources of atmospheric BTEX requires the existence of emission inventories, which will help to develop efficient pollution control strategies and assist in the protection of the human health. The results for BTEX source apportionment would be useful in the control of pollution sources and the development of effective air quality control strategies based on urban site type, both on a regional and local scale. It is recommended that the other receptor models be used to identify and determine the contributions BTEX compounds. Control technologies and regulatory approaches that lead to effective controls are well-established and can serve as a basis for Tehran to develop a set of evaporative control regulations optimised and best suited for its local vehicle fleet, climate, and driving patterns.

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417 **Table 1.** Abbreviation organic species included in the UNMIX model and their major sources.

Species	Location	Identified major source categories	Reference
Hourly concentrations of C ₂ – C ₉ volatile organic compounds (VOC)	Downtown Atlanta, GA, USA	Emissions from vehicles in motion; evaporation of whole gasoline; gasoline headspace vapour	Henry (1994)
Ambient VOCs	Central EL Paso, TX, near The US–Mexico Border	Three sources: motor vehicle exhaust, gasoline vapour evaporation, liquefied Propane gas	Mukerjee et al. (2004)
VOCs in urban areas	Center of the capital on the flat roof of a building in Tehran, University of Medical Sciences	Five sources: vehicle exhaust; industrial solvents and painting; biogenic source; fuel evaporation; City gas and CNG	Sarkhosh et al. (2013)
Non-methane hydrocarbon (NMHC)	Helsinki, Finland	Four sources: gasoline exhaust; liquid gasoline; distant sources; others	Hellen et al. (2013)

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Table 2. Meteorological parameters for during sampling period.

Sampling period	Mean Temperature (⁰ C)	Relative Humidity (%)	Mean wind speed (km h ⁻¹)
November	8±2	39 %	8±0.6
December	12±4	41 %	11±0.8
January	10±4	35 %	9±0.5
February	10±3	37 %	9±0.7
March	15±1	33 %	14±0.5

Table 3. Statistical summary of BTEX concentrations at the receptor site.

Species name	Mean±standard deviation
Benzene	28.96±9.12
Toluene	29.55±9.73
Ethylbenzene	28.61±12.2
Xylene (m)	23.34±9.30
Xylene (o)	28.40±12.01
Xylene (p)	25.30±10.44

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Table 4. Comparison of BTEX compounds ($\mu\text{g m}^{-3}$) in various cities of the world.

Location	BTEX compounds					Reference
	Benzene	Toluene	Ethylbenzene	o,p-Xylene	m-Xylene	
Tehran,Iran	28.96	29.55	28.61	26.85	23.34	Present study
Cairo, Egypt	27.37	56.87	10.00	32.52	17.04	(Khoder 2007)
Izmir, Turkey	17.50	27.80	8.60	19.10	19.50	(Muezzinoglu et al. 2001)
Ahvaz,Iran	1.78	5.19	0.51	1.13	1.13	(Rad et al. 2014)
Giza, Egypt	14.47	29.74	5.26	17.24	8.37	(Khoder 2007)
London	22.0	36.5	6.69	6.62	13.5	(Monod et al. 2001)
Prague	62.7	98.1	25.6	25.00	50.5	(Monod et al. 2001)
Hannover,	4.00	22.2	2.8	9.7	9.7	(Ilgen et al. 2001)

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451

452 **Table 5.** Source profiles extracted from the UNMIX model ($\mu\text{g m}^{-3}$).

Compounds	solvent and painting sources	Vehicle exhaust and evaporation fuel	The mixed sources
Benzene	0.017	8.230	20.600
Toluene	3.530	4.970	21.100
Ethyl benzene	-3.200	7.510	24.400
Xylene(m)	5.570	1.720	19.100
Xylene(o)	2.410	1.950	27.100
Xylene(p)	-0.607	4.100	21.700

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Fig. 1. Geographical location of Tehran and the measurement site. Also are shown key industrial sources around the sampling location.

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460

461 Fig. 2. Percentage of species apportioned to each source obtained by UNMIX receptor models

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Fig. 3. Source contributions to the BTEX compound mass (%) at the receptor site.

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Fig. 4. The weekly variations in source apportionment to ambient BTEX compounds ($\mu\text{g m}^{-3}$).

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Fig. 5. Edge plots in the UNMIX Receptor model: (a) BTEX mass concentrations in source 2 attributed to source 1, (b) BTEX mass concentrations in source 3 attributed to source 1.

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