



VOCs and PM concentrations in underground parking garages of the commercial center and high-rise residential buildings

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Abstract

In China, due to the high density of population and vehicles in megacities, the commercial center and high-rise residential buildings have been widely equipped with large underground parking garages (UPGs). Volatile organic compounds (VOCs) and particulate matters (PM) have been confirmed as major pollutants in underground parking garages. And therefore, exposure to these air pollutants, especially ultrafine particles and BTEX (benzene, toluene, ethylbenzene, and xylenes), is closely associated with the health of the car owners or the garage workers. This study sampled or online monitored the concentrations of benzene, toluene, xylenes, formaldehyde, TVOC (total volatile organic compounds), PM_{2.5}, and PM₁₀ in the underground garage of three commercial center and high-rise residential buildings. The time-weighted average (TWA) concentrations of pollutants in the underground garage in the commercial center/high-rise residential buildings were as follows: benzene 0.12/0.11 mg·m⁻³, toluene 0.33/0.31 mg·m⁻³, xylenes 0.23/0.20 mg·m⁻³, TVOC 1.04/1.19 mg·m⁻³, PM_{2.5} 0.20/0.19 mg·m⁻³, and PM₁₀ 0.24/0.22 mg·m⁻³. Indoor PM concentrations were slightly higher than outdoor PM concentrations, and there was a significant positive relationship between the difference of I/O PM concentrations and traffic volume. This paper also compared and analyzed BTEX concentration ratios (e.g., toluene/benzene and xylenes/benzene) at every sampling point of different underground garages. These ratios were compared and analyzed to evaluate the influence of vehicle emissions and outdoor air quality on the spatial distribution of indoor air quality quantitatively.

Keywords BTEX · Indoor pollutant concentrations · Underground parking garage · Natural ventilation · Inter-species ratio

Introduction

With rapid urbanization and rising incomes in China, the booming number of private vehicles has led to tight parking spaces in megacities in recent years, especially in areas with high-rise residential and large commercial complexes. To meet the demand for parking, large underground garages, including many mechanical multistory underground garages, have occupied large areas underground and standard in

shopping malls and residential areas in the last decade. Due to the building's enveloped structure, many parking vehicles, and inadequate ventilation design, severe air pollution has been found in the underground garage (Batterman et al. 2006). Air pollutants can also affect the above-ground part of the building and the surrounding surface through diffusion (Dodson et al. 2008; Nirvan et al. 2012). Although the general population spends relatively little time (entering parking spaces, going out of the vehicles, approaching off-ground parking lot exits), human health may be affected because of high concentrations of pollutants in the garage air (Zhang et al. 2008; Glorennec et al. 2010).

The air pollutants of the commercial and high-rise residential UPGs in China partly came from the light gasoline vehicle emissions with kinds of pollutants, including not only a large number of CO, PM, and NO_x but also organic matters with various VOCs components, such as ethylene, ketones, benzene, and aromatic hydrocarbons (Dallmann et al. 2014; Hao et al. 2018). Cao et al. (2015) systematically studied VOCs' emission characteristics from 30 light gasoline vehicles

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(LDDV) in use in Beijing. It was found that alkanes, aromatic hydrocarbons, and carbonyl compounds are the main components in the exhaust gas, accounting for 36.4%, 33.1%, and 17.4%. For fresh motor vehicle exhaust, the concentration of aromatic hydrocarbons is high, and fresh motor vehicle exhaust is distributed in the air inside the garage, which is the primary source of benzene series in the ambient air. The US EPA has classified benzene as a human carcinogen (group A). The assessments in non-smoking populations have repeatedly identified benzene as a significant contributor to the cumulative risk of cancer.

The air pollutants of the UPGs monitoring indicators in previous studies usually only included CO and PM. Simultaneously, there was a lack of quantitative assessment of volatile organic compounds (VOCs) such as benzene, toluene, ethylbenzene, and xylenes (BTEX) in UPGs, which was more toxic and posed a severe threat to human health. BTEX belongs to the toxic and harmful air pollutants announced by US-EPA and International Agency for Research on Cancer (IARC), including benzene, toluene, ethylbenzene, and xylenes. Recognized as a class I carcinogenic compound by international carcinogenic institutions, benzene has neurotoxicity (causing neurasthenia, headache, insomnia, dizziness, the fatigue of lower limbs, and other symptoms) and genotoxicity (destroying DNA). Long-term exposure can lead to anemia and leukemia (<https://www3.epa.gov/ttn/atw/188polls.html>). Fossil fuels are the primary source of the monoaromatic organic compounds benzene, toluene, ethylbenzene, and xylenes (BTEX). From the environmental perspective, the BTEX plays a vital role as primary pollutants, considered some of the leading ozone precursors, contributing to severe possible health problems (Pariselli et al. 2009; Hu and Jiang 2014). Many research revealed that the air quality index in long tunnels, subways, and underground shopping areas was several times higher than those above ground (Colberg et al. 2005; Zhang et al. 2016). BTEX levels in private garages are 5 to 18 times higher than in adjacent living areas of single-family houses (Marć et al. 2016). For large commercial underground garages and high-rise residential subdivisions, traffic will be much more frequent than for individual garages; therefore, the pollution is likely to be more severe.

Scholars have studied the air quality of underground space (underground garage, underground shopping mall, subway); the research methods include numerical simulation and field measurement. For example, Song et al. simulate the particle concentration distribution of transient natural wind passing through UPG entrance by a three-dimensional numerical model (Song and Zhao 2017). A CFD model was developed to simulate a typical central garage's CO content under natural and mechanical ventilation to study the ventilation effect (Papakonstantinou et al. 2003). To estimate the pollution level and its potential impact on health, Zhang et al. take air samples

from 802 new cars parked in well-ventilated underground parking lots in Beijing, China, and found that the concentration of all pollutants in new vehicles was higher than that in old ones (Zhang et al. 2008). Lee et al. used parallel factor analysis to monitor and explain the periodic daily and seasonal changes of indoor air pollutants in subway stations online (Lee et al. 2014). Hun et al. studied indoor BTEX and MTBE concentrations in houses with attached garages, independent garages, or carports for natural ventilation. The results show that the volatile organic compounds related to gasoline in parked cars, especially in the connected garages, will reduce the indoor air quality (Hun et al. 2011). Mariuszmar et al. adopted passive sampling technology—Radiello diffusion passive sampler and graphitized charcoal box as an adsorption medium. A thermal desorption-gas chromatography-flame ionization detector (TD-GC-FID) system was used to carry out the release stage and final determination of collected analytes. Researchers monitored and analyzed BTEX in two-story underground parking lots and independent residential garages in a closed state; this was to verify that underground parking garages may be regarded as “hot spots” and particular emission sources BTEX to urban air. Moreover, they studied the relationship between the number of parked cars and BTEX concentration inside parking garages (Marć et al. 2016). According to the literature and the investigation, the pollutants in most underground garages exceed the standard, especially for VOCs emitted by motor vehicles in underground garages under natural ventilation (Shinohara et al. 2009). The main VOCs pollutants in the underground parking garages are benzene, toluene, and xylenes (Batterman et al. 2007; Kim et al. 2007; Gloennec et al. 2010). It is hard to diffuse and to dilute them thoroughly, and therefore the concentration there is much higher than outside.

Natural ventilation was the prevalent and sole method for air exchange in the residential and regular commercial underground garages in Xi'an, China. Although more severe air pollution could be found in naturally ventilated underground garages than mechanically ventilated ones, studies in several northern Chinese cities have shown that though CO concentrations of vehicle exhaust pollution hardly exceed the World Health Organization (WHO) long-term (8 h) CO exposure limit (25 ppm) for garages, PM and VOCs concentrations often exceeded the healthy range (Chan et al. 1997; Wang et al. 2008).

The mechanisms of the formation and diffusion of the traffic-related particulate matter and volatile organic chemicals were complex. Some particle formation and dilution studies had shown that the processes of formation and distribution of particles were strongly influenced by traffic volume and background conditions. TVOC concentrations were mainly influenced by vehicle emissions and closely related to traffic volume. As PM pollution in the outdoor environment had become severe, especially in northern China

cities, outdoor particles became critical particle sources in semi-enclosed environments such as underground garages (Li and Xiang 2013; Tong et al. 2016; Tham et al. 2018). It has been reported that PM pollutants of underground garages in northern Chinese cities not only comes from the exchange with a high concentration of polluted outdoor air but also related to the number of motor vehicles passing through the garage and emissions (Wang et al. 2008).

Although a considerable amount of research has been conducted on the air quality in underground garages, including CO, CO₂, PM, and TVOC, invalid research could be found on numerous sizable commercial complexes and high-rise residential areas in Chinese metropolitans. Moreover, reliable results were found blank concerning the concentration and spatial distribution of BTEX in UPGs. This study envisages the spatial and temporal distribution of pollutants such as TVOC, BTEX, PM_{2.5}, and PM₁₀ in natural ventilated underground garages of commercial centers and residential buildings. Statistical analysis of the main influencing factors, such as traffic volume, air exchange rate, and outdoor concentrations, was carried out. The study results indicate an indication of future human exposure to VOCs pollutants in underground spaces (tunnels and garages) and define design criteria for ventilation systems. TVOC and BTEX concentrations in underground garages should be considered essential pollution indicators when analyzing underground garage exposure levels and garage ventilation design. Conducting various VOCs concentrations in underground garages was vital for determining population health exposures and ventilation system design.

Experiment and methods

Sampling site

To determine the current status and daily variation characteristics of VOCs and PM pollution in underground garages, researchers have conducted observations shown in Fig. 1 at the Hanshen Shopping Plaza (site 1, N34° 20' 20.51", E108° 56' 12.26"), Long Shou Yuan District (site 2, N34° 17' 27.97", E108° 56' 55.67"), and Nansha Community (site 3, N34° 14' 36.29", E108° 59' 34.38") in Xi'an, China. The general information of three underground garages is shown in Table 1.

Mainly surrounded by residential areas with no apparent point source pollution, the first experimental sampling site was the Han Shen Shopping Plaza Commercial Complex (HS), located between the North Second Ring Road and the North Third Ring Road in Xi'an. The underground garage was located on the B2 floor with approximately 1000 spots and was ventilated only by natural means, with one entrance and two exits (as shown in Fig. 2a). The primary parked vehicles were private gasoline vehicles, with a high volume of traffic

and pedestrian traffic in the afternoon and evening (14:00–22:00). The garages were all level parking spaces with plastic flooring. The parking area was 21256 m², and the height was 3.0m. There were three sampling points located near the entrance, elevator entrance, and exit. The test height was 1.5m, and the observation period was 8:00–21:00 on 1–10 January 2020.

The second sampling site was Longshouyuan Gong Yuan residential subdistricts (GY), located in the central area of Xi'an City. The surrounding commercial and residential buildings were dense, with no apparent point pollution source. The underground parking garage was located on the B1 floor and had 200 mechanical and 500 flat parking spaces. The parking garage had a shared entrance and exit; ventilation was only natural, and the vehicles parked were private gasoline vehicles. The underground parking garage covers an area of 12792.8 m² and had a height of 5.0 m (as shown in Fig. 2b). Three sampling points were set up in the parking garage, located at the shared entrance and exit, the garage's central elevator of the garage, and the garage's garage corner. The observation period was 8:00–21:00 on 11–15 January 2020.

The third sampling site was the underground parking garage of the Nansha community, located at the southeast corner of the Second Ring Road in Xi'an City. There was a high density of commercial and residential buildings surrounded, with no distinct point source of air pollution. The underground garage was located on the B1 floor and had 230 mechanical parking spaces on three levels in a tightly arranged layout (as shown in Fig. 2c). The parking garage had heavy traffic flows, and the mechanical parking spaces was operated frequently. There was only one shared entrance and exit to the garage, and its ventilation was only by natural way. The vehicles parked here were private gasoline vehicles. This underground parking garage covers an area of 1968.3 m² with a height of 4.7 m. Two sampling points were set at the entrance and the central elevator in the garage, respectively. The observation period was 8:00–21:00 on 16–20 January, 2020.

The emission sources in underground garages of high-rise residents and commercial centers are mainly light-duty gasoline vehicles. Only a few domestic SUVs and light trucks are diesel vehicles (accounting for less than 5%). Local gasoline vehicles mostly use No. 92 or No. 95 gasoline, and the emission standard is China's national four or national five standard vehicles. The emission standard can be consulted by the Ministry of Ecology and Environmental Protection of China (<https://www.mee.gov.cn/>).

Parameters and instruments

Meteorological data was collected from the mobile weather station (WS30, Honeywell), including speed and direction of the wind at parking garage entrances and exits and

Table 1 General information of the research subjects—underground garages attached to the commercial center and high-rise residential buildings

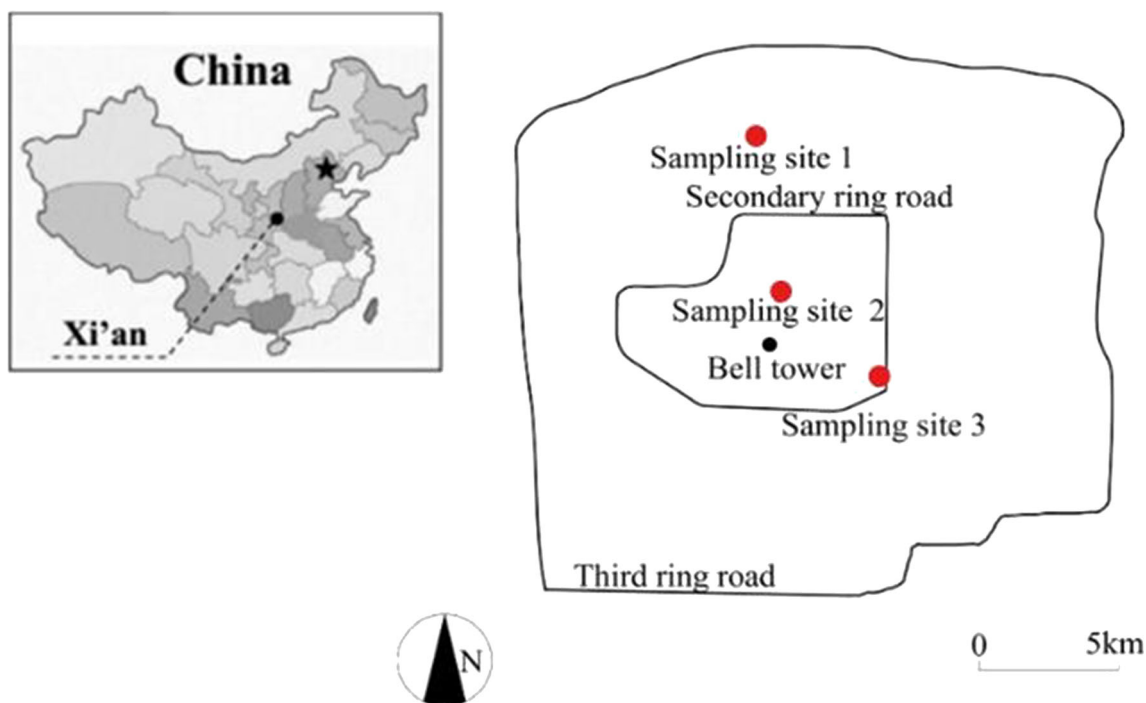
Garages	Internal diameters	Temperature	Vehicle characteristics	Garages situation
A. Hanshen shopping plaza	302.2m×70.3m×3.0m 63734.0 m ³	Average: 9.0°C Maximum: 15.4°C Minimum: 2.9°C	Vehicle classification: mostly car segment C (small family car) Fuel type: gasoline (~90%) and diesel (~10%)	Type of garages: ~1000 flat garages spot (with car washing place) Entrance and exit: 1 entrance and 2 exits Traffic flow: ~120 vehicles per hour
B. Gongyuan residential subdistricts	182.2m×70.2m×5.0m 63952.2 m ³	Average: 11.0°C Maximum: 17.2°C Minimum: 4.5°C	Vehicle classification: mostly car segment C (small family car) Fuel type: gasoline	Garages type: 700 underground garages, including little mechanical multistory garages (~30%) Entrance and exit: 1 shared entrance and exit Traffic flow: ~13 vehicles per hour
C. Nansha community	97.1m×20.2m×4.7m 9218.7 m ³	Average: 12.0°C Maximum: 17.1°C Minimum: 8.7°C	Vehicle classification: mostly car segment C (small family car) Fuel type: gasoline	Type of garages: 300 mechanical multistory garages Garages entrance and exit: 1 shared entrance and exit Traffic flow: ~13 vehicles per hour Internal air condition: the mechanical garages operated frequently, and the car owners generally have the habit of preheating the car in the garage lot. Therefore, the smell of gasoline was evident during the peak traffic period

temperature and humidity at sampling points. The meteorological station was monitored at an altitude of 1.5 m from the ground.

The concentrations of PM₁₀ and PM_{2.5} in UPGs were obtained from an aerosol monitor (DustTrak 8530, TSI) with an accuracy of 1.0 µg·m⁻³, and the set of test intervals was 30 min. The instrument was calibrated before the experiment. Briefly, we used this instrument with another membrane concentration monitoring device (MiniVol TAS, Airmetrics) to monitoring for 24 h and calibrated it by comparing the data

from two devices. This method meets the standards of the Chinese environmental protection department standards, similar to the USEPA-designated Federal Reference Method (FRM). The ambient concentrations of PM₁₀ and PM_{2.5} were obtained from the China national environmental monitoring center (<http://www.cnemc.cn/>), including the Xiaozhai region, Economic Development region, and Municipal People's Stadium in this study, with a time resolution of 1 h.

TVOC concentration data were measured by a hand-held in-line detection device (PGM-7340, RAE) based on a PID

**Fig. 1** Locations of three sampling sites in Xi'an, China

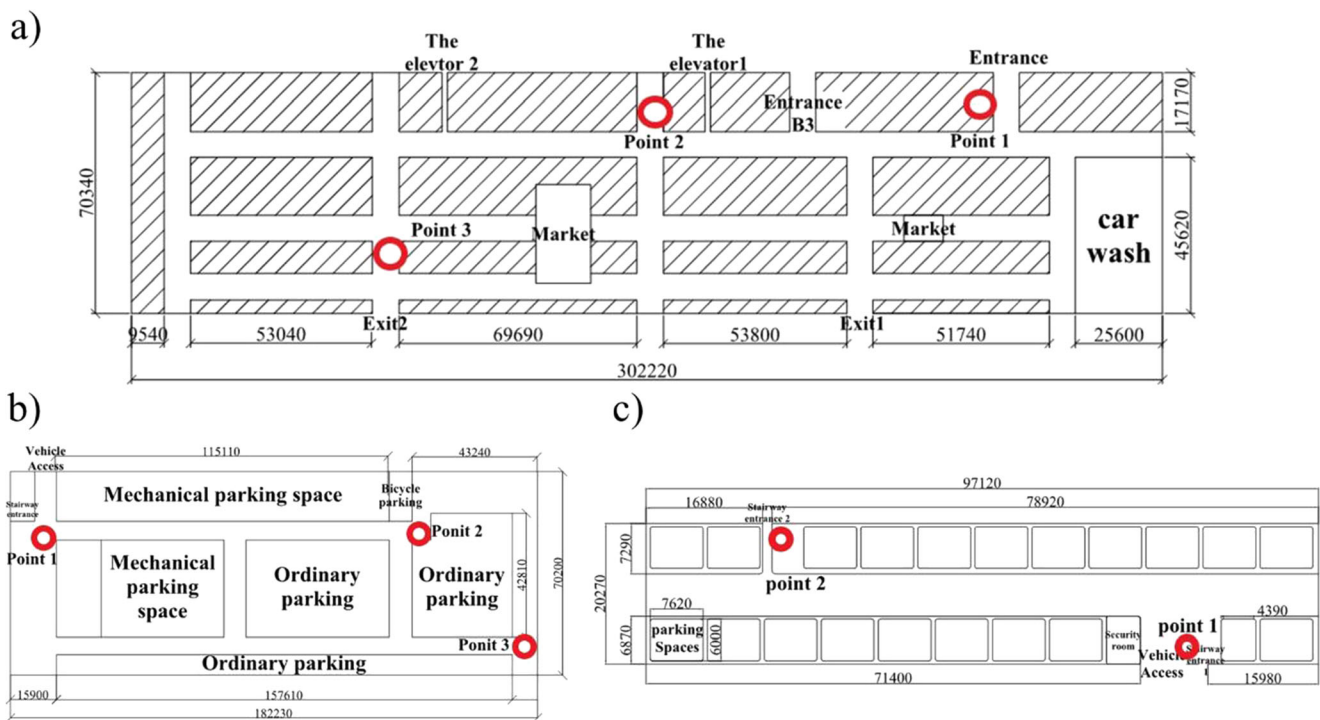


Fig. 2 (a) Plan of the Hanshen Shopping Plaza Commercial Complex (HS) underground parking garage. (b) Plan of the Longshouyuan Gong Yuan residential subdistricts (GY) underground parking garage. (c) Plan of the Nansha community (NS) underground parking garage (lengths are in mm)

sensor with a high degree of precision (ppb), calibrated to a ppb accuracy before use. HCHO concentration data were measured by a hand-held in-line detection device (FP-30MK2, Riken) based on the dipstick photoelectric photometric method. The devices can meet the national standard of the People’s Republic of China GB9663 “Public Places Formaldehyde Determination Method” completely. The time resolution of the above two online monitoring devices is set at 30 min.

BTEX was collected by sampling tubes (MK-G20). Sample tubes contained 100 mg of activated coconut charcoal. For the fabrication of the sampling tubes, coconut charcoal was heated in an oven at 200 °C overnight to facilitate the adsorbed compounds to escape and then cooled in desiccators. At each sampling site, sampling tubes, including one field blank, were exposed at the height of ~1.5 m. The charcoal was packed in glass tubes, 6 mm id×80 mm sealed from one side with a Teflon cap. Glass wool plugs on either side supported the charcoal bed. The other side of the tube was left open and kept in the oven overnight at 250 °C for conditioning, and then the other end of the tube was sealed with a Teflon cap. The tubes were then opened only at the sampling site. The sampling tubes were filled after exposure. The charcoal samples were transferred from the sealed tubes into a glass vial and sealed immediately to prevent further adsorption of compounds and were stored at 4°C until analysis.

A programmable minipump was calibrated using a bubble flow meter. A breakthrough experiment was conducted by pumping air at a flow of 0.5 L/min in 50 min through the sampling tube. Sample tubes were sealed with polypropylene caps and sent to the laboratory. The activated carbon in the sampling tube was poured into a stop-scale test tube, and 1.0 ml of carbon disulfide was added to tighten the tube plug. The tube was placed for 1 h and vibrated from time to time. Take a 1.0 μL sample to analyze qualitatively with retention time and quantitatively with peak height. Simultaneously, an unsampled activated carbon tube was taken and operated as a sample tube to measure the average peak height of the blank tube. Samples were analyzed within 1 week after the sampling.

All samples were analyzed by a system consisting of a Thermal Desorber coupled with a gas chromatography (GC) (HP 7890A, USA) fitted with a flame ionization detector (FID) (Agilent, USA). The sampling tubes were thermally desorbed for 10 min at 300 °C with a flow of pure helium gas passing through and carrying the desorbed gases to a pre-concentration trap at -10 °C. Following the tube desorption, the trap was thermally desorbed at 300 °C for 3 min, and gases were transferred to the GC/FID for determination. While high-purity helium gas was used as the carrier gas with a flow rate of 1.0 mL min⁻¹, high-purity nitrogen gas was used as the make-up gas at a 7 mL min⁻¹. The flow rates for H₂ and air were 30 mL min⁻¹ and 350 mL min⁻¹, respectively. The GC

oven temperature was set initially to 45 °C for 2 min, increasing at a rate of 10 °C min⁻¹ to 240 °C and then holding for 5 min.

An external standard calibration quantified the concentrations of individual BTEX species. A BTEX mixture (BTEX-1RPM, ChemService, USA) in methanol with purity greater than 98% was used to prepare the calibration solution. A five-point calibration (1, 5, 10, 50, and 200 ppmv) was performed for quantifying the BTEX species. These standard solutions were used to produce calibration curves and were analyzed with the samples. A good correlation was found for all measured BTEX species for benzene, toluene, m,p-xylenes, and o-xylene, respectively.

The number of traffic flow and vehicle types was captured by video for latter count and analysis.

Sampling methods

The sampling points were located in areas with a high frequency of crowd activity or vehicle activity. Therefore 2-3 sampling points were set up in each parking garage at the height of ~1.5 m, measuring both PM, TVOC, and BTEX concentrations simultaneously. The time intervals of PM and TVOC were set at 5 min. BTEX was sampled by absorbent tubes at one-hour intervals and three-periods per day, respectively 8:00-12:00, 14:00-17:00, and 18:00-21:00. Two days of sampling were conducted at each sampling site, located next to a high traffic corridor or places where people enter or exit. Such places represent the main areas of human activity. Online monitoring of outdoor conventional pollutant concentrations was also set up during the period of the experiment.

Quality assurance and quality control

The quality assurance and quality control (QA/QC) measures included laboratory and field blanks and triplicate samples measurements. As determined from replicate analyses of the standards and samples, precision was within ±15%, and those exceeding were discarded. For laboratory blank, unexposed charcoal tubes were analyzed for VOCs similar to the exposed ones. For the BTEX ranking concerning their contribution to O₃ formation, MIR coefficients as established by Carter (1994) and the rate constants of VOC–OH reactions as studied by Atkinson (1997) were used.

Results and discussion

According to the standard of indoor ambient air quality suggested by Chinese government and US Environmental Protection Agency, the standard daily average limits of PM₁₀ and PM_{2.5} mass concentrations were 0.15 mg·m⁻³ and 0.075 mg·m⁻³, respectively (US Environmental Protection

Agency 1990; China. MEP GB/T18883–2002 2002). The concentration limits of TVOC, benzene, toluene, and xylenes suggested by Chinese government were set at 0.60 mg·m⁻³, 0.11 mg·m⁻³, 0.20 mg·m⁻³, and 0.20 mg·m⁻³, respectively (China. MEP GB/T18883–2002 2002). The daily average concentration of PM_{2.5} and PM₁₀ suggested by the WHO Air Quality Guideline (AQG) were set at 0.025 mg·m⁻³ and 0.05 mg·m⁻³, respectively (World Health Organization 2006, 2010), as well as EU set value (European Union 2008), which was three times lower than Chinese and US standard. Garages and parking facilities were specific buildings of buildings and were among the most frequent places that residents will enter.

The concentrations of PM_{2.5}, PM₁₀, total VOCs, benzene, toluene, xylenes, and formaldehyde in each underground parking garage were measured in this study. To compare with VOCs' standards, researchers determined the average percentages of VOCs' main components in parking facilities and converted the data from ppb units to mg·m⁻³ units. According to the previous studies, the eight most abundant VOCs emitted in vehicle exhaust have been identified in many parking facilities (Batterman et al. 2006, 2007; Glorennec et al. 2010). Benzene, toluene, ethylbenzene, and xylenes (BTEX) are aromatic hydrocarbons that were categorized as hazardous air pollutants (HAP) by the US EPA (US Environmental Protection Agency 1999). The concentrations of PM and target components of VOCs in garages presented in this study were shown in Table 2.

Characteristics of average air pollutant concentrations of underground parking garages in commercial and residential buildings

In this study, the underground parking garages were all naturally ventilated with low ventilation efficiency, and pollutants' concentrations were uneven inside the garages. There were differences between high-frequency areas of human and vehicular activity. When natural ventilation was not optimal, there was an accumulative increase in pollutant concentrations in parts of the garage that eventually reached the concentration that will threaten human health.

The summary of the daily average target pollutant concentrations and over standard rate in all samples were listed in Table 2. The comparison of the indoor and outdoor concentrations of PM_{2.5} and PM₁₀ was shown in Figs. 3 and 5. Compared with indoor and outdoor PM_{2.5} concentrations, it is found that the indoor concentration is generally higher than the outdoor, with the I/O (indoor/outdoor) value of 1.15. The result is close to Chan et al. (I/O=1.16) (Chan et al. 2004). The daily average indoor PM_{2.5} concentrations of HS-3, HS-1 NS-1, and NS-2 were highest among all samples that were 0.21 mg·m⁻³, followed by the HS-3, GY-2, GY-3, and GY-1 that were 0.20 mg·m⁻³, 0.16 mg·m⁻³, 0.16 mg·m⁻³, and 0.14 mg·m⁻³, respectively, while the traffic volume of HS, GY, and NY were 121.9,

Table 2 Comparison of target pollutant concentrations and over standard rates in each sample

Case no.	Indoor				LOQ	Outdoor	Indoor-outdoor	Reference values	Over standard rate	
	Mean	SD	Min	Max					GB	AQG
TVOC ($\text{mg}\cdot\text{m}^{-3}$) $N=200(\text{HS})/100(\text{GY and NS})$										
HS-1	0.77	0.23	0.55	1.25	ppb			GB=0.60 $\text{mg}\cdot\text{m}^{-3}(\text{hour})$	28.1%	
HS-2	1.15	0.43	0.50	1.75					91.7%	
HS-3	1.23	0.43	0.70	2.00					104.2%	
GY-1	1.19	0.18	1.00	1.50					97.6%	
GY-2	1.08	0.10	1.00	1.20					80.2%	
GY-3	1.31	0.16	1.10	1.50					119.0%	
NS-1	1.18	0.48	0.60	2.00					95.8%	
NS-2	1.22	0.35	0.75	1.75					103.5%	
Benzene ($\text{mg}\cdot\text{m}^{-3}$) $N=100(\text{HS})/50(\text{GY and NS})$										
HS-1	0.06	0.05	0.01	0.15	$1.5 \mu\text{g}\cdot\text{m}^{-3}$			GB=0.11 $\text{mg}\cdot\text{m}^{-3}(\text{hour})$	<i>N</i>	
HS-2	0.11	0.13	0.04	0.40				Europe=0.005 $\text{mg}\cdot\text{m}^{-3}(\text{year})$	<i>N</i>	
HS-3	0.21	0.21	0.03	0.50					88.6%	
GY-1	0.04	0.02	0.02	0.06					<i>N</i>	
GY-2	0.05	0.02	0.03	0.08					<i>N</i>	
GY-3	0.09	0.06	0.03	0.20					<i>N</i>	
NS-1	0.17	0.14	0.05	0.40					52.9%	
NS-2	0.15	0.12	0.05	0.40					40.2%	
Toluene ($\text{mg}\cdot\text{m}^{-3}$) $N=100(\text{HS})/50(\text{GY and NS})$										
HS-1	0.22	0.11	0.05	0.43	$1.5 \mu\text{g}\cdot\text{m}^{-3}$			GB=0.20 $\text{mg}\cdot\text{m}^{-3}(\text{hour})$	7.8%	
HS-2	0.35	0.16	0.10	0.65					76.6%	
HS-3	0.43	0.29	0.18	1.00					117.2%	
GY-1	0.28	0.14	0.09	0.50					42.1%	
GY-2	0.18	0.11	0.05	0.40					<i>N</i>	
GY-3	0.27	0.14	0.02	0.50					35.7%	
NS-1	0.35	0.17	0.20	0.80					72.9%	
NS-2	0.38	0.14	0.20	0.70					91.7%	
Xylenes ($\text{mg}\cdot\text{m}^{-3}$) $N=100(\text{HS})/50(\text{GY and NS})$										
HS-1	0.15	0.07	0.03	0.27	$1.5 \mu\text{g}\cdot\text{m}^{-3}$			GB=0.20 $\text{mg}\cdot\text{m}^{-3}(\text{hour})$	<i>N</i>	
HS-2	0.23	0.11	0.06	0.37					14.2%	
HS-3	0.31	0.21	0.13	0.67					56.3%	
GY-1	0.16	0.09	0.03	0.27					<i>N</i>	
GY-2	0.16	0.11	0.02	0.33					<i>N</i>	
GY-3	0.20	0.09	0.13	0.40					<i>N</i>	
NS-1	0.23	0.13	0.10	0.53					14.6%	
NS-2	0.25	0.09	0.13	0.40					25.0%	
PM _{2.5} ($\text{mg}\cdot\text{m}^{-3}$) $N=200(\text{HS})/100(\text{GY and NS})$										
HS-1	0.21	0.04	0.15	0.27	$0.001 \text{ mg}\cdot\text{m}^{-3}$	0.038		GB/EPA=0.075 $\text{mg}\cdot\text{m}^{-3}(\text{day})$	177.3%	732.0%
HS-2	0.15	0.05	0.12	0.28		0.17		AQG=0.025 $\text{mg}\cdot\text{m}^{-3}(\text{day})$	100.0%	500.0%
HS-3	0.24	0.05	0.12	0.30				Europe=0.025 $\text{mg}\cdot\text{m}^{-3}(\text{year})$	219.3%	858.0%
GY-1	0.14	0.06	0.06	0.20					90.9%	472.7%
GY-2	0.16	0.06	0.09	0.23		0.13	0.034		118.2%	554.7%
GY-3	0.16	0.07	0.07	0.23			0.033		117.1%	551.4%
NS-1	0.21	0.02	0.19	0.25			0.002		182.8%	748.4%
NS-2	0.20	0.02	0.17	0.24		0.21	-0.002		177.1%	731.3%
PM ₁₀ ($\text{mg}\cdot\text{m}^{-3}$) $N=200(\text{HS})/100(\text{GY and NS})$										
HS-1	0.26	0.05	0.19	0.33	$0.001 \text{ mg}\cdot\text{m}^{-3}$	0.067		GB/EPA=0.15 $\text{mg}\cdot\text{m}^{-3}(\text{day})$	71.4%	414.3%

Table 2 (continued)

Case no.	Indoor				LOQ	Outdoor	Indoor-outdoor	Reference values	Over standard rate	
	Mean	SD	Min	Max					GB	AQG
HS-2	0.18	0.06	0.14	0.35		0.19	-0.006	AQG=0.05 mg·m ⁻³ (day)	22.5%	267.6%
HS-3	0.28	0.06	0.14	0.34			0.093	Europe=0.050 mg·m ⁻³ (year)	88.3%	465.0%
GY-1	0.17	0.08	0.07	0.26			0.005		10.0%	230.0%
GY-2	0.19	0.08	0.10	0.28		0.16	0.033		28.8%	286.3%
GY-3	0.20	0.09	0.08	0.29			0.037		31.2%	293.7%
NS-1	0.26	0.02	0.22	0.29			0.029		72.7%	418.0%
NS-2	0.26	0.03	0.22	0.30		0.23	0.030		73.6%	420.9%
Formaldehyde (mg·m ⁻³) N=200(HS)/100(GY and NS)										
HS-1	0.26	0.05	0.19	0.33	0.01 mg·m ⁻³		0.067	GB=0.10 mg·m ⁻³	160.0%	160.0%
HS-2	0.18	0.06	0.14	0.35		0.19	-0.006	AQG=0.10 mg·m ⁻³	80.0%	80.0%
HS-3	0.28	0.06	0.14	0.34			0.093		180.0%	180.0%
GY-1	0.17	0.08	0.07	0.26			0.005		70.0%	70.0%
GY-2	0.19	0.08	0.10	0.28		0.16	0.033		90.0%	90.0%
GY-3	0.20	0.09	0.08	0.29			0.037		100.0%	100.0%
NS-1	0.26	0.02	0.22	0.29			0.029		160.0%	160.0%
NS-2	0.26	0.03	0.22	0.30		0.23	0.030		160.0%	160.0%

AQG WHO Air Quality Guideline, GB Chinese National Standard, EPA US Environmental Protection Agency; Europe: 2008/50/EU (<https://ec.europa.eu/environment/air/quality/standards.htm>)

13.0, and 13.3 vehicles per hour, respectively. Besides, the daily indoor average concentration of PM₁₀ was also slightly higher than outdoors, with the average *I/O* value of 1.19. Figure 3 showed the spatial distribution of PM concentrations in the HS commercial UPG. The average daily PM_{2.5} and PM₁₀ concentrations of three sampling points in Hanshen commercial center were not close, with 0.21–0.24 mg·m⁻³ at the entrance (HS1) and exit (HS3), and only 0.15 mg·m⁻³ at the elevator point (HS2), respectively. In this study, the monitoring period is

January, and the ambient concentration of PM_{2.5} was as high as 0.13–0.21 mg·m⁻³. The UPGs are mainly filled with light gasoline vehicles, whose primary PM_{2.5} emission concentration was only about 0.2–1.0 mg/km (Shen et al. 2014). Besides, the ultraviolet light intensity in the underground space is close to zero, which leads to the low speed of secondary reactions. Therefore, the pollutants exhausted by vehicles were primary. When the outdoor PM_{2.5} concentration was high (0.17–0.21 mg·m⁻³), the PM_{2.5} concentrations at the UPGs entrance and exit

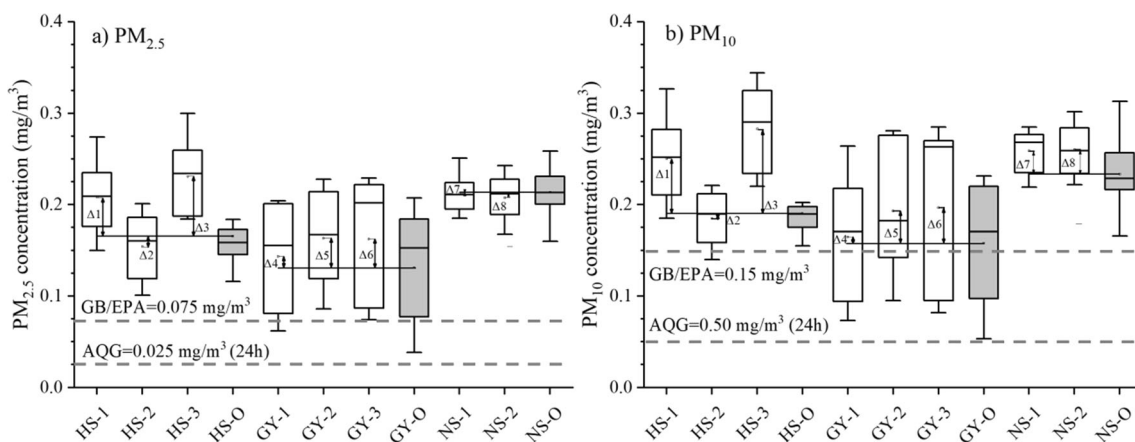


Fig. 3 Comparison of indoor and outdoor PM_{2.5} concentrations in the three UPGs of all samples. HS-1, HS-2, and HS-3 indicate three sampling points near the entrance, elevator entrance, and exit of the Hanshen commercial complex garage. GY-1, GY-2, and GY-3 indicate three sampling points near the shared entrance and exit, elevator entrance,

and Gongyuan residential garage corner. NS-1 and NS-2 mark the shared entry and exit and the elevator entrance of the Nansha residential garage. HS-0, GY-0, and NS-0 indicate the outdoor concentration of PM. Δ1~Δ8 indicates the differences between indoor and outdoor PM concentrations at different monitoring points

points (HS-1, HS-3, NS-1) were higher than that at the internal point (HS-2, NS-2). When the outdoor $PM_{2.5}$ concentration was relatively low ($0.13 \text{ mg}\cdot\text{m}^{-3}$), the concentrations at the entrance and exit point (GY-1) were lower than that at the internal point (GY-2, GY-3). The ratios of indoor and outdoor $PM_{2.5}$ and PM_{10} concentrations ranged from 0.88 to 1.41 and 0.95 to 1.47. In general, $PM_{2.5}$ concentration in underground parking lots in Xi'an depends more on ambient $PM_{2.5}$ concentration in winter.

Among all the samples, the daily average concentration of total VOCs was $1.14 \text{ mg}\cdot\text{m}^{-3}$, and that of GY-3 was the highest, which was $1.31 \text{ mg}\cdot\text{m}^{-3}$, followed by HS-3 and NS-2. Meanwhile, the daily average concentration of benzene, toluene, and xylenes was $0.11 \text{ mg}\cdot\text{m}^{-3}$, $0.31 \text{ mg}\cdot\text{m}^{-3}$, and $0.32 \text{ mg}\cdot\text{m}^{-3}$, respectively; GY-3 was the highest, followed by HS-3 and NS-2. HS-3 was located near the exit of the commercial UPG; both GY-3 and NS-2 were situated at the corner of residential UPGs. Figure 4 showed that the VOCs concentrations were the highest at the exit ($1.23 \text{ mg}\cdot\text{m}^{-3}$ for TVOC and $0.43 \text{ mg}\cdot\text{m}^{-3}$ for toluene, respectively), followed by the elevator inside the garage ($1.15 \text{ mg}\cdot\text{m}^{-3}$ for TVOC and $0.35 \text{ mg}\cdot\text{m}^{-3}$ for toluene, respectively) and the lowest concentration at the entrance ($0.77 \text{ mg}\cdot\text{m}^{-3}$ for TVOC and $0.22 \text{ mg}\cdot\text{m}^{-3}$ for toluene, respectively).

On the other hand, VOCs pollution came mainly from vehicle emissions in the parking garage. The higher VOC concentrations were found at the commercial center parking garage's exit because the engine could not reach the optimum operation parameters at the beginning period. The use of a "cold start engine" (unfavorable conditions for burning liquid fuels in engines) in underground parking garages resulted in high concentrations of PM and VOCs in the air. Additionally, the high density of vehicles at the exit due to the queues for collecting parking fees combined with the cold start of engines and the idling process together resulted in significantly higher

emission of exhaust pollutants at this time. There was no such problem at the entrance because of the perturbation of vehicle operation, a large amount of incoming outdoor ambient air, and better ventilation. However, traffic flow was also high. The concentration of organic matter in the air was lower than the exit from 42.2 to 80.0% at the entrance.

The entrance and exit to the two residential areas' underground parking garages were shared (shown in Fig. 2). Its natural ventilation was smaller than that of the underground parking garage in the commercial center, resulting in significantly different spatial distribution characteristics of pollutant concentrations. The NS parking garage was relatively small, with approximately 230 mechanical parking spaces on three levels. Although there was only one natural vent, pollutant concentrations were evenly distributed, with no significant statistical differences in pollutant concentrations at any point. GY's parking garage area and height value were larger than NS, with approximately 700 level parking spaces and only one natural vent.

Due to the lower outdoor ambient PM concentrations during the monitoring period, PM pollutant concentrations were lower at the entrance point with better ventilation than the garage's point. Figure 4 demonstrated the characteristics of VOCs concentrations as below: garage corners were higher than entrances, and entrances were higher than a central garage. The garage's interior of the garage was a complex space with an average wind speed of less than 0.1 m/s per minute in the corners of the garage year round. Poor air diffusion conditions determined the accumulation of vehicle fuel volatiles and exhausted pollutants and resulted in higher TVOC, benzene, toluene, and xylenes.

When the number of vehicles parked in the parking garage was large, the diffusion was not effective when the natural ventilation was the single mode and made the apparent prominent accumulation of the pollutants in the garage's corner.

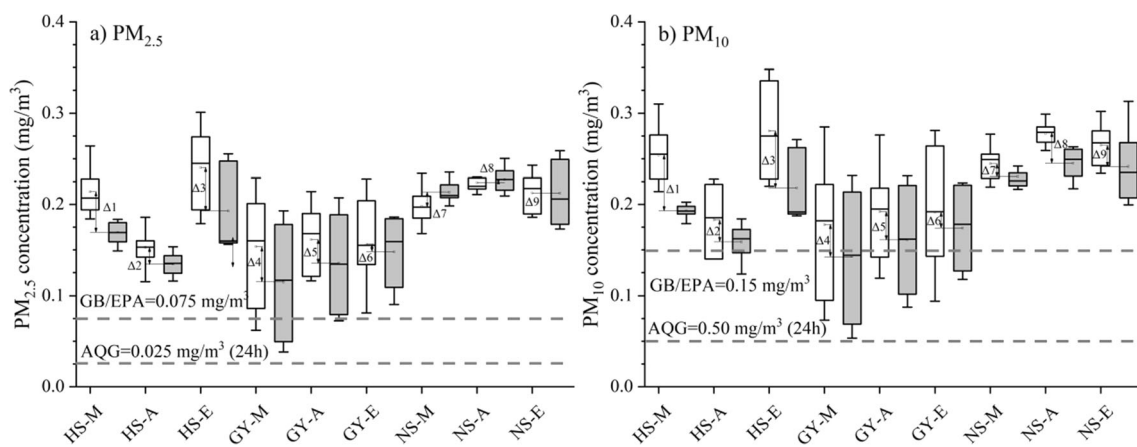


Fig. 5 Comparison of indoor and outdoor $PM_{2.5}$ concentrations in the three periods of all samples. The white boxes showed the PM concentration in the underground garage, and the gray boxes showed the outdoor ambient PM concentration during the same period. M, A,

and E indicate the morning, afternoon, and evening. $\Delta 1\sim\Delta 9$ indicates the differences between indoor and outdoor PM concentrations at different monitoring points

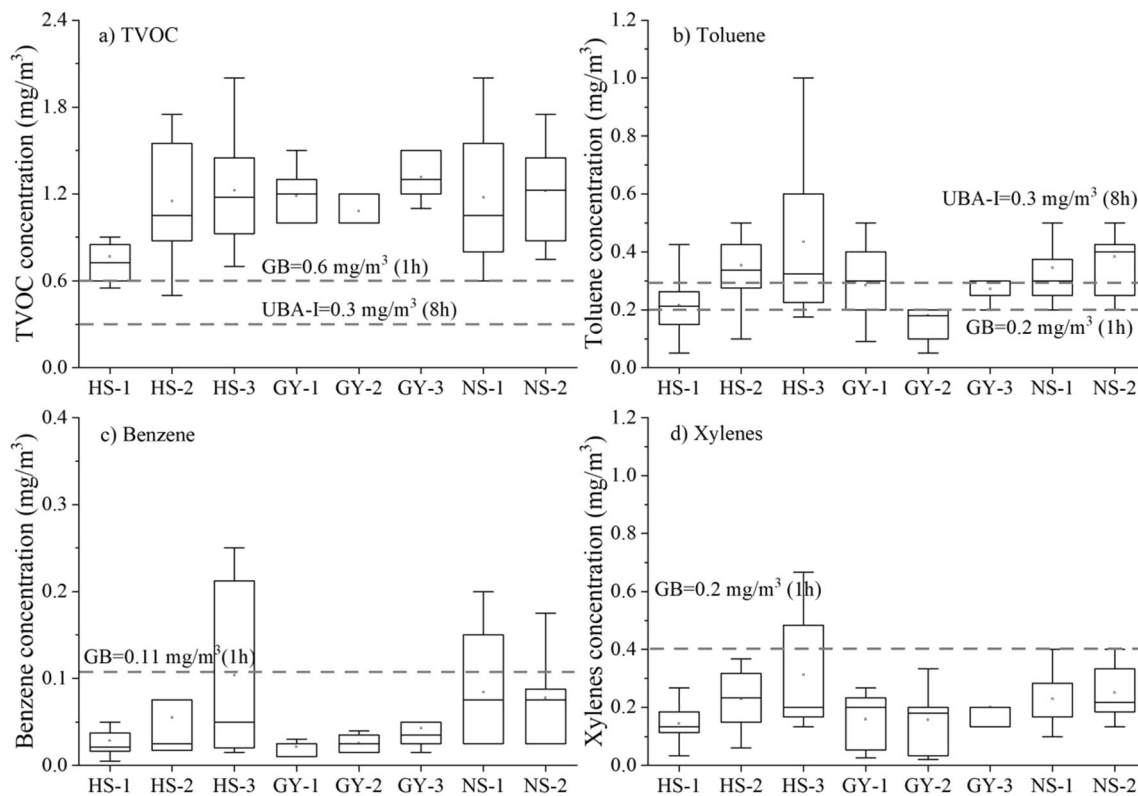


Fig. 4 The average concentrations of benzene, toluene, xylenes, and TVOC of all sampling sites in three UPGs. GY-1, GY-2, and GY-3 indicate three sampling points located at the shared entrance and exit,

the garage's central elevator, and the garage's corner, respectively. NS-1 and NS-2 indicate the two sampling sites located at the shared entrance and exit and the garage's central elevator, respectively

During the monitoring periods, at the interior or corner of the garage, benzene, toluene, xylenes, and TVOC, all exceed the indoor air quality standards. The average time-weighted rate of exceeding the standard was 13.4%, 50.5%, 17.7%, and 91.8%, respectively. Therefore, elevators in garages with frequent human activities should not be located in corners with poor ventilation, for high concentrations of BTEX may pose a higher health risk.

Analysis and comparison of pollutants at different points in commercial and residential underground garages at different periods

Figure 5 demonstrated the average PM concentration values for the commercial center underground garage and two residential underground garages over three periods of the day. The three periods were 8:00–12:00 a.m., 14:00–17:00 p.m., and 18:00–21:00 p.m., respectively. We also compared the indoor and outdoor concentrations of $PM_{2.5}$ and PM_{10} during three periods. It was noteworthy that the $PM_{2.5}$ concentrations in commercial and residential garages exceeded $0.075 \text{ mg}\cdot\text{m}^{-3}$ at all times (average daily values were 0.20, 0.16, and $0.21 \text{ mg}\cdot\text{m}^{-3}$, respectively) and PM_{10} concentrations exceeded $0.15 \text{ mg}\cdot\text{m}^{-3}$ at all times (average daily values were 0.24, 0.18, and $0.26 \text{ mg}\cdot\text{m}^{-3}$, respectively). The PM concentrations

do not vary significantly in different periods. Wang et al. (2008) showed that the main influence factor of winter fine PM pollution in naturally ventilated underground parking garages in northern Chinese cities was ambient atmospheric PM. To determine the severity of fine PM pollutants in underground parking garages, we compared PM concentrations in underground parking garages with those in the outdoor ambient atmosphere. During the monitoring period, the daily average outdoor ambient $PM_{2.5}$ concentrations in three underground parking garages were 0.15, 0.11, and $0.19 \text{ mg}\cdot\text{m}^{-3}$, and the daily average PM_{10} concentrations were 0.16, 0.12, and $0.21 \text{ mg}\cdot\text{m}^{-3}$, respectively. The garages' PM concentrations were equal or slightly higher than the outdoor ambient PM concentrations when little cars were entering or leaving the UPGs. However, with the increase in traffic volume, the difference between indoor and outdoor PM concentrations increased considerably. Moreover, the difference between the PM concentrations in the commercial garages with higher traffic flow and the outdoor ambient PM concentrations was 2 to 4 times higher than the other two residential garages in the evening, which is the peak traffic volume period. Therefore, it can be concluded that the variation trend of the difference between indoor air and outdoor PM concentrations depended on that of traffic volume to a large extent, which is a similar result to Liu et al. (Liu et al. 2019).

Figure 6 showed the average concentration values of VOCs, (a) TVOC, (b) toluene, (c) benzene, and (d) xylenes, in one commercial center underground parking garage and two residential underground parking garages for three periods of the day. The concentration values of VOCs in the two types of underground parking garages showed differences in temporal distribution. Firstly, TVOC concentrations of the underground garage at the commercial center were lower (average value was $0.91 \pm 0.23 \text{ mg} \cdot \text{m}^{-3}$) in the morning and higher (average value was $1.42 \pm 0.44 \text{ mg} \cdot \text{m}^{-3}$) in the evening. The TVOC concentrations were higher in residential garages (average values were $1.28 \pm 0.47 \text{ mg} \cdot \text{m}^{-3}$) in the morning and evening and lower in the afternoon (average value was $1.08 \pm 0.35 \text{ mg} \cdot \text{m}^{-3}$), which is due to the temporal distribution of the traffic flow. Observations showed that the commercial center's underground parking garage's average traffic flow was only 71.7/h in the morning. The values increased to 131.7/h and 162.2/h in the afternoon and night, which exceeded the morning traffic flow by 83.7% and 126.2%, respectively. The TVOC concentration exceeded the traffic flow in the morning by 68.4% and 73.8%, respectively. The average traffic of underground parking garages in residential areas in the morning and night was 18.1/h and 15.0/h, compared to 6.3/h in the afternoon. Traffic exceeded in the afternoon by 187.3% and 138.1%, respectively. In the morning, TVOC concentrations exceeded in the afternoon by 28.4%

and 29.7%, respectively. Traffic volume becomes a significant influence factor on the concentration of VOCs in underground parking garages.

Inter-species correlation

In the present study, Pearson's correlation (Table 3, 2-tailed, $p < 0.01$) of the concentrations of TVOC, BTEX members, $\text{PM}_{2.5}$ in the UPGs, outdoor ambient $\text{PM}_{2.5}$, and the difference between indoor and outdoor $\text{PM}_{2.5}$ concentrations was checked for different monitoring sites. Xylenes were significantly correlated ($R^2 > 0.85$, $p < 0.01$) with toluene and benzene in commercial and residential UPGs. Since xylenes come mainly from a traffic source, it can be used to indicate other aromatic compounds in heavy traffic areas. Besides, UPGs' $\text{PM}_{2.5}$ concentration was significantly positively correlated with the ambient $\text{PM}_{2.5}$ concentration. The correlation coefficient of indoor $\text{PM}_{2.5}$ and outdoor ambient $\text{PM}_{2.5}$ is as high as 0.69 and 0.66 ($p < 0.01$) in commercial UPG and residential UPGs. Therefore, the UPGs' $\text{PM}_{2.5}$ were dominated by the outdoor ambient $\text{PM}_{2.5}$. Meanwhile, the $\delta\text{PM}_{2.5}$ concentration (outdoor-indoor) was positively correlated with BTEX and indoor $\text{PM}_{2.5}$ concentration but negatively correlated with outdoor $\text{PM}_{2.5}$ concentration.

Table 3 Summary of the concentration of air pollutant correlation coefficients (R^2) at different UPGs

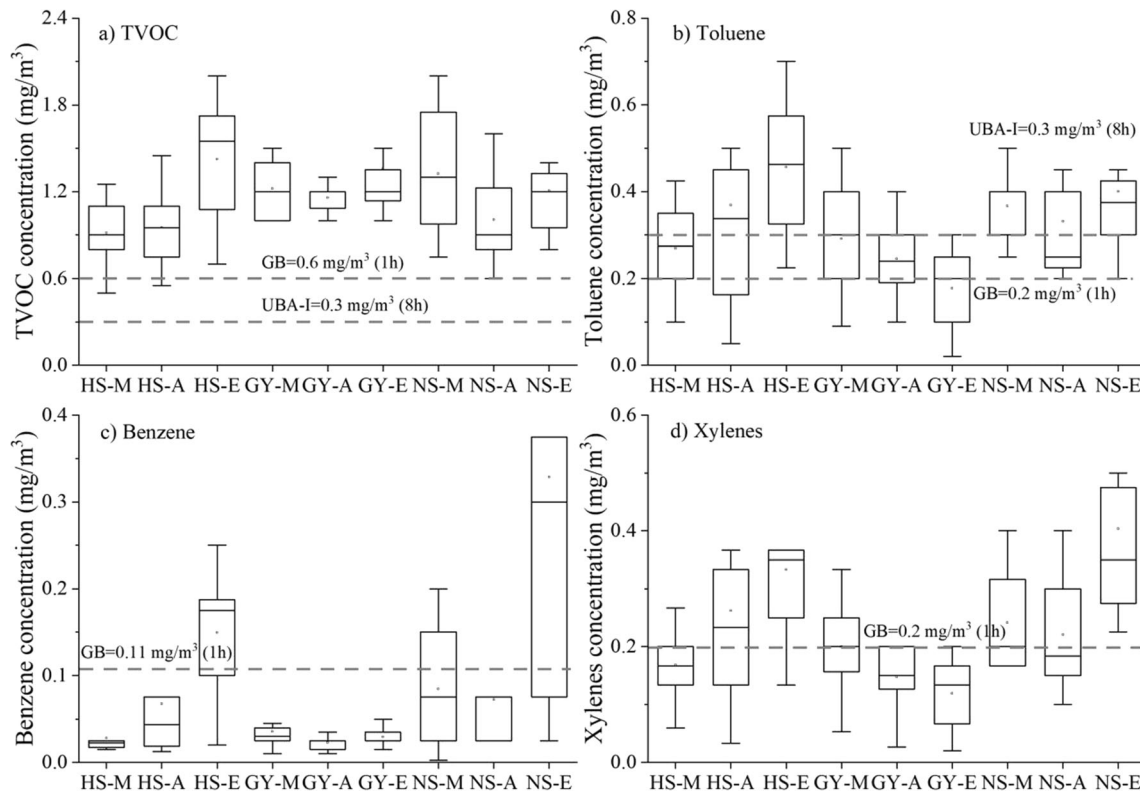


Fig. 6 The average concentration of benzene, toluene, xylenes, and TVOC in three underground garages for three periods of the day. M, A, and E indicate the morning, afternoon, and evening

Commercial UPG (<i>N</i> =200/100)	TVOC	Benzene	Toluene	Xylenes	HCHO	PM2.5-In	PM2.5-Out	δPM2.5
TVOC	1.00							
Benzene	0.67**	1.00						
Toluene	0.74**	0.79**	1.00					
Xylenes	0.69**	0.85**	0.92**	1.00				
HCHO	−0.15	−0.07	−0.03	0.08	1.00			
PM2.5-In	−0.12	0.53*	0.50*	0.33	0.34	1.00		
PM2.5-Out	0.13	0.56*	0.01	−0.05	−0.22	0.69**	1.00	
δPM2.5	−0.35	0.06	0.54*	0.33	0.53**	0.45	−0.33	1.00
Residential UPGs (<i>N</i> =200/100)								
TVOC	1.00							
Benzene	0.82**	1.00						
Toluene	0.74**	0.88**	1.00					
Xylenes	0.80**	0.91**	0.96**	1.00				
HCHO	0.04	0.10	0.13	0.16	1.00			
PM2.5-In	0.20	0.17	0.16	0.05	−0.07	1.00		
PM2.5-Out	0.09	−0.07	−0.08	−0.12	−0.30	0.66**	1.00	
δPM2.5	0.18	0.32	0.29	0.20	0.23	0.56**	−0.21	1.00

BTEX concentration ratios in the commercial and residential underground garages

The studies showed that the concentration ratio of toluene to benzene in semi-enclosed spaces, where air quality was mainly affected by internal combustion engine's emissions, ranges from 1.3 to 4.3 (Chan et al. 2004; Khoder 2007). The lower the toluene to benzene ratio was, the greater the effect of motor vehicle activity on BTEX compound concentrations. For BTEX ratios, the document shows 1:3:5 (benzene: toluene: xylenes) in the air in areas mainly affected by traffic.

Figure 7 demonstrated the BTEX inter-species ratio data for different points and periods, which allowed us to assess

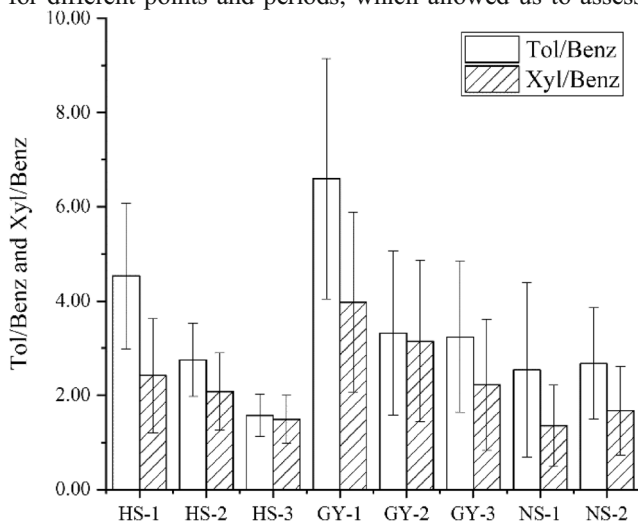


Fig. 7 BTEX inter-ratios (toluene/benzene and xylenes/benzene) determined in three different vehicle garages attached to commercial and residential building

how much an impact motor vehicle movement had on air quality in the monitored semi-closed areas. The three monitoring points' ratios were reduced sequentially in the commercial center's underground garage, with the lowest toluene/benzene ratio (1.5) at the exit and 2.7 and 4.5 at the internal garage and entrance, respectively. The toluene/benzene and xylenes/benzene ratios were also low at 1.9 and 1.4, respectively, at the peak of evening-time traffic in commercial centers, with only 53.6 to 57.8% of the hourly morning traffic.

Due to the high frequency of vehicle activity at the exit point and the long idle time after starting a cold vehicle engine, the total amount of pollutants emitted by motor vehicles was the highest. The toluene/benzene ratio was the lowest. It indicated that the organic matter at the exit was emitted to the underground parking garage through "fresh" and high-intensity motor vehicle exhaust.

The toluene/benzene ratio in residential garages was also significantly negatively correlated with traffic volume ($p < 0.05$), with the GY underground garage having the highest traffic flow at night (18.5/h) and only 56.5–62.2% of the lowest traffic flow period in the afternoon (7.3/h). In the corner of garages with poor diffusion conditions, the pollutants mainly came from the exhaust from motor vehicle activities; the ratio was only 49.1–66.1% of the entrance and exit with right diffusion conditions. Therefore, it can be concluded that air quality in underground parking garages was primarily affected by motor vehicle activity.

The inter-species concentration ratios shown in Fig. 7 varied considerably across garages. However, they were essentially within the range provided in the references, indicating that vehicle activity with internal combustion engines was a

significant source of emissions in underground parking garages in daily use. The high ratio of T/B at some points (entrance) was due to better diffusion conditions, where the ambient air was able to mix well with the basement air and affect the BTEX ratio. In turn, this fact illustrated that well-ventilated underground garages spread pollutants through the ventilation system into the surrounding parking garage and may become an essential source of urban air pollution, especially in densely populated areas.

Conclusions

Field measurements in commercial and residential UPGs under natural ventilation were carried out to identify the spatial and temporal distribution of indoor pollutants in UPGs and examine the indoor air pollution determinants. Based on the above analysis, several following conclusions can be drawn:

- (1) The time weighted average (TWA) concentrations of pollutants in the underground garage in the commercial center/high-rise residential buildings were as follows: benzene 0.12/0.11 $\text{mg}\cdot\text{m}^{-3}$, toluene 0.33/0.31 $\text{mg}\cdot\text{m}^{-3}$, xylenes 0.23/0.20 $\text{mg}\cdot\text{m}^{-3}$, TVOC 1.04/1.19 $\text{mg}\cdot\text{m}^{-3}$, $\text{PM}_{2.5}$ 0.20/0.19 $\text{mg}\cdot\text{m}^{-3}$, and PM_{10} 0.24/0.22 $\text{mg}\cdot\text{m}^{-3}$. Daily average air pollutant concentrations in all samples exceeded the standard obviously (sometimes the pollution was even severe). The pollution was most serious at night when the traffic flow in the UPGs was highest. However, due to the large difference in size and diffusion conditions, the peak pollution periods and the most painful areas were significantly different.
- (2) As for the garages, the spatial distribution of TVOC and BTEX concentrations was uneven. Concentrations of pollutants in the exit locations of the garage were significantly higher than in other locations. The concentration of BTEX and TVOC in the commercial center parking garage exit was 44.3 to 203.2% higher than the other locations. However, the PM concentration was similar to that at the entrance. It was also found that due to the high traffic volume and long average idling time of vehicles at the exit of the underground parking garage of the commercial center, the VOCs in this location were the most serious, so the collection of parking fees should speed up, and the average speed of vehicles in this area should increase.
- (3) Fuel combustion in motor vehicle engines was a significant source of VOCs pollution in garages. Outdoor ambient air dispersion was a significant source of PM pollution in garages (in winter).
- (4) Although the duration of human exposure to heavily polluted air in underground garages was relatively short, it was shown that air quality in this semi-closed

environment remains a vital problem and that the total amount of air pollutants inhaled by staff members throughout the day needs to be assessed and concentration standards should be developed accordingly.

At present, most garages in megacities in China were located below ground level. It was necessary to provide excellent and adequate ventilation to remove pollutants from underground garages. Natural ventilation measures currently used do not allow the garage air quality to meet the indoor air quality standards. Pollutant treatment facilities should be installed in the ventilation system; otherwise, the parking garage will become a severe air pollution source in the surrounding area and inner the building. However, the number and types of underground garages monitored in this study are not comprehensive enough, and the time accuracy needs to be improved. The environmental health risks of different age groups have not been effectively identified and need to be further monitored and investigated.

Furthermore, by explaining the results obtained, the air quality in underground areas used for parking can significantly impact the population's health and comfort entering such places.

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Author contribution Conceptualization, L.Z. and Y.Z.; experiment, Y.H. and J.W.; methodology, J.T.Z.; validation, X.H., Y.B., and J.Z.; formal analysis, L.Z. and J.W.; writing (original draft preparation), L.Z.; writing (review and editing), Y.Z. and Y. H.; and visualization, Y.Z. All authors have read and agreed to the published version of the manuscript.

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Data availability All data generated or analyzed during this study are included in this published article.

Declarations

Competing interests The authors declare no competing interests.

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