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Volatile organic compounds (VOCs) source profiles of on-road vehicle emissions in China

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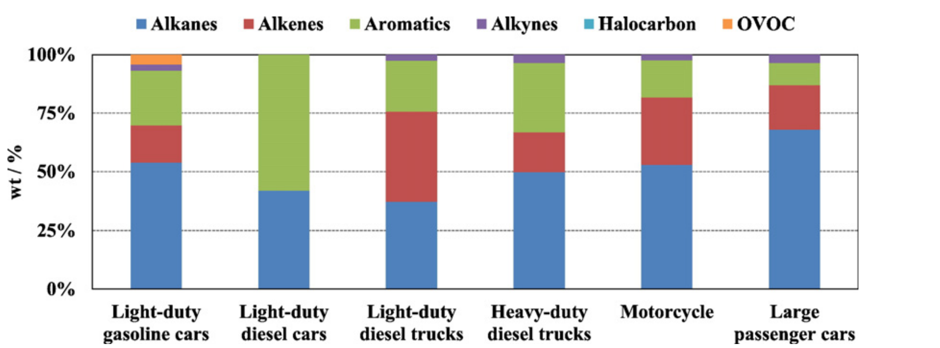
HIGHLIGHTS

- Studies of VOCs source profiles from on-road vehicles from 2001 to 2016 were summarized.
- Variations of VOCs source profiles in different studies were observed.
- 6 factors were extracted and their impact to VOCs source profile of on-road gasoline vehicles was discussed.

GRAPHICAL ABSTRACT



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ABSTRACT

Volatile Organic Compounds (VOCs) source profiles of on-road vehicles were widely studied as their critical roles in VOCs source apportionment and abatement measures in megacities. Studies of VOCs source profiles from on-road motor vehicles from 2001 to 2016 were summarized in this study, with a focus on the comparisons among different studies and the potential impact of different factors. Generally, non-methane hydrocarbons dominated the source profile of on-road vehicle emissions. Carbonyls, potential important components of vehicle emission, were seldom considered in VOCs emissions of vehicles in the past and should be paid more attention to in further study. VOCs source profiles showed some variations among different studies, and 6 factors were extracted and studied due to their impact to VOCs source profile of on-road vehicles. Vehicle types, being dependent on engine types, and fuel types were two dominant factors impacting VOCs sources profiles of vehicles. In comparison, impacts of ignitions, driving conditions and accumulated mileage were mainly due to their influence on the combustion efficiency. An opening and interactive database of VOCs from vehicle emissions was critically essential in future, and mechanisms of sharing and inputting relative research results should be formed to encourage researchers join the database establishment. Correspondingly, detailed quality assurance and quality control procedures were also very important, which included the information of test vehicles and test methods as detailed as possible. Based on the community above, a better uncertainty analysis could be carried out for the VOCs emissions profiles, which was critically important to understand the VOCs emission characteristics of the vehicle emissions.

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1. Introduction

Volatile organic compounds (VOCs) are important precursors in atmospheric chemistry, contributing to the formation of photochemical smog and secondary organic aerosols (Hao et al., 2015; Lu et al., 2006; Seinfeld and Pandis, 2006; Tang et al., 2006)that can impact atmospheric visibility and are hazardous to human health, living and production (Fiore et al., 2009; Gao et al., 2012). For example, BTEX (i.e. benzene, toluene, ethylbenzene, xylenes) can stimulate the human respiratory system and causes central nerve injury. Formaldehyde, acetaldehyde and benzene can increase the risk of cancer. In addition, VOCs can damage crops and reduce yield (Cao et al., 2016). In recent years, most of urban areas in China are suffering from serious haze, many of which are related to the second organic aerosols in the atmosphere (Guo et al., 2014; Huang et al., 2014). VOCs emission abatement has become one of the priorities for China's air quality improvement.

On-road motor vehicle emissions, solvent use and industrial processes are three dominant sources of VOCs emissions in China (Bo et al., 2008; Li et al., 2014; Wei et al., 2008; Wei et al., 2001; Wei et al., 2014).Among the total anthropogenic sources of VOCs, motor vehicle emissions account for >25% (Wei et al., 2001) in 2010 in China, which is larger in urban area (Wang et al., 2013a; Wang et al., 2015). In recent decades, the vehicle population in China has rapidly grown, especially in megacities (Editorial Board of China Auto Market Almanac, 2013), which shows increasing importance to air pollution. Hence, it is important to investigate VOCs emissions from on-road motor vehicles.

Source profiles, expressed as the weight fraction of each species relative to the total amount of mass emissions, are essential to separate total VOC emissions into individual species. Accurate and reliable VOCs source profiles are important for researches on the source apportionment, assessments of human exposure to toxic and hazardous substances, and the simulation of regional air quality (Ov et al., 2014; Yao et al., 2012). Many studies have been done on the emission characteristic of VOCs species from vehicles (Cao et al., 2016; Dong et al., 2014; Mo et al., 2016; Pang et al., 2014; Wang et al., 2013b; Yao et al., 2015a, 2015b; Yue et al., 2017; Watson et al., 2001), which involved various categories of vehicles with different ignitions, different fuel types, different test methods and VOCs species, leading to variations of VOCs source profiles.

The purpose of this study was to further understand VOCs source profiles of on-road motor vehicle emissions. Studies of VOCs source profiles from on-road motor vehicles from 2001 to 2016 were summarized in this study, focusing on the comparisons among different studies and the potential factors affecting VOCs source profiles of on-road vehicle emissions, which might provide guidance for future studies and for mitigation of vehicle emissions.

2. Studies of VOCs emissions of on-road vehicles

Twenty-seven publications were searched and selected from Chinese National Knowledge Infrastructure (CNKI) and Web of science about VOCs source profile of on-road motor vehicle emissions from 2001 to 2016 in all. As shown in Fig. 1, the vehicle emission of VOCs was always one hot research topic in the past years. There were publications each year, and each publication included dozens of tests on average.

Test methods of VOCs from motor vehicle emissions can be divided into 4 categories, including bench test, roadside sampling, tunnel test and vehicle exhaust measuring. Tunnel test is the most popular and used in 8 literatures.

For method of vehicle exhaust measuring, there are usually two kinds of tests. One is test on chassis dynamometers measured by constant volume sampler (CVS) exhaust gas analyzer. Another one is on-road emission test measured by a combined portable emissions measurement system (PEMS). VOCs samples are collected after dilution system that cooling the exhaust as it leaves the tailpipe and eliminates the problem ofwater condensation during sampling. Generally, this method is employed to study the emission curves and emission factors of pollutants of specific vehicle. Bench test, actually, is test of engines rather than testof real vehicles. VOCs sampling is similar to that of vehicle exhaust measuring. Bench test stimulates the engine on-road operating conditions, such as idling, acceleration and cruise, which, therefore, is usually employed to study vehicle emissions.

For tunnel test, VOCs samples are collected in tunnel. Tunnel test can be used to study both the emission factor and VOCs source profile. Generally, tunnel's supply ventilation system is not operated during the test, and thus emission factors can be calculated through the VOCs concentrations at inlet and outlet, vehicle flowrate, and the air flow in the tunnel resulted entirely from the vehicle traffic. Kind of similar to tunnel test, roadside sampling is also about the microenvironment full of vehicle emissions, thus is usually employed to study VOCs source profiles of vehicles. However, roadside sampling is influenced more easily by other sources compared with the tunnel test, which is not suitable to study emission factors.

Based on the source classification method of the National Guidelines for VOCs Inventories Preparation (China) (MEP, China, 2014), five categories of on-road vehicle VOCs sources were defined as light-duty passenger cars, light-duty trucks, heavy-duty trucks, large passenger cars and motorcycles, respectively. Passenger cars and trucks can be further divided into gasoline and diesel ones. Among them, VOCs emissions of gasoline and diesel vehicles were studied most, and 13 publications were searched, respectively. Overall, 165 tests were carried out for the VOCs emissions of gasoline vehicles in the publications, which was the hottest topic among all the research of on-road vehicle VOCs emissions.

For the region distribution, most research was studied in USA and eastern China. Studies in China mainly concentrated in Beijing, Shanghai and Guangzhou.

3. VOCs source profiles of on-road vehicle emissions

Sixteen of twenty-seven publications reported detailed datasets of VOCs source profiles. There were 90 VOCs species in source profile in the datasets, as shown in Table 1S, which can be divided into alkanes, alkenes, alkynes, aromatics, halocarbons (x-VOC) and oxygenated VOCs (OVOCs). Based on the reported data, the present study further

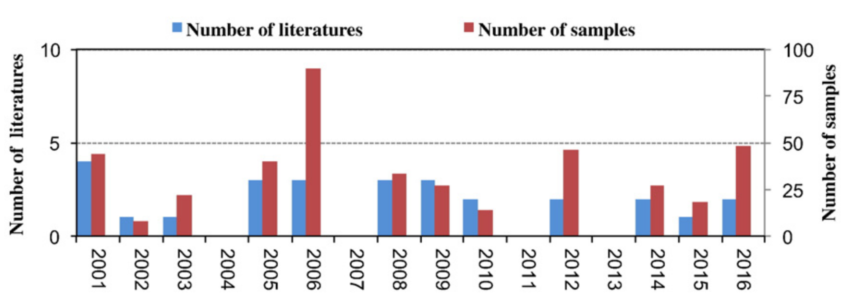

Fig. 1. Numbers of literatures and samples in different years.

summarized the VOCs source profile of on-road vehicle emissions focusing on the comparisons among different studies and the potential impact of fuels, ignitions, vehicle types, test methods, driving conditions and accumulated mileage on the source profiles.

There were two considerations for the 6 factors extracted in the present study. Firstly, VOCs emissions of vehicles were mainly from the fuel evaporation and incomplete combustion. VOCs emitted from fuel evaporation were similar to the fuel components and VOCs from combustion process contained many small molecular compounds. Thus, VOCs source profiles of vehicles were mainly impacted by the fuel type, internal combustion engine as well as the exhaust treatment system. Vehicles types were dependent on the types of internal combustion engines which included gasoline engine and diesel engine. Different type of engine resulted in difference of combustion efficiency (John, 1988) which impacted VOCs source profiles of vehicles. Besides the type of engine, ignitions, driving conditions, and accumulated mileage could influence the combustion efficiency and thus impacted VOCs emissions. Secondly, these factors have been more or less studied in dribs and drabs in previous studies (eg. Cao et al., 2016; Dai et al., 2010; Wang et al., 2006; Yao et al., 2015a; Yao et al., 2015b), which were regarded as important factors influencing VOCs source profiles of vehicles. The present study summarized all the published data and aimed to give a more comprehensive study of VOCs source profiles of vehicles and the potential impact of different factors.

The vehicles were divided into five categories according to the source classification methods mentioned above. The sub-categories contain vehicles with gasoline or diesel fuel. Large passenger cars include buses and coaches. Motorcycles have three-stroke or four-stroke

Table 1
Summary of literatures of VOCs source profile from different motor vehicle emissions.

Class	Class (Number of literatures/samples)	Category	Subcategory	Test method	Ignition	Driving condition	Accumulated mileage	Number of literatures	Number of samples	Number of species	Literatures
Mobile source	On-road motor vehicles(27/418)	Light-duty passenger cars	Gasoline	Bench test	EFI	NP	<100 thousand km	1	5	58	(Gao et al., 2012)
				Vehicle exhaust measuring	NP	Uniform speed	NP	1	21	59	(Ov et al., 2014)
						NP	NP	2	9;5	51;4	(Cao et al., 2016; Wang et al., 2008)
						NP	>100 thousand km	1	30	74	(Cao et al., 2016)
						Idle speed	NP	2	22;9	59;22	(Lu et al., 2003; Wang et al., 2006)
					EFI	Uniform speed	>100 thousand km	2	8;5	47;59	(Dai et al., 2010; Fu et al., 2008;)
							<100 thousand km	2	8;5	47;59	(Dai et al., 2010; Fu et al., 2008;)
					Carburator	Uniform speed	>100 thousand km	2	8;5	47;59	(Dai et al., 2010; Fu et al., 2008;)
							<100 thousand km	2	8;5	47;59	(Dai et al., 2010; Fu et al., 2008;)
			Diesel	Vehicle exhaust measuring	NP	Idle speed	NP	1	9	22	(Wang et al., 2006)
		Light-duty trucks	Diesel	Vehicle exhaust measuring	NP	NP	NP	1	7	64	(Yao et al., 2015a, 2015b)
						Uniform speed	<100 thousand km	1	1	47	(Fu et al., 2008)
						Idle speed	NP	2	3;1	4;22	(Wang et al., 2006; Yao et al., 2015a, 2015b)
				Bench test	NP	Uniform speed	NP	1	4	59	(Ov et al., 2014)
		Heavy -duty trucks	Diesel	Vehicle exhaust measuring	NP	NP	NP	3	3;6;9	4;64;51	(Wang et al., 2008; Yao et al., 2015a; Yao et al., 2015b)
					NP	Idle speed	NP	1	22	59	(Lu et al., 2003)
		Large buses	Buses	Vehicle exhaust measuring	EFI	Uniform speed	>100 thousand km	1	2	47	(Fu et al., 2008)
					NP	Idle speed	NP	1	9	22	(Wang et al., 2006)
						NP	NP	1	6	4	(Yao et al., 2015a, 2015b)
		Motorcycles	Two/four stroke	On-site measuring	NP	Uniform speed	NP	1	36	34	(Kawashima et al., 2006)
				Vehicle exhaust measuring	NP	Uniform speed	NP	1	5	59	(Ov et al., 2014)
				Vehicle exhaust measuring	NP	Idle speed	NP	1	9	22	(Wang et al., 2006)
		Tunnel			NP	NP	NP	5	15;8;23;9	79;62; 56;108;	(Fu et al., 2005; Ho et al., 2009; Hwa et al., 2006; Lu et al., 2010; Wang et al., 2001)

"NP" indicates no mention in the original literatures.

engines. Studies of different types of vehicles were summarized in Table 1, including fuel types, test methods, number of samples and VOCs species, and other detailed information of the vehicles and the tests.

3.1. VOCs source profiles of different vehicle types

VOCs source profiles of different vehicle types were obtained by calculating the arithmetic mean of different data from similar studies listed in Table 1S. As shown in Fig. 1S, alkanes, alkenes and aromatics take up most of VOCs composition regardless of the vehicle types. The dominant species were various among the source profiles measured from different vehicle categories, as shown in Fig. 2. Most of light-duty passenger cars used gasoline (Ov et al., 2014; Gao et al., 2012; Cao et al., 2016; Wang et al., 2008; Lu et al., 2003; Dai et al., 2010; Fu et al., 2008; Liu et al., 2008), and only one publications reported light-duty passenger car powered by diesel (Wang et al., 2006; Liang and Zhou, 2005; Qiao et al., 2012), as listed in Table 1. Light-duty gasoline cars emissions were composed of alkanes (52.1%), aromatics (22.8%), alkenes (15.4%),OVOCs (4.1%), x-VOCs (3.2%), and acetylene (2.4%).The five most abundant species from light-duty gasoline cars was ethylene, with a proportion of 7.4%, followed by toluene (5.8%), 3-Methylnonane (4.2%), 1,3-Dimethylcyclopentane (3.9%), isoPentane (3.8%) and ethylcyclohexane (3.6%). VOCs emissions of light-duty diesel cars was mainly composed of alkenes and alkanes, and ethene and propene were the two most dominant species with proportions of 32.6% and 31.2%, respectively. In terms of diesel trucks, oxygenated VOCs, mainly carbonyls, were very important components of VOCs emissions, with a mass fraction of 12.4% for light-duty diesel trucks and 20.1% for heavy-duty diesel trucks. Carbonyls, as the key intermediates in atmospheric chemistry, should be paid more attention to of their primary emissions from vehicles in future (Ban-Weiss et al., 2008; Grosjean et al., 2001; Kean et al., 2001; Mo et al., 2016; Yao et al., 2015a 2015b). Compared with emissions from heavy-duty diesel trucks, light-duty diesel trucks emitted more alkenes, i.e. more ethene and propene.

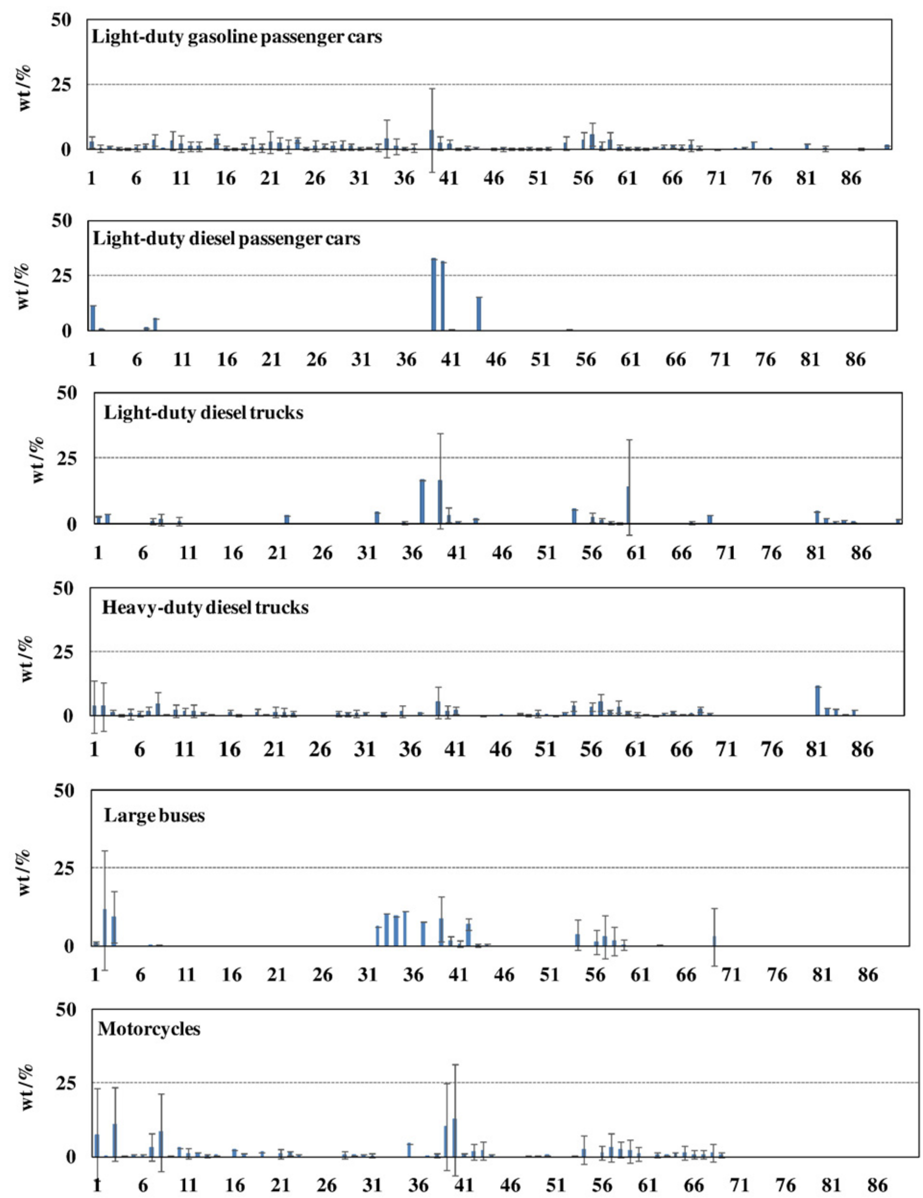

Fig. 2. Source profiles of different motor vehicles (Number of the X axis is the ID of each VOCs species, being same with that in Table 1S. the same below).

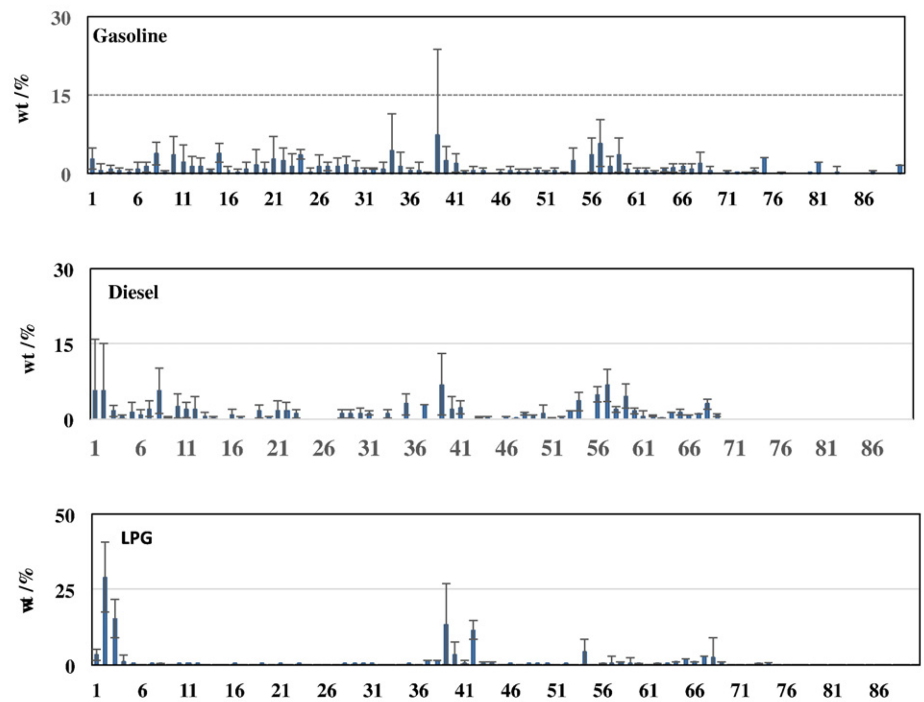

Fig. 3. Source profiles of motor vehicles with different fuels.

Large buses usually used diesel and emitted more alkanes than other diesel vehicles, such as propane, butane, nonane and decane. Motorbike emissions were characterized by high proportions of ethane, butane, i-pentane, ethylene, and propene.

3.2. Impact of fuel type on VOCs source profiles

Fuel type is one of the key factors affecting VOCs source profiles of motor vehicle emissions, and gasoline, diesel and LPG (Lai et al., 2009) are three main fuel types used in China. As shown in Fig. 2S, contributions of alkanes were similar among the three fules, ranging from 49.7% to 58.9%. Diesel vehicle emissions contained the largest contribution of aromatics, i.e. 29.6%, while LPG vehicle emissions contained the largest contribution of alkenes, i.e. 34.2%. In terms of particular species, less kinds of species were observed in the emissions of LPG vehicles, as shown in Fig. 3. Characteristic species including propane, n-butane, ethene, and i-butene were dominated in the source profile of LPG vehicle emissions, which accounted for 69.5% of VOCs. Emissions from gasoline and diesel vehicles were more complex, but sharing similar species. Diesel vehicles emitted higher percentages of ethane, propane, n-decane and undecane, but lower percentages of C4–C5 alkanes, in comparison with that of gasoline vehicle, as shown in Fig. 3 and Table 1S.

3.3. Impact of test methods on VOCs source profiles

In general, there were mainly four test methods of vehicle emissions study, including roadside sampling, tunnel test, bench test and vehicles exhaust measuring. All of them were employed to study VOCs emissions from light-duty gasoline cars, as listed in Table 1, which provided possibilities to discuss the impact of test methods to VOCs source profiles in this section.

Fig. 3S illustrated VOCs compositions measured by different test methods. VOCs compositions measured by roadside sampling and tunnel test were very similar with each other, which were dominated by alkanes (i.e. 39.5% and 34.3%, respectively), followed by alkenes and aromatics. While, more OVOCs and x-VOCs were observed in emissions by roadside sampling compared to those by tunnel test, probably due to the mixture of other sources in ambient air. VOCs composition obtained by bench test showed the highest weight percentage of aromatics, i.e. 43.7%, followed by alkanes and alkenes. In comparison, VOCs measured in the exhausts was dominated by alkanes, which accounted for about 57.2%.

To be more specific, as shown in Fig. 4, VOCs source profiles obtained by roadside sampling and tunnel test were characteristic of C2–C3 species, n-butane, i-pentane, C6–C8 aromatics. VOCs source profile measured by bench test was similar to those by roadside and tunnel sampling, but with higher percentages of C6–C8 aromatics and lower percentages of C2–C3 species. By comparison, different species showed comparable contributions to total VOCs measured in vehicle exhausts, as shown in Fig. 4, but, uncertainties of source profiles of vehicle exhausts were the largest, probably due to the differences of the testing vehicles.

It should be pointed out that VOCs emissions should be more dependent on the testing vehicles than that on test methods. Differences measured by test methods here should be mainly due to the different testing vehicles, such as fuel types, vehicle types, driving conditions and so on. Generally, VOCs source profiles obtained by tunnel tests indicated the emissions mixture of various vehicles in the tunnel, which were dependent on the composition of vehicles in the tunnel or on the road. Method of roadsied sampling was similar to tunnel test. Consequently, tunnel test and roadside sampling might be not suitable to study VOCs source profile of a specific vehicle type. While, bench test and exhaust measuring methods were usually employed to study specific vehicles emissions, such as emission curves, factors and compositions. Thus, bench test and exhaust measuring were suitable to study source profiles of specific vehicle types, but sample size should be large enough to get a more representative source profile.

3.4. Impact of ignitions on VOCs source profiles

The impact of ignitions were discussed only to VOCs source profile from light-duty gasoline cars in this section. Electronic Fuel Injection (EFI) and carburetor were two major kinds of ignitions reported. Compared with vehicles ignited by EFI, those by carburetor emitted more VOCs due to lower air-fuel ratio (John, 1988), and more gasoline components were emitted directly due to the incomplete combustion for

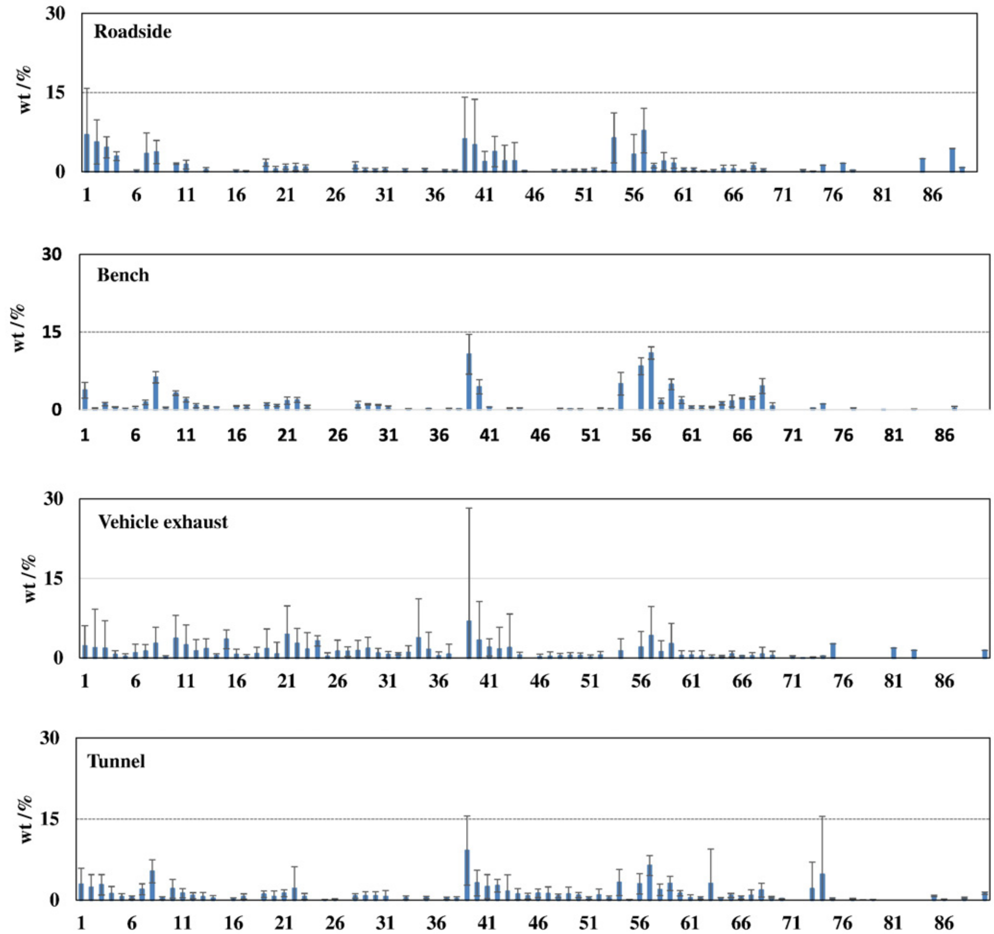

Fig. 4. Source profiles of emissions from light-duty gasoline vehicles by different test methods.

vehicles with carburetor. Most of vehicles are ignited by EFI and those by carburetor are out of use gradually.

As shown in Fig. 4S, VOCs emissions from vehicles with carburetor contained more alkanes but less aromatics than those from vehicles with EFI, and alkenes proportions were similar. To be more specific, much higher contribution of C5–C7 alkanes, being components of gasoline, were observed in emissions of vehicles with carburetor (37%) than that of vehicles with EFI (24.4%), which were emitted probably due to the incomplete combustion, as shown in Fig. 5. For emissions from light-duty gasoline cars with EFI, the top three species in terms of abundance were 3-methylnonane, toluene and ethylene, with ratios of 10.3%, 7.0% and 6.8%, respectively. For light-duty gasoline cars with carburetor, 2-methylhexane, toluene and 2-methylpentane are top three species, contributing7.7%, 7.4% and 6.5% of the total VOCs emissions.

3.5. Impact of accumulated mileage on VOCs source profiles

This section is focused on the impact of accumulated mileage of vehicles on VOCs source profiles, based on the emission data of light-duty gasoline cars with the most samples. The data were grouped into two categories, i.e. vehicles with accumulated mileage large than 100 thousand kilometers or not, as shown in Fig. 5S.

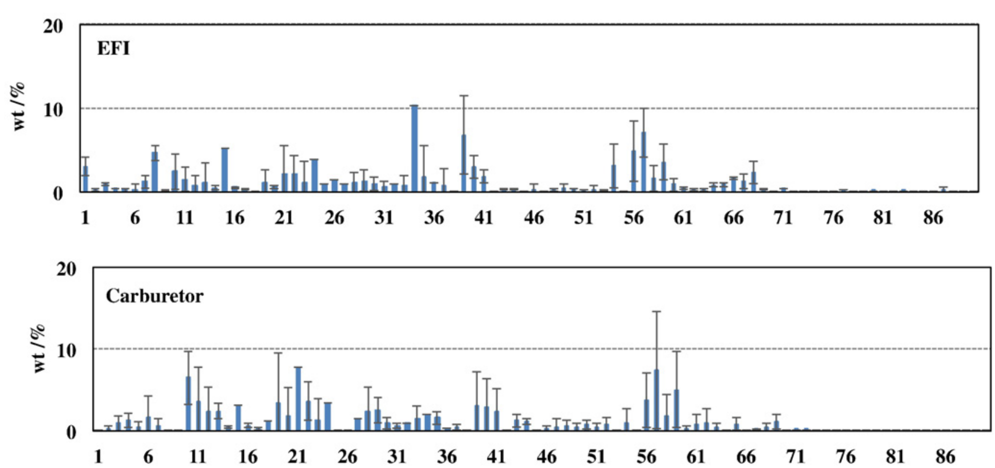

Fig. 5. Source profiles of light-duty gasoline vehicles with different ignitions.

Significant differences were observed between VOCs source profiles from two kinds of vehicles mentioned above. More alkanes (i.e. 68.9%) and less aromatics (i.e. 16%) were observed in source profiles of vehicles with accumulated mileage larger than 100 thousand kilometers, compared to those of <100 thousand kilometers, as shown in Fig. 5S. The relative contributions of alkenes were similar between two kinds of vehicles, accounting for about 14% of VOCs.

Generally, vehicles with longer accumulated mileage were more aged, with higher fuel consumption and worse three-way catalytic converter, which resulted in more VOCs emissions due to incomplete combustion (John, 1988). As shown in Fig. 6, it can be seen that much higher percentage of C5–C7 alkanes, being major components of gasoline, were observed in emissions of vehicles with longer accumulated mileage, in comparison with those with shorter accumulated mileage. For vehicles with long accumulated mileage, source profile was characteristics of high percentages of 2-Methylhexane (8.5%), 3-methylnonane (6.0%), 2-methylpentane (5.8%), toluene (5.3%), and ethylcyclohexane (4.2%). For vehicles with shorter accumulated mileage, the top five species were toluene (8.2%), ethylene (7.9%), benzene (5.8%), i-pentane (5.3%), and m,p-xylenes (4.2%).

It should be pointed that VOCs source profile rather than the emission factor was focused on in the present study due to the limited data searched in publications. Further study were essential to investigate the impact of vehicle accumulated mileage on VOCs emissions, in particular, both on source profiles and on emission factors, which might be very helpful for policy-makers to control emissions of in-use vehicles.

3.6. Impact of driving conditions on VOCs source profiles

Studies of light-duty gasoline cars in Table 1 were selected to discuss the impact of driving conditions to VOCs source profile in this section as the considerable samples. Generally, there were two kinds of driving conditions reported in the publications, i.e. running in uniform speed of 20 km/h and idling.

As shown in Fig. 6S, VOCs composition of vehicle emissions of running was largely different from that of idling. Specifically, alkanes dominated emissions during vehicles running, being with a proportion of 66.3%, followed by aromatics (17.4%) and alkenes (15.1%). Alkenes contributed >50% of VOCs emissions during idling, followed by alkanes (37.5%) and aromatics (10.6%). VOCs source profile during vehicle idling was characteristic of high contributions of ethylene, propene, propane, cis-2-butene, i-butane, and ethane, accounting for 55.5% of VOCs in total, as shown in Fig. 7. By comparison, mass fractions of different species were more comparable in VOCs source profile during vehicle running in a uniform speed. Especially, C4–C7 alkanes and C6–C8 aromatics were very important species emitted during vehicle running at a uniform speed.

Vehicles during idling or running with high speed were usually with low ratio of air to fuel and much incomplete combustion (John, 1988), probably resulting in high emissions of C5–C7 alkanes which were important components of gasoline. While, C2–C3 hydrocarbons dominated VOCs emitted during vehicles idling with large error bar, as shown in Fig. 7, which meant further study was essential to investigate the impact of driving conditions on VOCs source profiles.

4. Conclusions

VOCs source profiles of on-road vehicles were widely studied as their critical roles in VOCs source apportionment and abatement measures in megacities. The present study reviewed publications of VOCs emissions from on-road vehicles over the past 15 years, focusing on the comparisons among different studies and the potential factors affecting VOCs source profiles of on-road vehicle emissions.

Generally, non-methane hydrocarbons dominated the source profile of on-road vehicle emissions. Carbonyls were also important components of VOCs from heavy-duty diesel vehicle exhaust, which were seldom considered in VOCs emissions of gasoline vehicles in the past. VOCs source profiles of vehicles including carbonyls should be paid more attention to in further study.

VOCs source profiles showed some variations among different studies, and 6 factors were extracted and studied due to their impacts to VOCs source profile of on-road vehicles. The top two important factors were vehicle type and fuel type, which were usually reported in literatures. Generally, diesel vehicles emitted more alkenes than gasoline vehicles, and heavy-duty diesel vehicles were characterized by C9–C11 straight-chain alkanes, and light-duty gasoline vehicles were characteristic of i-pentane and C6–C8 branched alkanes. Vehicles powered by LPG usually emitted high percentages of C2–C4 hydrocarbons. Driving conditions, accumulated mileages (standing for the age of vehicles), and ignition ways also largely impacted VOCs composition emitted from vehicles, mainly due to their influence on the combustion efficiency. Generally, vehicles with longer accumulated mileage, ignited by carburetor, during idling emitted more C5–C7 alkanes probably due to higher incomplete combustion. VOCs source profiles obtained by tunnel tests indicated the emissions mixture of various vehicles in the tunnel, which were dependent on the composition of vehicles in the tunnel or on the road. Consequently, tunnel test and roadside sampling might be not suitable to study VOCs source profile of a specific vehicle type. While, bench test and exhaust measuring methods were usually employed to study specific vehicles emissions, such as emission curves, factors and compositions. Thus, bench test and exhaust measuring were suitable to study source profiles of specific vehicle types, but sample size should be large enough to get a more representative source profile. It should be pointed that all the factors discussed in the manuscript might impact VOCs source profiles of vehicles emissions together, and the crossed impact of these factors should be further studied by more detailed information of the tests and larger sample size.

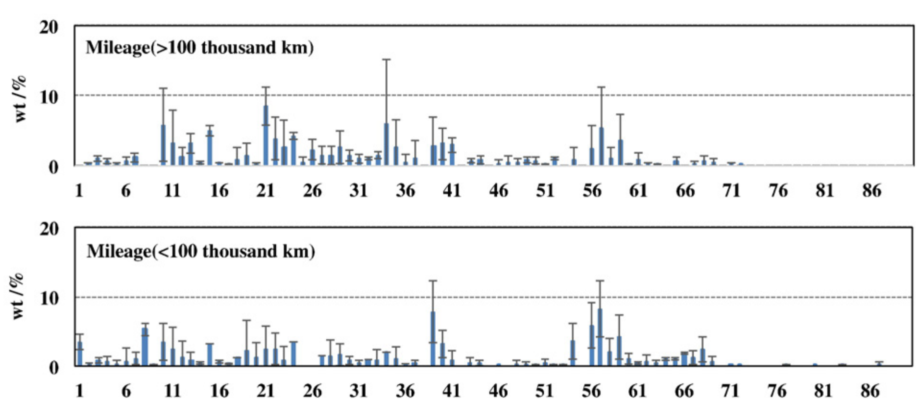

Fig. 6. Source profiles of light-duty gasoline cars with different accumulated mileage.

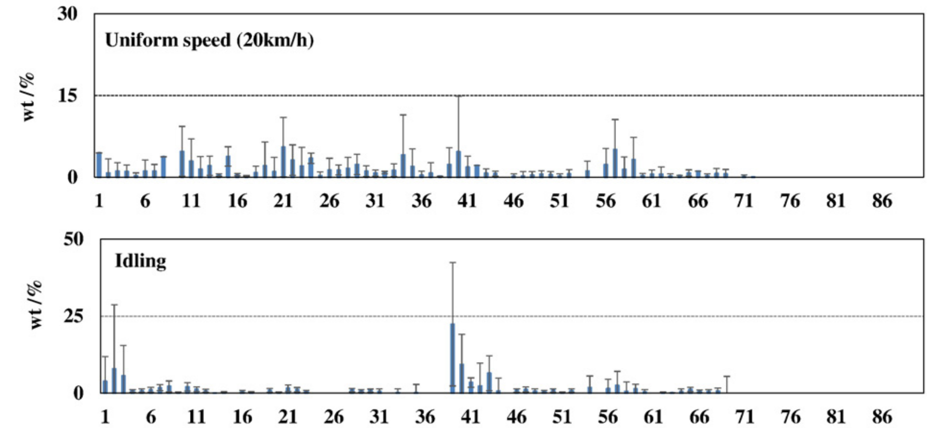

Fig. 7. Source profiles of emissions of light-duty gasoline cars under different driving conditions.

An opening and interactive database of VOCs from vehicle emissions was critically essential in future, and mechanisms of sharing and inputting relative research results should be formed to encourage researchers join the database establishment. Correspondingly, detailed quality assurance and quality control procedures were also very important, which included the detailed information such as vehicle type, fuel type, driving condition, accumulated mileage, test method, ignition way, even VOCs species list and other related information of vehicles and methods as detailed as possible. Based on the community above, a better uncertainty analysis could be carried out for the VOCs emissions profiles, which was critically important to understand the VOCs emission characteristics of the vehicle emissions.

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References

Ban-Weiss, G.A., Mclaughlin, J.P., Harley, R.A., 2008. Carbonyl and nitrogen dioxide emissions from gasoline- and diesel-powered motor vehicles. Environ. Sci. Technol. 42, 3944–3950.

Bo, Y., Cai, H., Xie, S.D., 2008. Spatial and temporal variation of emission inventoriesfor historicalanthropogenicNMVOCs in China. Atmos. Chem. Phys. 8, 11519–11566.

Cao, X.Y., Yao, Z.L., Shen, X.B., Ye, Y., Jiang, X., 2016. On-road emission characteristics of VOCs from light-duty gasoline vehicles in Beijing, China. Atmos. Environ. 124, 146–155.

Dai, T.Y., Wang, W., Ren, L.H., Chen, J.H., Liu, H.J., 2010. Emissions of non-methane hydrocarbons from cars in China. SCIENCE CHINA Chem. 53, 264–272.

Dong, D., Shao, M., Li, Y., Lu, S.H., Wang, Y.H., Ji, Z., Tang, D.G., 2014. Carbonyl emissions from heavy-duty diesel vehicle exhaust in China and the contribution to ozone formation potential.J. Environ. Sci. 26, 122–128.

Editorial Board of China Auto Market Almanac, 2013. 2013 China Auto MarketAlmanac. 11 pp. 222–226 (in Chinese).

Fiore, A.M., Dentener, F.J., Wild, O., Cuvelier, C., Schultz, M.G., 2009. Multimodel estimates of intercontinental source-receptor relationships for ozone pollution. J. Geophys. Res. 114 (D4), 83–84.

Fu, L.L., Shao, M., Liu, Y., Liu, Y., Lu, S.H., Tang, D.G., 2005. Tunnel experimental study on the emission factors of volatile organic compounds (VOCs) from vehicles. Acta Sci. Circumst. 25, 879–885 (in Chinese).

Fu, X.Q., Weng, Y.B., Qian, F.Z., Ye, W.B., Jin, Y.X., Chen, Y.Y., 2008. Study of the VOC source profile and benzene compounds emission of various motor vehicles. Acta Sci. Circumst. 28 (6), 1057–1062 (in Chinese).

Gao, S., Jin, L.M., Shi, J.W., Han, B., Wang, X.Y., Peng, Y., Zhao, L.J., Bai, Z.P., 2012. VOCs emission characteristics and emission factors oflight-duty gasoline vehicles with bench test. China Environ. Sci. 32, 397–405 (in Chinese).

Grosjean, D., Grosjean, E., Gertler, A.W., 2001. On-road emissions of carbonylsfrom light-duty andheavy-dutyvehicles.Environ. Sci. Technol. 35, 45–53.

Guo, S., Hu, M., Zamora, M.L., Peng, J.F., Shang, D.J., Zheng, J., Du, Z.F., Wu, Z.J., Shao, M., Zeng, L.M., Molina, M.J., Zhang, R.Y., 2014. Elucidating severe urban haze formation in China. Proc. Natl. Acad. Sci. 111, 17373–17378.

Hao, J.M., Lv, Z.F., Chu, B.W., Wu, S., Zhao, Z., et al., 2015. Characterization, Experimental Study and Modeling of Atmospheric Secondary Organic Aerosol[M]. Beijing: China. Science Publishing House (in Chinese).

Ho, K.F., Lee, S.C., Ho, W.K., Blake, D.R., Cheng, Y., Li, Y.S., Kung, K., Louie, P.K.K., Park, D., 2009. Vehicular emission of volatile organic compounds(VOCs) from a tunnel study in Hong Kong. Atmos. Chem. Phys. Discuss. 9, 12645–12674.

Huang, R.J., Zhang, Y.L., Bozzetti, C., Ho, K.F., Cao, J.J., Han, Y.M., Daellenbach, K.R., Slowik, J.G., Platt, S.M., Canonaco, F., Zotter, P., Wolf, R., Pieber, S.M., Bruns, E.A., Crippa, M., Ciarelli, G., Piazzalunga, A., Schwikowski, M., Gülcin Abbaszade, G., Schnelle-Kreis, J., Zimmermann, R., An, Z.S., Szidat, S., Baltensperger, U., Haddad, I., Pre'vôt, A., 2014. High secondary aerosol contribution to particulatepollution during haze events in China. Nature 514, 218–222.

Hwa, M.Y., Hsieh, C.C., Wu, T.C., Chang, L.F.W., 2006. Real-world vehicleemissions and VOCs profile in theTaipei tunnel locatedat Taiwan Taipei area. Atmos. Environ. 36, 1993–2002.

John, B.H., 1988. Internal Combustion Engine Fundamentals. McGraw-Hill Company.

Kawashima, H., Minami, S., Hanai, Y., Fushimim, A., 2006. Volatile organic compound emission factors from roadsidemeasurements. Atmos. Environ. 40, 2301–2312.

Kean, A.J., Grosjean, E., Grosjean, D., Harley, R.A., 2001. On-road measurement of carbonyls in California light-duty vehicle emissions. Environ. Sci. Technol. 35, 4198–4204.

Lai, C.H., Chang, C.C., Wang, C.H., Shao, M., Zhang, Y.H., Wang, J.L., 2009. Emissions of liquefied petroleum gas (LPG) from motorvehicles. Atmos. Environ. 43, 1456–1463.

Li, M., Zhang, Q., Streets, D.G., He, K.B., Cheng, Y.F., Emmons, L.K., Huo, H., Kang, S.C., Lu, Z., Shao, M., Su, H., Yu, X., Zhang, Y., 2014. Mapping Asian anthropogenic emissions of non-methane volatile organic compounds to multiple chemical mechanisms. Atmos. Chem. Phys. 14, 5617–5638.

Liang, B.S., Zhou, Y., 2005. Study on emission characteristics of volatile organic compounds in the exhaust of different types of vehicles. Environmental Monitoring in China. 21, pp. 9–11 (in Chinese).

Liu, Y., Shao, M., Fu, L.L., Lu, S.H., Zeng, L.M., Tang, D.G., 2008. Source profiles ofvolatile organic compounds(VOCs)measured in China: part I. Atmos. Environ. 42, 6247–6260.

Lu, S.H., Bai, Y.H., Zhang, G.S., Ma, J., 2003. Study on the characteristics of VOCs source profiles of vehicle exhaust and gasoline emission. Acta Sci. Circumst. 39, 508–511 (in Chinese).

Lu, S.H., Bai, Y.H., Zhang, G.S., Li, T.T., et al., 2006. Source apportionment of anthropogenic emissions of volatile organic com pounds[J]. Acta Sci. Circumst. 26 (5), 758–763 (in Chinese).

Lu, J., Wang, H.L., Cheng, C.H., Huang, C., Li, L., Cheng, Z., Liu, J., 2010. The composition and chemicalreactivity of volatile organic compounds (VOCs) fromvehicleexhaust in Shanghai, China. Environ. Poll. Cont. 32, 19–26 (in Chinese).

MEP, China, 2014. The Technical Guide for the Compilation of Emission Inventory of Volatile Organic Compounds (in Chinese).

Mo, Z.W., Shao, M., Lu, S.H., 2016. Compilation of a source profile database for hydrocarbon and OVOC emissions in China. Atmos. Environ. 143, 209–217.

Ov, J.M., Feng, X.Q., Liu, Y.C., Gao, Z.J., Yang, Y., Zhang, Z., Wang, X.M., Zheng, J.Y., 2014. Source characteristics of VOCs emissions from vehicular exhaust in the Pearl River Delta region. Acta Sci. Circumst. 34, 826–834 (in Chinese).

Pang, Y.B., Fuentes, M., Rieger, P., 2014. Trends in the emissions of volatile organic compounds (VOCs) from light-duty gasoline vehicles tested on chassis dynamometers in Southern California. Atmos. Environ. 83, 127–135.

Qiao, Y.Z., Wang, H.L., Huang, C., Chen, C.H., Su, L.Y., Zhou, M., Xu, H., Zhang, G.F., Chen, Y.R., Li, L., Chen, M.H., Huang, H.Y., 2012. Source profile and chemical reactivity of volatile organic compounds fromvehicleexhaust. Environ. Sci. 33, 1072–1079 (in Chinese).

Seinfeld, J.H., Pandis, S.N., 2006. Atmospheric Chemistry and Physics: From Air Pollution to Climate Change[M]. Second edition. Wiley-Interscience, USA, p. 2006.

Tang, X.Y., Zhang, Y.H., Shao, M., 2006. Atmos. Environal Chemistry[M]. second edition. Higher Education Press, Beijing (in Chinese).

Wang, W., Liang, B.S., Zeng, F.G., Liu, H.J., Ye, H.H., Pan, Z., 2001. Study on pollution characteristics and emission factors of volatile organic compounds (VOCs) in Tanyugou highway tunnel. Res. Environ. Sci. 14, 10–12 (in Chinese).

Wang, M., Shao, M., Chen, W., Lu, S., Liu, Y., Yuan, B., Zhang, Q., Chang, C.C., Wang, B., Zeng, L., Hu, M., Yang, Y., Li, Y., 2015. Trends of non-methane hydrocarbons (NMHC) emissions in Beijing during 2002–2013. Atmos. Chem. Phys. 15 (3), 1489–1502.

Wang, B.G., Shao, M., Zhang, Y.H., Lv, W.M., Liu, H.X., Zhang, N., 2006. A study of volatile organic compounds and its emission factors from city vehicles. Res. Environ. Sci. 19, 75–80 (in Chinese).

Wang, B.G., Zhang, Y.H., Shao, M., Zhou, Y., Feng, Z.C., 2008. Sources apportionment of anthropogenic C2–C9 non-methanehydrocarbons in the atmosphere of Guangzhou, China. Acta Sci. Circumst. 28, 1431–1440 (in Chinese).

Wang, H.L., Chen, C.H., Wang, Q., Huang, C., Su, L.Y., Huang, H.Y., Lou, S.R., Zhou, M., Li, L., Qiao, L.P., Wang, Y.H., 2013a. Chemical loss of volatile organic compounds and its impact on the source analysis through a two-year continuous measurement. Atmos. Environ. 80, 488–498.

Wang, J., Jin, L., Gao, J., Shi, J., Zhao, Y., Liu, S., Jin, T., Bai, Z., Wu, C., 2013b. Investigation of speciated VOC in gasoline vehicular exhaust under ECE and EUDC test cycles. Sci. Total Environ. 445-446, 110–116.

Watson, J.G., Chow, J.C., Fujita, E.M., 2001. Review of volatile organic compound source apportionment by chemical mass balance. Atmos. Environ. 35, 1567–1584.

Wei, W., Wang, S., Hao, J., Cheng, S., 2001. Projection of anthropogenic volatile organic compounds (VOCs) emissions in China for the period 2010–2020. Atmos. Environ. 45 (38), 6863–6871.

Wei, W., Wang, S.X., Chatani, S., Klimont, Z., Cofala, J., 2008. Emission and speciation of non-methane volatile organic compounds from anthropogenic sources in China. Atmos. Environ. 42, 4976–4988.

Wei, W., Wang, S., Hao, J., Cheng, S., 2014. Trends of chemical speciation profiles of anthropogenic volatile organic compounds emissions in China, 2005–2020. Front. Environ. Sci. Eng. 8, 24–42.

Yao, Z., Zhang, Y.Z., Shen, X.B., Wang, X.T., He, K.B., 2012. Impacts of temporary traffic control measures on vehicular emissions during the Asian games in Guangzhou, China. J. Air Waste Manage. Assoc. 63, 11–19.

Yao, Z.L., Shen, X.B., Cao, X.Y., Jiang, X., Zhang, Y.Z., He, K.B., 2015a. On-road emission characteristics of VOCs from diesel trucks in Beijing, China. Atmos. Environ. 103, 87–93.

Yao, Z.L., Wu, B.B., Shen, X.B., Cao, X.Y., Jiang, X., Ye, Y., He, K.B., 2015b. On-road emission characteristics of VOCs from rural vehicles and their ozone formation potential in Beijing, China. Atmos. Environ. 105, 91–96.

Yue, T.T., Yue, X., Chai, F.H., Hu, J.N., Lai, Y.T., He, L.Q., Zhu, R.C., 2017. Characteristics of volatile organic compounds (VOCs) from the evaporative emissions of modern passenger cars. Atmos. Environ. 151, 62–69.