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Decrease of VOC emissions from vehicular emissions in Hong Kong from 2003 to 2015: results from a tunnel study

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Abstract

Vehicular emissions are one of major anthropogenic sources of ambient volatile organic compounds (VOCs) in Hong Kong. During the past twelve years, the government of the Hong Kong Special Administrative Region has undertaken a series of air pollution control measures to reduce vehicular emissions in Hong Kong. Vehicular emissions were characterized by repeated measurement in the same roadway tunnel in 2003 and 2015. The total net concentration of measured VOCs decreased by 44.7% from 2003 to 2015. The fleet-average VOC emission factor decreased from 107.1 ± 44.8 mg veh^{-1} km^{-1} in 2003 to 58.8 ± 50.7 mg veh^{-1} km^{-1} in 2015, and the total ozone (O_3) formation potential of measured VOCs decreased from 474.1 mg O_3 veh^{-1} km^{-1} to 190.8 mg O_3 veh^{-1} km^{-1}. The emission factor of ethene, which is one of the key tracers for diesel vehicular emissions, decreased by 67.3% from 2003 to 2015 as a result of the strict control measures on diesel vehicular emissions. Total road transport VOC emissions is estimated to be reduced by 40% as compared with 2010 by 2020, which will be an important contributor to achieve the goal of total VOC emission reduction in the Pearl

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River Delta region. The large decrease of VOC emissions from on-road vehicles demonstrates the effectiveness of past multi-vehicular emission control strategy in Hong Kong.

Keywords:
Volatile organic compounds; Vehicular emissions; Tunnel; Emission factor.

1 Introduction

Volatile organic compounds (VOCs), which can be primarily emitted from natural or anthropogenic sources (Atkinson and Arey, 2003; Watson et al., 2001), are important precursors of ground level ozone (O_3) formation and have adverse effects on human health (Sillman, 2002; von Schneidemesser et al., 2010). Vehicular emission is one of major anthropogenic sources of ambient VOCs in Hong Kong. In 2002 – 2003, vehicle- and marine vessel-related sources contributed 31 – 48% of ambient VOC concentrations in Hong Kong, and this contribution increased to 40 – 54% in 2006 – 2007 (Lau et al., 2010). Moreover, traffic-related sources including vehicle exhaust, gasoline evaporation and LPG usage contributed 30–60% of ambient VOCs in Hong Kong from 2005 to 2013 through a multi-year study in Hong Kong (Ou et al., 2015).

Over the past twelve years, a series of air pollution control strategies have been undertaken by the government of the Hong Kong Special Administrative Region to reduce vehicular emissions in Hong Kong. These measures mainly include: (1) tightening vehicle emission standards, (2) updating vehicle fuel standards, (3) switching diesel vehicles to liquefied petroleum gas (LPG) vehicles, and (4) retrofitting emission control devices (Lau et al., 2015). Euro 3 standard was used before 2003 in Hong Kong, and it was tightened to Euro 5 standard from 2012. Vehicle fuel standard for diesel was ultra-low sulfur diesel (ULSD, 50 ppm of sulfur) before 2003 and it was updated to Euro 5 diesel standard (10 ppm of sulfur) from 2012. Gasoline fuel was also updated to Euro 5 gasoline standard from 2010. In order to control diesel vehicular emission, nearly 99% taxies were changed from diesel to LPG from August 2000 to the end of 2003. Light buses were also switched from diesel to LPG from August 2002, and about 65% light bus fleets used LPG in 2006. As a result, road transport VOC emissions in Hong Kong decreased gradually from 2003 (7,600 tonnes) to 2015 (4,800 tonnes) even the total vehicle kilometers traveled (VKT) in Hong Kong increased by 18.7% from 2003 to 2015.

Direct measurements in roadway tunnels is one of the methods to determine vehicular emission factors and profiles. Repeated measurements performed at the same tunnel can be used to assess the effectiveness of local vehicular emission control strategies and/or new technologies to reduce vehicular emissions (Stemmler et al., 2005). For instance, a tunnel study was conducted in Gubrist tunnel in Switzerland in 1993 and 2002, and it was found that the emission factors of particular VOCs significantly decreased after nine years, indicating the effectiveness of modern car fleets on reducing VOCs emissions (Staehelin et al., 1995; Stemmler et al., 2005). In Hong Kong, vehicular emission factors and profiles of different VOC groups (alkanes, alkenes, alkynes, and aromatic hydrocarbons) were obtained through a tunnel study in

the Shing Mun Tunnel by our group in 2003 (Ho et al., 2009). In this study, experiments were repeated in the Shing Mun tunnel in 2015 to evaluate the vehicular emission reduction after 12 years in Hong Kong. Emission factors of individual VOCs are also updated in this study, which will be useful for speciated VOC inventories and related modelling studies.

2 Methodology

2.1 Sampling site

The tunnel measurements were conducted in the south bore of Shing Mun tunnel at two sampling sites: one at the inlet (Point A in **Figure 1**), and the other one at the outlet (Point B in **Figure 1**). The cross section area of the tunnel is 70.0 m^2, and the vehicle speed limit is 80 km h^{-1}. The tunnel has a 1% grade from the entrance to the exit. The actual vehicle speed was in the range of 60 – 70 km h^{-1}. The ventilation fans were not turned on during this study. Therefore, the air movement in the tunnel was mainly inducted by the piston effect of vehicle movement. More details of the sampling site was presented elsewhere (Ho et al., 2009). The inlet and outlet sampling sites were 686 m from the entrance and 350 m from the exit of the tunnel, respectively. The selection of the sampling position was based on the following three reasons, (1) inlet and outlet sampling points were set at the same position in 2015 as in 2003 to maintain the consistency, (2) the selection of sampling points should acquire the permission of related managing department, and (3) there can provide sufficient spaces for setting up other sampling instruments.

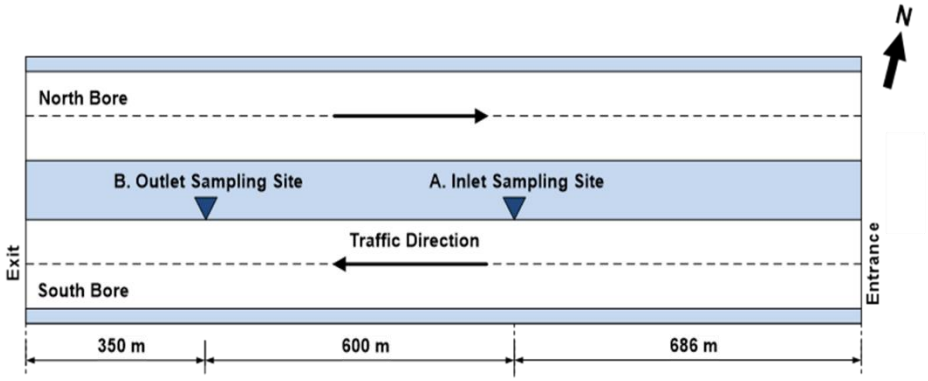


Figure 1. Schematic diagram of Shing Mun Tunnel and sampling sites in the south bore.

2.2 VOC collection, analysis, and gas measurement

The experiment was conducted from 19 January to 31 March, 2015. From late evening to early morning (23:00 – 06:00 local standard time [LST]) of Monday through Thursday, one of the tunnel bores was closed for cleaning and maintenance and the traffic was routed to the other bore. On-line data collected during the maintenance period were excluded in this study.

A total of 46 pairs of VOC samples were collected in stainless steel canisters during the sampling period. One pair of samples was collected at the inlet and outlet sites simultaneously. Tunnel air was sampled from inlets located at 1.5m above the ground through Teflon tubing and collected into pre-cleaned and pre-evacuated 2 L

stainless steel canisters at a flow rate of 27.3 mL min^{-1} for 2 hours by a multi-port canister sampler (Model 8001, ATEC, California, USA). The sampling periods covered morning rush hour (8:00 – 10:00), midday (11:00 – 13:00 and, 14:00 – 16:00), and afternoon rush hour (17:00 – 19:00). Air samples inside canisters were firstly preconcentrated in a pre-concentrator (Model 7100, Entech Instruments Inc., California, USA) and then analyzed by a gas chromatography-mass selective detector/flame ionization detector (GC-MSD/FID) system (Model 5973N, Agilent Technologies, California, USA). Detailed analysis procedures are described elsewhere (Wang and Wu, 2008; Zhang et al., 2012). Briefly, preconcentrated air samples were firstly separated by a HP-1 column (60 m × 0.32 mm × 1.0 μm, Agilent Technologies, USA), and then separated into two streams: one went through a PLOT-Q column (30 m × 0.32 mm × 2.0 μm, Agilent Technologies, USA) which was detected by FID detector, and the other one went through a 65 cm × 0.1 mm stainless steel line followed by the MSD detector. The accuracy of the measurement was 0.5 – 5%, and the detection limit was 3 – 57 pptv for individual species.

In addition to integrated canister samples, carbon monoxide (CO), and nitrogen oxides ($NO/NO_2/NO_x$) concentrations were measured continuously every minute) at both inlet and outlet sampling sites. CO was monitored with CO analyzers (Model 300E, Teledyne API, California, USA), and $NO/NO_2/NO_x$ was measured by $NO/NO_2/NO_x$ analyzers (Model T200, Teledyne API, California, USA). The sampling inlets of trace gas analyzers were set at 1.5m above the ground with the sampling flow rates both at 1.0 L min^{-1}.

2.3 Emission factor calculation

Emission factors (in mg veh^{-1} km^{-1}) of specific pollutants from vehicular emissions is the mass of the pollutants emitted over a certain distance normalized by the vehicle number and distance. (Pierson and Brachaczek, 1982; Pierson et al., 1996)

$$EF_{veh} = \frac{(C_{outlet} - C_{inlet}) \cdot A \cdot U \cdot t}{N \cdot L} \qquad (1),$$

where EF_{veh} is the average emission factor in mg veh^{-1} km^{-1}; C_{outlet} and C_{inlet} are the mass concentrations on of specific pollutants at the outlet and inlet sampling sites in mg m^{-3}; A is the cross section area of the tunnel in m^2; U is the wind speed in m s^{-1}; t is the sampling duration in s; N is the total number of vehicles pass through the tunnel during the sampling period; and L is the distance between outlet and inlet sampling sites in km.

The emission factors of VOCs calculated by the above equation reflect the fleet-average emission factors of specific VOCs from mixed vehicle types during the sampling period. Multiple linear regressions (MLR) method can be used to estimate the EFs of individual VOCs for non-diesel vehicles (NDVs) and diesel vehicles (DVs) according to the following equation (Grosjean et al., 2001; Ho et al., 2007; Pierson et al., 1996),

$$EF_i = \alpha_i EF_{NDV} + \beta_i EF_{DV} \qquad (2),$$

where EF_i is the measured individual VOC EFs in time interval i; α_i, β_i are the fractions of NDV and DV passing through the tunnel during the time interval i, respectively, and

$\alpha_i + \beta_i = 1$; EF_{NDV} and EF_{DV} are estimated EFs for NDV and DV, respectively.

2.4 Ozone formation potential

VOCs are significant precursors of O_3 formation and different VOC species have different photochemical reactivity. The maximum incremental reactivity (MIR) value developed by Carter (2009)) is used in this study to assess the O_3 formation potential (OFP) of individual VOCs according to the following equation,

$$OFP_j = EF_j \cdot MIR_j \quad (3),$$

where OFP_j is the ozone formation potential of specie j in mgO_3 veh^{-1} km^{-1}; EF_j is the emission factor of specie j in mg veh^{-1} km^{-1}; and MIR_j is the maximum incremental reactivity value of specie j in grams of O_3 per gram of organic compound.

2.5 Traffic count analysis

Traffic compositions and volume were determined by manual counts at the entrance of the tunnel tube at 15-minute intervals during the sampling periods. Video-recording was also taken for data validation and review purposes. The vehicle types were classified into three major categories, namely diesel-fueled vehicle (heavy goods vehicle, light goods vehicle, double deck bus and single deck bus), gasoline-fueled vehicle (motor cycle and private car), and LPG-fueled vehicle (taxi and public light bus). The age and mileage distribution of vehicles were not obtained in this study.

3 Results and discussion

3.1 Characteristics of traffic pattern and trace gas concentrations

About 99.7% of total on-road fleets in Hong Kong used the folowing three types of fules, namely LPG (3.1%), gasoline (77.3%) and diesel (19.3%), by the end of Mar 2015 (HKTD, 2015). Total vehicle fleets were classified into NDVs and DVs in this study.

Figure 2 shows time series of vehicle numbers, wind speed and CO, NO, and NO_2 concentrations at the outlet sampling site of Shing Mun tunnel during the campain in 2015. The total vehicle number cross the tunnel south bore ranged from 500 to 2300 veh h^{-1} during the daytime. Apparent double-peak pattern was found for the total vehicle number on weekdays, but the morning peak was not obvious on weekends. Moreover, the total vehicle number on weekdends was less than that on weekdays, beacuase there were much less DVs (e.g. sigle deck buses, double deck buses and goods vehicles) on weekends, while the total number of NDVs kept at a similar level both on weekdays and on weekends as shown in **Figure 2**. The average wind speed at the outlet of Shing Mun tunnel was about 4.2 m s^{-1}. It can be found that the wind speed maintained at relative stable high level (5.0 m s^{-1}) during the daytime and decreased in the evening when there were less vehicles, and this is consitent with the study of Stemmler et al. (2005)). Apparent double-peak pattern was also found for NDVs during weekdays with the same morning peak (around 8:00) and late afternoon peak (around 18:00) as total vehicle numbers, corresponding to the morning and afternoon rush hours. For DVs, the two peaks were smoother than NDVs' with a later morning peak (9:00 – 10:00) and

earlier afternoon peak (15:00 – 16:00).

The temporal pattern of CO is similar to the NDV counts because spark-ignition vehicles have higher CO emission (Kean et al., 2003). NO_x ($NO + NO_2$) concentrations are influenced by both the larger number of NDVs with lower emission factors and the lower number of DVs with higher emission factors. Therefore, their diurnal variations have mixed contributions from NDVs and DVs (Ho et al., 2013; Huang et al., 2015; Wang et al., 2002). The absence of afternoon NO peak on some days could be resulted from the reaction with ozone.

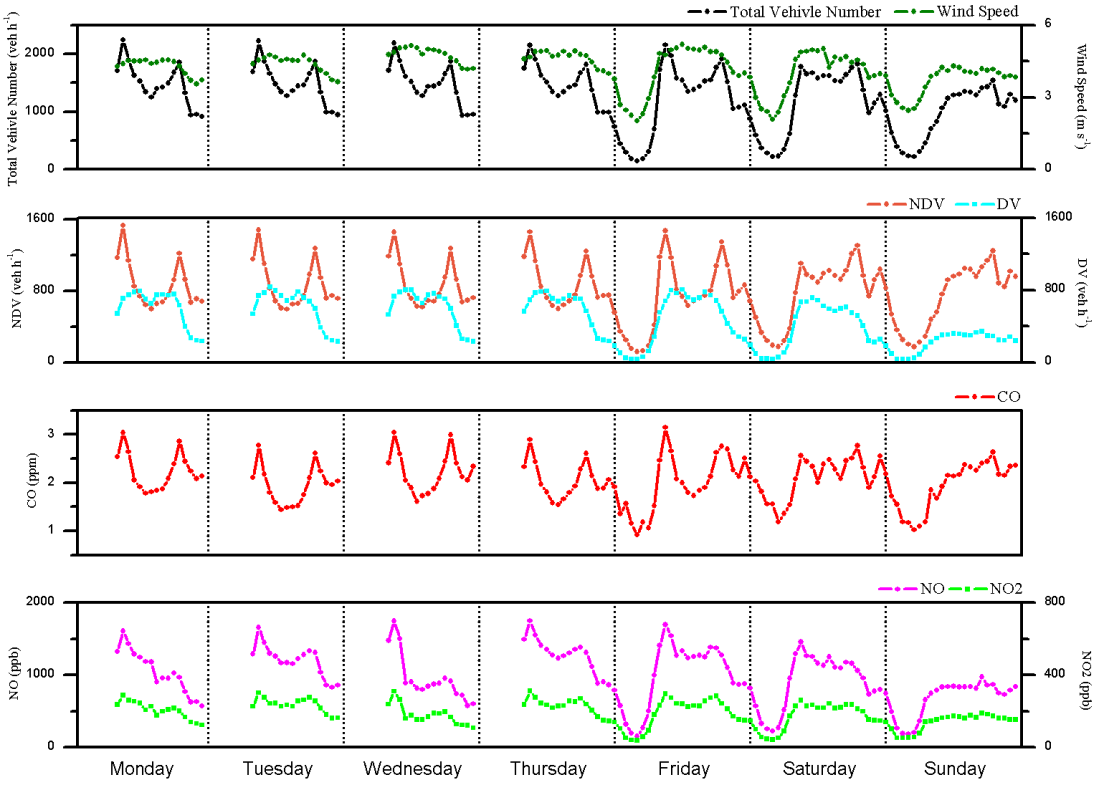


Figure 2. Time series of vehicle numbers, wind speed, and trace gases at the outlet site of Shing Mun tunnel in January - March 2015

3.2 Diurnal pattern of VOCs at the outlet site

Figure 3 shows the diurnal pattern of VOCs at the outlet site of Shing Mun tunnel. The highest concentration of total measured VOCs was found during 8:00 – 10:00 at 78.6 ppb. During 17:00 – 19:00, the concentration of total measured VOCs was 65.0 ppb. Relative lower levels for total measured VOCs were found during 11:00 – 13:00 and 14:00 – 16:00 at 53.3 ppb and 49.4 ppb, respectively. This pattern is consistent with the diurnal pattern of total vehicle numbers, not only the two peaks occurred at the same periods, but also the morning peak was higher than the late afternoon peak.

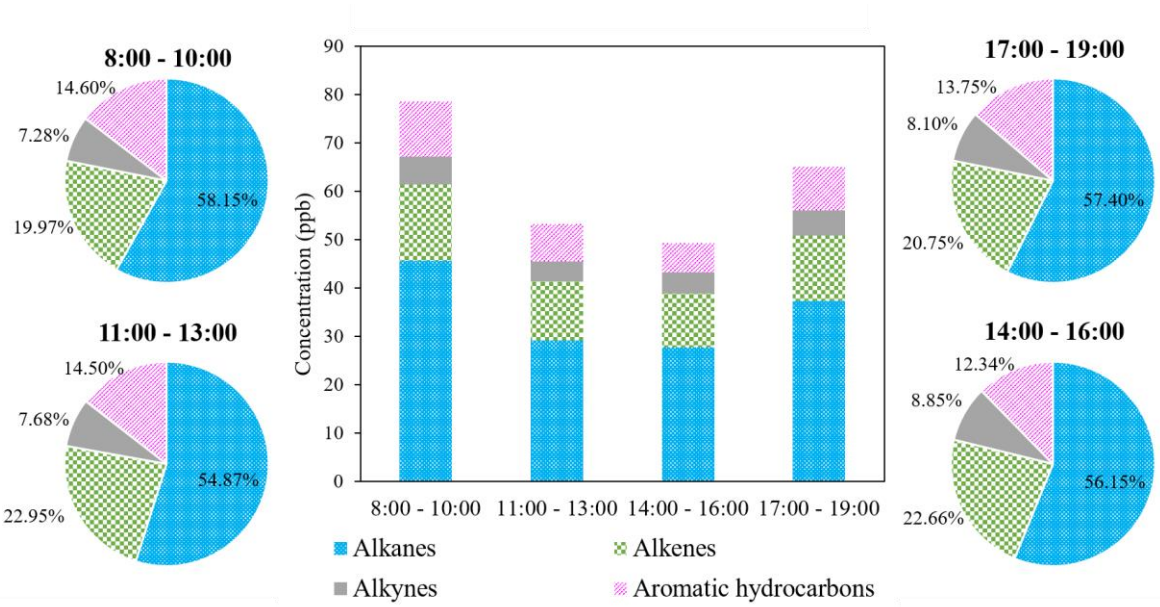


Figure 3. Diurnal patterns of VOCs at the outlet site of Shing Mun tunnel and their group compositions

The composition of different VOC groups at different sampling periods are plotted in **Figure 3**. Similar group abundances were found at different sampling periods, alkanes were the most abundant group, followed by alkenes, aromatic hydrocarbons and alkyne. Alkanes had much higher concentrations and abundances during 8:00 – 10:00 (45.71 ppb) and 17:00 – 19:00 (37.33 ppb) than the other two periods 11:00 – 13:00 (29.23 ppb) and 14:00 – 16:00 (27.73 ppb). Aromatic hydrocarbons had the similar diurnal pattern with alkanes, 11.48 ppb during 8:00 – 10:00, 8.94 ppb 17:00 – 19:00, 7.73 ppb during 11:00 – 13:00, and 6.09 ppb during 14:00 – 16:00. The traffic volume of NDV during the early morning peak (1277 veh h^{-1}) and late afternoon peak (1218 veh h^{-1}) were also much higher than the other two periods (760 – 780 veh h^{-1}). The much higher concentration of alkanes and aromatic hydrocarbons during early morning peak and late afternoon peak could be due to high traffic volume of NDV (about 70% of total vehicles) during these periods as well as NDV's high emission factors of alkanes (56.3 mg veh^{-1} km^{-1}) and aromatic hydrocarbons (12.5 mg veh^{-1} km^{-1}). In contrast, the alkene concentrations were very similar over the four sampling periods; the high volume of NDVs did not cause obviously higher alkene concentrations during 8:00 – 10:00 and 17:00 – 19:00. This is likely due to the relatively constant DV traffic volume over the four sampling periods (1325 veh h^{-1}, 1351 veh h^{-1}, 1365 veh h^{-1}, and 1160 veh h^{-1}, respectively) and higher alkenes emission factors from DVs (9.8 mg veh^{-1} km^{-1}) than NDVs (8.9 mg veh^{-1} km^{-1}).

3.3 Emissions of VOCs at Shing Mun tunnel from 2003 to 2015

In this study, 66 VOC species were quantified in the tunnel air samples. These include 27 C_2 – C_{10} alkanes, 21 C_2 – C_{10} alkenes, 1 alkyne, and 17 aromatic hydrocarbons.

3.3.1 Concentrations of VOCs

The average concentrations and standard deviations of all the measured VOCs at the inlet and outlet sites of Shing Mun tunnel in 2015 are listed in **Table 1**. In 2015, the concentrations of total measured VOCs at the inlet site and outlet sites of the tunnel were 41.83 ppb, and 60.76 ppb, respectively. Among the 66 compounds, n-butane was the most abundant VOC, followed by ethene, propane and i-butane. For both the inlet site and outlet site of Shing Mun tunnel in 2015, alkanes were the most abundant group with the percentage of 57.4% and 56.9%, respectively, followed by alkenes (inlet: 20.3%, outlet: 21.6%), aromatic hydrocarbons (inlet: 13.9%, outlet: 13.7%) and alkyne (inlet: 8.4%, outlet: 7.9%). The inlet and outlet relative contribution were similar indicating that vehicular emissions was the dominant VOC source at the both sampling locations (Ho et al., 2009). Net concentrations (average concentrations of outlet minus inlet concentrations for each pair of canister sample) of the measured VOCs are also listed in **Table 1**. For the net concentrations, n-butane was still the most abundant species with the value of 3.74 ± 2.82 ppb, followed by ethene (2.96 ± 1.74 ppb), i-butane (2.56 ± 2.09 ppb), and propane (2.29 ± 1.44 ppb). This indicates that n-butane, ethene, i-butane and propane have the highest emission rates in the vehicular emissions. Based on the net concentrations, the four most abundant species comprised about 60% of the total measured VOC emissions.

Table 1 also lists the average concentrations and standard deviations of all the measured VOCs at the inlet and outlet of Shing Mun tunnel in 2003 for comparison (Ho et al., 2009). For individual VOC species, **Figure 4(a)** shows the net concentration of each compound in 2003 and 2015. The net concentration decreased from 2003 to 2015 for almost all measured VOCs. The net concentrations of ethene, which was the most abundant species in 2003, decreased by 65.2% from 2003 to 2015. The net concentrations of ethyne, 1-butene, and 1-pentene, which are also marker species of diesel fleets exhaust, decreased by 57.1%, 55.7%, and 89.7%, respectively. However, the net concentrations of i-butane and n-butane, which are key components of LPG, were 50.9% and 33.4% higher in 2015 than in 2003, respectively. This should be due to that about 99.8% of taxies were changed from diesel to LPG from 2000 to 2004, and more than 65% of public light buses were changed from diesel to LPG from 2002 to 2007 (Lau et al., 2015). Based on the licensed number of vehicles by fuel type from the Hong Kong Transport Department, the rate of adoption from diesel to LPG for public light buses increased from 7.5% before the 2003 study to 68.2% before the 2015 study, the rate of adoption from diesel to LPG for taxis increased from 93.4% before the 2003 study to 100% before the 2015 study. Moreover, one previous study at a roadside environment in Hong Kong found the ambient concentrations of propane, i-butane and n-butane increased from 2003 to 2011 with the percentages of 23%, 42%, and 61%, respectively (Huang et al., 2015), which was consistent with the increase of the net concentration of i-butane and n-butane in this study. However, the net concentration of propane, which is another key LPG component, decreased by 4.5% in this study. This may be resulted from the LPG catalytic converter replacement programme for LPG taxies and public light buses from 2013 to 2014 in Hong Kong.

The net concentration of total measured VOCs decreased by 44.7% from 2003 to

2015. For individual VOC groups, the net concentrations of alkanes, alkenes, alkyne, and aromatic hydrocarbons all decreased from 2003 to 2015 with the percentage of 10.8%, 64.9%, 57.1%, and 63.3%, respectively (shown in **Figure 5(a)**). The net concentration of alkanes did not decrease as significantly as the other groups because of the increase of i-butane and n-butane levels.

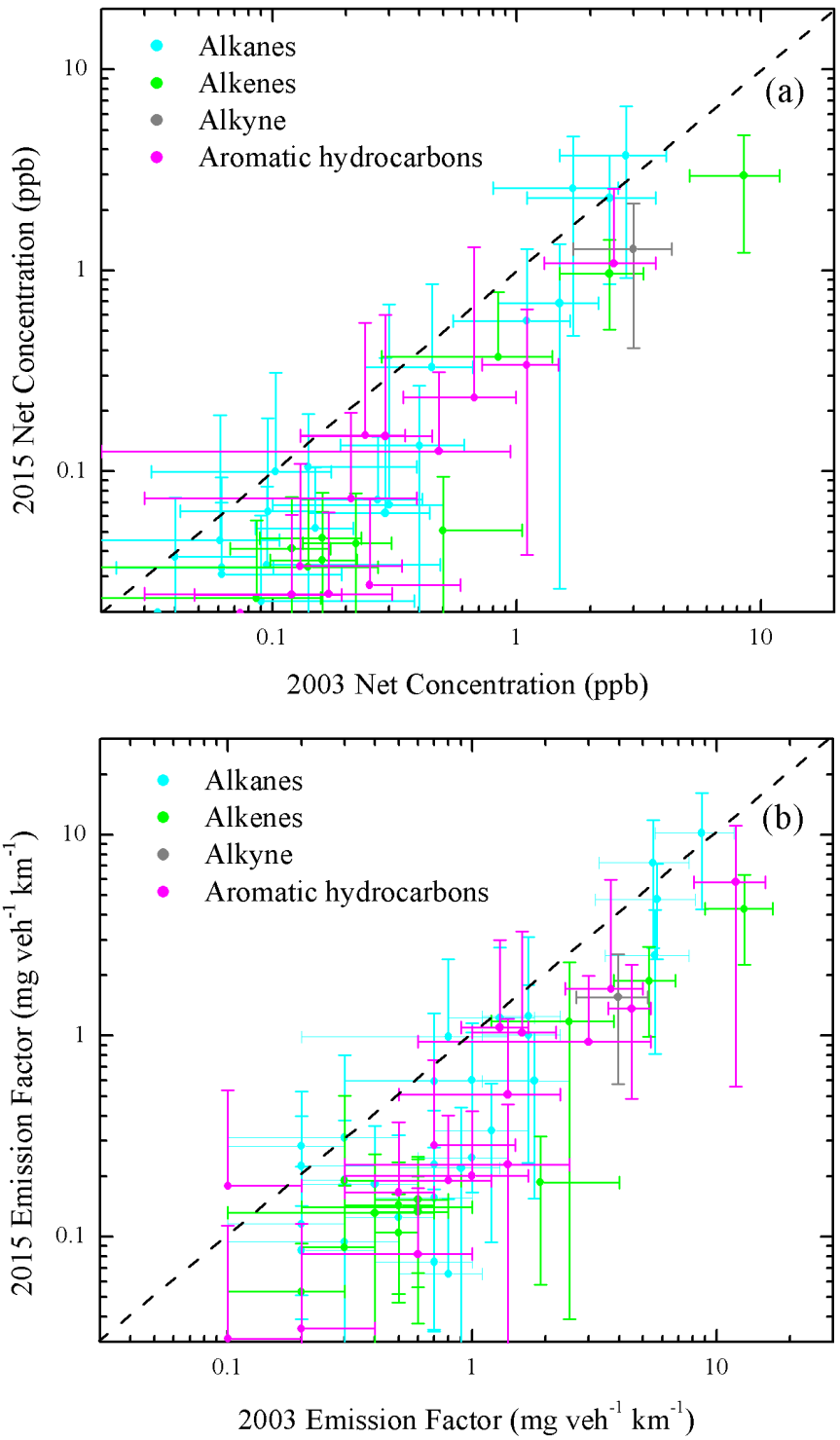
306 **Table 1.** Concentrations and emission factors of VOCs at Shing Mun tunnel in 2003 and 2015

VOCs	SMT 2003 (Ho et al., 2009)				SMT 2015 (this study)			
	Concentration (ppb)			Emission Factor (mg veh^{-1} km^{-1})	Concentration (ppb)			Emission Factor (mg veh^{-1} km^{-1})
	Inlet	Outlet	Outlet minus inlet		Inlet	Outlet	Outlet minus inlet	
Alkane								
ethane	4.4±1.5	5.5±1.8	1.1±0.55	1.7±0.6	3.32±0.86	3.86±0.95	0.56±0.71	1±0.8
propane	5.8±2.8	8.2±4	2.4±1.3	5.7±2.5	5.21±1.82	7.42±2.43	2.29±1.44	4.8±2.4
i-butane	3.5±1.8	5.3±2.6	1.7±0.9	5.5±2.2	4.64±2.26	7.06±2.94	2.56±2.09	7.3±4.5
n-butane	5.7±2.5	8.5±3.7	2.8±1.3	8.7±3.1	6.1±2.52	9.62±4.06	3.74±2.82	10.2±5.9
i-pentane	3.2±1.1	4.6±1.6	1.5±0.66	5.6±2.1	1.45±0.73	2.09±0.94	0.69±0.66	2.5±1.7
n-pentane	0.98±0.31	1.4±0.48	0.45±0.21	1.7±0.6	0.52±0.32	0.83±0.59	0.33±0.53	1.3±1.8
2,2-dimethyl-butane	0.1±0.04	0.13±0.05	0.03±0.05	0.2±0.2	0.04±0.03	0.06±0.03	0.02±0.02	0.1±0.1
cyclopentane	0.53±0.21	0.81±0.34	0.29±0.15	1±0.4	0.05±0.03	0.11±0.29	0.06±0.3	0.2±0.9
2,3-dimethylbutane	0.14±0.06	0.2±0.09	0.06±0.05	0.3±0.2	0.09±0.11	0.13±0.11	0.05±0.15	0.3±0.5
2-methylpentane	0.94±0.33	1.3±0.47	0.4±0.21	1.8±0.7	0.28±0.19	0.41±0.22	0.13±0.13	0.6±0.4
3-methylpentane	0.67±0.22	0.94±0.31	0.27±0.14	1.2±0.5	0.19±0.13	0.25±0.14	0.07±0.08	0.3±0.2
n-hexane	0.68±0.27	0.98±0.41	0.3±0.2	1.3±0.5	0.9±0.83	0.95±0.73	0.07±0.61	1.2±1.5
methyl-cyclopentane	0.32±0.11	0.47±0.17	0.15±0.07	0.7±0.2	0.1±0.06	0.15±0.07	0.05±0.05	0.2±0.2
2,4-dimethylpentane	0.07±0.03	0.11±0.04	0.03±0.02	0.2±0.1	0.04±0.02	0.06±0.03	0.02±0.02	0.1±0.1
cyclohexane	0.18±0.09	0.24±0.1	0.06±0.04	0.3±0.1	0.1±0.08	0.13±0.09	0.03±0.06	0.2±0.2
2-methyl-hexane	0.39±0.34	0.48±0.18	0.09±0.29	0.7±0.3	0.08±0.07	0.1±0.06	0.02±0.04	0.2±0.1
2,3-dimethyl-pentane	0.07±0.04	0.12±0.07	0.05±0.04	0.3±0.2	0.04±0.03	0.06±0.03	0.01±0.02	0.1±0.1
3-methyl-hexane	0.1±0.03	0.14±0.04	0.04±0.02	0.2±0.1	0.2±0.23	0.18±0.1	-0.02±0.2	0.3±0.2
2,2,4-trimethyl-pentane	0.35±0.28	0.49±0.33	0.14±0.25	1±0.7	0.15±0.07	0.25±0.13	0.11±0.09	0.6±0.4
n-heptane	0.43±0.46	0.53±0.2	0.1±0.39	0.9±0.4	0.08±0.05	0.12±0.06	0.03±0.05	0.2±0.2

methyl-cyclohexane	0.22±0.17	0.28±0.11	0.06±0.13	0.4±0.2	0.07±0.04	0.1±0.05	0.03±0.04	0.2±0.2
2,3,4-trimethyl-pentane	0.09±0.05	0.13±0.09	0.04±0.05	0.2±0.2	0.05±0.03	0.09±0.05	0.04±0.04	0.2±0.2
2-methyl-heptane	0.39±0.34	0.48±0.18	0.09±0.29	0.7±0.3	0.03±0.03	0.03±0.02	0±0.03	0.1±0.1
3-methyl-heptane	0.49±0.76	0.49±0.18	0±0.72	0.8±0.3	0.01±0.01	0.02±0.01	0.01±0.02	0.1±0.1
n-octane	0.15±0.05	0.23±0.08	0.08±0.04	0.5±0.2	0.03±0.02	0.05±0.03	0.02±0.03	0.1±0.2
n-nonane	0.14±0.08	0.24±0.11	0.1±0.05	0.7±0.4	0.08±0.08	0.15±0.14	0.06±0.12	0.6±0.7
n-decane	0.14±0.1	0.25±0.12	0.1±0.07	0.8±0.6	0.17±0.15	0.26±0.19	0.1±0.21	1±1.4
Alkene								
ethene	16±4.2	25±7.1	8.5±3.4	13±4	5.62±1.43	8.59±1.91	2.96±1.74	4.2±2
propene	4.5±1.1	6.9±1.9	2.4±0.9	5.3±1.5	1.61±0.44	2.57±0.64	0.97±0.46	1.9±0.9
1-butene	1.7±0.51	2.6±0.85	0.84±0.56	2.5±1.3	0.56±0.25	0.91±0.42	0.37±0.41	1.2±1.1
1,3-butadiene	0.41±0.39	0.36±0.34	-0.05±0.36	0.3±0.6	0.06±0.09	0.08±0.15	0.01±0.14	0.2±0.3
trans-2-butene	0.4±0.13	0.62±0.21	0.22±0.09	0.6±0.2	0.06±0.03	0.11±0.04	0.04±0.03	0.1±0.1
cis-2-Butene	0.29±0.1	0.45±0.16	0.16±0.06	0.5±0.1	0.06±0.03	0.1±0.04	0.04±0.03	0.1±0.1
3-methyl-1-butene	0.11±0.03	0.17±0.05	0.06±0.02	0.2±0.1	0.02±0.01	0.04±0.02	0.02±0.01	0.1±0
1-pentene	0.46±0.34	0.95±0.75	0.5±0.55	1.9±2.1	0.08±0.03	0.13±0.05	0.05±0.04	0.2±0.1
2-methyl-1-butene	0.21±0.08	0.33±0.13	0.12±0.05	0.5±0.2	0.06±0.02	0.1±0.05	0.04±0.03	0.1±0.1
isoprene	0.16±0.34	0.06±0.06	-0.11±0.36	0±0	0.06±0.07	0.05±0.07	-0.02±0.09	0.2±0.2
trans-2-pentene	0.27±0.1	0.44±0.17	0.16±0.07	0.6±0.2	0.06±0.03	0.11±0.05	0.05±0.03	0.2±0.1
cis-2-pentene	N.A.	N.A.	N.A.	N.A.	0.03±0.01	0.05±0.02	0.02±0.02	0.1±0.1
2-methyl-2-butene	0.26±0.18	0.4±0.24	0.14±0.13	0.6±0.4	0.06±0.02	0.09±0.05	0.03±0.04	0.1±0.1
cyclopentene	N.A.	N.A.	N.A.	N.A.	0.02±0.01	0.04±0.02	0.02±0.01	0.1±0
4-methyl-1-pentene	0.09±0.03	0.17±0.06	0.07±0.04	0.3±0.1	0.01±0.01	0.02±0.03	0.01±0.03	0.1±0.1
1-hexene	0.13±0.06	0.21±0.11	0.09±0.07	0.4±0.3	0.04±0.02	0.07±0.03	0.02±0.03	0.1±0.1
trans-2-hexene	0.08±0.03	0.14±0.05	0.06±0.02	0.2±0.1	0.01±0.01	0.01±0.01	0±0.01	0±0
cis-2-hexene	0.04±0.02	0.07±0.03	0.03±0.01	0.1±0	0±0.01	0.01±0.01	0±0.01	0±0

3-hexene	0.06±0.02	0.08±0.03	0.03±0.02	0.1±0	0±0.01	0.01±0.01	0±0.01	0±0
a-pinene	0.01±0.01	0.01±0.01	0±0.01	0±0	0.02±0.07	0.02±0.03	0±0.07	0.1±0.2
b-pinene	0.01±0	0.01±0	0±0	0±0	0±0.01	0±0.02	0±0.02	0±0.1
Alkyne								
ethyne	7.4±1.8	10±2.6	3±1.3	4±1.3	3.53±1.07	4.81±1.41	1.29±0.87	1.5±1
Aromatic hydrocarbon								
benzene	2.5±0.62	3.5±0.92	1.1±0.38	4.5±0.9	0.81±0.31	1.15±0.37	0.34±0.3	1.4±0.9
toluene	6.1±2	8.7±2.8	2.5±1.2	12±3.9	2.4±2.15	3.48±2.14	1.09±1.46	5.8±5.2
ethyl-benzene	0.59±0.21	0.82±0.27	0.24±0.11	1.3±0.4	0.48±0.63	0.64±0.85	0.15±0.4	1.1±1.9
m/p-xylene	1.45±0.46	2.16±0.71	0.67±0.33	3.7±1.3	1.24±2.67	1.56±3.17	0.23±1.07	1.7±4.2
o-xylene	0.64±0.18	0.94±0.31	0.29±0.16	1.6±0.6	0.06±0.09	0.08±0.13	0.01±0.12	0.3±0.5
styrene	N.A.	N.A.	N.A.	N.A.	0.41±0.7	0.59±0.98	0.15±0.45	1±2.3
isopropylbenzene	0.03±0.01	0.05±0.02	0.02±0.01	0.1±0.1	0.03±0.04	0.05±0.06	0.02±0.05	0.2±0.4
n-propylbenzene	0.12±0.04	0.2±0.06	0.07±0.04	0.5±0.2	0.03±0.02	0.05±0.04	0.02±0.03	0.2±0.2
m-ethyltoluene	0.32±0.13	0.53±0.2	0.21±0.18	1.4±0.9	0.08±0.07	0.16±0.15	0.07±0.12	0.5±0.7
p-ethyltoluene	0.13±0.06	0.27±0.23	0.13±0.21	0.7±0.8	0.03±0.03	0.07±0.08	0.03±0.08	0.3±0.5
1,3,5-trimethyl-benzene	0.18±0.07	0.3±0.09	0.12±0.07	0.8±0.4	0.03±0.02	0.06±0.04	0.02±0.04	0.2±0.2
o-ethyltoluene	0.21±0.1	0.38±0.14	0.17±0.14	1±0.7	0.03±0.03	0.06±0.05	0.02±0.04	0.2±0.2
1,2,4-trimethylbenzene	0.72±0.4	1.2±0.46	0.48±0.46	3±2.4	0.14±0.11	0.27±0.25	0.13±0.19	0.9±1
1,2,3-trimethylbenzene	0.28±0.15	0.48±0.19	0.25±0.34	1.4±1.1	0.04±0.04	0.07±0.06	0.03±0.05	0.2±0.2
m-diethylbenzene	0.03±0.02	0.05±0.02	0.03±0.03	0.2±0.2	0±0	0.01±0.01	0±0.01	0±0.1
p-diethylbenzene	0.07±0.06	0.15±0.07	0.08±0.08	0.6±0.4	0.01±0.01	0.02±0.02	0.01±0.02	0.1±0.1
o-diethylbenzene	0.02±0.01	0.03±0.01	0.01±0.01	0.1±0.1	0±0.01	0±0.01	0±0.01	0±0.1
Total	76.16±28.05	111.26±39.20	35.02±19.95	107.1±44.80	41.83±21.40	60.76±27.96	19.36±19.14	58.8±50.7

307 * N.A. – not available



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309 **Figure 4.** Comparison of net concentrations and emission factors of individual VOCs
310 at Shing Mun tunnel in 2003 and 2015 (error bars stand for one standard deviation)

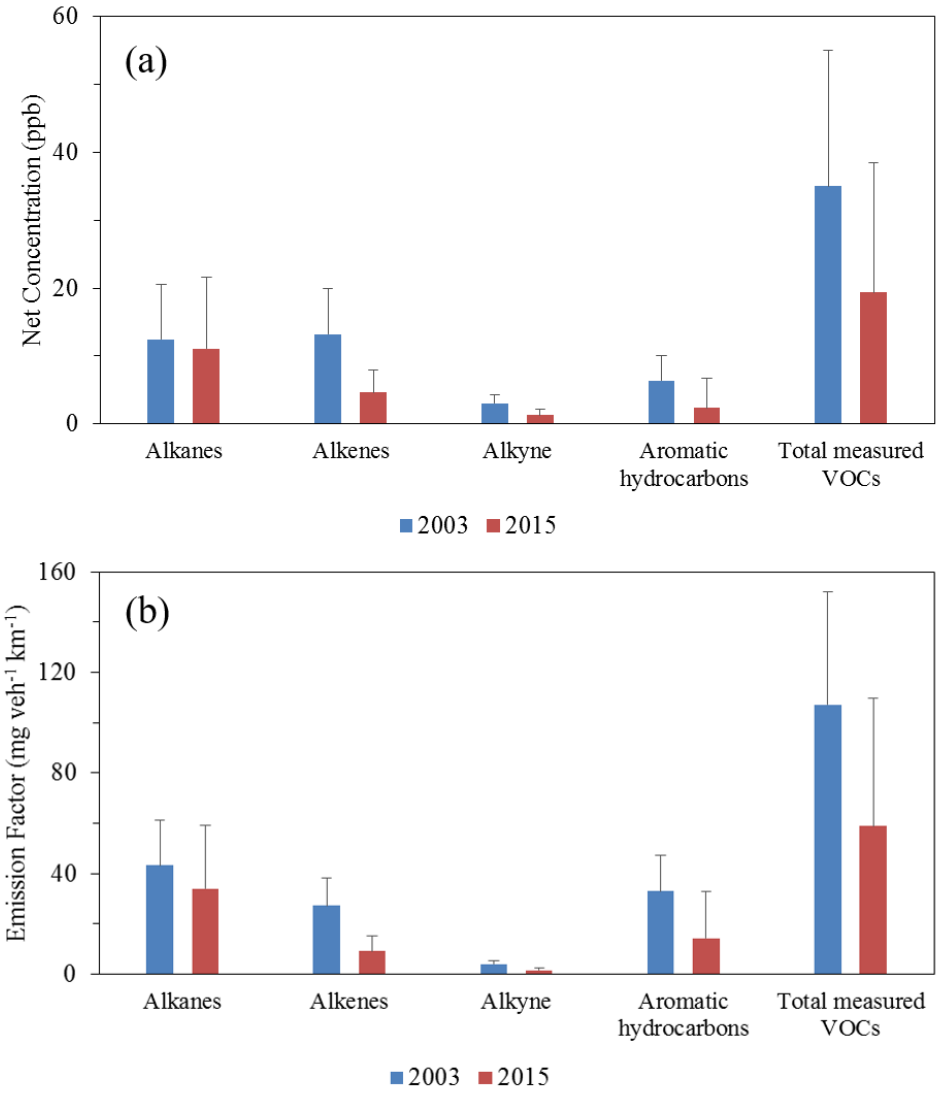

Figure 5. Comparison of net concentrations and emission factors of different VOC groups at Shing Mun tunnel in 2003 and 2015 (error bars stand for one standard deviation)

3.3.2 Emission factors of VOCs

The average emission factors of all measured VOCs are presented in **Table 1**. The emission factors of several VOCs were zero because their concentrations at the outlet site were less or equal to that at the outlet site. In 2015, n-butane, toluene, i-butane, propane and ethene had the highest emission factors of 10.2 ± 5.9 mg veh^{-1} km^{-1}, 5.8 ± 5.2 mg veh^{-1} km^{-1}, 7.3 ± 4.5 mg veh^{-1} km^{-1}, 4.8 ± 2.4 mg veh^{-1} km^{-1}, and 4.2 ± 2.0 mg veh^{-1} km^{-1}, respectively. In contrast, the five most abundant VOC species for emission factor in 2003, in decreasing order were: ethene (13.0 ± 4.0 mg veh^{-1} km^{-1}), toluene (12.0 ± 3.9 mg veh^{-1} km^{-1}), n-butane (8.7 ± 3.1 mg veh^{-1} km^{-1}), propane (5.7 ± 2.5 mg veh^{-1} km^{-1}), and i-pentane (5.6 ± 2.1 mg veh^{-1} km^{-1}), respectively.

Figure 4(b) compares the emission factors of individual VOCs in 2003 and 2015. The emission factors decreased from 2003 to 2015 for almost all measured VOCs. As one of the key tracers for diesel vehicular emissions, ethene had the highest emission

factor in 2003, and its emission factor decreased by 67.3% from 2003 to 2015. However, the emission factors of i-butane and n-butane increased from 2003 to 2015 by 32.1% and 17.2%, respectively, while the magnitude of change of their emission factors were much lower than their net concentrations.

Because of the series of air pollution control measures, the total vehicular emission factors of measured VOCs decreased from 107.1 ± 44.8 mg veh^{-1} km^{-1} in 2003 to 58.8 ± 50.7 mg veh^{-1} km^{-1} in 2015 as shown in **Figure 5(b)**. The emission factor of alkenes decrease the most among the five groups with the percentage of 66.3%, followed by alkyne (61.4%), aromatic hydrocarbons (57.0%), and alkanes (21.2%).

3.4 Reconstructed emission factor for NDVs and DVs

The reconstructed emission factors of individual VOCs for NDVs, and DVs based on the MLR method are presented in **Table 2**. The top 4 species of EF_{NDV} were n-butane (18.7 ± 3.9 mg veh^{-1} km^{-1}), i-butane (13.4 ± 3.0 mg veh^{-1} km^{-1}), propane (7.9 ± 1.6 mg veh^{-1} km^{-1}), and i-pentane (4.8 ± 1.1 mg veh^{-1} km^{-1}), respectively. Among the four species, n-butane, i-butane, and propane are major tracers of LPG vehicular emissions (Ling et al., 2011; Liu et al., 2008; Ou et al., 2015), and i-pentane is one of major tracers of gasoline vehicular emissions (Ou et al., 2015; Tsai et al., 2006). **Figure 6** shows the EF_{NDV} and EF_{DV} of specific VOC species in 2003 and 2015. Even though n-butane, i-butane, propane, and i-pentane had the highest EF_{NDV} in 2015, EF_{NDV} of n-butane, i-butane, propane, and i-pentane decreased by 62.3%, 60.6%, 78.4%, and 61.0%, respectively, from 2003 to 2015. The large decrease of EF_{NDV} of these LPG tracers could be due to the effectiveness of reducing LPG vehicle emissions by the LPG catalytic converter replacement programme even though the total LPG vehicle fleets significantly increased during the past twelve years in Hong Kong. The top 4 species of EF_{DV} were toluene (8.3 ± 5.6 mg veh^{-1} km^{-1}), ethene (6.0 ± 2.1 mg veh^{-1} km^{-1}), n-pentane (2.1 ± 2.0 mg veh^{-1} km^{-1}), and ethyne (1.8 ± 1.0 mg veh^{-1} km^{-1}), respectively. As one of the key tracers of diesel vehicular emissions, ethene had the highest EF_{DV} (25.70 ± 2.62 mg veh^{-1} km^{-1}) in 2003, but the EF_{DV} of ethene has been decreased to 6.0 ± 2.1 mg veh^{-1} km^{-1} (by 76.8%) in 2015. The large decrease of EF_{NDV} and EF_{DV} for these key tracers of LPG, gasoline and diesel demonstrates the effectiveness air pollution control measures in Hong Kong from 2003 to 2015.

Table 2. EF_{NDV} and EF_{DV} of VOCs at Shing Mun tunnel in 2015

VOCs	EF_{NDV}		EF_{DV}		r^2
	Average	S.D.	Average	S.D.	
Alkane					
ethane	0.8	0.5	1.3	0.8	0.6
propane	7.9	1.6	0.0	2.4	0.8
i-butane	13.4	3.0	0.0	4.5	0.7
n-butane	18.7	3.9	0.0	5.9	0.8
i-pentane	4.8	1.1	0.0	1.7	0.7
n-pentane	0.7	1.3	2.1	2.0	0.3
2,2-dimethyl-butane	N.A.	N.A.	N.A.	N.A.	N.A.

cyclopentane	0.1	0.7	0.4	1.1	0.0
2,3-dimethylbutane	N.A.	N.A.	N.A.	N.A.	N.A.
2-methylpentane	0.9	0.3	0.1	0.5	0.6
3-methylpentane	0.5	0.2	0.1	0.3	0.7
n-hexane	3.4	1.2	0.0	1.8	0.4
methyl-cyclopentane	0.2	0.1	0.2	0.2	0.6
2,4-dimethylpentane	N.A.	N.A.	N.A.	N.A.	N.A.
cyclohexane	0.1	0.1	0.3	0.2	0.5
2-methyl-hexane	0.1	0.1	0.2	0.1	0.6
2,3-dimethyl-pentane	N.A.	N.A.	N.A.	N.A.	N.A.
3-methyl-hexane	0.2	0.2	0.4	0.3	0.6
2,2,4-trimethyl-pentane	N.A.	N.A.	N.A.	N.A.	N.A.
n-heptane	0.4	0.2	0.0	0.3	0.5
methyl-cyclohexane	0.1	0.1	0.3	0.2	0.5
2,3,4-trimethyl-pentane	N.A.	N.A.	N.A.	N.A.	N.A.
2-methyl-heptane	0.2	0.1	0.0	0.1	0.4
3-methyl-heptane	0.1	0.1	0.0	0.1	0.3
n-octane	0.3	0.1	0.0	0.2	0.3
n-nonane	0.3	0.5	1.0	0.8	0.4
n-decane	2.9	1.1	0.0	1.6	0.4
Alkene					
ethene	3.1	1.4	6.0	2.1	0.8
propene	2.2	0.6	1.3	0.9	0.8
1-butene	1.1	0.9	1.4	1.3	0.5
1,3-butadiene	N.A.	N.A.	N.A.	N.A.	N.A.
trans-2-butene	0.1	0.0	0.1	0.1	0.8
cis-2-Butene	0.2	0.0	0.0	0.1	0.8
3-methyl-1-butene	0.1	0.0	0.0	0.0	0.7
1-pentene	0.1	0.1	0.3	0.1	0.7
2-methyl-1-butene	0.2	0.1	0.0	0.1	0.7
isoprene	0.1	0.2	0.2	0.3	0.3
trans-2-pentene	0.3	0.1	0.0	0.1	0.7
cis-2-pentene	0.2	0.0	0.0	0.1	0.7
2-methyl-2-butene	0.3	0.1	0.0	0.1	0.7
cyclopentene	0.1	0.0	0.1	0.0	0.7
4-methyl-1-pentene	0.0	0.1	0.2	0.1	0.5
1-hexene	0.2	0.1	0.1	0.1	0.5
trans-2-hexene	0.0	0.0	0.0	0.0	0.3
cis-2-hexene	0.0	0.0	0.0	0.0	0.6
3-hexene	0.1	0.0	0.0	0.0	0.3
a-pinene	0.4	0.1	0.0	0.2	0.4
b-pinene	0.2	0.1	0.0	0.1	0.1
Alkyne					
ethyne	1.4	0.7	1.8	1.0	0.7

Aromatic hydrocarbon					
benzene	1.2	0.6	1.6	0.9	0.7
toluene	4.1	3.7	8.3	5.6	0.5
ethyl-benzene	0.9	1.5	1.4	2.4	0.2
m/p-xylene	3.6	3.3	0.0	5.2	0.1
o-xylene	0.5	0.4	0.1	0.6	0.2
styrene	0.5	1.7	1.8	2.7	0.1
isopropylbenzene	0.0	0.3	0.4	0.4	0.2
n-propylbenzene	0.3	0.1	0.0	0.2	0.4
m-ethyltoluene	0.5	0.5	0.5	0.7	0.3
p-ethyltoluene	0.2	0.3	0.5	0.5	0.2
1,3,5-trimethyl-benzene	N.A.	N.A.	N.A.	N.A.	N.A.
o-ethyltoluene	0.3	0.2	0.1	0.2	0.4
1,2,4-trimethylbenzene	N.A.	N.A.	N.A.	N.A.	N.A.
1,2,3-trimethylbenzene	N.A.	N.A.	N.A.	N.A.	N.A.
m-diethylbenzene	0.1	0.1	0.0	0.1	0.2
p-diethylbenzene	0.2	0.1	0.0	0.1	0.4
o-diethylbenzene	0.1	0.1	0.0	0.1	0.1

* N.A. – not available

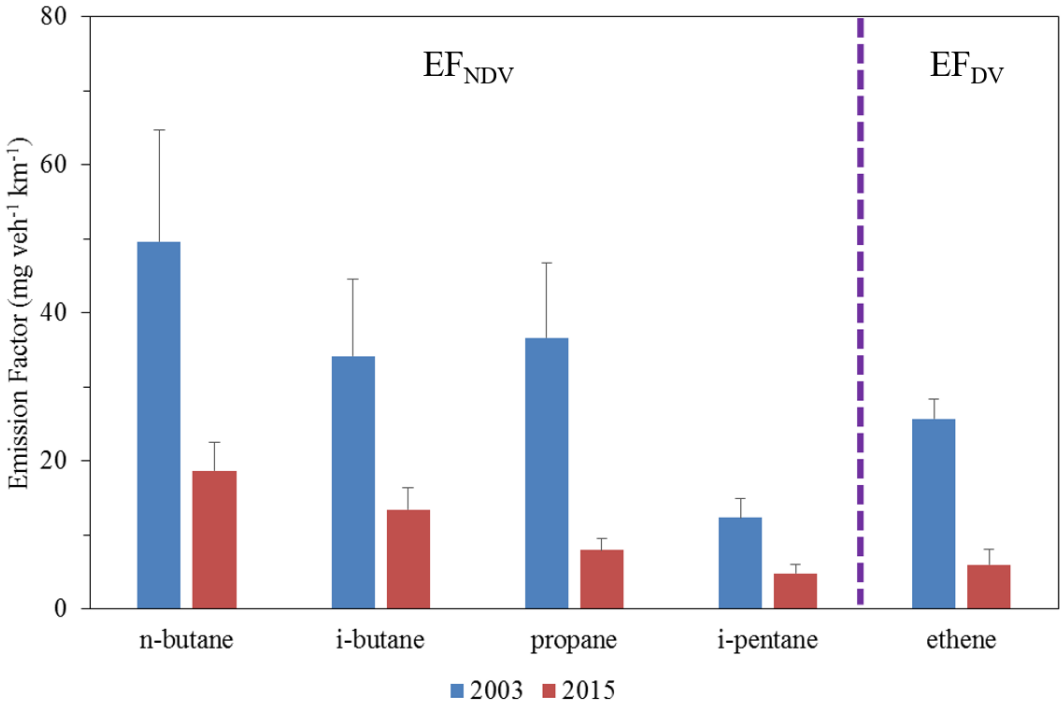

Figure 6. EF_{NDV} (left panel) and EF_{DV} (right panel) of specific VOC species through the tunnel study in 2003 and 2015 (error bars stand for one standard deviation)

3.5 Ozone formation potential of VOCs

MIR method (Carter, 1994) was applied to evaluate the contribution of VOCs from vehicular emissions to OFP in Hong Kong. The top ten species with the highest OFP

from vehicular emission in 2003 and 2015 are listed in **Table 3**. Ethene contributed the most to OFP of the measured VOCs in both 2003 (24.0%) and 2015 (19.5%). Due to the large decrease of diesel vehicular emissions, OFP of ethene decreased from 113.9 mgO_3 veh^{-1} km^{-1} in 2003 to 37.2 mgO_3 veh^{-1} km^{-1} in 2015. n-Butane and i-butane, which were in top ten list of OFP in 2015, were not major contributors to total OFP in 2003, because of the increase of their concentrations in 2015 and the decrease of other species as explained in section 3.3. The total OFP of measured VOCs decreased by 59.8% from 474.1 mg O_3 veh^{-1} km^{-1} to 190.8 mg O_3 veh^{-1} km^{-1} during the past 12 years (shown in **Figure 7**). The contributions of different groups to total OFP in 2003 and 2015 are also shown in **Figure 7**. The contributions by VOC groups to total OFP were different in 2003 and 2015 with the large decrease of alkenes' contribution from 53.7% in 2003 to 45.1% in 2015. However, alkenes and hydrocarbon aromatics were always the two major contributors to total OFP from vehicular emissions in Hong Kong in both 2003 and 2015.

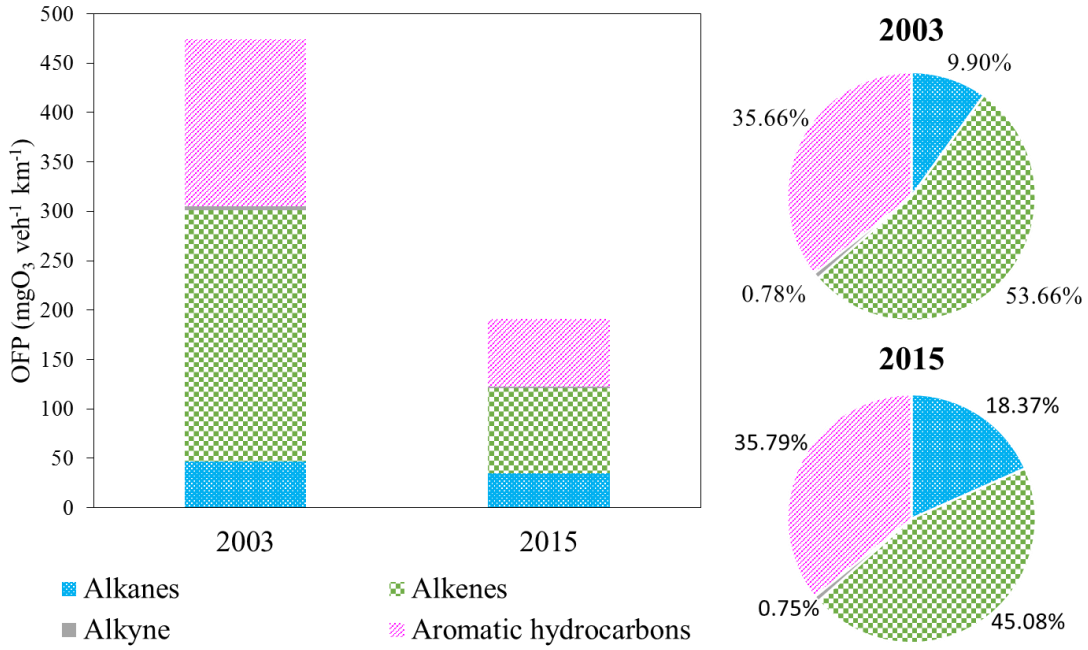

Figure 7. OFP of different VOC groups at Shing Mun tunnel in 2003 and 2015

388 **Table 3.** Top ten species with the highest OFP from vehicular emission in 2003 and 2015 through the tunnel study

2003					2015				
VOCs	**MIR ($gO_3\ g^{-1}$)**	**OFP ($mgO_3\ veh^{-1}\ km^{-1}$)**		**Percentage (%)**	**VOCs**	**MIR ($gO_3\ g^{-1}$)**	**OFP ($mgO_3\ veh^{-1}\ km^{-1}$)**		**Percentage (%)**
		Average	**S.D.**				**Average**	**S.D.**	
ethene	8.76	113.88	35.04	24.0%	ethene	8.76	37.23	17.55	19.5%
propene	11.37	60.26	17.06	12.7%	toluene	3.88	22.43	20.25	11.8%
toluene	3.88	46.56	15.13	9.8%	propene	11.37	21.22	10.02	11.1%
m/p-xylene	7.57	28.01	9.84	5.9%	m/p-xylene	7.57	12.94	32.14	6.8%
1,2,4-trimethylbenzene	8.64	25.92	20.74	5.5%	1-butene	9.42	11.05	10.68	5.8%
1-butene	9.42	23.55	12.25	5.0%	n-butane	1.08	11.01	6.42	5.8%
1,2,3-trimethylbenzene	11.66	16.32	12.83	3.4%	i-butane	1.17	8.50	5.30	4.5%
1-pentene	6.97	13.24	14.64	2.8%	1,2,4-trimethylbenzene	8.64	8.07	9.07	4.2%
o-xylene	7.44	11.90	4.46	2.5%	o-xylene	7.44	7.72	16.80	4.0%
3-ethyltoluene	7.21	10.09	6.49	2.1%	m-ethyltoluene	7.21	3.69	5.05	1.9%

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3.6 Estimation of road transport VOC emission factor in Hong Kong

From Hong Kong Environmental Protection Department (EPD)'s emission inventory results, total VOC emissions from road transport in Hong Kong was reduced by 36.8% from 2003 (7,600 tonnes) to 2015 (4,800 tonnes) (HKEPD, 2017). A linear trend analysis in **Figure 8(a)** shows that the annual reduction of road transport VOC emission in Hong Kong was -234 tonnes per year (r^2 = 0.97). **Figure 8(b)** shows that the annual total VKT in Hong Kong has been linearly increasing at a rate of 1.73×10^8 veh km per year (r^2 = 0.94) (http://www.td.gov.hk/). Road transport VOC emission factor of the entire Hong Kong vehicle fleet can be obtained through total VOC emissions from road transport and VKT data based on following equation.

$$EF = \frac{RTVE \times 10^9}{VKT} \qquad (4),$$

where EF is emission factor, mg veh^{-1} km^{-1}; RTVE is road transport vehicle emission, tonnes; VKT is vehicle kilometers traveled, veh km.

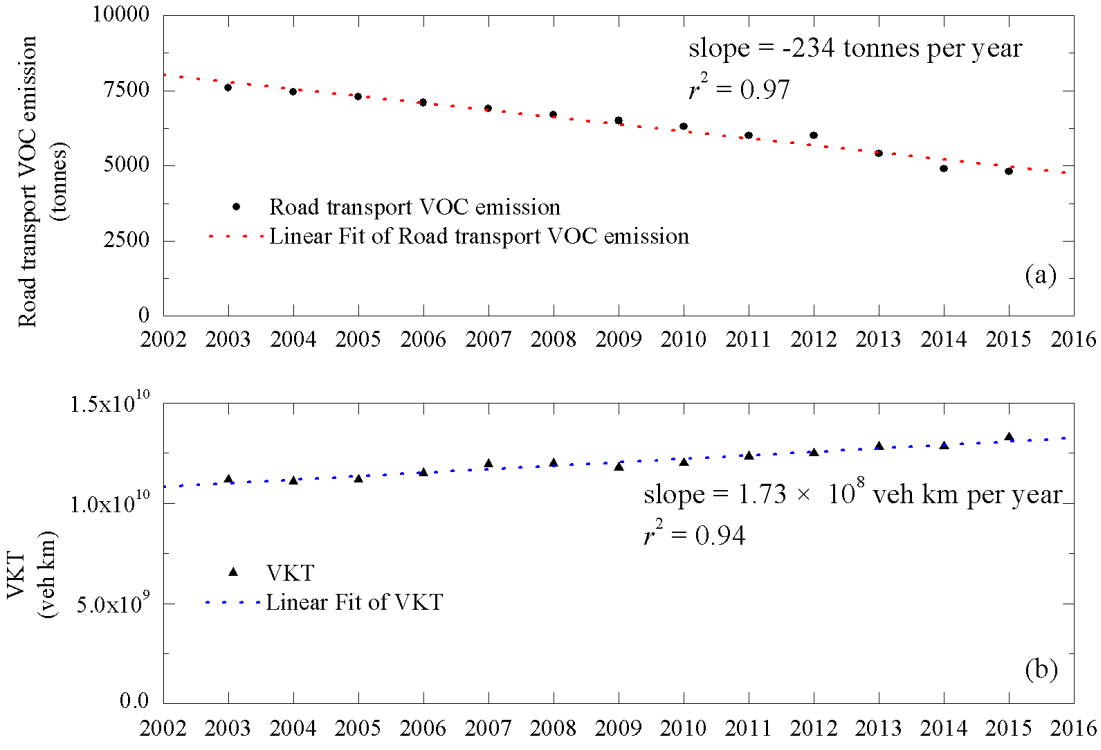

Figure 8. Total road transport VOC emission and VKT in Hong Kong and their linear regression with years

Total road transport VOC emission factor in Hong Kong from 2003 to 2020 is plotted in **Figure 9**. The total road transport VOC emission factor in Hong Kong from 2016 to 2020 is estimated by the linear regression in **Figure 8** and Equation (4). It can be found that total road transport VOC emission factor in Hong Kong decreased by 46.8% from 2003 (679.2 mg veh^{-1} km^{-1}) to 2015 (361.2 mg veh^{-1} km^{-1}). Meanwhile, from the tunnel results in this study, the emission factor of total measured VOCs decreased from 107.1 ± 44.8 mg veh^{-1} km^{-1} in 2003 to 58.8 ± 50.7 mg veh^{-1} km^{-1} in

2015 with the percentage of 45.1%. The decrease percentage was comparable between simulation result (46.8%) and tunnel experiment result (45.1%). However, the estimated total road transport VOC EF in Hong Kong is about 6 times of tunnel experiment result. It should be pointed out that, the tunnel experiments only provide the EFs of vehicles under the tunnel conditions: warmed up engines and cruising speed mostly in the range 60 – 70 km h^{-1}, while on-road vehicles have a larger varieties of driving conditions including cold starts and stop-and-go. It has been reported that the lower vehicle speed, the higher VOC EFs (Int Panis et al., 2006; Kean et al., 2003). Guo et al. (2011)) reported that the total VOC EFs of gasoline and LPG vehicles under idle state were more than 10 times of that under 70 km h^{-1} in Hong Kong. Considering the huge traffic density in Hong Kong, the average vehicle speed should be less than 60 – 70 km h^{-1}, which can explain the 6 times difference between entire fleet estimation and tunnel experiment results. To 2020, the total road transport VOC emission and emission factor is estimated to be 3,803 tonnes, and 279.6 mg veh^{-1} km^{-1}, respectively. Hong Kong committed to reduce total VOC emissions by at least 15% by 2020 as compared to 2010, as part of an emission reduction plan for the Pearl River Delta (PRD) region. Assuming the emission trend will remain as found in this study, it is estimated that road transport VOC emission, one of the major sources of VOCs in Hong Kong, can be reduced by 40% from the 2010 level by 2020. Therefore, the reduction of road transport VOC emissions will be a driving contributor for achieving the goal of total VOC reduction in Hong Kong.

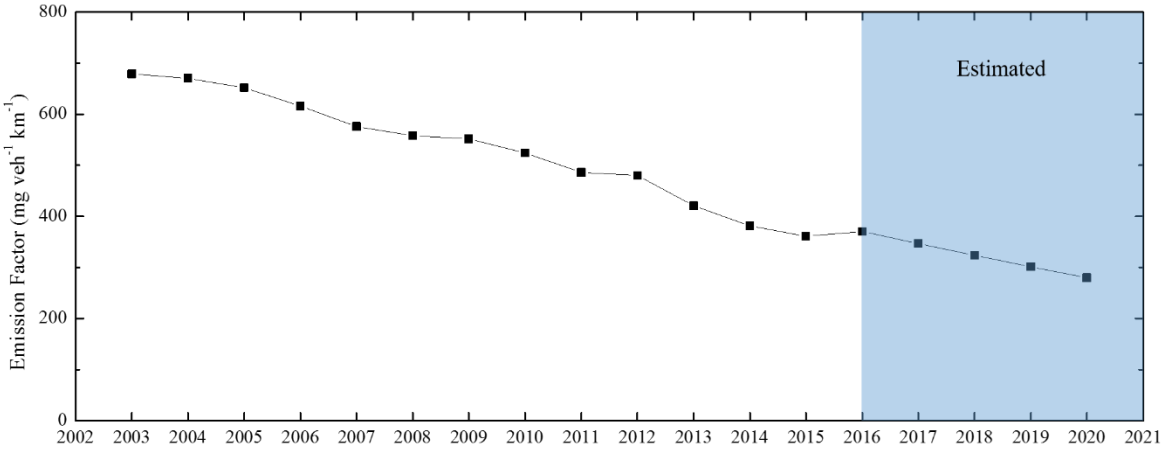

Figure 9. Total road transport VOC emission factor in Hong Kong from 2003 to 2020

4 Conclusions

Large decrease of VOC emissions from on-road vehicles was found in Hong Kong from 2003 to 2015 with the total measured VOC emission factor decreased from 107.1 ± 44.8 mg veh^{-1} km^{-1} in 2003 to 58.8 ± 50.7 mg veh^{-1} km^{-1}. The emission factor of ethene decreased by 67.3% from 2003 to 2015 as a result of diesel vehicular emissions controls over the past twelve years in Hong Kong. The total OFP of measured VOCs decreased by 60% from 2003 (474.1 mgO_3 veh^{-1} km^{-1}) to 2015 (190.8 mgO_3 veh^{-1} km^{-1}). Total road transport VOC emission has been reduced by more than 36.8% from 2003 to 2015, demonstrating the effectiveness of air pollution control strategies in Hong

Kong. To 2020, total road transport VOC emissions is estimated can be reduced by 40% as compared with 2010, which will help achieve total VOC emission reduction goal set by the emission reduction plan for the PRD region.

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