Comparison of Vehicle Emissions by EMFAC-HK Model and Tunnel 1 Measurement in Hong Kong 2 3 Xiaoliang Wanga*, Lung-Wen Antony Chenb, Kin-Fai Hoc, Chi Sing Chand, Zhuozhi 4 Zhang^d, Shun-Cheng Lee^d, Judith C. Chow^a, and John G. Watson^a 5 6 7 ^aDesert Research Institute, Reno, Nevada, U.S.A. ^bUniversity of Nevada-Las Vegas, Las Vegas, Nevada, U.S.A. 8 ^cThe Chinese University of Hong Kong, Hong Kong, China 9 ^dHong Kong Polytechnic University, Hong Kong, China 10 11 12 Submitted to: 13 14 15 Atmospheric Environment 16 Original Submission: December 21, 2020 17 18 Revision 1: March 24, 2021 19 Revision 2: April 21, 2021 20

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Abstract

On-road vehicular emissions were measured in a Hong Kong roadway tunnel and compared to those from a mobile emission model (EMFAC-HK) for the 2003 and 2015 fleets to assess emission changes and effectiveness of emission controls. EMFAC-HK results compared well with tunnel measurement for the 2015 fleet with differences ≤50% for most pollutants. Both measurement and modeled estimates show that non-methane hydrocarbon (NMHC) and PM_{2.5} decreased by over 40% from 2003 to 2015. Tunnel measurements also show that nitrogen dioxide (NO₂) and liquefied petroleum gas (LPG) -related NMHC emissions increased due to expanded installation of diesel oxidation catalysts and LPG fueling. Motorcycles and LPG-fueled public light buses (3.7% of the fleet) contributed disproportionally high fractions of carbon monoxide (CO; 27%) and NMHC (49%) and deserve additional emission controls. Fuel economy improvements did not result in expected carbon dioxide (CO₂) emission reductions, indicating that more aggressive CO₂ reductions, particularly from heavy-duty vehicles, are needed.

Keywords: tunnel study; emission model; EMFAC-HK; vehicle emissions; emission factors; air pollution

1 Introduction

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On-road vehicle emissions are major air pollution sources in urban areas, causing adverse health effects to the public, particularly for those who live or work close to busy roads (HEI, 2010; Krzyzanowski et al., 2005). Emissions from the road transport sector in Hong Kong contributed to 51% of carbon monoxide (CO), 18% of nitrogen oxides (NO_x) and volatile organic compounds (VOCs), and 10% of primary PM_{2.5} (particulate matter with an aerodynamic diameter ≤2.5 µm) in 2015 (HKEPD, 2017a). The transportation sector also contributed to 18% of total greenhouse gas emissions (HKENB, 2020). Vehicle emissions are often estimated using mobile source emission models such as EMission FACtors (EMFAC) by the California Air Resources Board (CARB, 2018), MOtor Vehicle Emission Simulator (MOVES) by the United States Environmental Protection Agency (U.S. EPA, 2018), and COmputer Programme to calculate Emissions from Road Transport (COPERT) by the European Environment Agency (Ntziachristos et al., 2009). These models have been widely used for assessing mobile source emissions at national to project levels to generate emission inventories, develop pollution control implementation plans, evaluate effectiveness of emission control regulations, analyze/demonstrate project conformity, and predict future mobile emission trends. Estimates from mobile emission models may deviate from real-world conditions. Emissions are calculated by summing the products of vehicle activities and emission rates (Fujita et al., 2012). Different models use different methods to quantify and stratify vehicle activities and to determine baseline emission factors (EFs) as well as adjustment factors. The base EFs and their adjustment factors are often derived from limited testing data and may not be representative of real-world vehicle emissions. The U.S. National Research Council (NRC, 2000) pointed out

that adequate validation and evaluation of these models is needed to ensure their accuracy. A review of mobile models by Smit et al. (2010) concluded that the mean prediction errors were generally a factor of 1.3 of the observed values of carbon dioxide (CO₂), within a factor of 2 for hydrocarbon and NO_x, and within a factor of 3 for CO and particulate matter (PM).

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Real-world vehicle emission measurements to validate model estimates include remote sensing, roadside measurements, portable emission measurement system (PEMS) onboard testing, on-road plume chasing, and tunnel measurements (Franco et al., 2013; Smit et al., 2010). Tunnel studies have the advantage of sampling a large number of vehicles, adequately representing local real-world fuels and fleet compositions (El-Fadel and Hashisho, 2001; HEI, 2010). Well-defined parameters (i.e., air temperature [T], relative humidity [RH], wind speed and direction, and vehicle speed) in the tunnel simplify evaluation of emission models with these specific input parameters. Furthermore, repeating vehicle emission measurements in the same tunnel over years allows examination of long-term emission trends and the effectiveness of emission control measures implemented between tunnel studies. Several limitations of tunnel measurements should be kept in mind when interpreting or generalizing results (HEI, 2010; Marinello et al., 2020). Vehicles in the tunnels represent a limited range of operating conditions (e.g., hot-stabilized); therefore, study results provide snapshots of emissions under the vehicle fleet, driving speeds, and road conditions, which may not fully represent other driving and road conditions. Owing to confinement of the vehicles, fresh emissions measured in tunnels do not experience as much atmospheric dilution and mixing as on an open roadway.

The vehicle emission model EMFAC-HK, developed by Hong Kong Environmental Protection Department (HKEPD, 2020b), has been officially used to develop Hong Kong-wide as well as high-resolution vehicle emission inventories, to model street-level vehicle emissions

and dispersion, and to evaluate vehicle emission control effectiveness (Che et al., 2020; Leung, 2019; Li et al., 2020a; Wang et al., 2019; Xing and Brimblecombe, 2018; Xing et al., 2019). EMFAC-HK calculates vehicle EFs and emission rates based on localized vehicle classes, fuels, and activities. However, limited studies have validated the model performance. Lau et al. (2011) compared EMFAC-HK estimates for liquefied petroleum gas (LPG) taxi emissions with onboard measurements by PEMS. Although EMFAC-HK estimates agreed with measurements for CO and total hydrocarbon (THC) within ~10%, the model underestimated NO_x EFs by a factor of ~3. Brimblecombe et al. (2015) found that EMFAC-HK EFs for CO and NO_x were lower than tunnel measurements by factors of 2–3.

This study aims to: (1) compare vehicle emissions measured in a Hong Kong roadway tunnel with the EMFAC-HK model to assess model performance; (2) compare measured and modeled emissions between 2003 and 2015 to evaluate changes of vehicle emissions and effectiveness of emission controls during this period; and (3) examine modeled emission changes by vehicle classes and fuels to inform future emission control policies.

2 Method

2.1 The Shing Mun Tunnel (SMT)

The SMT is a two-bore highway tunnel that connects the two towns of Sha Tin and Tsuen Wan in the New Territories of Hong Kong. Each bore has one-way traffic with two traffic lanes (Supplemental Figure S1). Vehicle emissions were measured in the south bore of the SMT east section, which is 1.6 km long with a rising grade of 1.054% and a cross-sectional area of 70 m². Approximately 53,000 vehicles pass through the tunnel every day in both directions, with an average gasoline, LPG, and diesel fleet mix of ~41%, 9%, and 50% in 2003 and 45%, 13%, and 42% in 2015 (HKTD, 2016). None of the air ventilation fans in the tunnel were turned on during the study; therefore, ventilation in the tunnel was induced by wind and traffic flow. Most

vehicles were travelling at ~80 km hr⁻¹, the tunnel speed limit. The tunnel is sufficiently distant from local neighborhoods (the closest highway ramp is about 2.5 km from the entrance) so that vehicles passing through the tunnel are likely hot-stabilized.

2.2 Vehicle Emission Measurements in the Shing Mun Tunnel

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Vehicle emissions were measured during August 2003 and January–February 2004 (referred to as the 2003 study) as well as during January–March 2015. Detailed measurements are described in the Supplementary Materials and in previous publications (Cheng et al., 2006; 2010; Cui et al., 2018; Ho et al., 2007; 2009a; 2009b; Niu et al., 2020; Wang et al., 2018; 2019). Simultaneous measurements were conducted with identical or similar instruments (Supplemental Figure S1 and Table S1) at inlet and outlet sites separated by distance of 600 m. Concentrations of carbon dioxide (CO₂), CO, nitric oxide (NO), nitrogen dioxide (NO₂), sulfur dioxide (SO₂), and PM_{2.5} (by light scattering) were measured with near real-time instruments at time resolutions of 1 s to 1 min. Integrated one- to two-hour samples of non-methane hydrocarbon (NMHC) and PM_{2.5} were collected in canisters and on filters, respectively. Wind speed, wind direction, T, and RH were measured by two weather stations every five minutes. The traffic flow was recorded by video cameras installed at the tunnel entrance and the outlet site. The time-integrated samples were collected during morning and evening rush hours (e.g., 0800–1000 and 1700–1900 local standard time [LST]), midday hours (e.g., 1100–1300 and 1400–1600 LST), and late evening hours (e.g., 2100–2300 LST). The real-time continuous gaseous, PM, meteorological, and traffic camera data were averaged over each integrated sampling period.

The distance-based emission factor (EF_D; in g veh⁻¹ km⁻¹) calculation uses the mass balance principle (Pierson et al., 1996):

$$EF_D = \left(C_{i,out}U_{out} - C_{i,in}U_{in}\right)A\Delta t/(N \times L) \tag{1}$$

where C_i (g m⁻³) is the average concentrations of pollutant i, U (m s⁻¹) is the wind speed; subscripts in and out denote the values measured at the inlet and outlet sampling sites, respectively; A (m²) is the tunnel cross-sectional area (70 m² for SMT); Δt (s) is the sampling period; N is the number of vehicles passing through the tunnel section during Δt ; and L (m) is distance between the inlet and outlet sampling sites (0.6 km).

During the 2003 study, the traffic videos were used to manually classify the fleet into seven categories (i.e., motorcycle [MC], private car [PC], taxi, light goods vehicle [LGV], heavy goods vehicle [HGV], light bus, and big buses). Vehicles were counted for every 15-minute video segment of each one or two-hour sampling period. The 2015 study classified traffic videos into nine categories similar to those in 2003, except that HGV was further separated into medium goods vehicle [MGV] and HGV, and big buses were further separated into single and double deck buses. The total vehicle numbers from manual counting in 2015 differed by <4% from hourly vehicle counts from the SMT toll booths.

2.3 EMFAC-HK Emission Estimates

Following the methodology of CARB's EMFAC2002 model, modifications were made in EMFAC-HK to account for the fleet and fuel compositions, emission control regulations, and inspection and maintenance (I/M) programs in Hong Kong (HKEPD, 2020b). Emission factors are calculated from zero mile (when the engine is brand new) emission factors (ZMEFs; based on emission standards) with several adjustment factors, including technology group (i.e., emission control, inspection and maintenance), vehicle age (i.e., deterioration rate; DRs), and vehicle speed as well as ambient T and RH. The HKEPD has been updating ZMEFs, DRs, and speed correction factors using on-board emission measurement by PEMS (Papadopoulos et al., 2020). The types of roadway links (e.g., highways and urban arterial streets) or vehicle driving

modes (e.g., free flowing, stop-and-go, acceleration, or deceleration) are not distinguished in EMFAC-HK. Vehicle start-up emissions are estimated by the number of trips, taking into account the duration of vehicle sitting prior to starting.

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EMFAC-HK assigns the vehicle fleet to 16 classes based on license data from the transportation department (Supplemental Table S2). However, it was challenging to categorize vehicles into all 16 EMFAC-HK classes from the traffic videos. Instead, the seven and nine manual traffic counting classes from the 2003 and 2015 studies, respectively, were assigned to one or more EMFAC-HK vehicle class in Table S2 based on the EMFAC-HK default Vehicle Kilometers Traveled (VKT) proportions in Table S3. As discussed in the Supplementary Materials, when VKT ratios of vehicle classes (e.g., medium vs. heavy-goods vehicle and single vs. double deck buses) were available from tunnel traffic videos, the observed VKT ratios were used to adjust the fleet assignment, assuming that the tunnel-measured vehicle ratios were similar between 2003 and 2015. Figure S2 compares the VKT compositions between territory-average values used in EMFAC-HK and those observed in the SMT, showing similar differences for 2003 and 2015. Compared with the territory-averages, SMT fleets consisted of higher percentages of private cars (PC), heavy-goods vehicles (HGV7), and franchised double deck buses (FBDD), but with lower taxi and HGV8 fractions (see vehicle class description in Table S2). The default breakdowns of vehicles into technology groups, model years, and fuel types by the EMFAC-HK were used, as this information could not be obtained from the tunnel traffic videos. Table S4 illustrates percentages of registered vehicles and VKT by fuel and technologies (i.e., gasoline without catalytic converter, gasoline with catalytic converter, diesel, and LPG) for each vehicle class.

Vehicle emissions were modeled using the "Emfac" mode with EMFAC-HK V4.2. The model generates a matrix of EFs at a range of temperatures (0–40 °C), RH (0–100%), and vehicle speeds (0–130 km hr⁻¹) for each vehicle class-fuel combination (Wang et al., 2019). Running exhaust EF_D (in g veh⁻¹ km⁻¹), i.e., emissions from the vehicle tailpipe while it is travelling on the road, were calculated for methane (CH₄), THC, CO, CO₂, NO₂, NO_x, PM_{2.5}, and PM₁₀. EF_D for NO (expressed as NO₂) was calculated by subtracting NO₂ EF_D from that of NO_x (expressed as NO₂). Additionally, evaporative running loss EFs (in g veh⁻¹ min⁻¹) while the vehicle is operating were calculated for CH₄ and THC. These EFs were organized as lookup tables associated with combinations of T, RH, and vehicle speed values.

Vehicle emissions between the inlet and outlet sampling sites in SMT for each 15-minute vehicle counting period were calculated as follows:

$$Emissions = Emission Factor \times Source Activity$$
 (2)

The emission factors were determined from the lookup table assuming a constant vehicle speed of 80 km hr⁻¹ (speed limit) and average measured ambient T and RH over each period. For running exhaust emissions, the source activity is the VKT (i.e., the number of vehicles in each class multiplied by 0.6 km distance between the inlet and outlet sites). For CH₄ and THC evaporative running losses, the source activity is the average transit time (0.45 min) through the tunnel section times the number of vehicles in each class. Since the EMFAC-HK does not directly output NMHC emissions, NMHC running exhaust emissions were calculated by subtracting CH₄ from THC emissions. The fleet emissions were calculated by summing emissions from all 16 vehicle classes; and the fleet-average EF_D were calculated by dividing the total emissions by tunnel VKT, either for every 15-min vehicle counting period or each one or two-hour integrated sample collection period.

3 Results and Discussion

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3.1 Measured and Modeled Emission Factor Comparisons

Table 1 compares the fleet-average modeled and measured EF_D for CO₂, CO, NMHCs, NO, NO₂, NO_x, and PM_{2.5}. Uncertainties are expressed as the standard errors of multiple measurements from all measurement periods. The Student's t-test p-value < 0.05 indicates statistically significant difference between the pair under comparison. For the 2003 fleet, the EMFAC-HK modeled and measured EF_D for CO₂ agreed within 3% (p = 0.58); with ratios of modeled over measured EF_D of 1.6-1.7 for CO, NO, and NO_x (p < 0.05); 2.3 for NHMC (p < 0.05) 0.05); 0.84 for NO₂ (p = 0.22); and 0.62 for PM_{2.5} (p = 0.19). The 2015 fleet had better agreement: the modeled EF_D for CO₂ and PM_{2.5} differed from the measurement by 3% (p = 0.18), while the modeled EF_D were <30% lower than measurements for CO (p < 0.05), NO (p =0.18), NO₂ (p < 0.05), and NO_x (p = 0.08). The ratio of modeled over measured EF_D for NMHCs was 1.5 (p < 0.05), partially due to different NMHC definitions for the EMFAC-HK and tunnel measurement results. The modeled NMHC was calculated by subtracting CH₄ from THC, where the EF_D was developed using a flame ionization detector calibrated with propane (U.S. EPA, 2005). The SMT-measured EF_D for NMHCs was calculated by summing 66 speciated NMHC species (Cui et al., 2018). The better agreement between modeled and measured EFD for 2015 than 2003 is attributed to EMFAC-HK refinements with more representative EFs and traffic composition information incorporated for the newer model years (HKEPD, 2020b). Different from the <30% lower EF_D for the 2015 SMT fleet in this study, Brimblecombe et al. (2015) found that EMFAC-HK EFs for CO and NO_x were typically 2-3 times lower than those measured from driving through several tunnels in Hong Kong (not including the SMT). Potential causes of these discrepancies include: (1) variations in fleet composition, vehicle operating conditions, and road gradients; (2) assumptions for the cold-start mode, vehicle

loadings, and inter-vehicle variability, etc.; (3) differences in sampled vehicle fleet between fixed-site monitoring and the drive-through method (e.g., small vehicle population causing larger statistical variability); and (4) differences in EF calculations as this study directly measures EF_D while Brimblecombe et al. (2015) measured fuel-based EFs and then converted to EF_D using assumed fuel consumption rates.

The modeled and measured 2015/2003 EF_D ratios (Table 1) were similar for CO_2 and NO_2 . However, the EF_D for CO, NMHCs, NO, and NO_x by the EMFAC-HK showed larger decreases than the tunnel measurement, likely due to higher EF_D values in 2003 by the model. On the other hand, the measured EF_D for $PM_{2.5}$ decreased more than that estimated by the model, probably caused by the lower modeled value in 2003.

Table 2 compares the 2015/2003 ratios of modeled and measured emission rates for the SMT fleet, Hong Kong road transport emission inventory (HKENB, 2020; HKEPD, 2017a), and roadside ambient concentrations acquired from three near-road urban monitoring stations (HKEPD, 2017b). The ratios of emission rates were calculated by multiplying the 2015/2003 emission factor ratios in Table 1 with the corresponding VKT ratio (1.043) observed in the SMT. Because the emission inventory was developed using EMFAC-HK, the ratios between model and inventories are similar. Even though the traffic conditions near the roadside stations differ from those in the SMT (hot-stabilized vehicles mostly free-flowing at ~80 km hr¹), the 2015/2003 ratios of modeled emission rates and roadside concentrations had reasonable agreements for CO, NO, NO₂, and NO_x. The roadside PM_{2.5} concentration reductions are less pronounced than those from the EMFAC-HK estimates and SMT measurements, probably due to non-transportation contributions from ambient background and other sources affecting the roadside stations.

Figure 1 presents time series of modeled and measured EF_D for the 2015 fleet, showing qualitatively similar temporal patterns. However, the measured EF_D exhibits greater variations than the modeled values. This is likely caused by vehicle-to-vehicle EF_D fluctuations that are not represented by the single average EF_D value for each vehicle class under specific environmental and operating conditions assumed by EMFAC-HK. Similar findings are reported in previous studies (Brimblecombe et al., 2015; Lau et al., 2015).

3.2 Vehicle Emission Changes and Control Effectiveness

The Hong Kong government has promulgated aggressive vehicle emission controls since 1990s, including adopting tighter vehicle emission and fuel standards, replacing diesel with LPG fuel for all taxis and a majority (70% in 2015) of public light buses (PLB), and retrofitting older diesel vehicles with advanced aftertreatment emission control devices (HKEPD, 2020a; Lau et al., 2015).

These emission control programs are effective in reducing fleet-average EF_D, especially for NMHC and PM_{2.5}. As compared to 2003, modeled and measured EF_D for NMHCs in 2015 were reduced by 63% and 44%, respectively (Table 1). Hourly vehicle number and the modeled hourly average EF_D for the 16 vehicle classes, three fuels, and fleet average are compared in Figure 2 and tabulated in Table S5. The tunnel fleet VKT for most vehicle classes did not change much over the decade with an overall increase of 4.3% in 2015. The exception was found for LPG-fueled vehicles (taxi and PLB) with a 35% increase in VKT. NMHC emissions decreased for most vehicle classes, resulting in an overall 65% and 72% reduction for gasoline and diesel vehicles, respectively. However, NMHC emissions from LPG vehicles increased by 37%, corresponding to their VKT increases. Speciated NMHC measurements by gas chromatography—mass spectrometry (GC-MS) measurement of canister samples confirm that while most gasoline and diesel related NMHC species decreased from 2003 to 2015 (e.g., 65% reduction in ethane

and propene), emissions of i-butane and n-butane, two LPG markers, increased by 32% and 17%, respectively (Cui et al., 2018; Wang et al., 2018). After replacing worn LPG catalytic converters in taxis and PLBs from October 2013 to April 2014, Cui et al. (2020) reported a 36.7% reduction in total VOCs. LPG vehicle contributions to NMHC would have been larger in 2015 without the implementation of the retrofit.

From 2003 to 2015, modeled and measured EF_D for PM_{2.5} decreased by 70% and 81%, respectively (Table 1). This is consistent with the 43% reduction in roadside concentrations, confirming the effectiveness of PM_{2.5} controls. Figure 2 shows over 35% PM_{2.5} emission reductions across all vehicle classes, with the highest PM_{2.5} reductions found for diesel vehicles (e.g., HGV, non-franchised buses [NFBs], franchised buses [FBs], and PLBs). PM_{2.5} emissions from PLBs were also significantly reduced by switching a large fraction from diesel to LPG.

It is surprising that both model and measurement show minor (3%) reductions in fleet-average EF_D for CO₂, but it is consistent with the road transport emission inventory (Table 2). Due to fuel economy improvements, a larger reduction of EF_D for CO₂ would be expected, irrespective of the 4.3% increase in VKT. Analysis by the Global Fuel Economy Initiative (GFEI, 2016) found that new car fuel economies improved by 2% per year globally between 2005 and 2013. In the U.S., light-duty vehicle fuel economy increased by 30% and CO₂ emissions decreased by 23% from Model Year 2004 to 2018 (U.S. EPA, 2020). However, a growing gap between the CO₂ emission rating and the real-world emissions is observed for European, Japanese, and Chinese vehicles, with on-road emissions being 25–45% higher than official ratings based on laboratory tests (Frey, 2018; Tietge et al., 2017). The 4% reduction in CO₂ EF_D for diesel vehicles was offset by the 4% and 11% increases for gasoline and LPG vehicles, respectively (Table S5). It seems that negligible CO₂ emission reductions due to fuel

economy improvements of newer vehicles is assumed in EMFAC-HK. In addition, aging of vehicles and increased vehicle power and weight may also counteract CO₂ reductions for newer vehicles (Schipper, 2008). As heavy-duty diesel vehicles have higher EF_D for CO₂ (Figure 2), it is important to reduce their emissions; regulations are underway in several countries (GFEI, 2016).

In contrast to the generally decreasing trends, EF_D for CO and NO_x for several vehicle classes increased from 2003 to 2015. For example, EF_D for CO increased by a factor of ~5.7 for PLB, and 3-9 fold for other private light buses (i.e., PV4 and PV5). Although EF_D for NO and NO_x decreased across all vehicles, EF_D for NO₂ increased by more than 70% for several diesel vehicle classes, with a 17% increase in the total diesel fleet (Table S5). The larger modeled CO and NO_x EF_D reductions compared with measurements (Table 1) were probably caused by measurement uncertainties and discrepancies between EMFAC-HK model and SMT conditions, particularly for 2003.

3.3 EMFAC-HK Modeled Emissions by Vehicle Classes and Fuels

Estimation of emissions by vehicle classes and fuels in the EMFAC-HK allows examination of source contributions and emission changes over time, offers insights on the effectiveness of emission controls, and provides information for future controls.

Figure 3 shows the modeled relative hourly average emission rates for CO₂, CO, NMHC (evaporation, tailpipe, and total), NO₂, NO_x, and PM_{2.5} by the nine manual traffic counting vehicle classes. Relative contributions by fuels are summarized in Table 3 and plotted in Figure S3. Distributions of CO₂ emissions among vehicle classes were similar between 2003 and 2015 (Figures 3a-b). Private cars and medium goods vehicles (MGV) were the largest contributors (20–26%) to CO₂ emissions, consistent with their large (~43% and 14–19%, respectively) fleet fractions. Double deck buses (DDB) contributed to ~17% of CO₂ emissions, as they had the

highest CO₂ EF_D among vehicle classes (Figure 2). For different fuel types, Table 3 shows that the largest (69% and 64% in 2003 and 2015, respectively) CO₂ contributions came from diesel-fueled vehicles, attributable to their lower fuel economies. Therefore, implementing fuel economy standards for heavy-duty diesel vehicles is important for reducing the transport sector CO₂ emissions (GFEI, 2016).

The modeled fleet-average CO emission rate decreased by 56% from 2003 to 2015 (Table 2), with large contribution variations among vehicle classes (Figures 3c-d). During this period, contributions from private cars decreased from 49% to 34% mainly because of the reduction of cars without a catalytic converter (from 4.4% to 0.4%) (Table S4) and the corresponding CO EF_D reduction by ~70% (Figure 2). On the other hand, CO contributions from light buses (LB) increased from 1% to 14%, due to conversion of a large fraction of PLBs from diesel to LPG. CO contributions from motorcycles and taxis (15–18%) were also significant. Overall, the gasoline contribution to CO reduced from 67% to 50%, while LPG contribution increased from 18% to 31% (Table 3).

EMFAC-HK assumes NMHC evaporation occurs only for gasoline vehicles, dominated by motorcycles and private cars (Figures 3e-f). While the evaporative NMHC emissions for cars decreased by 76% from 2003 to 2015, emissions from motorcycles were about the same (Table S5), resulting in a two-fold increase in motorcycle contributions from 35% to 70%. Figure 2 and Table S5 show that even though motorcycles only accounted for ~2% of the VKT, their EF_D tailpipe NMHCs were up to 40 times those of other vehicle classes. Therefore, motorcycles were one of the largest contributors (~22–25%) to tailpipe NMHCs emissions (Figures 3g-h). Consistent with higher NMHCs EF_D for LPG than diesel vehicles, tailpipe emissions from light buses increased from 3% in 2003 to 18% in 2015. The sum of emissions from motorcycles and

private cars contributed to over 50% of the total (evaporative + tailpipe) NMHCs emissions (Figures 3i-j). The total NMHC emission rates from LPG vehicles almost doubled (from 8.4 to 15.5 g hr⁻¹) from 2003 to 2015, resulting in increased contributions from 4% to 18% (Table 3). Note that the running evaporative losses were an important fraction (22% – 29%) of total NMHC emissions. While several studies have quantified tailpipe or total NMHC emissions, few studies have systematically quantified evaporative losses for the Hong Kong vehicle fleet (Cui et al., 2018; Ho et al., 2009b; Li et al., 2020b; Ou et al., 2015). In the interest of further reducing ozone and secondary organic aerosol pollution, future tunnel studies should incorporate measurements at different ambient temperatures (e.g., winter and summer), conduct source apportionment to quantify the evaporative emission contributions, and compare measurements with model estimates (Fujita et al., 2012; Ho et al., 2004; Wang et al., 2019).

Diesel vehicles remain as the dominant (>80%) NO_x contributors (Table 3) even though the EMFAC-HK estimated fleet-average NO_x emission rates were reduced by 50% from 2003 to 2015 (Figures 3m-n). Medium goods vehicles (MGVs) and double deck buses (DDBs), contributing to 28–35% and 17–23% of total NO_x emissions, respectively, were the largest NO_x emitters. Other diesel-powered vehicles (i.e., LGV, HGV, and single deck buses) contributed to 3–17% of NO_x emissions.

In contrast to the model estimated ~50% reduction in NO_x emissions, the modeled NO₂ emissions increased by 6% from 2003 to 2015 (Figures 3k-I); the corresponding fleet-average NO₂/NO_x emission ratio increased from 6.7% to 14.4%. This increasing trend is in agreement with roadside measurements in Hong Kong by Tian et al. (2011) that showed primary NO₂/NO_x increased from 2% in 1998 to 13% in 2008. These ratios are lower than those in London (Carslaw and Rhys-Tyler, 2013) and other European countries (Wild et al., 2017) because the

fraction of diesel cars, which have much higher NO₂/NO_x ratios than gasoline cars, is much lower in Hong Kong ($\lesssim 1\%$, as shown in Table S4)) as compared to ~40–50% diesel cars in the London car fleet (Carslaw and Rhys-Tyler, 2013). The tunnel measurements show that the volume ratios of NO₂/NO_x increased from $5.8 \pm 2.7\%$ and $9.5 \pm 2.0\%$ in 2003, to $16.6 \pm 2.2\%$ and $16.3 \pm 1.6\%$ in 2015 at the SMT inlet and outlet sampling sites, respectively (Wang et al., 2018). The 2015 ratios are similar to those observed in an Australian tunnel (15%) (Smit et al., 2017). The measured NO₂ emission rates increased by 16% from 2003 to 2015, ~10% higher than the EMFAC-HK estimate (Table 2). While the NO₂/NO_x emission ratios were similar for gasoline and LPG-fueled vehicles between 2003 and 2015, the ratios increased for several diesel vehicle classes. The increased NO₂ emissions are likely due to more diesel vehicles equipped with oxidation catalysts that convert NO to NO₂ to enhance the performance of selective catalytic reduction (SCR) and diesel particulate filters (Carslaw, 2005; Tian et al., 2011). Diesel vehicles were the dominant PM emitters, accounting for $\geq 95\%$ of PM_{2.5} emissions (Table 3). Figures 3o-p shows that MGVs, HGVs, and DDBs were the largest PM_{2.5} contributors (23–31%). Even though mandatory retrofitting with diesel particulate filters on DDBs had reduced PM emissions (Brimblecombe et al., 2015), DDB remained to be the largest PM contributor (27%) in 2015. EMFAC-HK assumes zero PM emissions from LPG-fueled taxies and PLBs. This assumption is questionable because particle emissions can originate from lubrication oil. PM emissions from LPG vehicles have been identified in several past studies (Cao et al., 2006; Ristovski et al., 2005); however, further studies are needed to quantify LPG vehicle emission factors, especially as the engines and aftertreatment systems deteriorate. The

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distributions of PM₁₀ emissions (not shown) among vehicle classes were similar to those for

PM_{2.5}, as EMFAC-HK currently does not estimate non-tailpipe emissions (including brake wear, tire wear, and resuspended road dust) that are the main contributors to coarse particles.

According to EMFAC-HK, significant emission reductions were achieved for CO, total NMHCs, NO_x, and PM_{2.5} from 2003 to 2015. Table 3 shows that gasoline vehicles continue to be the major contributors to CO and NMHC emissions, with diesel vehicles being the largest contributors to CO₂, NO_x, and PM_{2.5} emissions. Despite reduced NO_x emissions, NO₂ emissions from diesel vehicles have increased. Due to increased VKT, LPG vehicle contributions to CO₂, CO, total NMHCs, and NO_x increased. Future vehicle emission controls should continue targeting these main contributors while keeping reducing emissions from all vehicles.

Motorcycles and LPG-fueled PLB emit disproportionally high CO and NMHC. In 2015, motorcycles accounted for 2.3% of the fleet VKT, but contributed to 15% CO, 70% evaporative NMHC, 25% tailpipe NMHC, 38% total NMHC, and 41% CH₄ emissions (Figure 2 and Table S5). Similarly, LPG-fueled PLB accounted for 1.4% of the VKT, but contributed to 12% CO, 16% tailpipe NMHC, and 11% total NMHC emissions. Together, they accounted for 3.7% of the 2015 tunnel fleet, but contributed to 27% CO and 49% NMHC emissions. These EMFAC-HK estimates require validation with direct emission measurements. If the model is correct, more CO and NMHC controls should be placed on motorcycles and LPG vehicles to reduce ozone pollution, as recent controls have effectively reduced NMHC emissions from solvents and diesel vehicles in Hong Kong (Lyu et al., 2017).

On-board emissions measurements by Lau et al. (2011) attributed 15-18% of total VKT to taxis between 2003 and 2009, higher than the 10-12% (Table S5) in this study. The EMFAC-HK estimates by Lau et al. (2011) attributed 28-32% of CO, 20-30% of THC, and 19-31% of NO_x to taxis, suggesting disproportionally high contributions. These levels are much higher than

the 17-18% CO, 3-5% NMHC, and 6-12% NO_x found in this study (Figure 3). Given the large percentage (over 20%) of fleet VKT in Hong Kong (Table S3), emissions from taxis warrant further investigation.

4 Conclusions and Discussion

The present study compares vehicle emissions modeled by EMFAC-HK with those measured from the Shing Mun Tunnel (SMT) fleets in 2003 and 2015. Studying the same tunnel over a decade apart elucidates changes in vehicle emissions and allows an assessment of the effectiveness of emission control policies during this period. It also enables evaluation of the EMFAC-HK model for older and newer fleets under the tunnel operating conditions. The detailed estimates of emissions by vehicle classes and fuels in the EMFAC-HK provide insights into causes of emission changes and offer guidance on priorities for future controls. Significant findings are:

- (1) EMFAC-HK modeled EF_D differed from SMT measurements by <50% for most pollutants emitted by the 2015 fleet, in line with other models reviewed by Smit et al. (2010). The differences were larger for the 2003 fleet: ratios of modeled over measured EF_D were 1.6–2.3 times for CO, NO_x, and NMHCs, and 0.62 for PM_{2.5}. The exception was for CO₂, agreeing within 3%.
- (2) Vehicle emission controls in Hong Kong effectively reduced NMHC and PM_{2.5} emissions, as is evident from ~40–80% reduction from 2003 to 2015 in EMFAC-HK estimates, SMT measurements, emission inventory, and roadside ambient concentrations.
- (3) CO₂ emissions has not decreased from 2003 to 2015 as expected from the increased vehicle fuel economy. While the actual causes warrant further investigation, more

stringent fuel economy standards for heavy-duty vehicles would accelerate the vehicular CO₂ emission reduction.

- (4) While tunnel measurements did not show as much reduction as the EMFAC-HK model or roadside concentrations for CO, NO, NO_x, both model and measurement show increased primary NO₂ emissions from 2003 to 2015, likely due to the increased diesel oxidation catalysts in diesel vehicles.
- (5) Gasoline vehicles are the major contributors to CO and NMHC emissions, while diesel vehicles are the largest contributors to CO₂, NO_x, and PM_{2.5} emissions. Future vehicle emission controls should continue targeting these main contributors while keeping reducing emissions from all vehicle classes.
- (6) Motorcycles and LPG fueled public light buses (PLB) were disproportionally high CO and NMHC emitters, deserving additional emission controls. Motorcycles accounted for 2.3% of the SMT fleet, but contributed 15% of CO, 38% of total NMHC, and 41% of CH4 emissions. LPG-fueled PLB accounted for 1.4% of the fleet, but contributed to 12% CO and 11% total NMHC emissions. Direct emission measurements with these vehicles are needed to validate the model estimates.
- (7) NMHC running evaporative losses contributed to 20-30% of total NMHC emissions, according to the EMFAC-HK. While several studies have quantified tailpipe NMHC emissions, evaporative losses have not been systematically studied for the Hong Kong fleet. Future tunnel studies should incorporate measurements at different ambient temperatures (e.g., winter and summer), conduct source apportionment to quantify the evaporative emission contribution, and compare measurements with model estimates.

Both tunnel measurements and EMFAC-HK model findings are applicable to the specific conditions during the SMT study: the fleet had ~45% gasoline, 13% LPG, and 42% diesel vehicles that drove at ~80 km h⁻¹ speed under hot-stabilized conditions; the road was nearly flat with a 1.054% uphill gradient; and ambient temperature and RH ranged 20–25 °C and 60–70%, respectively. As a consequence, generic statements cannot be made about the accuracy of the EMFAC-HK based on this study and caution should be exerted when applying current findings to other conditions. Other studies (e.g., measurement in other tunnels with different traffic conditions, cold start emission characterization in parking garages, on-board measurement, driving-through or plume chasing, and remote sensing) are needed to further evaluate model performance. One major limitation of the EMFAC-HK and other emission models is the use of a single average emission factor for a specific condition, without considering the large variability of traffic emissions. Future model development should consider incorporating uncertainties in emission calculations (Smit et al., 2010). Furthermore, emission models should be evaluated over time as vehicle emissions, model input, and measurement technologies continue to change.

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Author Contributions

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- 472 Review & Editing, and Funding acquisition. Lung-Wen Antony Chen: Software, Data curation,
- 473 Visualization, Writing Review & Editing. **Kin-Fai Ho**: Conceptualization, Methodology, Data

- 474 Curation, Writing - Review & Editing. Chi Sing Chan: Methodology, Data Curation, Writing -
- 475 Review & Editing. Zhuozhi Zhang: Methodology, Data Curation, Writing - Review & Editing.
- 476 Shun-Cheng Lee: Conceptualization, Methodology, Resources, Writing - Review & Editing,
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- 478 Editing, and Funding acquisition. John G. Watson: Conceptualization, Methodology,
- 479 Resources, Writing - Review & Editing.

Declaration of Competing Interest 480

481 None.

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References

- Brimblecombe, P., Townsend, T., Lau, C.F., Rakowska, A., Chan, T.L., Močnik, G., Ning, Z. 483 484 (2015). "Through-tunnel estimates of vehicle fleet emission factors." Atmos. Environ. 123, 485 Part A:180-189.
- 486 Cao, J.J., Lee, S.C., Ho, K.F., Fung, K., Chow, J.C., Watson, J.G. (2006). "Characterization of 487 roadside fine particulate carbon and its eight fractions in Hong Kong." Aerosol Air Qual. Res. 488 6 (2):106-122.
- 489 CARB, (2018). "EMFAC2017 Volume I – User's Guide V1.0.2." Mobile Source Analysis Branch, 490 Air Quality Planning & Science Division, California Air Resource Board, Sacramento, CA. 491 Accessed on 3/20/2018. https://www.arb.ca.gov/msei/downloads/emfac2017-volume-i-users-492 guide.pdf
- 493 Carslaw, D.C. (2005). "Evidence of an increasing NO₂/NO_x emissions ratio from road traffic 494 emissions." Atmos. Environ. 39 (26):4793-4802.
- 495 Carslaw, D.C., Rhys-Tyler, G. (2013). "New insights from comprehensive on-road measurements 496 of NO_x, NO₂ and NH₃ from vehicle emission remote sensing in London, UK." Atmos. Environ. 497 81:339-347.
- 498 Che, W., Frey, H.C., Fung, J.C.H., Ning, Z., Qu, H., Lo, H.K., Chen, L., Wong, T.-W., Wong, 499 M.K.M., Lee, O.C.W., Carruthers, D., Cheung, F., Chan, J.W.M., Yeung, D.W., Fung, Y.H.,
- 500 Zhang, X., Stocker, J., Hood, C., Hohenberger, T.L., Leung, K.W., Louie, P.Y.K., Li, A.T.Y.,
- 501 Sun, L., Wei, P., Li, Z., Zhang, Y., Wang, M., Shen, Q., Huang, W., Lee, E., Patwary, A., Lei,
- 502 X., Cheng, S., Shakhaoat Hossain, M., Tang, K.T.J., Lao, X., Leung, R., Chan, D., Li, Y., Yuan,
- Z., Lau, A.K.H. (2020). "PRAISE-HK: A personalized real-time air quality informatics system 503
- 504 for citizen participation in exposure and health risk management." Sustainable Cities and 505 Society 54:101986.
- 506 Cheng, Y., Lee, S.C., Ho, K.F., Louie, P.K.K. (2006). "On-road particulate matter (PM_{2.5}) and
- 507 gaseous emissions in the Shing Mun Tunnel, Hong Kong." Atmos. Environ. 40 (23):4235-
- 508 4245.

- Cheng, Y., Lee, S.C., Ho, K.F., Chow, J.C., Watson, J.G., Louie, P.K.K., Cao, J.J., Hai, X. (2010).
 "Chemically-speciated on-road PM_{2.5} motor vehicle emission factors in Hong Kong." *Sci. Total Environ.* 408 (7):1621-1627.
- Cui, L., Wang, X.L., Ho, K.F., Gao, Y., Liu, C., Ho, S.S., Li, H., Lee, S.C., Wang, X., Jiang, B.,
 Huang, Y., Chow, J.C., Watson, J.G., Chen, L.-W.A. (2018). "Decrease of VOC emissions
 from vehicular emissions in Hong Kong from 2003 to 2015: results from a tunnel study."
 Atmos. Environ. 177:64-74.
- Cui, L., Li, H.W., Huang, Y., Zhang, Z., Lee, S.C., Blake, D.R., Wang, X.M., Ho, K.F., Cao, J.J.
 (2020). "The characteristics and sources of roadside VOCs in Hong Kong: Effect of the LPG catalytic converter replacement programme." *Sci. Total Environ*.:143811.
- 519 El-Fadel, M., Hashisho, Z. (2001). "Vehicular Emissions in Roadway Tunnels: A Critical Review." *Critical Reviews in Environmental Science and Technology* 31 (2):125-174.
- Franco, V., Kousoulidou, M., Muntean, M., Ntziachristos, L., Hausberger, S., Dilara, P. (2013).
 "Road vehicle emission factors development: A review." *Atmos. Environ.* 70:84-97.
- Frey, H.C. (2018). "Trends in onroad transportation energy and emissions." *J. Air Waste Manage*. *Assoc.* 68 (6):514-563.
- Fujita, E.M., Campbell, D.E., Zielinska, B., Chow, J.C., Lindhjem, C.E., DenBleyker, A., Bishop,
 G.A., Schuchmann, B.G., Stedman, D.H., Lawson, D.R. (2012). "Comparison of the
 MOVES2010a, MOBILE6.2 and EMFAC2007 mobile source emissions models with on-road
 traffic tunnel and remote sensing measurements." *J. Air Waste Manage. Assoc.* 62 (10):1134 1149.
- GFEI, (2016). "Fuel Economy State of the World 2016." Global Fuel Economy Initiative (GFEI),
 London, United Kingdom. Accessed on 11/23/2020.
 https://www.globalfueleconomy.org/media/203446/gfei-state-of-the-world-report-2016.pdf
 - HEI, (2010). "Traffic-Related Air Pollution: A Critical Review of the Literature on Emissions, Exposure, and Health Effects. HEI Special Report 17." Health Effects Institute Panel on the Health Effects of Traffic-Related Air Pollution, Boston MA. Accessed on 8/5/2014. http://pubs.healtheffects.org/getfile.php?u=553
 - HKENB, (2020). "Greenhouse Gas Emissions in Hong Kong by Sector." Environment Bureau, the Government of the Hong Kong Special Administrative Region Accessed on July 17, 2020. https://www.climateready.gov.hk/files/pdf/greenhouse_gas_emissions_by_sector.pdf
 - HKEPD, (2017a). "Hong Kong Air Pollutant Emission Inventory." Environmental Protection Department, The Government of the Hong Kong Special Administrative Region, Hong Kong. Accessed on 4/10/2017. http://www.epd.gov.hk/epd/english/environmentinhk/air/data/emission inve.html
- 544 HKEPD, (2017b). "Hong Kong Air Quality Monitoring Data." Environmental Protection 545 Department, The Government of the Hong Kong Special Administrative Region, Hong Kong. 546 Accessed on 3/10/2017. http://epic.epd.gov.hk/EPICDI/air/station/?lang=en
- 547 HKEPD, (2020a). "Cleaning the Air at Street Level." Hong Kong Environmental Protection
 548 Department. Accessed on 7/22/2020.
 549 https://www.epd.gov.hk/epd/english/environmentinhk/air/prob_solutions/cleaning_air_atroad
 550 html

554 HK%20%28Ver%204.2%29.pdf

533

534

535536

537

538

539

540541

542

- HKTD, (2016). "The Annual traffic Census 2015." TTSD Publication No. 16CAB1, Hong Kong
 Transport Department, Traffic and Transport Survey Division.
 https://www.td.gov.hk/en/publications and press releases/publications/free publications/the
 annual traffic census 2015/index.html
- Ho, K.F., Lee, S.C., Guo, H., Tsai, W.Y. (2004). "Seasonal and diurnal variations of volatile organic compounds (VOCs) in the atmosphere of Hong Kong." *Sci. Total Environ.* 322 (1):155-166.
- Ho, K.F., Ho, S.S.H., Cheng, Y., Lee, S.C., Zhen Yu, J. (2007). "Real-world emission factors of fifteen carbonyl compounds measured in a Hong Kong tunnel." *Atmos. Environ.* 41 (8):1747-1758.
- Ho, K.F., Ho, S.S.H., Lee, S.C., Cheng, Y., Chow, J.C., Watson, J.G., Louie, P.K.K., Tian, L. (2009a). "Emissions of gas- and particle-phase polycyclic aromatic hydrocarbons (PAHs) in the Shing Mun Tunnel, Hong Kong." *Atmos. Environ.* 43 (40):6343-6351.
- Ho, K.F., Lee, S.C., Ho, W.K., Blake, D.R., Cheng, Y., Li, Y.S., Ho, S.S.H., Fung, K., Louie,
 P.K.K., Park, D. (2009b). "Vehicular emission of volatile organic compounds (VOCs) from a tunnel study in Hong Kong." *Atmos. Chem. Phys.* 9 (19):7491-7504.
- Krzyzanowski, M., Kuna-Dibbert, B., Schneider, J. (2005). "Health effects of transport-related air pollution". WHO Regional Office Europe, Copenhagen, Denmark.
- Lau, C.F., Rakowska, A., Townsend, T., Brimblecombe, P., Chan, T.L., Yam, Y.S., Močnik, G., Ning, Z. (2015). "Evaluation of diesel fleet emissions and control policies from plume chasing measurements of on-road vehicles." *Atmos. Environ.* 122:171-182.
- Lau, J., Hung, W.T., Cheung, C.S. (2011). "On-board gaseous emissions of LPG taxis and estimation of taxi fleet emissions." *Sci. Total Environ.* 409 (24):5292-5300.

580

- Leung, K.W., (2019). "Development and assessment of high-resolution vehicle emission inventory in Hong Kong." M.Phil. Thesis, The Hong Kong University of Science and Technology.
 - Li, Z., Yim, S.H.-L., Ho, K.-F. (2020a). "High temporal resolution prediction of street-level PM2.5 and NOx concentrations using machine learning approach." *J. Cleaner Prod.* 268:121975.
- Li, Z., Ho, K.-F., Yim, S.H.L. (2020b). "Source apportionment of hourly-resolved ambient volatile organic compounds: Influence of temporal resolution." *Sci. Total Environ.* 725:138243.
- Lyu, X.P., Zeng, L.W., Guo, H., Simpson, I.J., Ling, Z.H., Wang, Y., Murray, F., Louie, P.K.K.,
 Saunders, S.M., Lam, S.H.M., Blake, D.R. (2017). "Evaluation of the effectiveness of air pollution control measures in Hong Kong." *Environ. Pollut.* 220:87-94.
- 587 Marinello, S., Lolli, F., Gamberini, R. (2020). "Roadway tunnels: A critical review of air pollutant concentrations and vehicular emissions." *Transportation Research Part D: Transport and Environment* 86:102478.
- Niu, X., Chuang, H.-C., Wang, X.L., Ho, S.S.H., Li, L., Qu, L., Chow, J.C., Watson, J.G., Sun, J.,
 Lee, S., Cao, J., Ho, K.F. (2020). "Cytotoxicity of PM2.5 vehicular emissions in the Shing
 Mun Tunnel, Hong Kong." *Environ. Pollut.* 263:114386.
- NRC (2000). "Modeling Mobile-Source Emissions". Transportation Research Board, National Research Council, The National Academies Press, Washington, DC.
- Ntziachristos, L., Gkatzoflias, D., Kouridis, C., Samaras, Z. (2009). "COPERT: A European Road Transport Emission Inventory Model." Information Technologies in Environmental Engineering: Berlin, Heidelberg, May 28-29, 2009.
- 598 Ou, J., Guo, H., Zheng, J., Cheung, K., Louie, P.K.K., Ling, Z., Wang, D. (2015). "Concentrations 599 and sources of non-methane hydrocarbons (NMHCs) from 2005 to 2013 in Hong Kong: A 600 multi-year real-time data analysis." *Atmos. Environ.* 103:196-206.

- Papadopoulos, G., Ntziachristos, L., Tziourtzioumis, C., Keramydas, C., Lo, T.-S., Ng, K.-L., Wong, H.-L.A., Wong, C.K.-L. (2020). "Real-world gaseous and particulate emissions from Euro IV to VI medium duty diesel trucks." *Sci. Total Environ.* 731:139137.
- Pierson, W.R., Gertler, A.W., Robinson, N.F., Sagebiel, J.C., Zielinska, B., Bishop, G.A., Stedman,
 D.H., Zweidinger, R.B., Ray, W.D. (1996). "Real-world automotive emissions Summary of
 studies in the Fort McHenry and Tuscarora mountain tunnels." *Atmos. Environ.* 30 (12):2233 2256.
- Ristovski, Z.D., Jayaratne, E.R., Morawska, L., Ayoko, G.A., Lim, M. (2005). "Particle and carbon dioxide emissions from passenger vehicles operating on unleaded petrol and LPG fuel." *Sci. Total Environ.* 345 (1-3):93-98.
- Schipper, L. (2008). "Automobile fuel; economy and CO2 emissions in industrialized countries: troubling trends through 2005/6." Transportation Research Board 87th Annual Meeting: Washington DC, United States, 2008-1-13 to 2008-1-17.
- Smit, R., Ntziachristos, L., Boulter, P. (2010). "Validation of road vehicle and traffic emission models A review and meta-analysis." *Atmos. Environ.* 44 (25):2943-2953.
- Smit, R., Kingston, P., Wainwright, D.H., Tooker, R. (2017). "A tunnel study to validate motor vehicle emission prediction software in Australia." *Atmos. Environ.* 151:188-199.
- Tian, L., Hossain, S., Lin, H., Ho, K., Lee, S., Yu, I.S. (2011). "Increasing trend of primary NO₂ exhaust emission fraction in Hong Kong." *Environ. Geochem. Health* 33 (6):623-630.
- Tietge, U., Diaz, S., Yang, Z., Mock, P., (2017). "From laboratory to road international: A comparison of official and real-world fuel consumption and CO2 values for passenger cars in Europe, the United States, China, and Japan." International Council on Clean Transportation, Berlin, Germany. Accessed on 11/25/2020. https://theicct.org/sites/default/files/publications/Lab-to-road-intl_ICCT-white-paper 06112017 vF.pdf
- U.S. EPA, (2005). "Conversion Factors for Hydrocarbon Emission Components" EPA420-R-05-015, Assessment and Standards Division, Office of Transportation and Air Quality, U.S. Environmental Protection Agency, Ann Arbor, Michigan. Accessed on 11/23/2020.
 https://19january2017snapshot.epa.gov/www3/otaq/models/nonrdmdl/nonrdmdl2005/420r05
 015.pdf
- U.S. EPA, (2018). "MOVES2014b: Latest Version of MOtor Vehicle Emission Simulator
 (MOVES)." U.S. Environmental Protection Agency, Ann Arbor, MI. Accessed on July 16,
 2020. https://www.epa.gov/moves
- U.S. EPA, (2020). "The 2019 EPA Automotive Trends Report Greenhouse Gas Emissions, Fuel Economy, and Technology since 1975." EPA-420-R-20-006, U.S. Environmental Protection Agency, Ann Arbor, MI. Accessed on 11/18/2020. https://nepis.epa.gov/Exe/ZyPDF.cgi?Dockey=P100YVFS.pdf
- Wang, X.L., Ho, K.-F., Chow, J.C., Kohl, S.D., Chan, C.S., Cui, L., Lee, S.-c.F., Chen, L.-W.A.,
 Ho, S.S.H., Cheng, Y., Watson, J.G. (2018). "Hong Kong vehicle emission changes from 2003 to 2015 in the Shing Mun Tunnel." *Aerosol Sci. Technol.* 52 (10):1085-1098
- Wang, X.L., Khlystov, A., Ho, K.F., Campbell, D., Chow, J.C., Kohl, S.D., Watson, J.G., Lee,
 S.C., Chen, L.-W.A., Lu, M., Ho, S.S.H., (2019). "Real-World Vehicle Emissions
 Characterization for the Shing Mun Tunnel in Hong Kong and Fort McHenry Tunnel in the
 United States." Research Report 199, Health Effects Institute, Boston, MA.
- https://www.healtheffects.org/system/files/WangRR199.pdf

- Wild, R.J., Dubé, W.P., Aikin, K.C., Eilerman, S.J., Neuman, J.A., Peischl, J., Ryerson, T.B.,
 Brown, S.S. (2017). "On-road measurements of vehicle NO₂/NO_x emission ratios in Denver,
 Colorado, USA." *Atmos. Environ.* 148:182-189.
- Xing, Y., Brimblecombe, P. (2018). "Dispersion of traffic derived air pollutants into urban parks."
 Sci. Total Environ. 622-623:576-583.
- Xing, Y., Brimblecombe, P., Ning, Z. (2019). "Fine-scale spatial structure of air pollutant concentrations along bus routes." *Sci. Total Environ.* 658:1-7.

Figure Captions

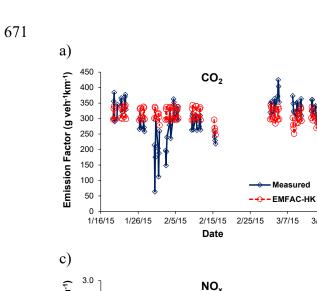
- Figure 1. Time series of EMFAC-HK estimates and measured emission factors (EF_D) during the 2015 Shing Mun Tunnel (SMT) 2-hour sampling periods.
- 657
 658 Figure 3
- Figure 2. Average hourly vehicle counts in the SMT and emission factors by vehicle class estimated by EMFAC-HK V4.2 for the 2003 and 2015 study periods. See Supplemental Table S2 for description of vehicle classes. The relative standard errors of emission factors from all
- measurement periods were <1%.
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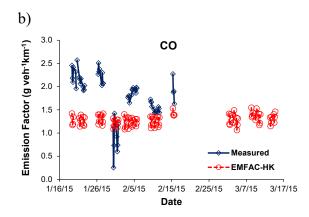
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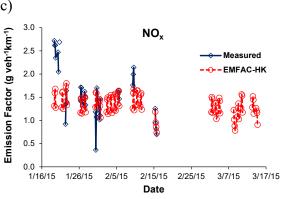
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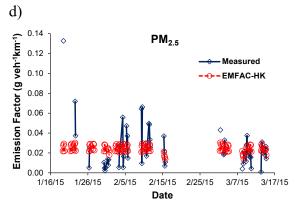
- Figure 3. EMFAC-HK estimations of SMT hourly average emission for CO₂, CO, evaporative NMHC, tailpipe NMHC, total NMHC, NO₂, NO_x, and PM_{2.5} during the 2003 (left) and 2015 (right) measurement periods by the nine vehicle classes counted from traffic videos. The total pie areas of the 2015 plots were scaled to those of 2003 plots based on emission rates to visualize fleet-average emission changes. (MC=motorcycle; LGV=light goods vehicle; MGV=medium goods
- average emission changes. (MC=motorcycle; LGV=light goods vehicle; MGV=medium goods vehicle; HGV=heavy goods vehicle; LB=light bus; SDB=single deck bus; DBB=double deck bus.)
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3/17/15



672 Figure 1.

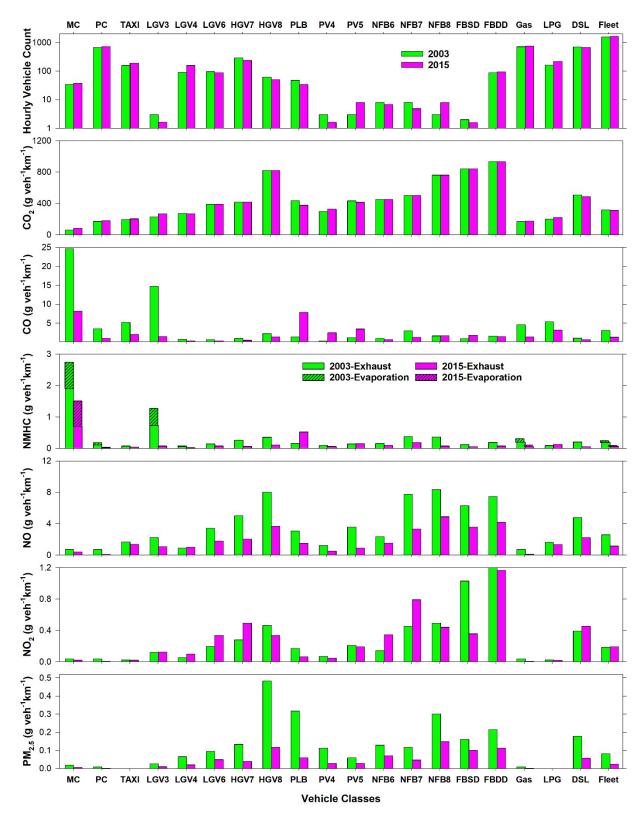
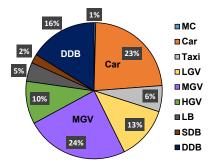
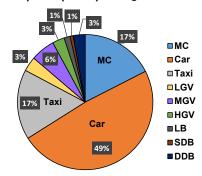


Figure 2.

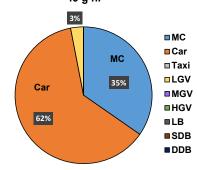
a) CO₂ (2003): 303 kg hr⁻¹



c) CO (2003): 2.9 kg hr-1



e) Evaporative NMHC (2003): 49 g hr⁻¹



g) Tailpipe NMHC (2003): 179 g hr-1

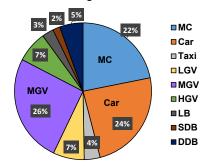
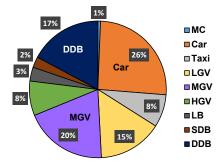


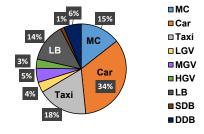
Figure 3 (color print).

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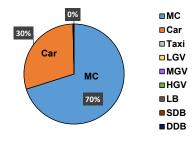
b) CO₂ (2015): 305 kg hr⁻¹



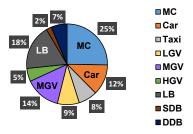
d) CO (2015): 1.3 kg hr-1



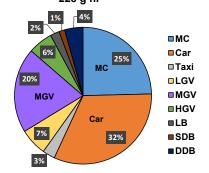
f) Evaporative NMHC (2015): 26 g hr⁻¹



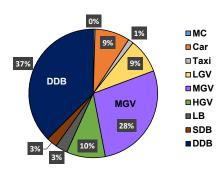
h) Tailpipe NMHC (2015): 62 g hr⁻¹



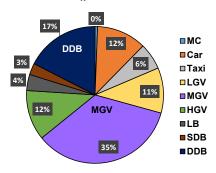
i) Total NMHC (2003): 228 g hr⁻¹



k) NO₂ (2003): 0.176 kg hr⁻¹



m) NO_x (2003): 2.6 kg hr⁻¹



o) PM_{2.5} (2003): 77 g hr⁻¹

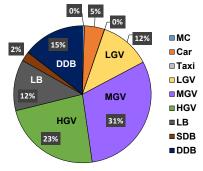
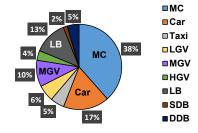
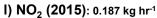
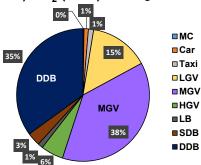


Figure 3 continued.

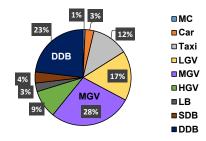
j) Total NMHC (2015): 88 g hr⁻¹



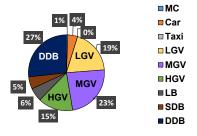




n) NO_x (2015): 1.3 kg hr⁻¹



p) PM_{2.5} (2015): 24 g hr⁻¹



680 Tables

Table 1. Comparison of the 2003 and 2015 fleet-average EMFAC-HK modeled and measured emission factors (EF_D; average \pm standard error) in the Shing Mun Tunnel (SMT). The p-values for Student's t-test for comparison pairs are listed in parentheses.

Species	Emission Factors (EF _D ; in g veh-1 km-1)				Ratios				
	2003		2015		Modeled : Measured		2015 : 2003		
	Modeled	Measured	Modeled	Measured	2003 (p-value)	2015 (p-value)	Modeled (p-value)	Measured (p-value)	
CO_2	320 ± 5	310 ± 17	311 ± 2	302 ± 6	1.03 (0.58)	1.03 (0.18)	0.97 (0.09)	0.97 (0.65)	
CO	3.07 ± 0.08	1.88 ± 0.11	1.29 ± 0.01	1.80 ± 0.13	1.63 (0.00)	0.72 (0.00)	0.42 (0.00)	0.95 (0.61)	
NMHCs	0.239 ± 0.003	0.106 ± 0.002	0.088 ± 0.001	0.059 ± 0.002	2.26 (0.00)	1.50 (0.00)	0.37 (0.00)	0.56 (0.00)	
NO (as NO ₂)	2.586 ± 0.093	1.501 ± 0.120	1.136 ± 0.023	1.332 ± 0.119	1.72 (0.00)	0.85 (0.18)	0.44 (0.00)	0.89 (0.38)	
NO_2	0.185 ± 0.006	$0.221 {\pm}~0.030$	0.192 ± 0.004	0.245 ± 0.023	0.84 (0.22)	0.78 (0.02)	1.04 (0.34)	1.11 (0.51)	
NO _x (as NO ₂)	2.771 ± 0.093	1.722 ± 0.130	1.328 ± 0.023	1.577 ± 0.141	1.61 (0.00)	0.84 (0.08)	0.48 (0.00)	0.92 (0.44)	
$PM_{2.5}$	0.082 ± 0.003	0.131 ± 0.037	0.024 ± 0.000	0.025 ± 0.003	0.62 (0.19)	0.97 (0.79)	0.30 (0.00)	0.19 (0.01)	

Table 2. Comparison of 2015/2003 ratios of modeled and measured emission rates in SMT, emission inventory of the road transport sector, and ambient concentrations monitored at roadside stations in Hong Kong.

Data Source	2015-to-2003 Ratio							
Data Source	CO_2	CO	NMHC	NO	NO_2	NO_x	SO_2	PM _{2.5}
Emission rates from EMFAC-HK model	1.01	0.44	0.39	0.45	1.06	0.49	NA	0.31
Emission rates from SMT measurement ^a	1.02	1.00	0.61	1.06	1.16	0.96	0.24	0.20
Road transport emission inventory	0.97	0.57	0.63	NA^{b}	NA^b	0.43	0.04	0.23
Roadside concentration	NA^b	0.62	NA^b	0.51	1.04	0.62	0.48	0.57

^aEmission rates were calculated by multiplying the 2015/2003 emission factor ratios in Table 1 with the 2015/2003

VKT ratio (1.043); note that the territory-average VKT ratio was slightly higher (1.19).

^bNA = not available.

Table 3. Relative contributions of SMT emissions by diesel, LPG, and gasoline vehicles in 2003 and 2015 as estimated by EMFAC-HK.

g :	Relative	Contribu	tion in 2003	Relative Contribution in 2015			
Species	Diesel	LPG	Gasoline	Diesel	LPG	Gasoline	
CO_2	69%	7%	24%	64%	10%	26%	
CO	15%	18%	67%	19%	31%	50%	
Evaporative NMHC	0%	0%	100%	0%	0%	100%	
Tailpipe NMHC	48%	5%	47%	37%	25%	38%	
Total NMHC	38%	4%	58%	26%	18%	56%	
NO_2	89%	1%	9%	98%	1%	1%	
NO_x	81%	6%	13%	83%	14%	3%	
$PM_{2.5}$	95%	0%	5%	96%	0%	4%	