Evaluating in-use vehicle emissions using air quality monitoring stations and on-road remote sensing systems

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Abstract

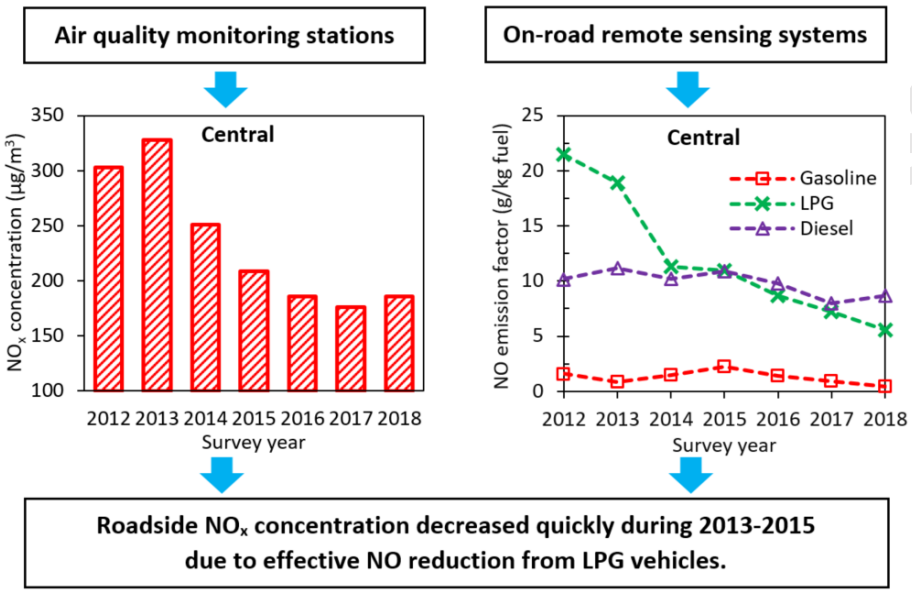
This study investigated real-world in-use vehicle emissions using two regulatory techniques simultaneously, namely on-road remote sensing (RS) systems and air quality (AQ) monitoring stations, aiming to provide a full pollution profile from tailpipe to roadside and atmosphere. Two large AQ and RS datasets collected during 2012-2018 were analysed. The effects of various emission control programmes on the trends of tailpipe emissions and air quality were evaluated. Correlations between tailpipe emissions and roadside and ambient air quality were also explored. The results showed a decreasing trend of NO_2 at both roadside and ambient AQ stations from 2013 to 2016, which was attributed to the intensive implementation of a series of vehicle emissions control programmes. Although NO_2 was decreasing, O_3 was generally increasing for all AQ stations. AQ data showed that O_3 had little correlation with either NO_2 or NO_x, but was mainly determined by NO_2/NO_x ratio. Roadside NO_2/NO_x ratio increased first and then decreased or stabilised after 2014, while ambient NO_2/NO_x ratio increased steadily. RS data showed that the overall NO decreased quickly during 2012-2015 and then decreased moderately after 2015. The decrease was mainly attributed to the effective NO reduction from LPG vehicles. However, diesel NO remained high and reduced relatively slowly during the studied period. Gasoline vehicles were relatively clean compared with LPG and diesel vehicles. Finally, good correlations were demonstrated between NO measured by RS sites and NO_x measured by roadside AQ stations, indicating that vehicle emissions were the major contributor to roadside NO_x pollution. Ambient NO_x emissions could be affected by various sources, leading to different correlations between RS and ambient AQ results.

Keywords: Real driving emissions; Air pollution; Vehicle emission control programmes; Tailpipe, roadside and ambient; Remote sensing

Highlights

- Real driving emissions were measured by both RS systems and AQ stations
- O_3 generally increased in spite of decreased NO_2 at both roadside and ambient AQ stations
- NO_x reduction was mainly attributed to emission control programmes on LPG vehicles
- On-road RS emissions data showed a good correlation with roadside AQ data

Graphical abstract



1. Introduction

Air pollution continues to be a major health hazard to the public. The World Health Organisation (WHO, 2020) estimated that ambient air pollution caused 4.2 million deaths per year globally. The criteria air pollutants include carbon monoxide (CO), lead (Pb), nitrogen dioxide (NO_2), ozone (O_3), particulate matter (PM) and sulfur dioxide (SO_2). Of great importance is NO_2 as an air pollutant, which has not yet well complied with the air quality standards globally (Grange *et al.*, 2017; Xu *et al.*, 2019). In addition, NO_2 and nitric oxide (NO), collectively known as nitrogen oxides (NO_x), are key precursors to the formation of secondary air pollutants of O_3 and PM (Anenberg *et al.*, 2017; Matthaios *et al.*, 2019) which are also major air pollution problems in many cities. Although many sources have contributed to the air pollution problem, road transport is widely believed to be the single largest source of atmospheric pollutants.

Real driving emissions (RDE) have attracted wide attention in recent years after the discovery of Volkswagen scandal which referred to the defeat devices installed on Volkswagen light-duty diesel vehicles that aimed to pass the certification tests in laboratory but emitted tens of times higher NO_x emissions in real-world driving (Anenberg *et al.*, 2017; Brand, 2016; Li *et al.*, 2018; Oldenkamp *et al.*, 2016; Schiermeier, 2015; Tanaka *et al.*, 2018; Zachariadis, 2016). However, it should be noted the problem of higher RDE than laboratory test is not only limited to Volkswagen diesel NO_x emissions, but also applies to other vehicle manufacturers (Schiermeier, 2015; Tanaka *et al.*, 2018), fuel types (e.g. gasoline, hybrid and CNG powered vehicles) (Duarte *et al.*, 2016; Huang *et al.*, 2019b; Rašić *et al.*, 2017) and emission species (e.g. CO and CO_2) (Fontaras *et al.*, 2017; Rašić *et al.*, 2017; Weiss *et al.*, 2011). To understand RDE characteristics, a variety of emission measurement techniques have been developed, including portable emissions measurement system (PEMS) (Vlachos *et al.*, 2014), plume chasing (Lau *et al.*, 2015), mobile monitoring platform (Kelp *et al.*, 2020), tunnel measurement (Song *et al.*, 2018), ambient measurement (Ke *et al.*, 2013) and on-road remote sensing (Huang *et al.*, 2018b). Each of these measurement techniques have their respective advantages and limitations that thus determine their applications. Among them, PEMS is a standard RDE test method with the highest accuracy. However, PEMS is only applicable for a small number of vehicles such as type approval tests due to its long setup time and high cost. On the other hand, ambient measurement and on-road remote sensing techniques are suitable and cost-effective for measuring RDE of a large number of vehicles and are well developed and widely adopted.

The ambient technique measures the pollutant mass or volume concentrations (i.e. $\mu g/m^3$ or ppm) at monitoring sites located in suitable proximity to the roads or other sources of interest. The pollutant concentrations data can be used for accessing vehicle emission trends and emission control programmes with little/simple treatment (Carvalho *et al.*, 2015; Matthaios *et al.*, 2019; Mavroidis and Ilia, 2012; Santos *et al.*, 2019) or after being processed by emission models (Carslaw and Ropkins, 2012; Chan *et al.*, 2011; Grange *et al.*, 2017; Ke *et al.*, 2013). A readily available data source is the air quality (AQ) monitoring stations which are widely employed in many cities as part of their air quality management. The main advantage is that all criteria air pollutants are being continuously measured on a long-term basis with little human supervision. However, ambient measurement technique cannot determine emissions of specific vehicle classes or individual vehicles due to the nature of indirect measurements. Besides, it measures non-vehicle emissions such as industry and household emissions alongside vehicle emissions.

On-road remote sensing (RS) technique measures tailpipe emissions, speed, acceleration and the license plate number of a vehicle in half a second when it passes by a measurement site. These data can be very useful for various applications, such as development of emission factors (Guo *et al.*, 2007; Ning and Chan, 2007), evaluation of vehicle emission control technologies and programmes (Bishop and Haugen, 2018; Chen and Borken-Kleefeld, 2016; Huang *et al.*, 2018c; Pujadas *et al.*, 2017), and identification of excessively-emitting vehicles for inspection and maintenance (I&M) programmes (HKEPD, 2020; Organ *et al.*, 2019). The major advantages of RS are that a large number of vehicles can be measured at a relatively low cost and the emissions data can be resolved for both individual vehicles and specific vehicle classes. Compared with AQ monitoring stations, however, the disadvantages of RS are that only limited emission species are measured (mostly CO_2, CO, HC and NO) and data is available for limited time periods (mostly work hours in dry weather) due to the requirement of constant human supervision (e.g. on-site calibration every two hours). Measurements are not conducted in servere wet weather including typhoons because rain droplets and tyre splatter will affect the RS accuracy. However, RS measurements are immune to haze because they are differential measurements with correction for the background pollution level. More details about the RS measurement mechanisms can be found in Huang *et al.* (2018b).

Previous studies have usually used either AQ or RS techniques in isolation, whereas this study investigates vehicle emissions by using the two techniques simultaneously, aiming to present a full picture from tailpipe to roadside and ambient emissions. The effects of various emission control programmes on the trends of vehicle emissions and air quality are also evaluated. Finally, this study analyses how RS tailpipe emissions correlate with the AQ data from neighbouring roadside and ambient monitoring stations, providing direct evidence on the contribution of vehicle emissions to air pollution.

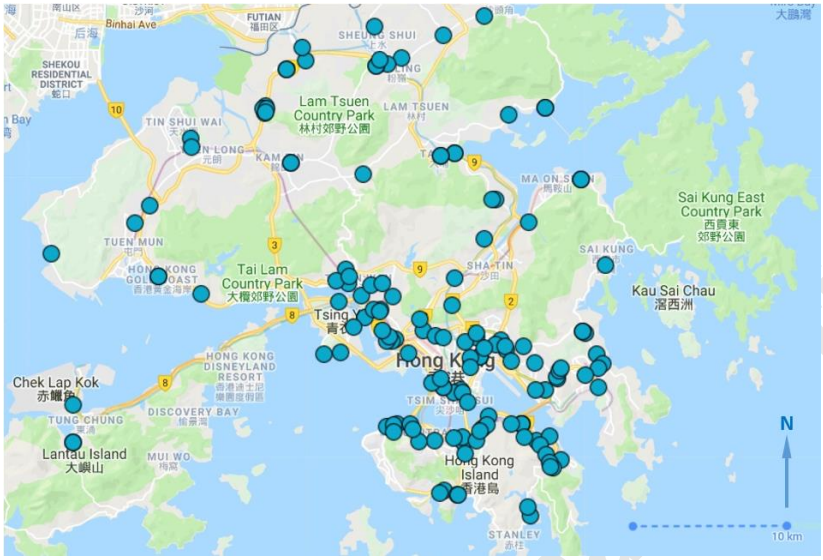
2. Methods

2.1. On-road RS systems and AQ monitoring stations

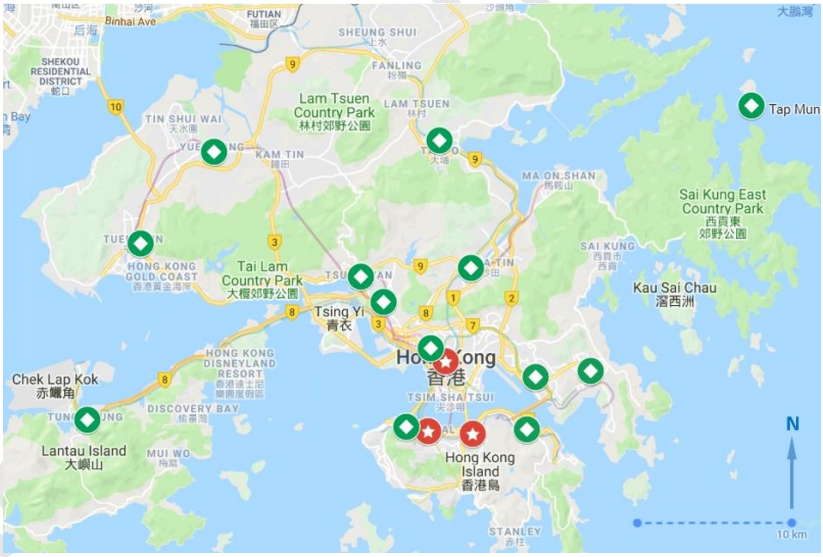
The Hong Kong Environmental Protection Department (HKEPD) is currently using two regulatory monitoring techniques for managing air quality, namely on-road RS systems and AQ monitoring stations:

An RS system consists of infrared (IR) and ultraviolet (UV) beam sources and detectors, speed and acceleration sensors, a retroreflector and a vehicle plate camera. The IR and UV beams are placed at the average height of vehicle tailpipes across a single-lane road and a measurement is triggered by the beams being blocked by a passing vehicle. CO, CO_2 and HC emissions are measured in the IR region and NO emissions are measured in the UV region. Since the effective plume path length and the amount of plume measured are influenced by a number of factors such as wind, turbulence, engine size and exhaust pipe height, RS can only determine the relative concentration ratios of pollutants over CO_2 (i.e. CO/CO_2, HC/CO_2 and NO/CO_2) (Huang *et al.*, 2020). Meanwhile, the speed, acceleration and an image of the license plate of the passing vehicle are measured by the RS system. The license plate number provides registration information of the passing vehicle, such as the maker, manufacture year, engine size, fuel type and emission standard. The RS data is being used by the HKEPD to detect gasoline and LPG excessively-emitting vehicles

for enforcement since 1 Sep 2014 and an enforcement program for diesel gross-emitters is under research and development (Huang *et al.*, 2018b). There are over 150 candidate RS sites across Hong Kong (**Fig. 1a**) and the HKEPD chooses 2-3 sites out of them for RS measurements each day from Monday to Friday if weather conditions permit. The RS emission sensors are calibrated every two hours using span gases to ensure data quality.



(a)



(b)

RS site **Ambient AQ station** **Roadside AQ station**

Fig. 1. Locations of on-road RS sites (a) and AQ monitoring stations (b).

The AQ monitoring programme uses 3 roadside stations and 13 ambient stations distributed across Hong Kong (**Fig. 1b**). Roadside AQ stations are placed at 3.0-4.5 meters above ground and ambient AQ stations are 11.0-28.0 meters above ground. These 16 fixed AQ stations continuously measure the regulated air pollutants in each hour, including CO, NO_2, NO_x, O_3, SO_2, and respirable and fine suspended particulates (RSP and FSP). The pollutant concentrations are determined continuously by automatic analysers. Among them, NO, NO_2 and NO_x are measured by

Chemiluminescence method using T-API 200A/T200 or TECO 42i instruments, and O_3 is measured by UV absorption method using T-API 400/400A/T400 instruments (HKEPD, 2017). The accuracy and precision of the emission analysers are ensured by following the international requirements such as the United States Environmental Protection Agency. Regular performance audits showed that the accuracy of AQ monitoring stations varied from -6.6 % to 6.7 % (95% probability limits) for gaseous pollutants, which was well within the performance goal of ±20%. In addition, regular precision checks showed that the precision (a measure of the repeatability) of AQ monitoring stations varied from -5.4% to 5.3% (95% probability limits), which was also within the goal of ±15%.

2.2. Selection of RS sites and AQ stations

In this study, suitable RS-AQ pairs are chosen for the investigation to explore the possible correlations between tailpipe emissions and air pollution. The 16 AQ monitoring stations (**Fig. 1b**) are in operation continuously throughout the year. However, there are only 2-3 RS measurement sites in operation per weekday with dry weather. These 2-3 RS sites are chosen from the candidate sites (**Fig. 1a**) by the HKEPD according to the enforcement needs. Therefore, although the total number of RS emission records collected per year is large, the number of records per RS site would be small and vary significantly among RS sites. To make the analysis statistically valid, the following two criteria are used to select the AQ stations and RS sites.

- An RS site is close to an AQ station (within 1 km);
- There are sufficient emission records at each RS site (>30000 records).

Table 1. Details of the selected AQ stations and RS sites.

Pair No.	Location of AQ station	Type of AQ station	Code of RS site	Distance between AQ station and RS site
1	Central, Hong Kong Island	Roadside	H0001	568 m
2	Causeway Bay, Hong Kong Island	Roadside	H0008	512 m
3	Tseung Kwan O, New Territories	Ambient	N0075	733 m
4	Eastern, Hong Kong Island	Ambient	H0019	645 m
5	Kwun Tong, Kowloon	Ambient	K0007	987 m
6	Kwai Chung, New Territories	Ambient	N00025	619 m

Therefore, six RS-AQ pairs are chosen for this study, including two roadside and four ambient AQ stations. **Table 1** shows the details of the six RS-AQ pairs and **Fig. S1** shows the locations of each AQ-RS pair on the map. Generally, the distances between the ambient AQ-RS pairs are farther than those between the roadside AQ-RS pairs. The AQ and RS emissions data collected during 2012-2018 is used for analysis. AQ monitoring stations report emission concentrations hourly. The selected six AQ stations collected 330331 emission records during 2012-2018. Meanwhile, RS programme was operated in a total of 221 days at the six selected RS sites during 2012-2018. The

RS dataset contains a total number of 506591 valid measurements, including petrol, diesel and LPG vehicles. The number of valid emission records and fleet compositions at each RS site are shown in **Table S1**. Central and Causeway Bay are the most frequently used RS sites due to the high traffic volumes and serious air pollution problems there.

3. Results and Discussion

3.1. Emission trends measured by AQ stations

NO_2 is a major regulated air pollutant that has adverse health and environmental effects and often exceeds the standards in many cities globally. **Fig. 2** shows the annual average NO_2 concentrations measured at the roadside and ambient AQ stations. Although with slight fluctuations, NO_2 concentrations show a decreasing trend for all the roadside and ambient stations during 2012-2018. Particularly, a faster NO_2 decrease was observed during 2013-2016. This would be the result of a series of vehicle emissions control programmes implemented during this period, including a free replacement programme of three-way catalysts (TWCs) and oxygen sensors for gasoline and LPG taxis and light buses (2013-2014), an RS enforcement programme for excessively-emitting gasoline and LPG vehicles (since 1 September 2014), a subsidised retirement programme for pre-Euro IV diesel commercial vehicles (since 1 Mar 2014), and the setup of three franchised bus low emission zones (LEZs) in Causeway Bay, Central and Mong Kok (since 31 December 2015) where the roadside AQ stations are (Hong Kong SAR Government, 2019). As compared with Causeway Bay, Central is affected more by the emissions of LPG and gasoline vehicles (**Table S1**). Therefore, greater NO_2 reductions in Central than Causeway Bay were observed (**Fig. 2**) after the implementation of RS enforcement programme for LPG and gasoline gross emitters in 2014.

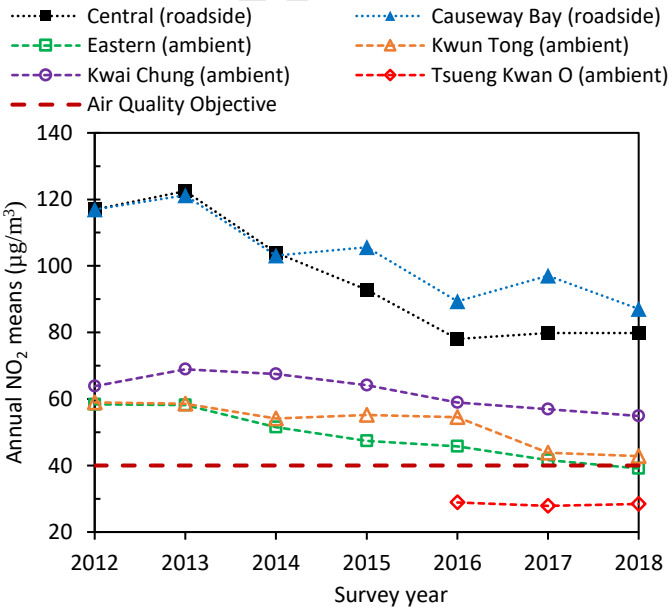


Fig. 2. Annual average NO_2 concentrations measured at roadside and ambient AQ stations.

NO_2 concentrations at roadside AQ stations are obviously higher than those measured at ambient AQ stations, due to their proximity to the emissions source, i.e. motor vehicles. The dashed line in **Fig. 2** indicates the Air Quality Objective (AQO) set by the Hong Kong government, which

is a maximum annual average of 40 μg/m^3 for NO_2. Despite the decreasing trend, roadside NO_2 concentrations are still significantly over the AQO, which were 99% and 118% higher than AQO for Central and Causeway Bay stations in 2018, respectively. Ambient NO_2 concentrations are gradually approaching the AQO, with Eastern station achieved AQO in 2018, while Kwun Tong and Kwai Chung stations were still 7% and 37% over AQO in 2018.

Although air quality regulations set limits for NO_2, automotive emission standards only have limits for total NO_x ($NO+NO_2$) but do not have specific limits for individual gases. This could lead to a counter-intuitive situation that while total NO_x emissions decreased over time, roadside NO_2 did not, as that observed in Europe (Carslaw *et al.*, 2011; Grange *et al.*, 2017). Therefore, NO_2/NO_x ratio is an important parameter that bridges the air quality and automotive emission standards. **Fig. 3** shows the annual NO_2/NO_x ratio measured by the roadside and ambient AQ stations. Eastern station had no NO_x measurement throughout the study period and thus NO_2/NO_x ratio was not available. As shown in **Fig. 3**, for roadside AQ stations, NO_2/NO_x ratio at Causeway Bay increases during 2012-2014 and then decreases after 2014, and NO_2/NO_x ratio at Central increases during 2012-2015 and becomes stable after 2015. For ambient AQ stations, NO_2/NO_x ratios increase throughout 2012-2018. This increasing trend is concerning as while vehicle manufacturers are reporting lower total NO_x emissions with their new models, the actual roadside and ambient NO_2 concentrations would not decrease at the same pace, or may even increase.

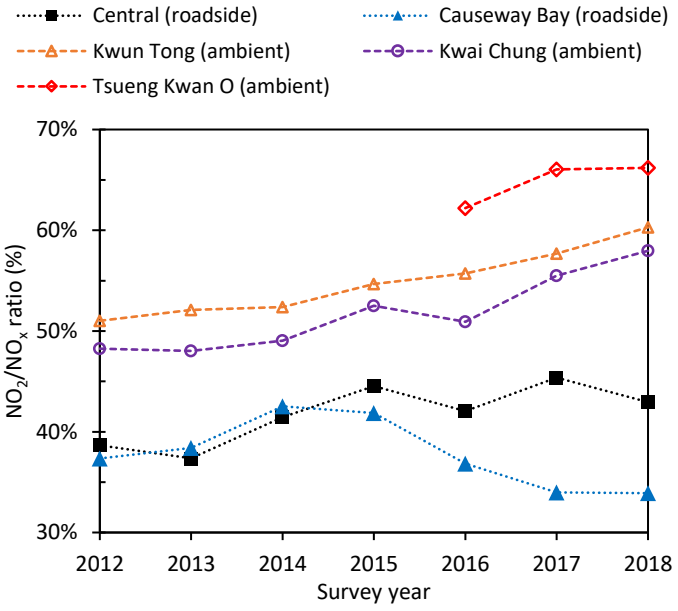


Fig. 3. Annual average NO_2/NO_x ratios measured at roadside and ambient AQ stations.

The increasing trend of NO_2/NO_x ratio would be caused by the change in engine after-treatment technologies. NO_x emissions are mainly formed in fuel combustion via a thermal NO (known as Zeldovich) mechanism under high-temperature oxygen-rich conditions. The majority of NO_x emissions (~90%) from uncontrolled diesel engines are emitted as NO while the primary NO_2 emissions in the engine exhaust only account for a small percentage (Gentner and Xiong, 2017). However, the adoption of exhaust after-treatment technologies such as diesel oxidation catalysts (DOCs) and diesel particulate filters (DPFs) can lead to an increase of primary NO_2 from engine exhaust. DOCs oxidise CO and HC emissions into harmless CO_2 and H_2O, and in the meantime

also oxidise NO into NO_2 (Huang *et al.*, 2019a). DPFs remove PM emissions and need to burn off the accumulated PM regularly via a regeneration process. NO_2 based regeneration is more popular than O_2 based regeneration, due to its low regeneration temperature and simple structure with DOCs (E *et al.*, 2016; Jiao *et al.*, 2017). However, the side effect of DPF regeneration is the increased NO_2 emissions as well as higher NO_x and NO_2/NO_x ratio (Ko *et al.*, 2019; Liu *et al.*, 2012). DOCs were mainly introduced to achieve the Euro 3 standards and DPFs were introduced for Euro 5 standards (Grange *et al.*, 2017). In Hong Kong, Euro 3/III and 5/V standard vehicles started entering the fleet in 2001-2003 and 2010-2013, respectively, depending on the vehicle classes (Huang *et al.*, 2018a). The increasing trends of NO_2/NO_x ratio after 2012 observed in **Fig. 3** could be well explained by the introduction of Euro 5/V vehicles in 2010-2013 which adopted DOC+DPF with NO_2-assisted regeneration for exhaust after-treatment. Moreover, as discussed later, NO_x titration (equation (3)) could also play an important role in the increase of NO_2/NO_x ratio. As shown in **Fig. 4**, O_3 concentrations were increasing during 2012-2018, which enhanced the conversion of NO to NO_2 and consequently increased the NO_2/NO_x ratio in ambient air.

Fig. 3 also shows that NO_2/NO_x ratios at roadside AQ stations are significantly lower compared with ambient AQ stations. This is because ambient AQ stations have longer reaction time and higher ambient O_3 level for NO_x titration. Modern diesel vehicles still emit the majority of NO_x emissions as NO even with DOC and DPF after-treatment devices. Laboratory chassis dynamometer experiments (Park *et al.*, 2019) showed that NO_2/NO_x ratios were 21%-28% and 12%-16% for Euro 5 and 6 diesel vehicles, respectively. As it disperses from vehicle tailpipes to roadside and ambient stations, NO is oxidised into NO_2 by O_3. Consequently, NO_2/NO_x ratio gradually increases from tailpipes to roadside and ambient AQ stations. Furthermore, ambient AQ stations have higher O_3 concentrations than roadside stations (**Fig. 4**), leading to stronger NO_x titration and consequently higher NO_2/NO_x ratios.

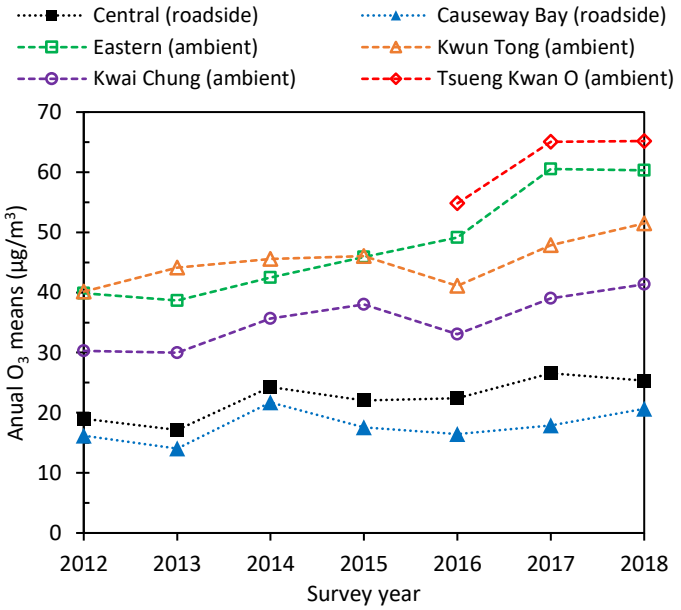


Fig. 4. Annual average O_3 concentrations measured at roadside and ambient AQ stations.

O_3 is another major regulated air pollutant that has not yet met the AQO which has a limit of 160 µg/m^3 for 8-h average and the allowed number of exceedances is nine per year. In 2018,

roadside AQ stations achieved the AQO while ambient AQ stations recorded a maximum number of exceedances of 20 (HKEPD, 2019) which was over double the AQO. **Fig. 4** shows the annual average O_3 concentrations during 2012-2018. Although with fluctuations, O_3 concentrations generally increase for both roadside and ambient stations from 2012 to 2018, despite the decreasing trend of NO_2 concentrations (the main precursor of O_3 formation) observed in **Fig. 2**. Similar trends were reported in eastern China where witnessed a rapid drop of NO_2 during 2012-2016 while O_3 was steadily deteriorating (Wang *et al.*, 2019). The opposite trends of O_3 and NO_2 were attributed to the change of O_3 formation from volatile organic compounds- (VOCs-) sensitive regime to mix-sensitive regime due to NO_x reductions (Wang *et al.*, 2019).

It is well known that tropospheric O_3 emission is a secondary pollutant formed through photochemical reactions of gaseous precursors (mainly NO_x and VOCs) in the presence of sunlight (Liu *et al.*, 2019; Wang *et al.*, 2019). The simple NO_x-O_3 chemistry can be described by equations (1-3), where *hv* represents a solar photon and M denotes a third body molecule that absorbs excess energy so that O and O_2 may recombine to form O_3 (Zhong *et al.*, 2016). The alkylperoxyl (RO_2) and hydroperoxyl (HO_2) from VOCs and CO lead to the oxidization of NO to NO_2, and thus promote the formation of O_3 (Wang *et al.*, 2019). The complexity of O_3 is linked to the lack of linearity in its formation pathways, as NO_x emissions are involved in both the formation (by NO_2 via equations (1-2)) and removal (by NO via equation (3)) of O_3 (Ripoll *et al.*, 2019).

$$NO_2 + h\nu \rightarrow NO + O \quad (1)$$

$$O + O_2 + M \rightarrow O_3 + M \quad (2)$$

$$O_3 + NO \rightarrow NO_2 + O_2 \quad (3)$$

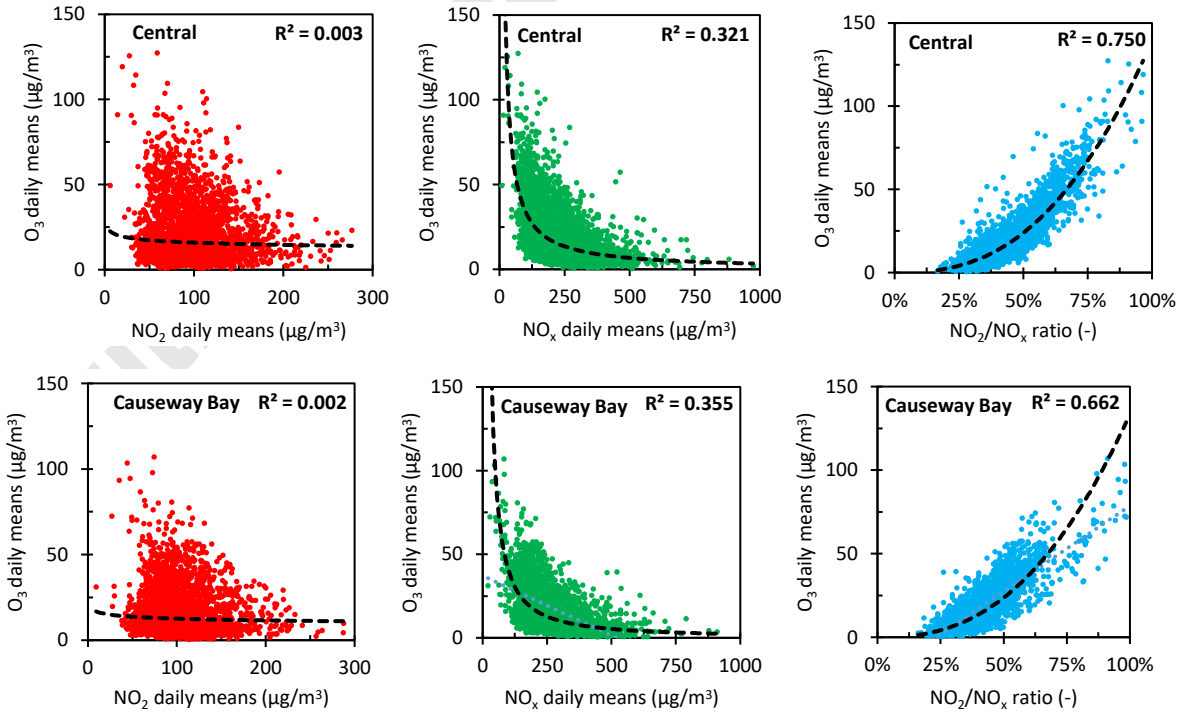


Fig. 5. Plots of daily mean concentrations of O_3 against NO_2, NO_x and NO_2/NO_x ratio at roadside AQ stations.

To explore the correlations between O_3 and NO_x emissions, **Figs 5** and **6** plot the daily mean concentrations of O_3 against NO_2, NO_x and NO_2/NO_x ratio for roadside and ambient AQ stations, respectively. Power trend lines are used to fit the plots as they produce the best overall R^2 values. As shown in **Figs 5** and **6**, O_3 shows little correlation with neither NO_2 nor NO_x at both roadside and ambient AQ stations, with R^2 values well below 0.10 for O_3 *vs* NO_2 and 0.50 for O_3 *vs* NO_x. Instead, strong correlations (R^2 values above 0.60) have been observed between O_3 and the relative ratio of NO_2 over NO_x (i.e. NO_2/NO_x ratio). The larger the NO_2/NO_x ratio, the higher the O_3 will be. This is because NO_2 promotes the formation of O_3 while NO reduces the formation of O_3. As a result, the net formation of O_3 increases with the NO_2/NO_x ratio.

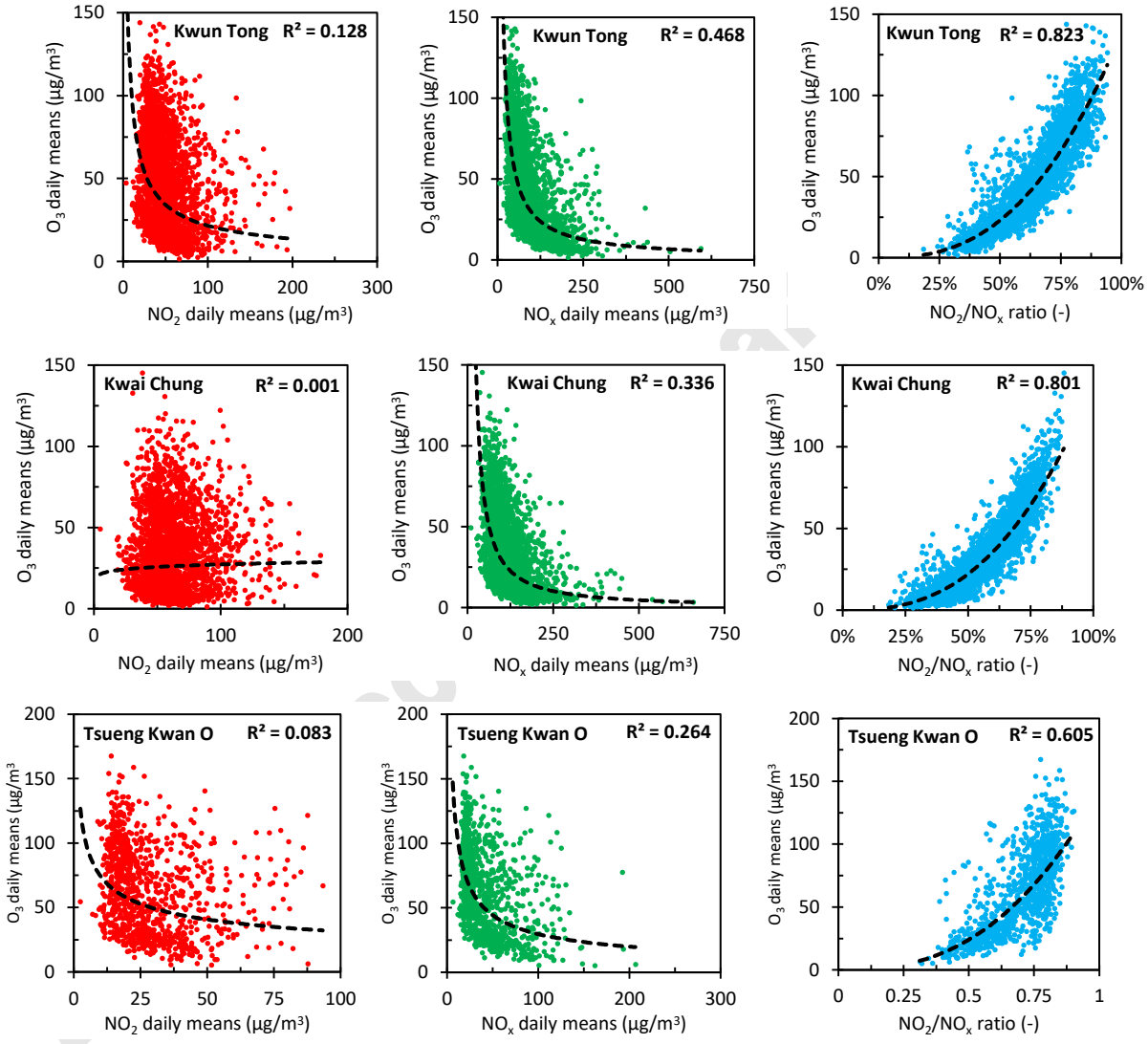


Fig. 6. Plots of daily mean concentrations of O_3 against NO_2, NO_x and NO_2/NO_x ratio at ambient AQ stations.

3.2. Emission trends measured by RS sites

Fig. 7 shows the average NO emission factors of all vehicles measured by on-road RS sites, including gasoline, LPG and diesel vehicles. Fuel based emission factors of NO (EF_{NO}) are calculated from the emission ratios measured by RS based on the principle of carbon balance using

equation (4), in which 30 is the molecular weight of NO (g/mol) and 0.014 is the molecular weight of diesel (kg/mol) assuming CH_2 for diesel.

$$EF_{NO} = \frac{30}{0.014} * \frac{NO/CO_2}{1+CO/CO_2+6*HC/CO_2} \quad [g/kg\ fuel] \qquad (4)$$

As shown in **Fig. 7**, NO emission factors decrease quickly during 2012-2015 and then decrease moderately after 2015. These emission trends measured by RS are in line with the emission trends observed at the roadside and ambient AQ stations (**Fig. 2**). The faster decrease of NO during 2012-2015 could be attributed to the intensive implementation of several vehicle emission control programmes during the same period, including the free replacement programme of TWCs and oxygen sensors for gasoline and LPG taxis and light buses (2013-2014), the RS enforcement programme for excessively-emitting gasoline and LPG vehicles (since 1 September 2014), the subsidised retirement programme for pre-Euro IV diesel commercial vehicles (since 1 March 2014), and the setup of franchised bus LEZs (since 31 December 2015), as discussed in section 3.1. **Fig. 7** also shows that different RS sites have very different NO emission levels before 2015, but then they all converge to a similar level after 2015. This is because the stringent vehicle emission control programmes have led to effective emission reductions at all RS sites across the city. Similar emission level convergence was also reported for different diesel vehicle types in an earlier study (Huang *et al.*, 2018c).

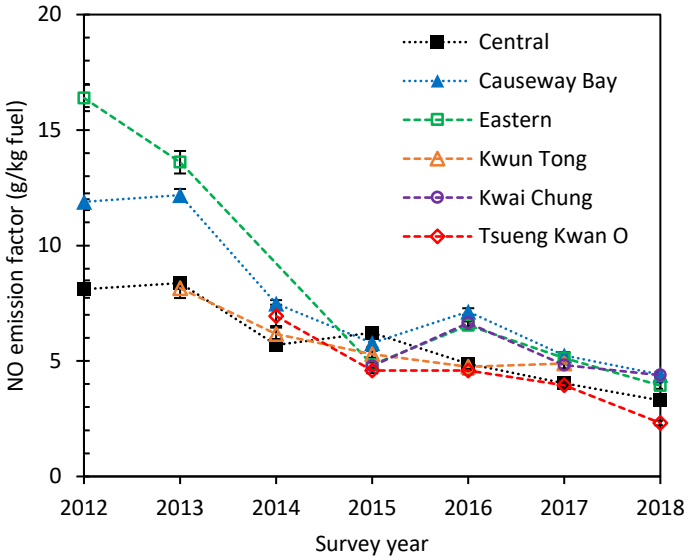


Fig. 7. Average NO emission factors of all vehicles measured at on-road RS sites. Error bars represent the 95% confidence interval over the mean.

Although RS measures fewer gaseous emission species at lower accuracy than AQ stations, a major advantage is that RS records the vehicle license plate number with each emission measurement, which can be used to identify the individual vehicle information such as its fuel type, make and manufacture year. This enables an analysis of the emission trends of individual fuel type fleets at each RS site to be performed. **Fig. 8** shows the breakdown of average NO emission factors by fuel types of gasoline, LPG and diesel measured at the six studied RS sites. As shown in **Fig. 8**, gasoline vehicles emit much lower NO emissions compared with LPG and diesel vehicles. This is mainly because gasoline vehicles are much less intensively used and are better maintained than

LPG and diesel vehicles. In Hong Kong, LPG vehicles are only used as taxis and light buses, and diesel vehicles are mostly used as goods vehicles and buses. These vehicles can accumulate a high mileage each year and thus their emission control systems deteriorate quickly. On the other hand, gasoline vehicles are mostly used as private cars which are moderately used and are usually well maintained.

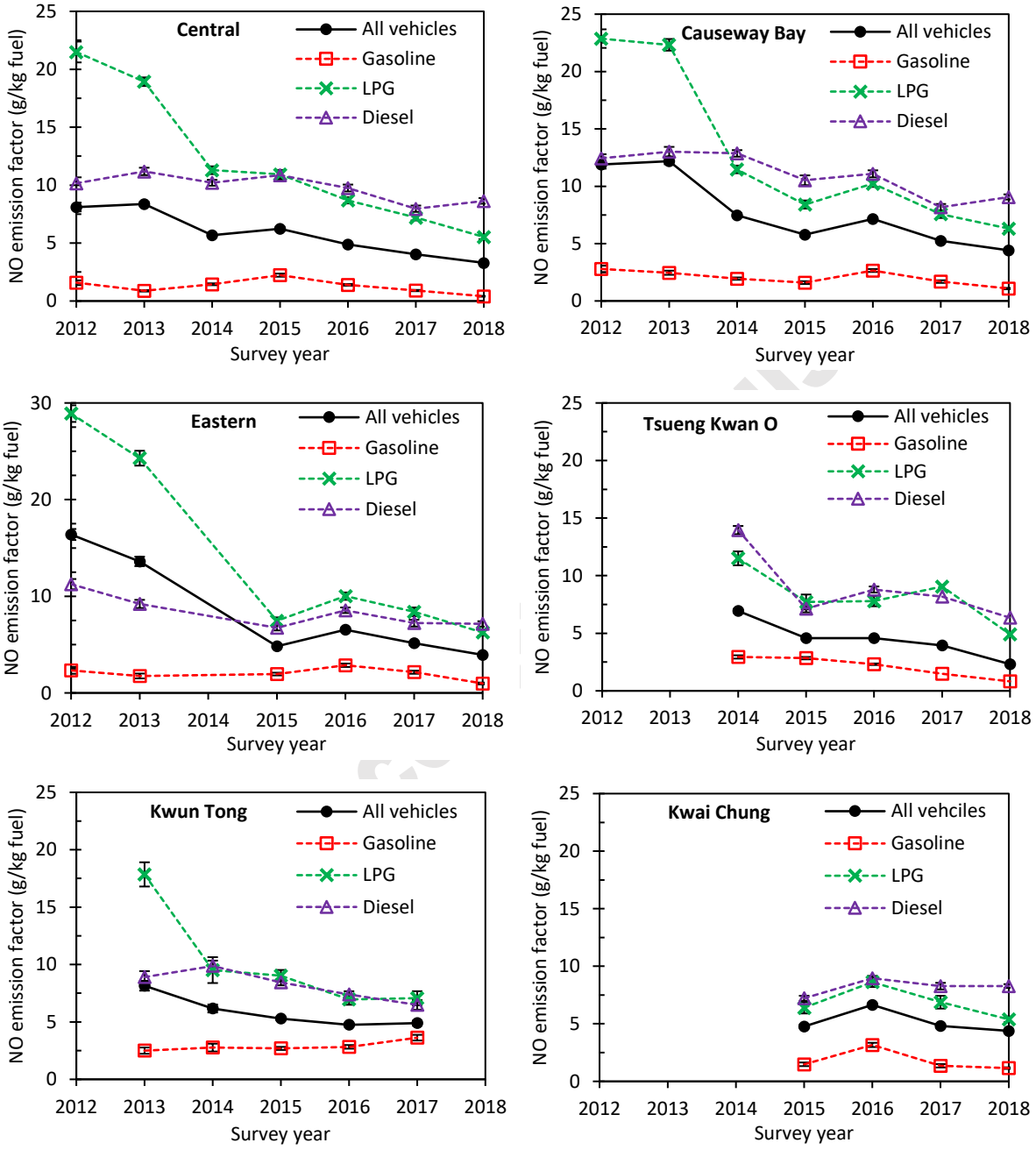


Fig. 8. Breakdown of average NO emission factors by vehicle fuel type measured at on-road RS sites. Error bars represent the 95% confidence interval over the mean.

In particular, LPG vehicles show a significant NO reduction between 2013 and 2015 at all RS sites (**Fig. 8**). This can be attributed to the free TWC and oxygen sensor replacement programme for LPG vehicles. Before introducing the RS gross-emitter enforcement programme on 1 September 2014, there was a one-off subsidy to taxi and light bus owners for replacing the TWCs and oxygen sensors. The free replacement programme was from August 2013 to April 2014, and

received 13942 taxis and 2881 light buses, reaching an overall participation rate of ~80% (Hong Kong SAR Government, 2018). The following introduction of the RS enforcement programme on 1 September 2014 also greatly reduced the number of excessively-emitting vehicles and facilitated the LPG fleet renewal rate (Organ *et al.*, 2018), contributing to lower NO emissions.

On the other hand, relatively small NO reduction is observed in **Fig. 8** for diesel and gasoline vehicles during 2012-2018. Gasoline vehicles are already relatively clean compared with diesel and LPG vehicles and further large reductions are not expected. However, diesel vehicles emit consistently high emissions throughout 2012-2018 although there were two emission control programs targeting diesel vehicles, i.e. retrofitting 1030 eligible Euro II and III diesel franchised buses with SCR (2014–2017) and phasing out pre-Euro IV diesel commercial vehicles (2014–2020). Two factors might have caused this trend. Firstly, there is increasing evidence reporting that the diesel RDE NO_x are significantly higher than laboratory test results and the standard limits, and are not reducing effectively with Euro standards (Degraeuwe and Weiss, 2017; Fu *et al.*, 2013; Kousoulidou *et al.*, 2013; Pujadas *et al.*, 2017; Schiermeier, 2015; Weiss *et al.*, 2011), despite the increasingly stringent automotive emission standards. Secondly, the two diesel emission control programs did not have high coverage rates as those for the LPG fleet (~80% for free TWC and oxygen sensors replacement programme and ~100% for RS gross-emitters enforcement programme).

3.3. Correlation between AQ stations and RS sites

It is widely believed that vehicle emissions are the major source of air pollutants in urban areas. With a large number of RS measurements, the average emissions represent roadside emissions and their contribution to urban air pollution. Therefore, it is hypothesized that the RS vehicle emissions data would agree well with the AQ monitoring stations data. **Fig. 9** shows the correlation between annual average NO emission factors measured by RS sites and NO_x concentrations measured by the roadside and ambient AQ stations. Comparisons at Eastern and Tseung Kwan O ambient AQ stations are not presented because of lack of NO_x data or insufficient data points.

As shown in **Fig. 9**, higher NO emission levels measured at RS sites are well correlated with higher NO_x emission levels measured at both roadside AQ stations (Central and Causeway Bay which are commercial and tourist areas with no major NO_x emission source other than vehicles). This indicates that vehicle emissions are the major contributor to roadside NO_x pollution. For ambient AQ stations, RS NO correlates well with AQ NO_x at Kwun Tong (R^2 = 0.518), but not at Kwai Chung (R^2 = 0.235). This implies that ambient NO_x pollution is more complex and is affected by multiple sources. Generally, roadside AQ stations show better correlations (0.549-0.853) with RS sites than ambient AQ stations (0.235-0.518). This is because the percentages of LPG vehicles at Kwun Tong and Kwai Chung are much smaller compared with roadside AQ stations (**Table S1**), indicating that vehicle emissions have less impact on AQ measurements at ambient stations. Relatively, emissions from other sources such as inter-regional pollutants transport and commercial and industrial sectors become significant, which are measured by AQ stations but not by RS sites. Besides, the ambient RS-AQ pairs are further apart (**Fig. S1**) and have fewer RS records (**Table S1**). Consequently, RS and ambient AQ do not demonstrate a strong correlation.

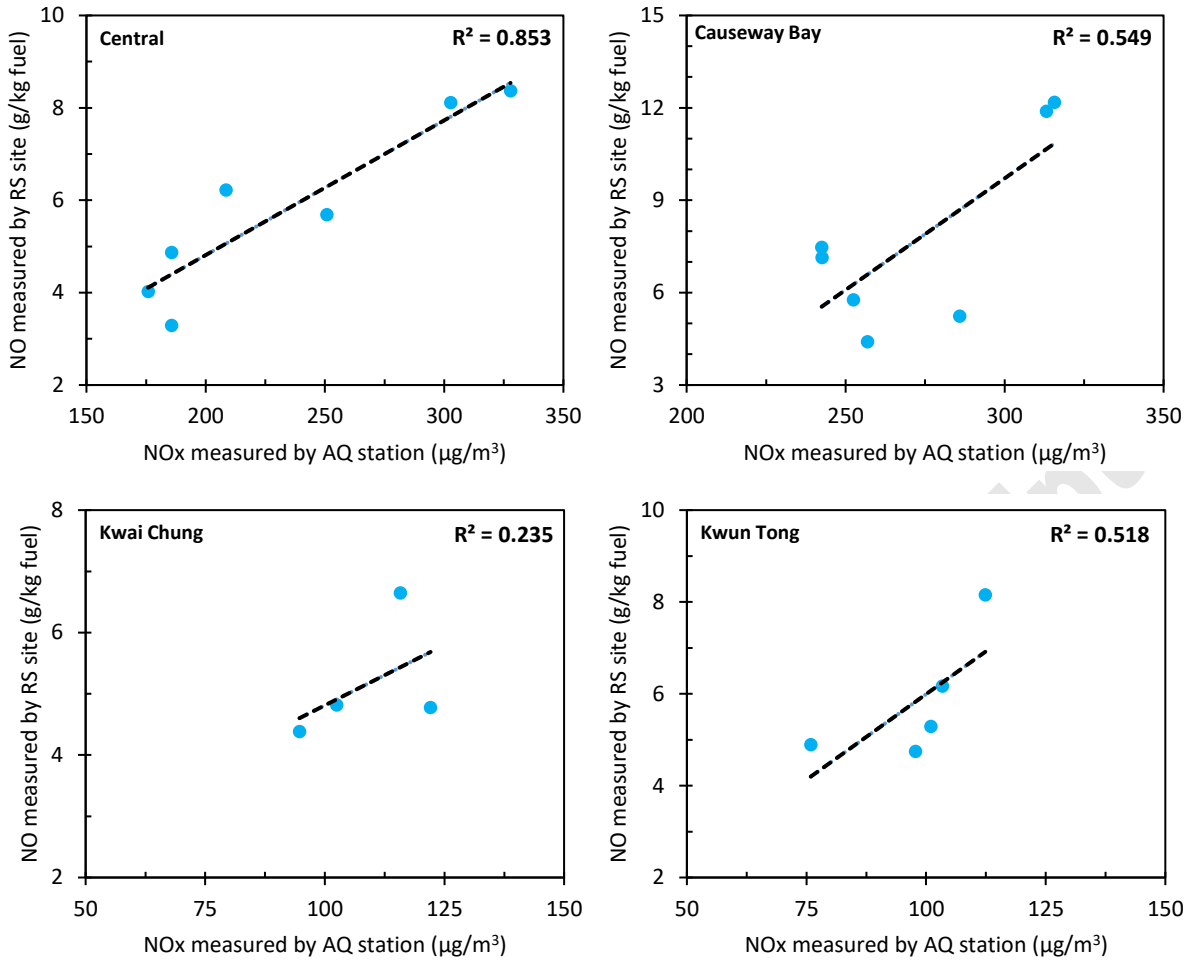


Fig. 9. Correlation between annual averages of NO emission factors measured by RS sites and NO_x concentrations measured by AQ stations.

4. Conclusions

This study investigated the real-world vehicle emissions using two regulatory monitoring techniques, namely on-road RS systems and AQ monitoring stations. Six pairs of RS sites and AQ stations were chosen which were located within 1 km, including two roadside and four ambient AQ stations. Two large datasets of AQ and RS emissions were collected during 2012-2018 and were analysed. The main results of this study can be summarised as follows:

1) All roadside and ambient AQ stations showed a decreasing trend in NO_2 concentrations during 2012-2018, in particular from 2013 to 2016 which could be attributed to the intensive implementation of a series of vehicle emissions control programmes during this period. Despite the decreasing trend observed, roadside NO_2 concentrations were still significantly over the AQO while ambient NO_2 concentrations were gradually achieving the AQO.

2) NO_2/NO_x ratios at ambient AQ stations increased from 2012 to 2018, while roadside NO_2/NO_x ratios increased first and then decreased or stabilised after 2014. The increasing trend of NO_2/NO_x ratio was caused by the adoption of DOC and DPF engine after-treatment systems which converted NO to NO_2 as well as more NO_x titration by the increasing ambient O_3. This led to a slower decreasing rate of NO_2 measured by AQ stations than that reported or expected

by the vehicle manufacturers and policymakers. In addition, roadside NO_2/NO_x ratios were significantly lower than those measured at ambient AQ stations.

3) While NO_2 concentrations (the main precursor of O_3 formation) were decreasing, O_3 concentrations were generally increasing for both roadside and ambient AQ stations from 2012 to 2018. AQ data showed that O_3 had little correlation with either NO_2 or NO_x. Instead, O_3 was mainly determined by the NO_2/NO_x ratio.

4) On-road RS data showed that overall NO emission factors decreased quickly during 2012-2015 and then decreased moderately after 2015. In addition, NO varied greatly among RS sites before 2015, but converged to a similar level after 2015. The decrease was mainly attributed to the effective NO reductions from LPG vehicles between 2013 and 2015 during which a free TWC and oxygen sensor replacement programme and an RS gross-emitters enforcement programme were conducted for LPG vehicles. On the other hand, diesel NO remained high and reduced slowly during 2012-2018. Gasoline vehicles were relatively clean compared with LPG and diesel vehicles.

5) Finally, NO_x emissions demonstrated good correlations between RS sites and roadside AQ stations, indicating that vehicle emissions were the major contributor to roadside NO_x pollution. However, ambient NO_x could be affected by various sources, leading to different levels of correlation between RS sites and ambient AQ stations.

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Supplementary information

Table S1. Number of valid emission records and fleet compositions at each selected RS site.

RS site	Fuel type	Survey year							Total
		2012	2013	2014	2015	2016	2017	2018	
Central	No. of records	4581	25906	30538	19377	26996	28150	36124	171672
	% of gasoline	60%	54%	56%	54%	54%	52%	50%	
	% of LPG	27%	35%	32%	34%	34%	38%	40%	
	% of diesel	13%	11%	12%	12%	12%	11%	10%	
Causeway Bay	No. of records	6030	17739	26934	10671	25142	14775	29866	131157
	% of gasoline	39%	44%	44%	45%	43%	42%	46%	
	% of LPG	31%	41%	39%	37%	37%	34%	37%	
	% of diesel	30%	16%	17%	19%	20%	24%	18%	
Eastern	No. of records	4201	4390		10987	15458	8512	13838	57386
	% of gasoline	36%	36%		45%	44%	49%	47%	
	% of LPG	47%	47%		34%	35%	34%	37%	
	% of diesel	17%	17%		21%	21%	17%	16%	
Tsueng Kwan O	No. of records			2687	14289	8758	12511	13373	51618
	% of gasoline			60%	61%	62%	66%	69%	
	% of LPG			16%	16%	16%	17%	17%	
	% of diesel			24%	22%	22%	18%	15%	
Kwai Chung	No. of records				7021	15843	8144	27905	58913
	% of gasoline				40%	39%	45%	45%	
	% of LPG				19%	20%	22%	25%	
	% of diesel				41%	42%	32%	31%	
Kwun Tong	No. of records		2692	2812	13406	10387	6548		35845
	% of gasoline		52%	51%	57%	56%	60%		
	% of LPG		29%	21%	20%	20%	19%		
	% of diesel		20%	28%	23%	24%	21%		
Total		14812	50727	62971	75751	102584	78640	121106	506591

(a) Pair 1 at Central

(b) Pair 2 at Causeway Bay

(c) Pair 3 at Tseung Kwan O

(d) Pair 4 at Eastern

(e) AQ-RS Pair 5 at Kwun Tong

(f) AQ-RS Pair 6 at Kwai Chung

AQ station

RS site

Fig. S1. Locations of AQ stations and RS sites selected for this study.