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Retrofit or renew the old diesel fleet: the NO_2 pollution in HK

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Summary

- Designed to control CO and PM, DOC increases NO_2 emission because it is intentionally produced as an oxidant.
- Designed to control NO_X, SCR may also increase NO_2 emission due to an increased share of NO_2 in NO_X.
- Euro IV or V buses performs no better than Euro III buses in terms of NO_2 emission. What is the point of retrofitting Euro III vehicles to meet Euro IV standard?
- It is NO_2 that is of public health relevance, not NO_X as regulated by Euro emission standards. In Euro VI, a separate NO_2 cap may be specified.
- The right way to control NO_2 is to drastically reduce NO_X to Euro VI level and the increased share of NO_2 in NO_X would be of less consequence.
- No more retrofit; quick replacement please.

Contents

1 NO_2

1.1 Roadside emissions and exposure

Roadside emissions and exposure

Roadside emissions from commercial vehicles, not Guangdong, are the major threat to the health of the HK population. Why?

- By emission, 53% of the time, the dominant source of HK air pollution is local [Lau et al., 2007].
- By exposure, a large population spend a large fraction of their time in the street canyons in HK.

Particulate matter and NO_2

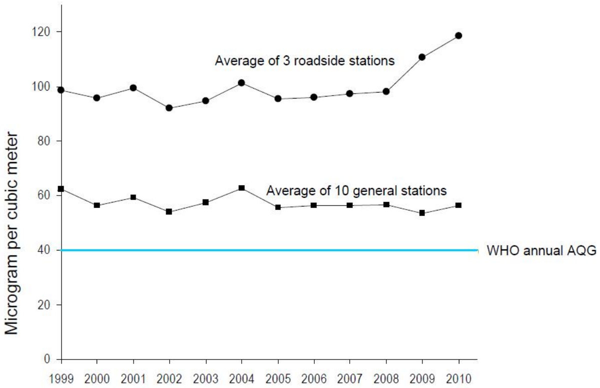
- Respirable suspended particles (RSP) and nitrogen dioxide (NO_2) have been the two pollutants that exceed the AQO limits.
- Of road vehicles, commercial diesel vehicles (buses & trucks) contribute 80-90% of roadside RSP and NO_X (EPD2006)
- Progress has been made in reducing roadside RSP levels
- Stagnant or even increasing trends are seen for NO_2.

NO_2 adverse health effects

- Aggravation of existing respiratory diseases (including asthma or other lung diseases)
- Reduction in lung function
- Increase the morbidity and death rates of cardiovascular and respiratory diseases

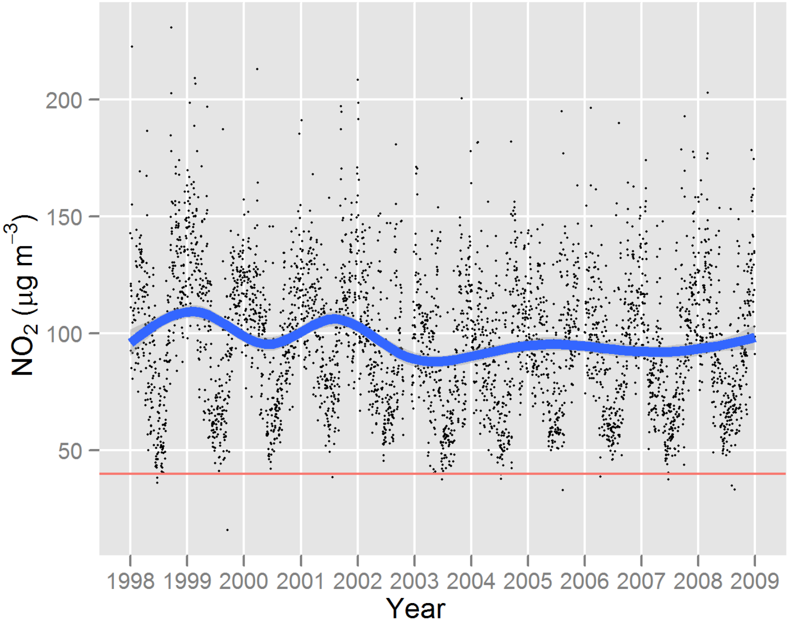
1.2 Trend of NO_2 concentrations

Annual mean NO_2 concentrations in 1999-2010



[Hedley, 2011]

Daily mean NO_2 concentrations in 1998-2008, in Causeway Bay



Let's look into one roadside station with complete data – the Causeway Bay (CB) roadside air monitoring station. The black dots denote the daily mean values of NO_2; the blue ribbon shows the smoothing trend; and the red line shows the World Health Organization (WHO) guideline value of 40 $\mu g/m^3$. Basically it is in every single day we are exposed to unacceptable levels of NO_2. And there is no sign of improvement in the past decade.

1.3 NO_2 sources and f-NO_2 trend

NO_2 sources and f-NO_2

- Directly from engine exhaust:

  $$N_2 + O_2 \rightarrow NO + NO_2$$

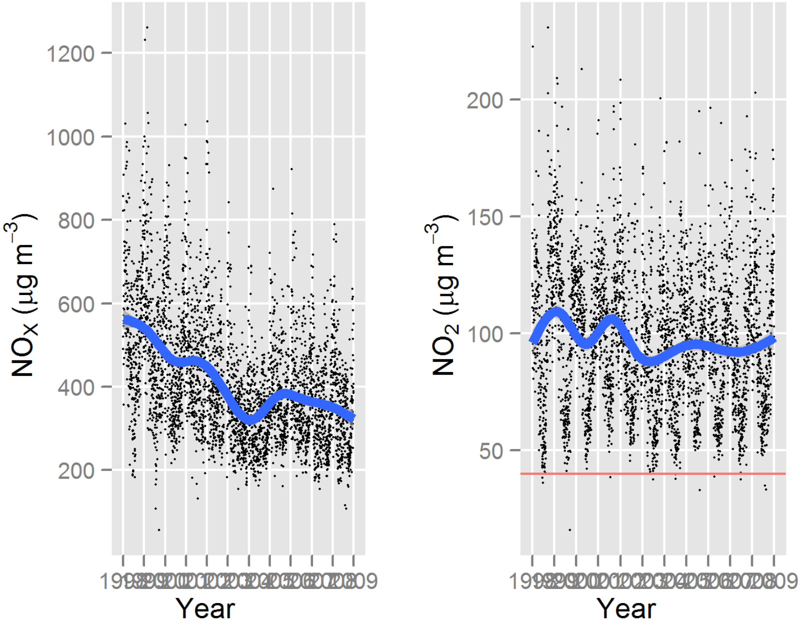
  The percentage of this primary NO_2 in the total ($NO + NO_2$), namely NO_X, from engine exhaust is known as f-NO_2. The f-NO_2 for petrol cars is around 5%, for diesel vehicles around 10% or higher.

- Converted from NO-O_3 reaction:

  $$NO + O_3 \rightarrow O_2 + NO_2$$

  O_3 is a regional air pollution issue.

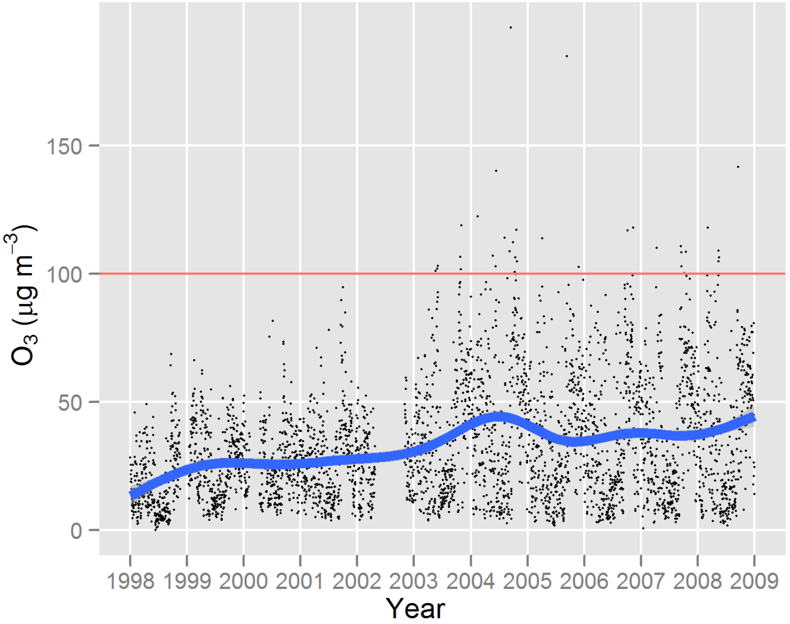
Daily mean NO_X concentrations in 1998-2008, in Causeway Bay



Interestingly, we notice NO$_X$ concentration has been decreasing continuously in the past decade. How do we explain the increasing trend of NO$_2$ despite the overall NO$_X$ decrease? Two possible reasons:

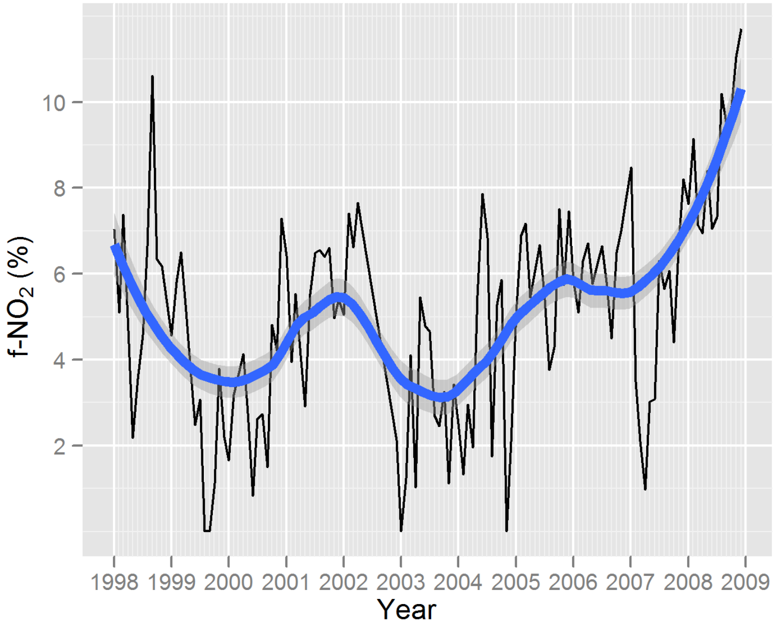
- Increased regional ozone concentrations cause higher oxidation of NO to NO$_2$.
- Higher share of NO$_2$ (f-NO$_2$) in the exhaust emissions of vehicles leads to increased NO$_2$.

Daily mean O$_3$ concentrations in 1998-2008, in Kwun Tong



We do observe a slightly increasing trend of O$_3$ in the general air monitoring station of Kwun Tong (KT), upwind of and close to the Causeway Bay roadside station. The red line in the figure shows the WHO guideline value (8-hour average) for O$_3$. The background O$_3$ concentration was taken into account when we calculate the f-NO$_2$ using a simple chemistry model developed by Carslaw and Beevers [Carslaw and Beevers, 2005] [Tian et al., 2011].

Trend of f-NO_2 in 1998-2008, in Causeway Bay

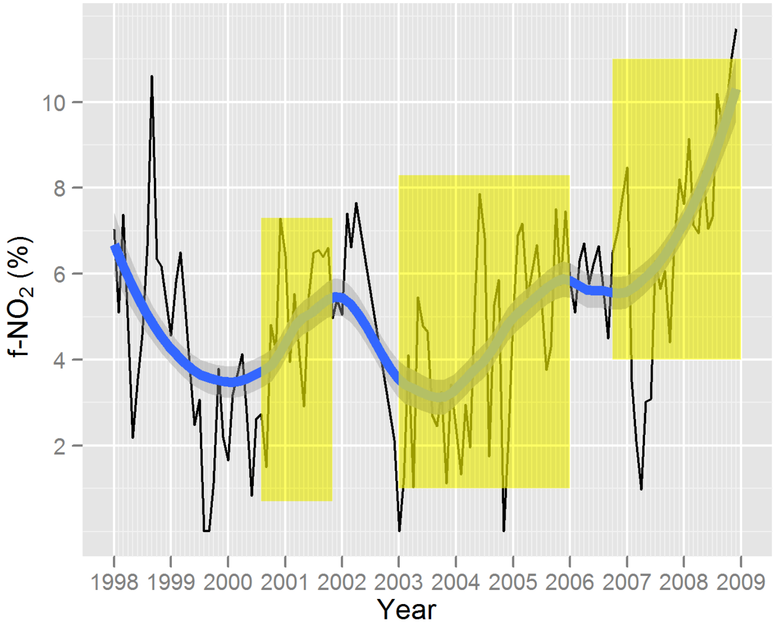


The f-NO_2 has also increased over the past decade. Therefore, besides background O_3 level increase, the increasing f-NO_2 in emission exhaust may be another reason for the persistently high concentration of NO_2 in HK [Tian et al., 2011].

2 DOC

2.1 DOC and f-NO_2 increase

Trend of f-NO_2 in 1998-2008, in Causeway Bay



There were ups and downs for the f-NO_2 in the Causeway Bay station. The three particular periods of rising f-NO_2 coincided with the implementation of three emission control measures, in 2000, 2003 and 2007, respectively (see Table below).

Measures to reduce vehicle emissions in Hong Kong, 1999-2008

- Advancement of smoke testing method:
  - *A(1)* - Dynamometer smoke test for light duty vehicles (Sep 1999)
  - *A(2)* - Dynamometer smoke test for heavy duty vehicles (Jan 2002)
- One-off grant for vehicle replacement:
  - *B(1)* - Diesel to LPG taxi (Aug 2000)
  - *B(2)* - Diesel to LPG light bus (Aug 2002)
  - *B(3)* - Replacement of Pre-Euro and Euro I commercial vehicles (Apr 2007)
- Retrofitting emission reduction device:
  - *C(1)* - Trap/DOC retrofitting for Pre-Euro LDV (Sep 2000)
  - *C(2)* - DOC retrofitting for Pre-Euro HDV (Jan 2003)
- Cleaner diesel:
  - *D(1)* - Ultra low sulfur disesle (ULSD, 0.005%) (Jul 2000)
  - *D(2)* - Euro V Diesel (Dec 2007)
- Stringent vehicle import standard:
  - *E(1)* - Euro III standard (Oct 2001)
  - *E(2)* - Euro IV standard (Oct 2006)
- Punishment for smoky vehicle:
  - *F* - The fine of fixed penalty ticket raised to $1000 (Dec 2000)

Correlation does not mean causation. However, the cause-effect relationship here is very likely because of similar experiences in Europe [Anttila et al., 2011, AQEG, 2006, Grice et al., 2009, Jenkin et al., 2008, Keuken et al., 2009, Velders et al., 2011] and the USA [Burgard and Provinsal, 2009, Millstein and Harley, 2010].

What was common in the three emission control measures?

Use of Diesel Oxidation Catalyst (DOC) in retrofit or new vehicles.

2.2 DOC equiped since Euro IV

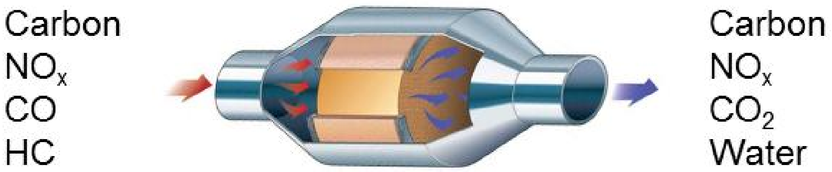
DOC equiped since Euro IV

A TNO report [Gense et al., 2006] says:

> "HD vehicles have not been equipped with oxidation catalysts before Euro IV. The introduction of PM traps on HD vehicles initiated the application of the oxidation catalysts in this segment. For HD it can therefore be stated that the introduction of exhaust gas aftertreatment (starting with Euro IV) significantly increases the direct NO2 emissions of the vehicles."

2.3 How DOC works

How DOC works



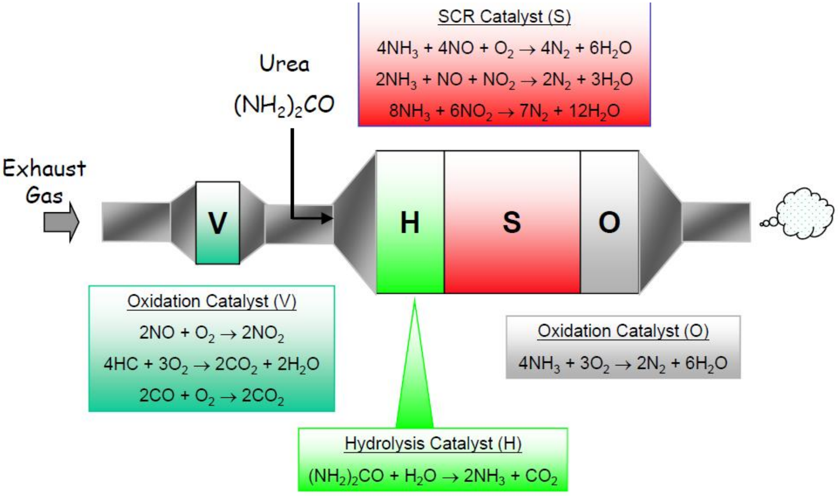
- A DOC is comprised of a stainless steel canister containing a honeycomb structure coated with a precious metal catalyst such as platinum or palladium.
- DOC oxidises a **portion** of the NO present in the exhaust gas into NO_2, and then NO_2 oxidize the exhaust into less harmful pollutants and the NO_2 itself reduced to NO again. It has been and will continue to be a method to control PM emissions.
- However, as this **portion** is difficult to manage (fine-tune) in practice, a surplus of NO_2 is inevitably formed, resulting in an increased NO_2 emission.

Designed to reduce emission of hydrocarbons (including polycyclic aromatic hydrocarbons), CO and particulate matter (PM), the particulate trap, and diesel oxidation catalysts (DOCs) have the disadvantage that they may increase the NO_2 fraction of total NO_X emissions. The health benefit offered in the reduction in hydrocarbons and PM should not be offset by the adverse health effects of increased or stagnant NO_2 concentrations.

3 SCR

3.1 SCR to reduce NO_X, not NO_2

Selective catalytic reduction (SCR) devices



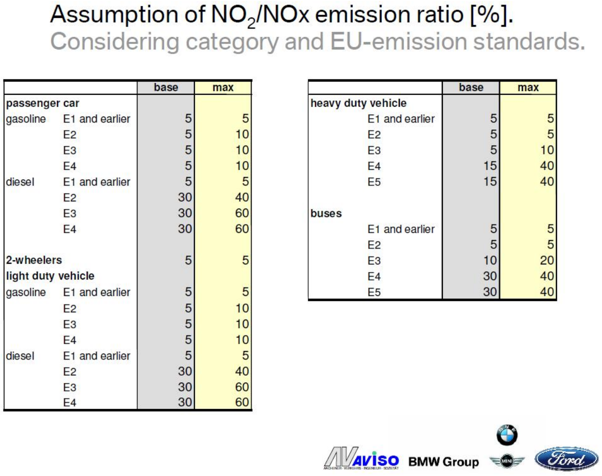
In recent years, selective catalytic reduction (SCR) has been developed for NO_X control.

Why DOC again is included in this SCR system? For a high deNOx efficiency, a NO/NO_2 ratio of about 1 is needed [Louis-Rose, 2007]. DOC is needed to convert some NO to NO_2. A consequence is that f-NO_2 will increase with SCR equipped vehicles despite total NO_X reduction. This explains the correspondence of the f-NO_2 rise with the Euro IV vehicle introduction since October 2006.

An important note

- SCR is designed to reduce NO_X emissions in order to meet Euro emission standards.
- SCR is not to reduce NO_2 which is of public health interest.
- A limit on f-NO_2 emission will not be specified until Euro VI – a cap of 20% is tentatively proposed.

f-NO_2 increase in newer models



Assumption of NO_2/NOx emission ratio [%].

Considering category and EU-emission standards.

		base	max
passenger car			
gasoline	E1 and earlier	5	5
	E2	5	10
	E3	5	10
	E4	5	10
diesel	E1 and earlier	5	5
	E2	30	40
	E3	30	60
	E4	30	60
2-wheelers		5	5
light duty vehicle			
gasoline	E1 and earlier	5	5
	E2	5	10
	E3	5	10
	E4	5	10
diesel	E1 and earlier	5	5
	E2	30	40
	E3	30	60
	E4	30	60

		base	max
heavy duty vehicle			
	E1 and earlier	5	5
	E2	5	5
	E3	5	10
	E4	15	40
	E5	15	40
buses			
	E1 and earlier	5	5
	E2	5	5
	E3	10	20
	E4	30	40
	E5	30	40

Here are some assumptions taken by the auto-industry for the f-NO_2 with different Euro emission standard vehicles [Kessler et al., 2006].

Let's look at the buses. Euro IV and Euro V vehicles have much higher f-NO_2 values.

3.2 Some simple math

Some simple math

Table 1: Direct NO_2 emissions from diesel buses in g/kWh

Tier	**NO_X** (g/kWh)	**f-NO_2** %	**NO_2** (g/kWh)
Euro III	5.0	10%	0.5
Euro IV	3.5	30%	1.05
Euro V	2.0	30%	0.6
Euro VI	0.4	20%	0.08

- From Euro III to Euro V, direct emission of NO_2 may actually increase despite decrease in NO_X.
- It is not until Euro VI, direct emission of NO_2 get lower than with Euro III.

This calculation is based on the assumption the on-road performance in NO_X control is the same as in the certification test cycle. In reality, the assumption is not true because of the poor performance of SCR equipped vehicles in urban driving conditions.

3.3 Test cycle vs. real world

Poor performance of SCR in urban driving conditions

- Vehicles fitted with SCR did result in considerably reduced NO_X emissions for motorway-type driving, but much higher emissions in off-cycle city-streets due to low exhaust temperature [Ligterink et al., 2009].
  - Compared with a Euro III truck, emissions of NO_X from SCR trucks on motorways was reduced from 11 g per kg CO_2 to 4 g per kg CO_2.
  - However, in urban areas the emissions changed from 13 to 10 g per kg CO_2.
- "The use of SCR on the bus fleet in London reduces NO_X emissions by about 65%. However, this reduction is seen as a 'best case' because the bus in question was had a small engine that would have been put under higher load than a larger bus engine" [Carslaw et al., 2011].

Poor performance of SCR in urban driving conditions

According to the International Council on Clean Transportation [ICCT, 2010]:

> "Recent research in Europe found that under low-load, low exhaust temperature conditions during urban driving SCR-equipped Euro V-compliant vehicles had NO_X emissions on the order of three times that allowed under the standard.
>
> Similar data collected by the Tokyo Metropolitan Government details an in-use HD truck with NO_X emissions between 1.8 and 3.8 times that measured on the new vehicle certification test cycle when tested on test cycles representative of real-world driving conditions in Tokyo."

4 Retrofit Euro III?

4.1 What's the point?

What's the point?

- As shown in Table 1, Euro IV or V buses performs no better than Euro III buses in terms of NO_2 emission. What is the point of retrofitting Euro III vehicles to meet Euro IV standard?

- Are we spending money to retrofit and protect public health? But it is NO_2 that is of public health relevance, not NO_X as has been regulated by Euro emission standards.
- Have we already forgotten the correspondence of the f-NO_2 rise with the Euro IV vehicle introduction since October 2006 in HK?
- Euro IV is not protective enough in terms of NO_2 emission. It is not until Euro VI a separate NO_2 cap may be specified.

Spend money to retrofit to Euro IV standard that is not protecting public health? Unacceptable.

4.2 Low feasibility anyway

Feasibility is also low for retrofitting to meet this questionable goal of Euro IV

- Lack of space: no sufficient space in individual bus models for adding a large urea storage tank and other sensors and devices.
- Lack of on-board failsafes (available in new models) to ensure that drivers properly fill urea tanks: without reagent, NOx emissions of a Euro V vehicle could be as poor as a Euro II vehicle. Inspection and maintenance (I/M) can be extremely challenging.
- Application of SCR needs very delicate calibration for proper operation in vehicles: usually it should be done by vehicle manufacturer.

5 Euro VI phase in

5.1 Early Euro VI phase in

Early Euro VI phase in

- It is NO_2 that is of public health relevance, not NO_X as has been regulated by Euro emission standards.
- In almost every single day we are exposed to unacceptable levels of NO_2. And there is no sign of improvement in the past decade.
- It is not until Euro VI a separate NO_2 cap may be specified.
- The right way to control NO_2 is to drastically reduce NO_X to Euro VI level and the increased share of NO_2 in NO_X would be of less consequence.
- The right choice is to replace old diesel vehicles with Euro VI ones or other less polluting vehicles as soon as possible.

5.2 Are we overregulating?

View from auto industry

A TNO report [Gense et al., 2006] says:

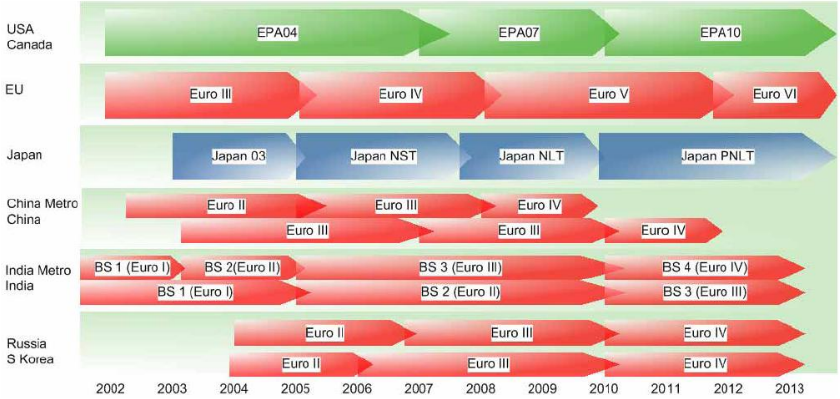
> “The industry is aware of the issue of increased direct NO_2 emissions and has stated that in principle there are technical solutions available for limiting the direct NO_2 emissions of vehicles with exhaust gas aftertreatment, but that in order to start this development the regulator should put adequate legislation in place.”

View from bus fleet operator

Mr. Kane Shum, KMB's Principal Engineer, said (SCMP, Feb 2, 2011):

> "The Euro VI vehicle is in the pipeline now. A prototype could be completed as early as 2013 or 2014, and then it will be rolled out to the market. We are lucky to be part of this and are working closely with the manufacturers.
>
> We are always ahead of the standard. When others were scrambling for Euro IV, we were already using Euro V. It will be the same for the Euro VI."

Emission legislation: where Hong Kong stands?



(Alex Woodrow, EngineExpo, 2007)

European Environmental Bureau [EEB, 2008] complains about lagging behind the USA and Japan in air quality control and insists that 1st April 2013 is too late a date to apply Euro VI regulation. How about Hong Kong? Should we continue to talk about retrofitting to meet Euro IV standard?

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ORIGINAL PAPER

Increasing trend of primary NO_2 exhaust emission fraction in Hong Kong

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Abstract Despite the successful reduction in roadside NO_x levels, no such decrease has been detected in roadside NO_2 concentration in Hong Kong. One underlying cause could be the rising primary NO_2 fraction of the total emission of NO_x. Primary NO_2 can be particularly detrimental to Hong Kong because a large fraction of the population are exposed to the traffic-related primary pollutants in the street canyons formed by congested high-rise buildings. In this study, hourly mean concentration data for roadside nitrogen oxides (NO_x), nitrogen dioxide (NO_2), and background ozone (O_3) were used to estimate the mean primary NO_2 fraction from vehicle exhausts in Hong Kong. An overall increasing trend was observed for the primary NO_2 fraction (f-NO_2) values in all the three roadside air monitoring sites. The primary NO_2 as a fraction of total NO_x (f-NO_2) increased approximately from 2% in 1998 to 13% in 2008 in Hong Kong. The two particular periods of rising f-NO_2 coincided with the two implementation periods of the diesel retrofit programs for the light-duty vehicles and heavy-duty vehicles. Future vehicle emission control strategies should target not only total NO_x but also primary NO_2. Health benefit or disease burden estimates should be taken into account and updated in the process of policy planning and evaluation.

Keywords Primary NO_2 · Trend · Diesel vehicle exhaust · DOC retrofitting · Health effects · Street canyon

Introduction

Exposure to nitrogen dioxide (NO_2) can give rise to adverse health effects including aggravation of existing respiratory diseases and reduction in lung function. People with asthma or lung diseases and children are more susceptible to the adverse effects of NO_2 exposure. Recent time series studies have found the associations of cardiovascular and respiratory mortalities with day-to-day changes in NO_2 levels (Wong et al. 2002b, 2008; Samoli et al. 2006). Environmental Protection Agency of USA (USEPA) has concluded that there is substantial evidence for a likely causal relationship between NO_2 and respiratory morbidities (U.S.EPA 2008).

In Hong Kong, the Air Quality Objectives (AQOs) has set NO_2 to an annual mean of 80 $\mu g/m^3$ and an hourly limit of 300 $\mu g/m^3$ not to be exceeded more than

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3 times per year (EPD 2008; CAI-Asia 2006). Even after various stringent emission control measures implemented by the Hong Kong government, NO_2 has been consistently violating the AQO since 1999 (EPD 2008; CAI-Asia 2006; EPD 2009, 2006, 2007a). The ambient NO_2 concentration in Hong Kong's general air quality monitoring stations (except the rural Tap Mun station) in 2008 exceeded the proposed 1-h AQO of 40 μg/m^3 for 84 times. No exceedance was recorded in Tap Mun, indicating that NO_2 in urban areas is largely contributed by local sources. All the three roadside air quality monitoring stations failed to achieve both hourly and yearly AQO in 2009 (EPD 2009). Road vehicles are the second largest source of air pollution in Hong Kong, contributing 27% of the NO_x emissions. In 2006, diesel commercial vehicles accounted for 23% of the vehicle fleet, but they contributed 80% of the total vehicular emission of NO_x (EPD 2007b). It is hoped that the implementation of vehicular emission control measures can reduce the levels of NO_2 to meet the new AQO (of Hong Kong SAR 2009).

A brief description of the roadside emission control policies (EPD 2010b) is given in Table 1. The control measure *B(1)* was a subsidy program started in August 2000 to offer a one-off grant of 40,000 HK dollars for each replacement of diesel taxis with one that operates on liquefied petroleum gas (LPG). The program was completed at the end of 2003, and nearly, all (about 99.9%) of the 18,000 diesel taxis switched to LPG. *B(2)* offered incentives to encourage the early replacement of the 6,000 diesel light buses with LPG or electric ones. It was completed at the end of 2005. In 2010, about 60% of the registered public light buses used LPG. *B(3)* provided a one-off grant to encourage vehicle owners to replace their pre-Euro and Euro I diesel commercial vehicles with Euro IV standard vehicles. *C(1)* and *C(2)* were two programs in Hong Kong to help the owners of pre-Euro light diesel vehicles and nonidling pre-Euro heavy diesel vehicles to retrofit their vehicles with particulate traps or oxidation catalysts, which were completed in 2001 and 2005, respectively (EPD 2010b). A total of 24,000 light diesel vehicles (more than 80%) and 34,000 heavy diesel vehicles (more than 96%) were fitted with these devices under the retrofit programm. A similar program retrofitted 2,500 or about 95% of long idling pre-Euro heavy diesel vehicles (i.e., concrete mixer, gully emptier, lorry crane, and pressure tanker) with oxidation catalysts and was completed in 2005 (EPD 2010b).

Table 1 Measures to reduce vehicle emissions in Hong Kong, 1999–2008

Advancement of smoke testing method
A(1)—Dynamometer smoke test for light-duty vehicles (Sep 1999)
A(2)—Dynamometer smoke test for heavy-duty vehicles (Jan 2002)
One-off grant for vehicle replacement
B(1)—Diesel to LPG taxi (Aug 2000)
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Punishment for smoky vehicle
F—The fine of fixed penalty ticket raised to $1,000 (Dec 2000)

The majority of roadside NO_2 are secondary pollutant derived from the chemical reaction between nitric oxide (NO) and ozone (O_3). However, a substantial proportion of NO_2 is emitted directly, which is known as "primary NO_2" (Boulter et al. 2007; Abbott and Stedman 2006; Carslaw and Beevers 2005; Jenkin 2004). A commonly accepted estimate for the proportion of primary NO_2 is 5% by volume of total NO_x emitted by motor vehicles, but recent studies have reported wide variation of this proportion (Carslaw and Beevers 2005, 2004a, b; Jenkin et al. 2008; Herner et al. 2009; Bishop et al. 2010). The current study aims to quantify the trend of primary NO_2 exhaust emission fraction in Hong Kong and explore the potential factors affecting this trend, such as road transport emission control measures.

Methodology

NO_x, NO_2, and O_3 air monitoring data were obtained from the Environmental Protection Department (EPD) of Hong Kong that currently operates 11 general (background) air monitoring stations and 3 roadside stations, namely Causeway Bay (CB), Central (CL), and Mong Kok (MK) (EPD 2008). Figure 1 shows the air monitoring stations in Hong Kong. Levels of NO_x and NO_2 were monitored in all the three road side stations and in the closest background station Kwun Tong (KT) from 1998-01-01 to 2008-12-31. Hourly NO_x and NO_2 were measured using chemiluminescence technique at the roadside stations; additionally, O_3 by UV absorption technique was monitored in the background stations (EPD 2008). Mann–Kendall test (Hirsch et al. 1982) was used to detect the presence of temporal trends in deseasonalized monthly mean concentrations of roadside NO_x, NO_2, and the slopes were calculated using the method of Sen (Gilbert 1987). Mann–Kendall test calculates Kendall's tau nonparametric correlation coefficient for any pair of X,Y data and examines its statistical significance. If X is time, it tests for trend in the Y variable. The method of Sen (Gilbert 1987), often used in conjunction with the Mann–Kendall test, calculates the Sen slope of the trend, which is the median slope of all slopes calculated using pairs of observations collected at the same time each year. Uncertainty in slopes is calculated using bootstrap methods (Carslaw and Ropkins 2010).

To quantify the fraction of primary NO_2, we adopted the method developed by Carslaw and Beevers (2005). The method is based on simple chemical equations that explain time-dependent change in NO, NO_2, and O_3 concentrations (Carslaw and Beevers 2005, 2004b). The increment in roadside NO_x and NO_2 against nearby background station is assumed to be contributed by two different origins of NO_2: one that is derived through the interaction between NO and O_3 and another that directly emitted by road vehicles. The primary NO_2 fraction in total NO_x emission is expressed as f-NO_2. NO_2 formed through the NO-O_3 reaction is dependent on the time available for the reaction to take place, termed as τ. By considering different values of f-NO_2 and τ, several combinations of NO_x–NO_2 relationship are predicted and root mean square error is calculated and then compared with the measured relationship. The value of τ is set to 60 s that provides a reasonably consistent calculation of f-NO_2 (Carslaw and Ropkins 2010). The value of f-NO_2 that attains good agreement between the measured and predicted NO_x–NO_2 relationship with minimum error is considered to be the best estimate of primary NO_2 fraction (Carslaw and Beevers 2005, 2004b, Carslaw 2005).

Fig. 1 Air monitoring stations in Hong Kong

This modeling method requires mean hourly roadside concentrations of NO_x and NO_2 in roadside and background monitoring stations. Concentration of O_3 is also needed for the background station. In this study, Kwun Tong (KT) was selected as the background station on the basis of two requirements: (1) the NO_x levels should be much higher in the roadside than in the background station and (2) the background site should be reasonably close to and upwind of the roadside station. The surface wind in Hong Kong is predominantly easterlies year-round, with a season pattern of more northerly winds in winter and southerly winds in summer. As seen in Fig. 1, the KT background station is upwind of the roadside stations.

The time trend of primary NO_2 fraction was compared with that of elemental carbon (EC) in PM_{10} measured in the MK roadside station. Measurement of EC has been routinely made at the MK station by the EPD using a thermal/optical carbon analyzer (Yu et al. 2004). Spearman correlation analysis was also performed for the relation between EC in PM_{10} and the primary NO_2 fraction.

For statistical analysis, we used R statistical software, version 2.11.0 (Team 2010) and the *openair* package (Carslaw and Ropkins 2010).

Results

Figure 2a shows the downward trend of NO_x level from 1998 to 2008 in road side stations. In contrast, NO_2 remained relatively constant over the same time period, as seen in Fig. 2b. By applying Mann–Kendall test (Hirsch et al. 1982), we found significant downward trend of NO_x at the $p = 0.001$ level. For NO_2, no statistically significant trend was detected. In addition to Mann-Kendall test, we use the method of Sen (Gilbert 1987) to calculate the trend of NO_x. The slope for NO_x showed that concentration decrease on average is 12.6 ppb/year.

Figure 3 shows the monthly trends in the modeled primary NO_2 fractions for the three roadside stations in Hong Kong (1998–2008). The gray ribbon in each station shows a smooth fit with 95% confidence intervals. The yellow and green rectangle shades indicate the periods of the diesel retrofit programs for the light-duty vehicles and heavy-duty vehicles, respectively. All of these three roadside stations indicated sustained upward trend of primary NO_2 fraction from 2003. The lowest f-NO_2 was estimated in CL station in 1999 that was about 2% and the highest was approximately 14% in MK site at the end of 2008. Among the three roadside stations, the CB station has the longest time series with complete data. There are two separate periods of rise in f-NO_2: the first one from late 2000 to the end of 2001 and the second one from early 2003 to the end of 2005. These

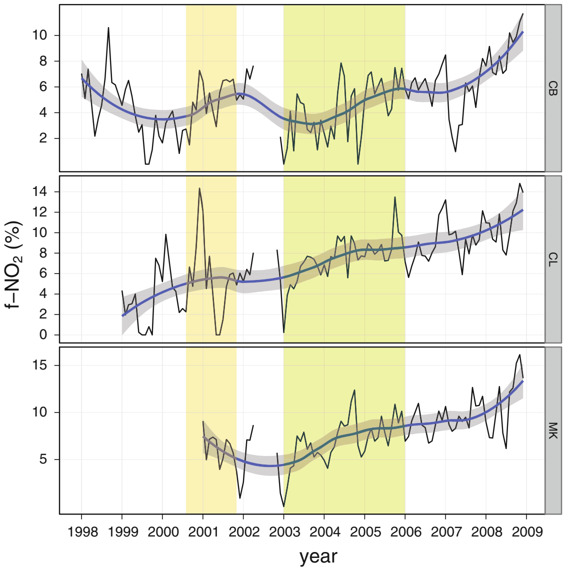

Fig. 3 Monthly trends in the modeled primary NO_2 fractions for the three roadside stations in Hong Kong (1998–2008). The *gray* ribbon in each panel shows a smooth fit with 95% confidence intervals. The *yellow* and *green rectangle* shades indicate the periods of the diesel retrofit programs for the light-duty vehicles and heavy-duty vehicles, respectively. (Color figure online)

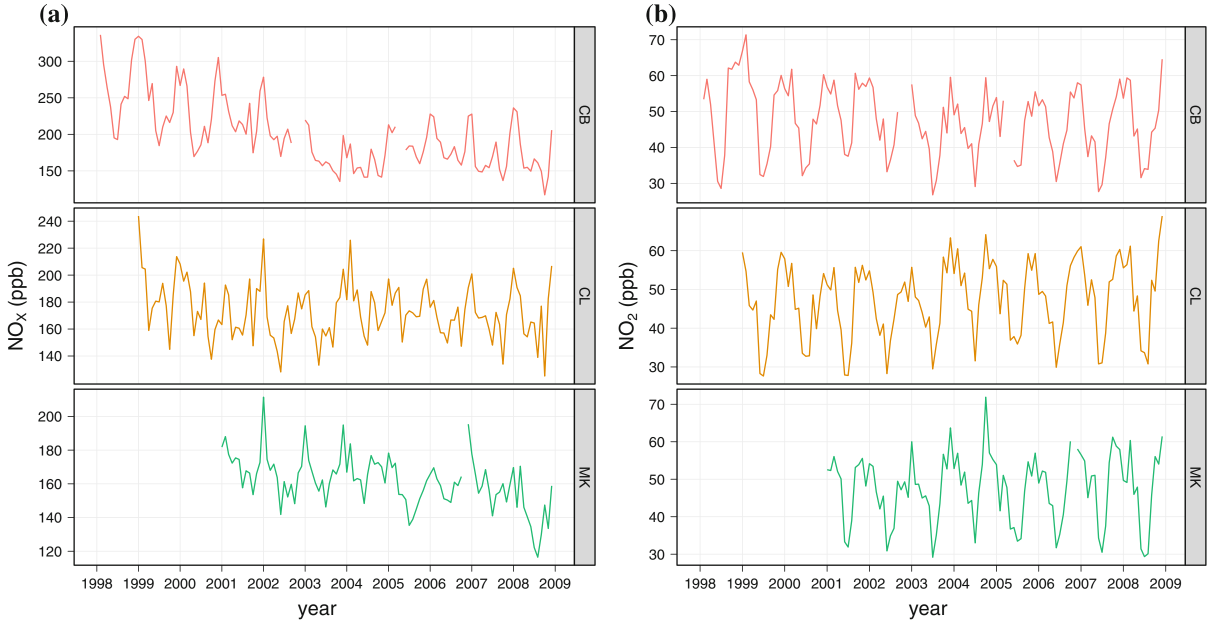

Fig. 2 Trends in monthly mean concentrations of NO_x (**a**) and NO_2 (**b**) at the roadside monitoring sites in Hong Kong (1998–2008)

two increase periods coincidently correspond to the two periods of the diesel retrofit programs for the light-duty vehicles and heavy-duty vehicles, respectively (Table 1).

Figure 4a shows the contrasting trends of EC and f-NO_2 in the MK roadside monitoring site. Figure 4b shows the inverse correlation between EC and f-NO_2. The Spearman correlation coefficient is −0.33, and the *p*-value is 0.002.

Discussion

Decreasing roadside NO_x reflects the reduction in vehicular emission due to successful implementation of various emission control measures. Major initiatives include replacing diesel taxis and light buses with liquefied petroleum gas (LPG) vehicles, introducing Euro IV emission standard, retrofitting pre-Euro diesel vehicles with particulate traps or oxidation catalysts, mandating ultra-low sulfur diesel for vehicle, and taking stronger enforcement actions against smoky vehicles (CAI-Asia 2006; EPD 2010a). Despite the reduction in NO_x, however, no such decrease was detected in roadside NO_2 concentration. One possible explanation could be the increase in directly emitted NO_2 from road vehicles. The trends of estimated primary NO_2 fraction in NO_x exhaust support this hypothesis. One new possible direction is to control primary NO_2 exhaust emission by legislation (Carslaw and Beevers 2004a).

An overall increasing trend was observed for the primary NO_2 fraction (f-NO_2) values in all the three roadside air monitoring sites. The f-NO_2 increased approximately from 2% in 1998 to 13% in 2008 in Hong Kong. This trend is similar to what was reported in other cities: Carslaw (2005) reported an increase in f-NO_2 from 5–6% to 17% in London, and Keuken et al. (2009) identified a rise in f-NO_2 from 9 to 13% in Rotterdam over the period 1986–2005. The absolute values in Hong Kong, estimated by using a simple constrained chemistry model (Carslaw and Beevers 2005), were higher than one actual measurement in 2005, 2% in the mid-sections of a tunnel in Hong Kong (Yao et al. 2005). In some situations where NO, NO_2, and O_3 concentrations are measured, it is possible to estimate the primary NO_2 fraction by considering the gradient in "total oxidant" defined as $NO_2 + O_3$ (Clapp and Jenkin 2001). However, O_3 is rarely measured in roadside monitoring sites. The

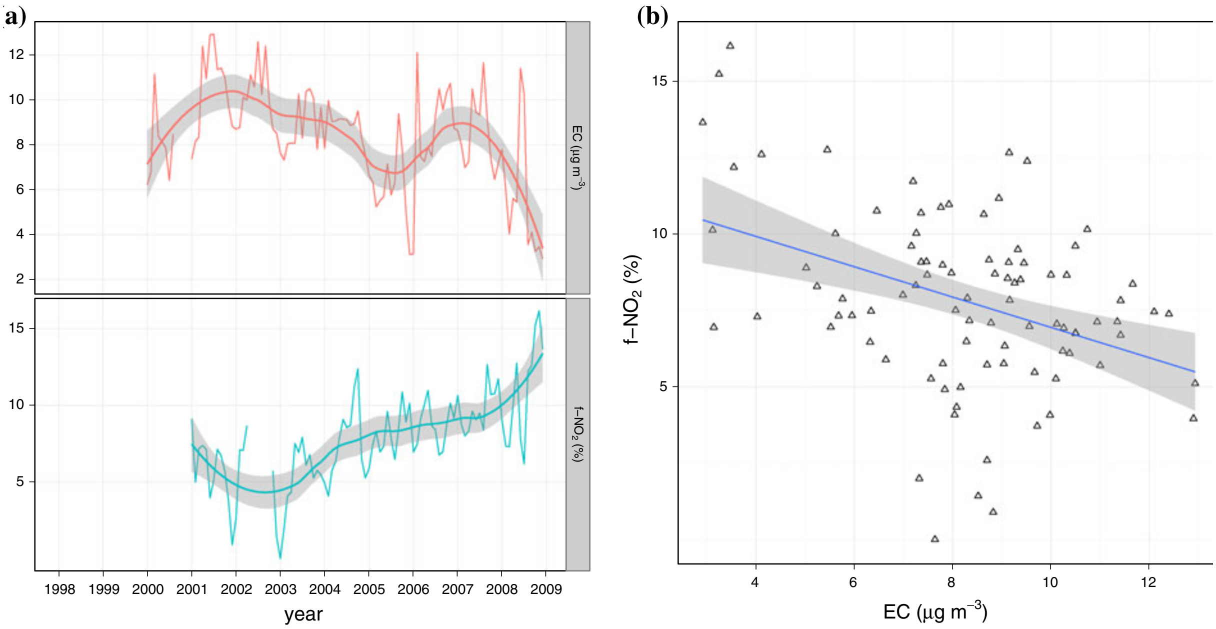

Fig. 4 Relationship between f-NO_2 and EC in PM_{10} in the MK roadside monitoring site in Hong Kong (2000–2008). **a** Contrasting trends of monthly EC and f-NO_2. **b** Scatterplot of monthly EC and f-NO_2. The *gray* ribbon in each panel shows a smooth fit with 95% confidence intervals. The Spearman correlation coefficient between the two variables is −0.33, and the *p*-value is 0.002

chemistry modeling method by Carslaw and Beevers (2005) remains an alternative in quantifying the level of primary NO_2 from the analysis of ambient monitoring data.

The two particular periods of rising f-NO_2 coincided with the introduction of oxidation catalysts for diesel vehicles, in 2000 and 2003, as part of vehicular emission control drive. A total of 24,000 light diesel vehicles (more than 80%) and 34,000 heavy diesel vehicles (more than 96%) were retrofitted by the end of 2001 and 2005, respectively. The rise of f-NO_2 in Causeway Bay and Central roadside stations during the end of 2000 corresponded to the retrofitting of light-duty diesel vehicle by DOC in 2001. Similarly, the subsequent increase in f-NO_2 during 2003 in all the roadside stations coincided with retrofitting of heavy-duty diesel vehicle in the same year. Previous studies have also established the link between fitting oxidation catalysts to the vehicular fleet and rising f-NO_2 (Carslaw 2005; Keuken et al. 2009; Carslaw et al. 2007; Rexeis and Hausberger 2009). Besides retrofitting the old diesel vehicles on the road, the Hong Kong government has also imposed stringent vehicle import standards since 2001. As newer diesel vehicles are usually equipped with DOCs or similar devices before coming into service, they may also have contributed to the general increase in primary NO_2 emissions at the roadside.

Designed to reduce emission of hydrocarbons (including polycyclic aromatic hydrocarbons), CO and particulate matter (PM), the particulate trap, and diesel oxidation catalysts (DOCs) have the disadvantage that they may increase the NO_2 fraction of total NO_x emissions. The DOCs oxidize CO and hydrocarbon emission catalytically by converting NO to NO_2. The NO_2 is then used to assist in the oxidation of trapped particles, therefore reducing emission of hydrocarbons, CO, and particulate matter (PM) (Carslaw 2005; Johnson 2009; Millstein and Harley 2010). The contrasting trends of decreasing elemental carbon and increasing f-NO_2 in the roadside, and the inverse correlation between these two measurements emphasize the importance of overall effect analysis in vehicle emission control measures. The overall public health impacts of the retrofit program should be evaluated. The health benefit offered in the reduction in hydrocarbons and PM should not be offset by the adverse health effects of increased or stagnant NO_2 concentrations.

Public health implications of the increasing f-NO_2 in the roadside warrant particular attention as the adverse health outcomes of NO_2 exposure have been well documented in Hong Kong. Time series studies have disclosed that ambient concentrations of NO_2 increase the daily mortalities of all causes (Wong et al. 2008), cardiovascular diseases (Wong et al. 2002b, 2008, 2001) and respiratory diseases (Wong et al. 2002b). Excess risks of hospital admission of respiratory diseases (Wong et al. 1999; Ko et al. 2007; Wong et al. 2002a) and cardiac diseases (Wong et al. 1999, 2002a) from NO_2 exposure have also been reported. Primary NO_2 can be particularly detrimental to Hong Kong because a large fraction of the population are exposed to the traffic-related primary pollutants in the street canyons formed by congested high-rise buildings. Street canyon situations in Hong Kong are not only commonly found in business areas, they are also found in some residential districts. Pollutants build up between the buildings that protect the street from the wind. Because of the high population density and congestion, the adverse health effect in the population per unit emissions may be magnified in street canyons. Health benefit or disease burden estimates should be taken into account and updated in the process of policy planning and evaluation.

Conclusion

Despite the successful reduction in roadside NO_x levels, no such decrease was detected in roadside NO_2 concentration in Hong Kong. One underlying cause could be the rising primary NO_2 fraction of the total emission of NO_x. Primary NO_2 can be particularly detrimental to Hong Kong because a large fraction of the population are exposed to the traffic-related primary pollutants in the street canyons formed by congested high-rise buildings. The two particular periods of rising f-NO_2 coincided with the two implementation periods of the diesel retrofit programs for the light-duty vehicles and heavy-duty vehicles. Future vehicle emission control strategies should target not only total NO_x but also primary NO_2.

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