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The use of tunnel concentration profile data to determine the ratio of NO_2/NO_x directly emitted from vehicles

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Abstract

Recently, it is reported that primary vehicular NO_2/NO_x ratio to be 10–30% and primary vehicular NO_2 has raised much interest and concern in the control of NO_2 in urban areas. In this study, primary vehicular NO_2/NO_x ratio in Hong Kong was investigated based on intensive long tunnel (3.7–4 km in length) experiments where concentration profiles of air pollutants along the entire lengths of the tunnels were obtained. Long tunnels were selected because of the inherent low O_3 concentrations in the partially enclosed environment. In addition the concentrations of pollutants from vehicles are high. Thus, the NO_2 measured inside long tunnels would be more representative of the primary NO_2 emitted by vehicles and contribution due to atmospheric transformation would be limited. This dataset was supported by a long-term on-road air quality dataset (June 2002–August 2003). Both datasets were obtained using the Mobile Real-time Air Monitoring Platform (MAP). The primary on-road vehicular NO_2/NO_x ratio was less than 2%, detected in the mid sections of tunnels investigated, where O_3 concentration was at a minimum. In sections of the tunnels (entrance and exit) where O_3 concentrations were relatively high, the NO_2/NO_x ratio could be as high as 19%. Long-term (annual average) on-road air quality data in open air yielded NO_2/NO_x ratios up to 28%. Thus, it is apparent that directly emitted NO_2 from vehicles is not significant in atmospheric NO_2 concentration. A simple model was used to segregate the contribution of background NO_2 and transformed NO_2 measured in vehicle plumes.

1. Introduction

NO_2 plays a central role in tropospheric chemistry. NO_2 in urban atmosphere originates mainly from primary emissions of combustion processes and oxidation of NO. Recently, primary vehicular NO_2 raised much interest and concern in NO_2 control in urban areas (Harrison and Shi, 1996; Carslaw and Beevers, 2004a, b, 2005; Soltic and Weilenmann, 2004). The reported primary vehicular NO_2/NO_x volume ratio varied from

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2–5% to ∼30% (Lenner and Lindqvist, 1983; Cariappa et al., 1994; Harrison and Shi, 1996; Clapp and Jenkin, 2001; Jimenez et al., 2001; Carslaw and Beevers, 2004a, b, 2005; Jenkin, 2004a, b; Soltic and Weilenmann, 2004). The lower ratio (2–5%) is the widely accepted value for gasoline engines under normal driving conditions (Hilliard and Wheeler, 1979). Two methods were used in these experiments: direct measurements using chassis dynamometers equipped with dilution system and remote sensing technology, and indirect measurements in tunnels at pre-determined spots, and conventional stationary (including roadside) monitoring. One of the difficulties encountered in these methods is how to isolate primary vehicular NO_2 from background and transformed NO_2. A more definitive primary NO_2/NO_x ratio would be very useful in the understanding of the transformation of this pollution in the atmosphere.

NO_x concentrations at tailpipes vary from a few ppm to hundreds of ppm depending on engine operating conditions and efficiency of the three-way catalytic converter installed (Turns, 1996). NO can be converted to NO_2 by a number of reactions, e.g.:

$$2NO + O_2 \rightarrow 2NO_2, \quad (1)$$

$$NO + O_3 \rightarrow NO_2 + O_2, \quad (2)$$

$$NO + HO_2 \rightarrow NO_2 + OH, \quad (3)$$

$$NO + RO_2 \rightarrow NO_2 + RO, \quad (4)$$

...

Reaction (1) is not important in NO_2 production compared to the O_3 and free radical reactions (Finlayson-Pitts and Pitts Jr., 2000), thus the ubiquitous O_3 in the atmosphere makes it difficult to determine the amount of NO_2 directly emitted from vehicles.

Reaction (2) is a fast reaction. The formation rate of NO_2 at 298 K by Reaction (2) is estimated to be 27% min^{-1} when O_3 is constant at 10 ppb. In urban area, it is reported that HO_2 concentration was as high as 0.01 ppb (Hard et al., 1992). The formation rate of NO_2 at 298 K by Reaction (3) is about 8% min^{-1} when HO_2 is constant at 0.01 ppb. If Reaction (2) is considered alone, the amount of NO_2 generated is same as the amount of O_3 reacted. Thus, (NO_2+O_3) is conserved. (NO_2+O_3) is not conserved when the

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NO_x and volatile organic carbon (VOC) reactions such as Reactions (3–4) take place resulting in a net increase of (NO_2+O_3) (Finlayson-Pitts and Pitts Jr., 2000; So and Wang, 2003).

In this study, intensive high-resolution air pollutant concentration profiles along the entire lengths of tunnels (3.7–4.0 km in lengths) were measured to estimate the primary vehicular NO_2/NO_x ratios and to study the oxidation of NO to NO_2 in vehicle plumes. The dataset was obtained using the Mobile Real-time Air Monitoring Platform (MAP). The advantages of the tunnel measurements are:

1. Tunnels are ideal for studying vehicular emissions because the space is confined and the conditions are "controlled". The main source of air pollutants is vehicular (fresh and aged) and the NO_x concentration can reach several ppm.

2. In the absence of solar irradiation inside tunnels, O_3 concentration is inherently low and Reaction (2) is not favored. This in turn inhibits the O_3-involved reactions. However, dark chemical reactions still can take place (Cariappa et al., 1994; Finlayson-Pitts and Pitts Jr., 2000). MAP concentration profile data allow the pinpointing of the exact location of the lowest O_3 concentration where HO_2 and RO_2 free radicals are also possibly the lowest, and, therefore, primary NO_2 is expected to be more prominent. In other words, the interference due to Reactions (2–4) could be reasonably isolated.

Notably, volatile organic compounds (VOC) are still present and in high concentrations. The tunnel results were supported by a long-term on-road air quality dataset (June 2002–August 2003), also obtained using MAP, for seasonal variation and statistical average of this ratio in open space.

2. Experimental

Two tunnels in Hong Kong were used in the study. The Tate Cairn's Tunnel (TCT) is ~4 km in length while the Tai Lam Tunnel (TLT) is 3.7 km long. The roadway grade

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is negligible in both tunnels. Both tunnels have two separated tubes, two-lane tubes in TCT and three-lane in TLT. The daily average traffic volumes in TCT and TLT were 62 600 and 45 400 vehicles day^{-1}, respectively, during the experiments. The ratio of diesel trucks to passenger vehicles in TCT was 25% while it was 43% in TLT. The difference in the daily averaged traffic flow and traffic compositions between the southbound and northbound tubes in the same tunnels at the same time was negligible, although a diurnal variation of traffic flow existed.

A longitudinal ventilation system with 16 fresh air fans is used in TCT with a total fresh air supply of 6 912 000 m^3 hr^{-1}, while a semi-transverse ventilation system with 24 fresh air fans and 15 exhaust air fans is used in TLT with a total fresh air supply of 42 200 000 m^3 hr^{-1} and a total exhaust air flux of 6 140 000 m^3 hr^{-1}. Ten sets of CO, NO_2, NO and visibility sensors are used to monitor air quality in the two tunnels to control the ventilation.

The Mobile Real-time Air Monitoring Platform (MAP) developed at the Hong Kong University of Science and Technology was used to make the tunnel measurements. The details have been reported in Yao et al. (2005a, b). Stand-alone NO_x, O_3 and CO gas analyzers (API, Inc.) onboard MAP were used to obtain the concentration profiles.

The intensive tunnel measurements were made in September 2004 and May 2005. For each tunnel, the average air pollutant concentrations were computed from five runs. The NO_x monitor reported concentration data in 10–20 s spans. The speed of vehicles in the tunnels was limited to 70 km hr^{-1}. Thus, the spatial resolution of NO_2 and NO_x concentrations in the tunnels was 300 m.

The long-term on-road dataset consists of a total of 126 days (runs) of air pollutant data (June 2002–August 2003). In each run, MAP measured air pollutants nominally from 10:00 to 16:00. Traffic conditions included low traffic country roads, highways heavily traveled by heavy-duty trucks, city streets and tunnels.

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3. Results

3.1. Concentration profiles of NO and NO_2 in tunnels

There was no evident difference in the measured air pollutant concentrations between the southbound and the northbound tubes of TCT at similar times (rush hour or non-rush hour in daytime). Averages of the two tubes of TCT will be discussed instead of treating them separately. However, a large difference was found in TLT between the two tubes probably due to fuels from different sources used in the vehicles. Ultra low sulfur-content fuel (<150 ppm for gasoline and <50 ppm for diesel) is used in Hong Kong while the sulfur-content is <800 ppm for gasoline and <2000 ppm for diesel across the border north of Hong Kong. When heavy-duty trucks return to Hong Kong, the tanks are usually filled with high sulfur-content fuels and the southbound TLT is the preferred truck route. The use of high sulfur-content fuels will eventually poison three-way catalytic converters resulting in high NO_x emissions. The average concentration profiles, with standard deviations, of NO and NO_2 in the tunnels are shown in Figs. 1a–c. The entrance of the tunnel is marked by 0 (zero) m.

Pollutant concentration profiles inside the tunnels are influenced by a combination of factors: sources, sinks, piston effect and mechanical ventilation. NO peak appeared at about two-thirds the length of the tunnel from the entrance, while lower NO concentrations occurred at the two ends as shown in Figs. 1a–c. It is reported that the peaks of NO_x and the less reactive species CO and SO_2 occurred almost at the same location in TCT (Yao et al., 2005). NO_2 peak was detected at the exit section while the lowest value occurred at 500–1500 m from the entrance section. The lowest O_3 occurred at 1000–2000 m from the entrance section and relatively high O_3 was detected at both ends (entrainment effect) as shown in Figs. 2a–c.

The respective NO, NO_2 and O_3 concentration profiles in the tunnels were similar only the concentrations varied. There are three sources of NO in the tunnels: ambient NO in front of the vehicle carried into the tunnels due to piston effect and relatively low background NO in fresh air brought into the tunnels due to mechanical ventilation,

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and primary NO emitted from vehicles. One of the NO sinks is the transformation of NO to NO_2 in the atmosphere. The highest NO_x concentration was detected in the southbound tube of the TLT tunnel and it will be used in the discussion to follow. The maximum NO concentration (averaged value of five runs) was 2720 ppb and the average for the entire tunnel was 1331 ppb. The maximum NO_2 concentration (averaged value of five runs) was only 82 ppb and the average for the entire tunnel was 52 ppb. NO concentration at the entrance was 214 ppb and was assumed to be the background NO outside of the tunnel. The high NO inside the tunnel indicates overwhelming contribution from fresh vehicle emissions.

There are four main NO_2 sources in the tunnel: ambient NO_2 in front of the vehicle carried into the tunnels due to piston effect and relatively low background NO_2 in fresh air brought into the tunnels due to mechanical ventilation, primary NO_2 emitted from vehicles, and transformed NO_2 from NO. NO_2 concentration at the entrance of the southbound tube of TLT was 50 ppb. The lowest NO_2 (28 ppb) occurred at ~1500 m. The higher NO_2 at the entrance was likely ambient NO_2 carried into the tunnel by piston effect. Furthermore, fresh air brought into the tunnel by mechanical ventilation would also contribute to the NO_2 concentration as well as diluting the emitted and transformed NO_2 in the entrance section. For the rest of the tunnel, accumulation of emitted NO_2 and transformed NO_2 overcame the dilution processes and NO_2 increased gradually to 82 ppb (an increase of 54 ppb).

In the 1500 to 2500 m section, both NO_x and NO_2 increased. From 2500 m to the exit section where end effect was experienced, NO_2 increased by 44 ppb while NO_x decreased by 1777 ppb. The concentration of the NO_2 originating from primary vehicular emissions in this section was expected to decrease with decreasing NO_x. The 44 ppb increase in NO_2 in this section was probably due to transformed NO_2. The wind speed inside the tunnel was estimated to be ~5 m s^{-1} based on ventilation data. It took the air ~240 s to travel from the 2500 m point to the exit of the tunnel. It should be noted that the average air mass movement in tunnels is much lower than vehicular speed. The NO concentration decreased from 2720 to 1001 ppb in this distance. A rough cal-

culation showed that NO_2 produced by Reaction (1) was ~6 ppb. The 44 ppb increase in NO_2 measured suggests that 86% of the NO_2 was not transformed by Reaction (1).

As shown in Fig. 3, the increase of (NO_2+O_3) from ~1500 m to the exit section of the tunnel by 63 ppb (to 96 ppb), suggests that the transformation reactions such as Reactions (3–4) were likely to be important.

3.2. The NO_2/NO_x ratio profiles in the tunnels

Higher NO_2/NO_x values were found at both ends of both tunnels as shown in Figs. 2a–c. The lowest NO_2/NO_x ratios (2 to 6%) with NO concentrations at 1006–2128 ppb, occurred at ~1000 m from the entrance of TCT and at ~2000 m in both tubes of TLT. These lowest NO_2/NO_x ratios should be closer to the primary vehicular NO_2/NO_x ratios. The apparent two-fold range in the lowest NO_2/NO_x ratio was probably due to vehicular conditions and ambient concentrations of these gases.

In the literature, higher primary NO_2/NO_x ratios have been reported for diesel engines than gasoline engines, and vehicles with higher NO_x emissions (Hilliard and Wheeler, 1979; Lenner and Lindqvist, 1983; Cariappa et al., 1994; Soltic and Weilenmann, 2004).

The NO_x concentration in the southbound tube of TLT doubled that in the northbound tube. As presented in the Experimental Section, traffic composition, traffic flow and ventilation in both tubes of TLT are similar. The difference in NO_x concentration between the two tubes was probably caused by low quality fuels used by the cross-border vehicles in the southbound tube of TLT. The primary vehicular NO_2/NO_x ratio was expected to be higher in the southbound tube of TLT.

TLT has more diesel vehicles, lower traffic flow and better ventilation system than TCT. The southbound tube of TLT had higher NO_x concentration than TCT, suggesting higher emission of this pollutant. Thus, the primary vehicular NO_2/NO_x ratio was expected to be higher in the southbound tube of TLT than TCT. The lowest NO_2/NO_x ratio at 2% detected in the southbound TLT was the upper limit of the average on-road primary vehicular NO_2/NO_x ratio in the two tunnels (four tubes) studied.

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Ventilated fresh air diluted all gases inside the tunnel and was probably responsible for NO_2 concentrations to decrease from the entrance to the minima. In this distance in the tunnels, the NO_2/NO_x ratio gradually decreased by 12–15%. It should be noted that the NO_2/NO_x ratio in ventilated air was higher than that in the vehicle exhaust, therefore the intrusion of ventilated air would increase the NO_2/NO_x ratio. Any oxidation of NO to NO_2 by Reactions (2–4) would also increase this ratio. This ratio increased by 8–13% from where the lowest NO_2 was detected to the exit section.

3.3. The NO_2/NO_x on-road ratio

In the open air, it is reported that NO_x concentration in vehicle plumes substantially decreased in a few minutes when vehicle plumes are diluted by ambient air (Seakins et al., 2002). During dilution processes, rapid transformation of NO to NO_2 can take place due to Reactions (2–4). Higher dilution extent of vehicle plumes by ambient air would yield a higher NO_2/NO_x ratio. We further examine the NO_2/NO_x ratio in on-road open air. The on-road NO_x concentrations are shown in Fig. 4a and the annual average was 251 ppb, indicating that the dilution extent of vehicle plumes by ambient air in open air is better than in tunnels. As expected, the annual average NO_2/NO_x ratio was as high as 28% (Fig. 4b). Using the primary vehicular NO_2/NO_x ratio determined in the tunnels (2%), background and/or transformed NO_2 would account for 26% of the ratio in open air. In addition, there was no significant seasonal variation of the NO_2/NO_x ratio in the long-term on-road dataset as shown in Fig. 4b. The correlation between NO_x and the ratio is poor (R^2=0.25).

Twenty-four-hour experiments conducted on 14–15 August 2002 was used as an example to examine the contribution of different sources and processes to the on-road NO_2/NO_x ratios. As shown in Figs. 5a, b, fairly good correlations exist between NO_2 and NO_x when NO_x was <300 ppb. The slopes are 0.35 and 0.32 in daytime and nighttime, respectively. However, for NO_x>300 ppb, correlations between NO_2 and NO_x are poor with the corresponding slopes at 0.21 and 0.19. Compared to the primary vehicular NO_2/NO_x ratio of 2%, background NO_2 or transformed NO_2 determined the

on-road NO_2/NO_x ratio regardless of time.

A simple model was used to isolate the emitted and transformed NO_2 from background NO_2. Assume that the measured NO_2 in vehicle plumes consisted of background NO_2($[NO_2]_{BG}$), primary vehicle emitted NO_2 ($[NO_2]_P$) and transformed NO_2 ($[NO_2]_T$)

Define NO_x<50 ppb to be background and NO_x≥300 ppb to be vehicle plume. For NO_x<50 ppb, the average NO_x, NO_2 and O_3 concentrations were 24, 12 and 37 ppb, respectively, and for NO_x≥300 ppb, they were 513, 115 and 6 ppb, respectively. In nighttime, the average NO_x, NO_2 and O_3 concentrations for NO_x<50 ppb were 25, 13 and 10 ppb, respectively, while for NO_x≥300 ppb, they were 463, 87 and 2 ppb.

The following was used to estimate $[NO_2]_P+[NO_2]_T$ from MAP data:

$$[NO_2]_P + [NO_2]_T = [NO_2]_{VP} - [NO_2]_{BG}. \quad (5)$$

where $[NO_2]_{VP}$ is the NO_2 measured in vehicle plumes.

The amount of NO_2 ($[NO_2]_{R2}$) produced by Reaction (2) alone (no VOC involved in O_3 formation) is:

$$[NO_2]_{R2} = [O_3]_{BG} - [O_3]_{VP}, \quad (6)$$

where $[O_3]_{BG}$ is O_3 concentration when NO_x<50 ppb, and $[O_3]_{VP}$ is O_3 concentration in vehicle plumes. Substituted Reaction (6) into (5),

$$[NO_2]_P + [NO_2]_T - [NO_2]_{R2} = [NO_2]_{VP} - [NO_2]_{BG} - ([O_3]_{BG} - [O_3]_{VP}), \quad (7)$$

Define:

$$\text{Ratio1} = ([NO_2]_P + [NO_2]_T)/([NO_x]_{VP} - [NO_x]_{BG}), \quad (8)$$

$$\text{Ratio2} = ([NO_2]_P + [NO_2]_T - [NO_2]_{R2})/([NO_x]_{VP} - [NO_x]_{BG}). \quad (9)$$

Ratio1 is the ratio of primarily emitted and transformed NO_2 to primarily emitted NO_x and they were 21% for daytime and 17% for nighttime. Ratio2 is the ratio of primarily

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emitted and transformed NO_2 excluding Reaction (2) to primarily emitted NO_x and it was 15% for both day and night. Considered the primary vehicular NO_2/NO_x ratio of 2%, the transformed NO_2 excluding Reaction (2) contributed 13% to Ratio2, however, the contribution cannot be isolated from the primary vehicular contribution based on the data measured in the open air alone.

Overall, the data measured in the open air may not be applicable for estimating primary vehicular NO_2/NO_x ratios due to the existence of background and/or transformed NO_2.

4. Summary

Intensive long-tunnel (3.7–4 km) air pollutant concentration profiles along the entire lengths of tunnels in Hong Kong were used to estimate primary vehicular NO_2/NO_x ratios under depleted O_3 conditions. The lowest NO_2/NO_x values were found in the middle of the tunnels at 2–6%. At the entrance sections of the tunnels, higher ambient NO_2 concentrations caused the NO_2/NO_x ratios to be as high as 20%. In the exit sections of the tunnels, more O_3 became available to rapidly oxidize NO to NO_2 and the NO_2/NO_x ratio was as high as 19%. These values, however, were still lower than the average annual NO_2/NO_x ratio on roads in Hong Kong at 28%, where atmospheric transformation was expected to be much more significant. Ozone and free radicals are ubiquitous and they interfere with the measurement of NO directly emitted from vehicles. Long tunnels, shielded from ultraviolet irradiation and have a constant flow of automobiles, provide an ideal and convenient laboratory for the assessment of primarily emitted vehicular NO_2 as demonstrated in this paper. Mobile platform measurements provide concentration profiles necessary for such analysis.

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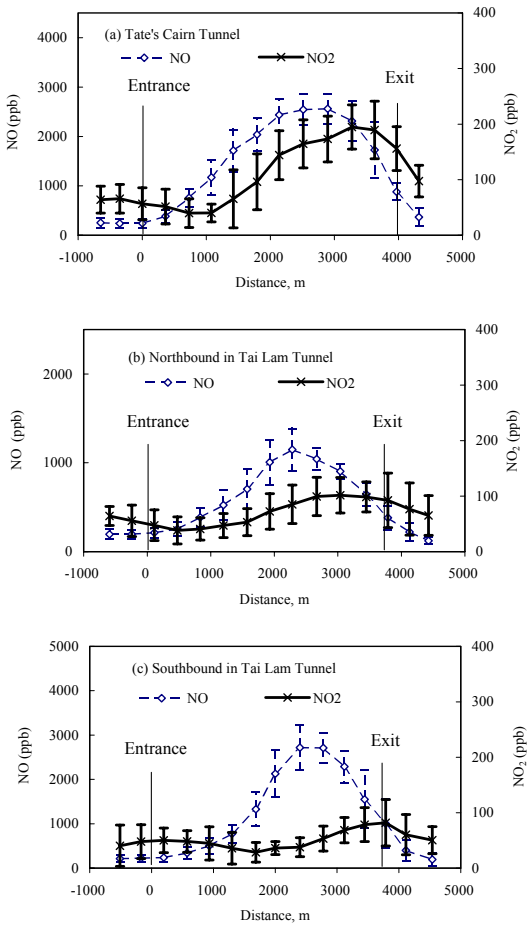

Fig. 1. Profiles of NO and NO_2 in tunnels.

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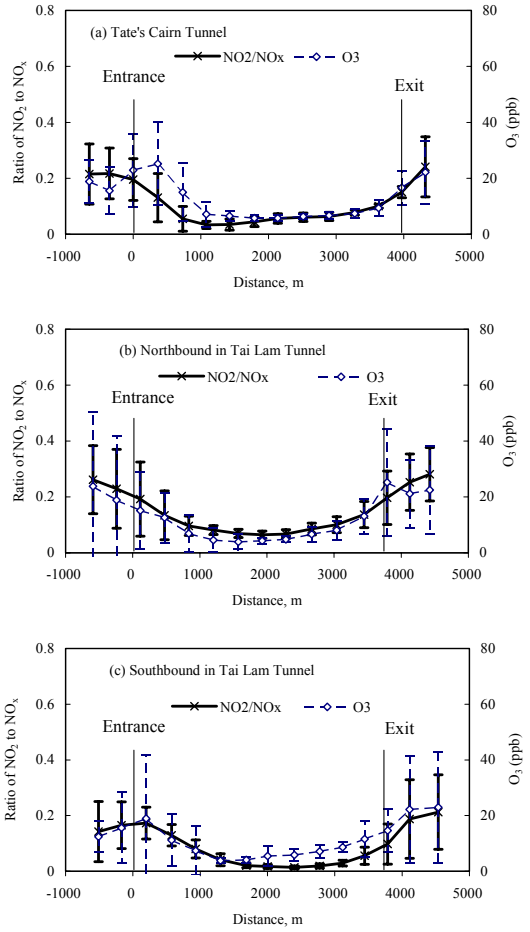

Fig. 2. Profiles of NO_2/NO_x and O_3 in tunnels.

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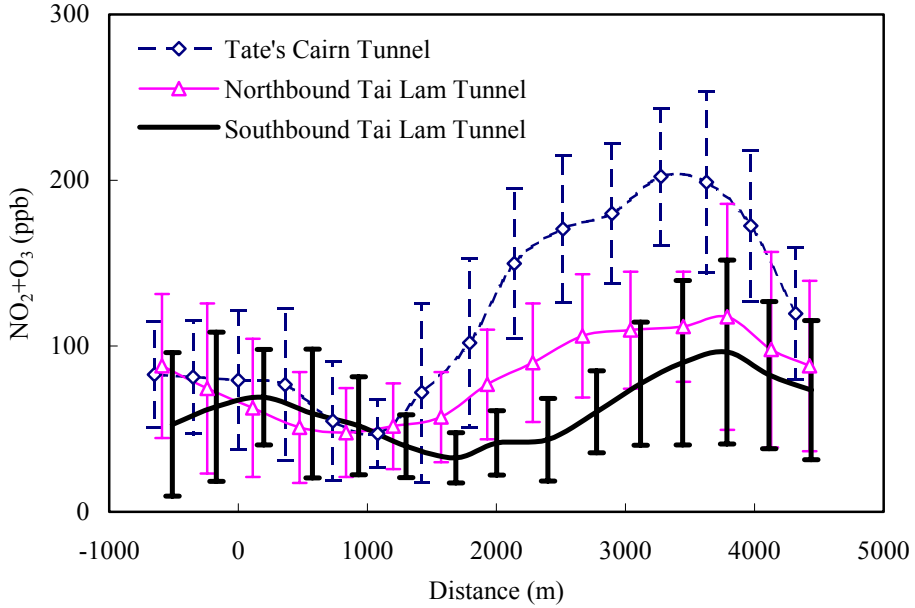

Fig. 3. (NO_2+O_3) profiles in tunnels.

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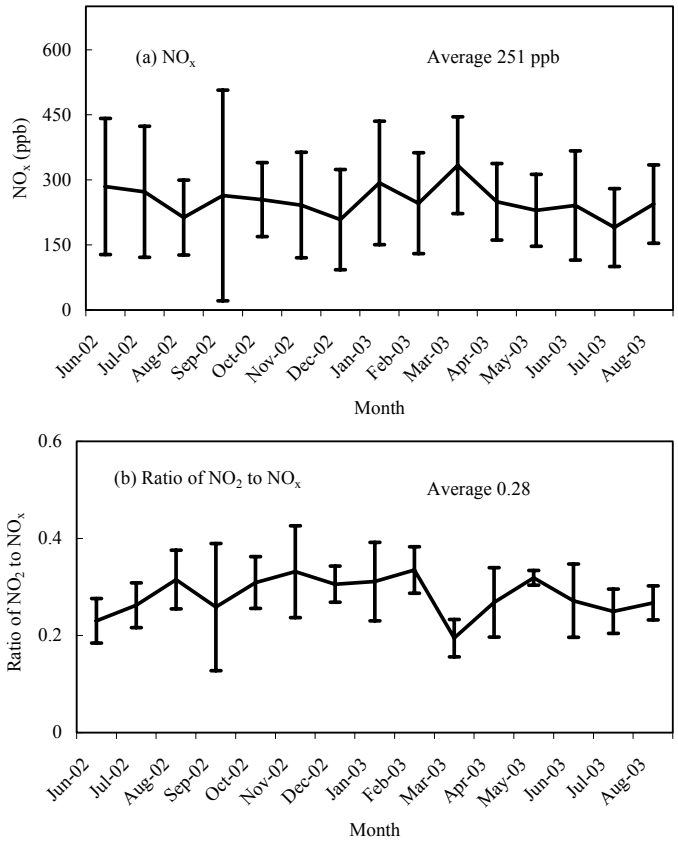

Fig. 4. Seasonal variations of NO_2/NO_x and NO_x in on-road vehicle plumes.

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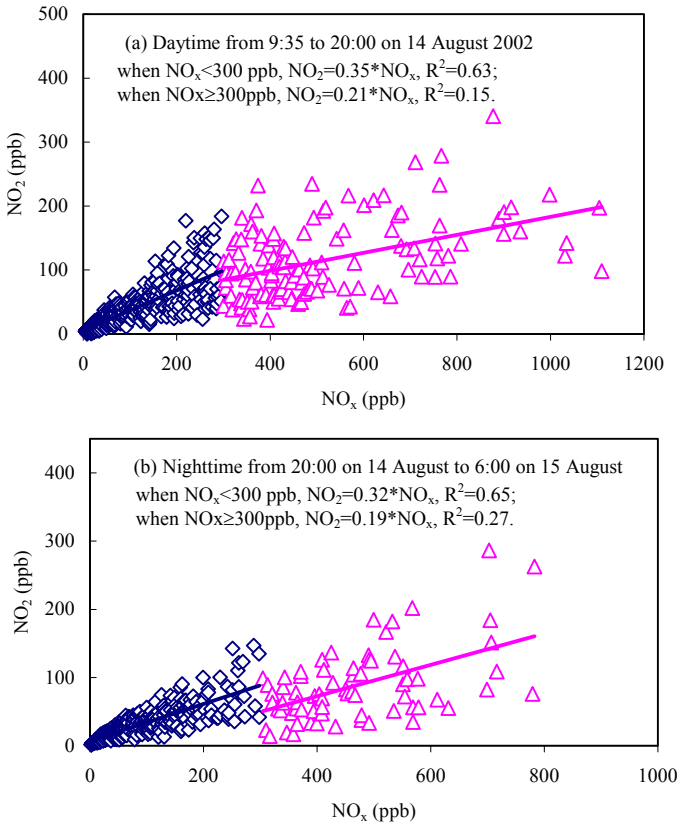

Fig. 5. Correlations between NO_2 and NO_x in daytime and nighttime measurements.

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