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Modeling mobile source emissions in presence of stationary sources

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Abstract

The impact of oxides of nitrogen (NO_x) emissions from motor vehicles to the air quality in city-state Singapore is analyzed using AIRVIRO, a regional scale dispersion model developed by the Swedish Meteorological and Hydrological Institute. In a predominantly urban location like Singapore, it is difficult to separate out the contribution of pollutants from mobile and point sources at different locations. In this work, a new approach is used by first modeling only the impact of point and area sources and then overlaying the traffic impact on air quality at different locations. Monthly scenario simulations are run with point, area and traffic sources of emissions for the Gaussian model validation. Street Canyon modeling is used for street segments surrounded by buildings on either side. A simplified photochemical model, which takes into account NO_x undergoing chemical transformations in the urban atmosphere, is used to account for variations in NO_x and ozone levels with respect to traffic data. The diurnal variation of NO_x concentration levels is studied as a function of ozone levels at site, hourly traffic counts and meteorological parameters. The impact on ambient air quality within the breathing zone of the public from mobile sources, is found to be about 40% at urban stations although overall emissions from mobile sources is only 24%. The proposed approach appears to predict the variations in NO_x as a function of traffic and meteorological conditions. © 2000 Elsevier Science B.V. All rights reserved.

Keywords: Mobile sources; Gaussian modeling; Street canyon modeling; NO_x; Ozone; Photochemical transformations

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1. Introduction

Urban air pollution problems and issues are of increasing concern to urban planners and policy makers. Air quality in most urban centers can reach levels high enough to cause substantial health impacts. Urban areas have several sources of emissions, depending on the type of activities. The major ones are point, area and mobile sources. Mobile source emissions are important since they are in the close vicinity of the city populace. Vehicle emissions are directly related to the variations in the traffic flow pattern, which vary in location and time. The establishment of time variant nature of emissions is difficult since it requires accurate dynamic emission database (EDB).

Key literature analysis of studies (Beaton et al. [4]; Bose [5]; Cernuschi et al. [6]; Cooper [7]; Derwent et al. [9]; Joumard et al. [17]; Lawson et al. [19]; Mitsoulis et al. [22]; Onursal and Gautam [23]; Riveros et al. [24]; Stein and Toselli [28]; Sturm et al. [30]) relating to traffic pollution in urban centers indicate that studies concerning detailed impact of vehicle emissions on the ambient air quality are few. This is due to the complexity in organizing and integrating information on:

- Emission of pollutants to the atmosphere to form a dynamic EDB,
- Meteorological conditions,
- Processes affecting pollutant concentration spatially at different points in time.

The objective of this work is to model the impact of oxides of nitrogen (NO_x) emissions from traffic in Singapore at urban locations and at an expressway site. The oxides of nitrogen (NO_x) emissions from mobile sources contribute to a significant proportion of urban pollution along with point sources. Since the ground level emissions are closer to the breathing zone of the public, their impact on human population is expected to be larger than the point source emissions of an equivalent amount. In a predominantly urban location like Singapore, it is difficult to isolate the contribution of mobile sources from point sources at different locations. Hence, a methodology needs to be developed that isolates and quantifies the impacts of stationary, line and mobile sources. In the modeling of emissions from mobile and nonmobile sources, hourly traffic counts, meteorological parameters, and photochemical conversion of NO_x undergoing chemical transformations in the presence of atmospheric ozone levels were taken into consideration.

2. Description of Singapore

The study area, Singapore, is located between latitudes 1°09′N and 1° 29′N and longitudes 103°36′E and 104°25′E. The land area of the main island, a predominantly urban location, is 42 km by 23 km with a coastline of nearly 150.5 km. The major NO_x sources are industrial stacks located on the off shore islands along the western coastline. Industrial estates are spread all over the main island and are characterized as area sources. Emissions of oxides of nitrogen (NO_x) and carbon monoxide (CO) are estimated from the fuel consumption data. About 97% of CO emissions come from

Table 1
Air pollution levels in Singapore

Pollutants	1995	1996	Air quality standards
Sulfur dioxide (mean)			80
Industrial	30	32	
Urban	19	27	
Suburban	17	22	
Nitrogen dioxide (mean)			100
Industrial	30	39	
Urban	31	33	
Suburban	18	23	

Source: YOS, 1997, Singapore. All units are in $\mu g/m^3$.

traffic. NO_x emissions from industrial sources are about 76% with the rest being from mobile ones. Industrial emissions are responsible for 65% of total emissions, whereas traffic contribution is 35%.

The impact of these emissions on the ambient air is measured at 12 urban and industrial monitoring stations and 2 roadside monitoring locations (canyons). Annual air quality values for SO_2 and NO_x for the period 1995 to 1996 (Table 1) indicate the levels well within the standards.

3. Description of model

For the current study, a regional scale dispersion model, called Indic Airviro, developed by the Swedish Meteorological and Hydrological Institute is used (SMHI [29]). This model is used by the Ministry of Environment, Singapore, through the Telemetric Air Quality Monitoring and Management System (TAQMMS) for monitoring ambient pollutant levels at the existing fifteen monitoring station sites. For this study, all of the three functional blocks of the model, namely the EDB, the Dispersion module, and the Indico for storing online ambient air quality data from the monitoring stations, are used. The ambient pollutant levels captured and stored in the Indico package are used for comparing simulated and measured levels. Modeling requires detailed collection of emission factors, static information, traffic flow variations as a function of speed and time, and a source coordinate system. All the information are stored and processed under the EDB database. The dispersion module of the model performs the dispersion of the pollutant with either measured or assumed meteorological conditions from the Indico module.

Indico uses a Gaussian dispersion model for dispersion of pollutants from stationary sources. This model considers the ground level reflection as well as the reflection from an inversion layer (SMHI [29]). The maximum ground level concentration is calculated as:

$$C(x,y,z,H) = \frac{Q}{2\pi\sigma_y\sigma_z U} e^{-\frac{y2}{2\sigma_y 2}} \left[\mathrm{e}^{-\frac{(z+h_\mathrm{e})^2}{2\sigma_z 2}} + \mathrm{e}^{-\frac{(z-h_\mathrm{e})^2}{2\sigma_z 2}} + \mathrm{e}^{-\frac{(z+h_\mathrm{e}-2h)^2}{2\sigma_z 2}} \right].$$

where C is concentration in $\mu g/m^3$, Q the emission rate in μ/m^3, U the wind speed in m/s, h_e the effective stack height, z the height above ground level, h the mixing layer height, σ_y the standard deviation in crosswind direction as a function of distance downwind, and σ_z the standard deviation in the vertical direction as a function of distance downwind.

For road sources used to describe traffic emissions, a Gaussian line source dispersion model for dispersion of pollutants from line sources is used. All roads specified are linked to a road type, which consists of tables describing the composition of different vehicles on the road as well as the time variation of traffic density over the year, month, day and hour. The road source location is built up by a number of straight links, and emissions for each link is expressed in $\mu g/m/s$.

Indico uses Street Canyon module for dispersion of NO_x from mobile sources within confined areas such as the space between tall buildings. This module, based on the ‘‘Stanford Model’’, originated from studies conducted in San Jose, USA, where measurements indicated a vertical eddy circulation whenever the wind blew perpendicular to the street directions (SMHI) [29]. The eddy circulation creates higher pollution concentrations on the leeward side of the road, because this air includes traffic exhausts. The upwind side showed much lower concentrations, as air originates from the roof level. The model has been tested and modified by many studies (EPA [11]; Stein and Toselli [28]; Yamartino and Weigand [35]). The model equations are:

$$\mathrm{CL} = \frac{KQ}{(U+0.5)\left[(x^2+z^2)^{1/2}+L_0\right]}, \tag{1}$$

$$\mathrm{CW} = \frac{KQ(H-z)}{W(U+0.5)H}, \tag{2}$$

where K is an empirical constant set to 10, Q is the total traffic emission in g/m s, U is the roof level wind speed in m/s, L_0 is a length scale of the individual cars set to 2 m, W is the effective width of the canyon, H is the typical building height in m, x is the horizontal distance from the street emission segment, z is the receptor height in m.

CL gives the concentration in the leeward side; CW gives the concentration in the windward side.

For the monthly scenario case, AIRVIRO uses a statistical approach for estimating monthly meteorological conditions. Wind direction is split in 30 classes and classified within six meteorological stability classes, namely very stable, stable, neutral negative, neutral positive, unstable and very unstable. These 180 cases thus established are computed from the meteorological conditions measured in Singapore through 1996. A critical analysis of the meteorological data revealed that for the computed stability classes, 75–80% falls in the neutral class, which are representative of overcast and cloudy conditions. For specific hourly simulations, the actual meteorological data for a given hour was used.

3.1. Traffic distribution and vehicle emission factors

Based on 1995–96 annual registration of vehicles (ROV [25]; YOS [36]) the composition of vehicle fleet on road is as shown in Fig. 1. Cars account for 55% of total

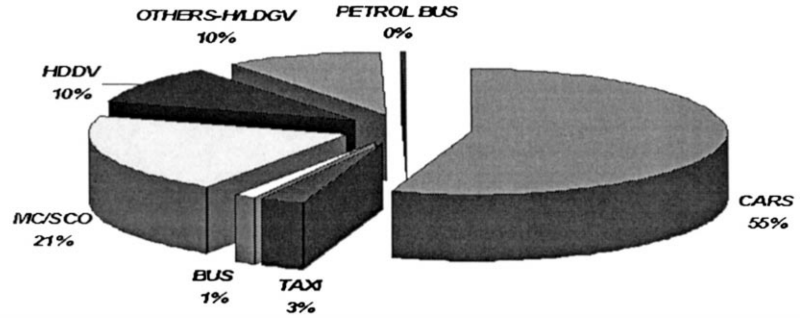

Fig. 1. Percent composition of vehicles on road (1995–1996).

vehicles. They are all petrol-driven with the majority having catalytic devices. This is followed by petrol-driven two wheelers, namely motorcycles and scooters which account for 21% of the fleet, and goods vehicles, running on petrol and diesel, each account for approximately 10% of the fleet. All the taxis run on diesel.

The traffic flow details on the expressway were measured on a 24-h basis covering weekdays and weekends. Cars constitute the largest proportion of vehicles at any given time of the day with typical morning and evening peaks. The morning peak is from 0700 until about 0945 h. The evening peak starts at around 1700 h and stretches to 2100 h. Goods vehicles follow the same peak and off-peak trends over the day, with the evening peak hours being much less than those for cars. Motorcycles show a relatively constant flow especially in the afternoon period through the evening, with a slight morning peak. During the morning peak, the flow of cars range between 5000 to 7500 vehicles/h. The off-peak hour speeds, especially at night, are nearly 18% higher than the average speed during peak traffic hours. Typical average traffic flow on any day in 1995 for the expressway was about 95,077 vehicles.

A set of emission factors was developed for the Singapore fleet. The estimates were derived based on EPA Mobile 5 A guidelines from AP 42 document on highway mobile source emission factor tabulations (AP 42[1]). Emission factors, estimated in milligrams of pollutants emitted per unit distance traveled, were based on the following sources of input:

- The zero mileage nontampered vehicle exhaust emission levels, at 31.6 kph, for each model year from 1980 until 1998 of all vehicles in different categories;
- Statistics on percentage age distribution for each vehicle category based on model years as obtained from statistical yearbooks (YOS [36]);
- Deterioration rate for each vehicle category as a function of the accumulated mileage, age and condition;
- The annual vehicle kilometers traveled by each vehicle categories, as obtained from past studies in Singapore (Ang et al. [2]; Ang et al. [3]; Chin [8]; LTA [18];ROV [25]) These were crosschecked with some isolated surveys.

The percentage distribution of different age groups (YOS [36]) and model years in the specific vehicle category were then used to compute a composite emission factor for the fleet on road in that vehicle category. Correction factors were applied, taking into account average speed of vehicles on a given road for a range of 20 to 105 kph.

4. Modeling approach

The study includes a *two-stage modeling approach*. In the *first stage*, NO_x emissions due to the impact of mobile and nonmobile sources are modeled at urban stations. NO_x here refers to nitric oxide (NO), since emission at source of NO_2 from point and area sources and those from traffic are negligible (Seinfeld and Flagan [26]). This is accomplished in four steps:

- Validation of the Gaussian point and area source model with SO_2 as a tracer and Gaussian line source model with CO as a tracer;
- Simulation of NO_x ambient air quality impact due to point and area sources at urban monitoring locations, using Gaussian point and area source models;
- Simulation of NO_x ambient air quality impact due to line sources (mobile) at urban monitoring locations with Gaussian line source model;
- Comparison of model predictions with measured NO_x values and isolating the impact due to point and traffic sources at urban locations.

In the *second stage*, NO_x at roadside locations is modeled with traffic emissions on road and background impact from point, area and neighboring traffic sources on an hourly basis. For street segments surrounded by buildings on either side, street level concentrations are simulated in a vertical plane perpendicular to the street section with Street Canyon modeling approach. This captures the impact of emissions due to traffic in the street canyon. The Gaussian line source model is used to estimate the impact due to surrounding roads at the site and the Gaussian point source model is used for the impact due to point and area sources at the site. The overall modeling is accomplished in five steps:

- Testing the validity of the Street Canyon model with CO as a tracer at the expressway location
- Simulation of NO_x due to traffic sources within the street canyon (Street Canyon model)
- Simulation of NO_x due to the major neighboring line sources (Gaussian Line source model)
- Simulation of NO_x due to point and area sources (Gaussian Point and Area source model)
- Validation of the model results with measured values captured at the roadside monitoring location (addition of all predicted values)

5. Results and discussions

5.1. NO_x at urban monitoring locations

The industrial sources of NO_x emissions are the same as those for SO_2. However with a 24% contribution from traffic, isolation of source impacts at the monitoring site becomes important. Although in a given urban area it is difficult to ascertain these impacts, a possible method is to model the emissions from nonmobile and mobile sources separately and then overlay the respective impact. This approach would provide a realistic way to model pollutants due to traffic in the presence of stationary sources.

The validity of the Gaussian model was tested with SO_2 as a tracer using monthly average concentration levels. Since SO_2 and NO_x are emitted from the same industrial sources and SO_2 is also a relatively stable pollutant (Seinfeld and Flagan [26]) compared to NO_x, this approach proved to be an ideal test bed for the applicability of Gaussian model taking into account the topographical conditions within the study area. The analysis also helped to identify selected urban monitoring stations within the study area. These stations represent the extent of air quality impact captured with the built up emissions database. For these stations, the model appears to predict the measured concentrations reasonably well (EPA [12]; Veal and Appleby [33]). The predicted and measured values agree to an accuracy of approximately $\pm 2\%$ with R^2 (Pearson Correlation coefficient) value of 0.89 for 117 monthly simulations.

The dispersion modeling was performed using a comprehensive database that contains information on stack height, exhaust gas velocity, gas temperature, pollutant concentration, stack location, and area source coordinates. To estimate the impact of NO_x from traffic at urban and roadside locations, twenty major roads with marked variations in traffic flow pattern were selected. These road sections are spatially spread across the island of Singapore but at the same time are in close vicinity of the urban and roadside monitoring stations. Though these roads capture nearly 30% of the total traffic emissions contributed by mobile sources, it is expected that the NO_x emissions from these roads are the only significant ones impacting the selected urban and roadside stations. The Gaussian line source model was run to estimate the impact of traffic at the urban locations on an average monthly concentration basis.

To validate the model for NO_x, the results of the simulations from point and area sources and those from the line sources were overlayed. These results were matched with the NO_x ambient readings at the urban stations. The model appears to predict the measured concentrations reasonably well (EPA [12]) (Fig. 2). These results also helped to predict the traffic impact contribution at different spatial locations in Singapore. On an average, 24% NO_x emissions at ground level from mobile sources appear to be responsible for nearly 40% ambient NO_x levels. The extent of the impact due to traffic sources varies at different urban locations depending on the respective vicinity to traffic sources and the variations in monthly meteorological conditions.

5.2. NO_x at roadside-expressway monitoring location

The average daily traffic volume on the expressway is 95,077 vehicles with vehicle speed of 60 kph. The expressway has an effective width of 37 m with six lanes and the

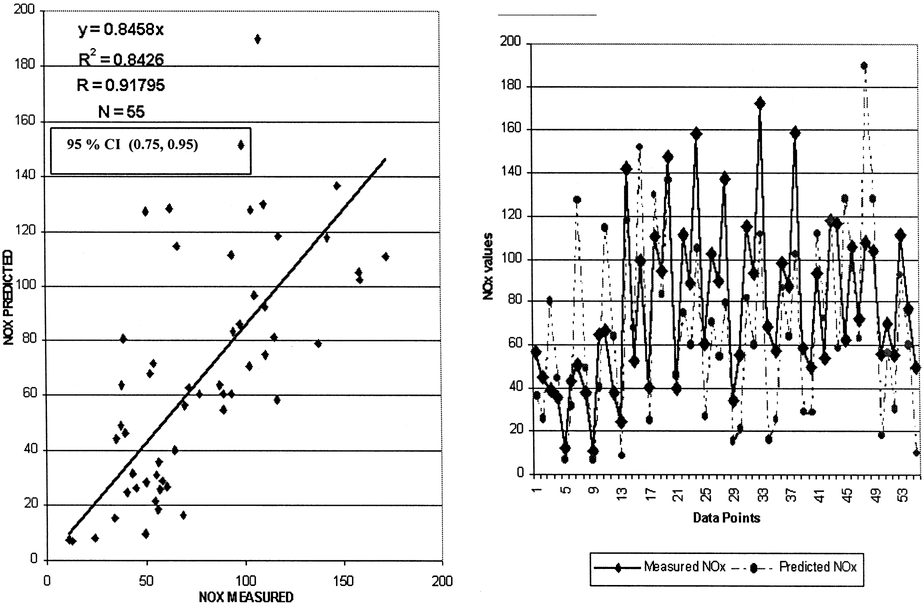

Fig. 2. NO_x (as NO) at urban stations.

canyon is at an angle of 45° to the north. The height of the buildings located on either side of the expressway is about 15 m. The canyon width to height ratio (SMHI [29]) is 1.8 for the expressway, which satisfies the criteria of 2 to 3 set by AIRVIRO model.

The applicability of the Street Canyon and Gaussian line source models were tested with hourly CO emissions. Since CO is exclusively emitted from traffic sources and relatively a stable pollutant, as compared with NO_x, it was selected as a tracer for validation of the above mentioned models. Six days in the year 1995 were chosen to capture different meteorological conditions that occur within this area. Each day, the simulations were run for 24 h. The impact of the nearby line sources was assumed as the approximate background levels at road-site location and was measured at the canyon rooftop level every hour (Stein and Toselli [28]; SMHI [29]; Yamartino and Weigand [35]). The Gaussian line source model was run for all the roads except the expressway site on which the background level needed to be estimated. This was computed on an hourly basis to capture the fluctuations in the background levels at the site as a function of hourly traffic on neighboring roads. For the expressway roadside station, the model appears to predict the measured CO concentrations reasonably well (EPA [12]; Veal and Appleby [33]). The predicted and the measured 1-h values agree to an accuracy of approximately ±19% with R^2 value of 0.67 for 99 simulations. The trend of the predicted value indicates that the model performs well (EPA [12]) and captures the changes in the meteorological characteristics every hour.

Based on the results for CO, NO_x simulations were run for the expressway street section using the same approach. To validate the model for NO_x, the results of the simulations from point and area sources, line sources and traffic within the canyon were overlayed. These results were matched with the NO_x ambient readings at the monitoring location. As shown in Fig. 3, the model overpredicts the measured concentrations significantly. Since NO_x is known both as a precursor and sink for ozone, it is important to look at other mechanisms such as photochemical transformations within the canyon.

5.3. Photochemical tranformations

The chosen model does not contain a chemical generation/depletion term and, hence, cannot be used for photochemical transformations. Since the model predictions for NO_x at urban locations on a monthly basis and for CO on an hourly basis are good, the relatively poorer predictions for NO_x within the canyon section could be attributed to photochemical reactions. The annual diurnal variation of ozone at site indicates ozone hours (sunlight hours) to be between 0900 and 1900 h. The correlation of predicted NO_x (NO) and measured values during the ozone hours is very poor as shown in Fig. 4. In addition, the trend of NO_x levels, such as decrease of NO_x with decrease in traffic are not captured very well. An examination of similar trends during non-ozone hours (Fig. 5) appears to be better than the ones seen from Fig. 4. This could be attributed to photochemical transformations within the Street Canyon, since NO_x is a known precursor of ozone formation (EPA [13]; Guardani et al. [14]; Glavas [15]; Hertel et al. [16]; Luecken et al. [20]; Mendez and Trevino [21]; Seinfeld and Pandis [27]; Strauss [31]; Uwe et al. [32]; Vukovich et al. [34]; Zannetti [37]).

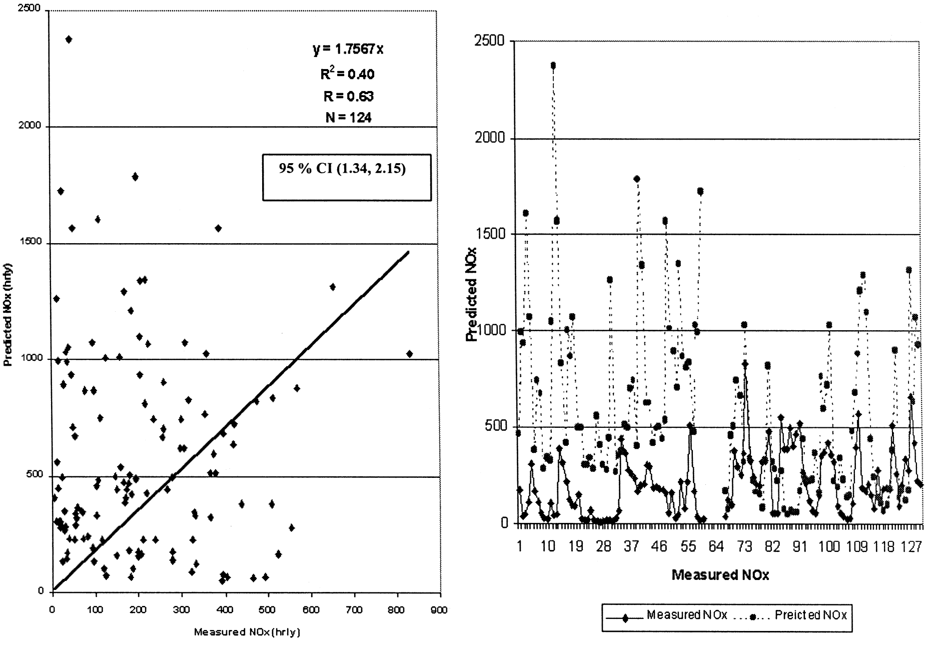

Fig. 3. NO_x (as NO) at expressway site.

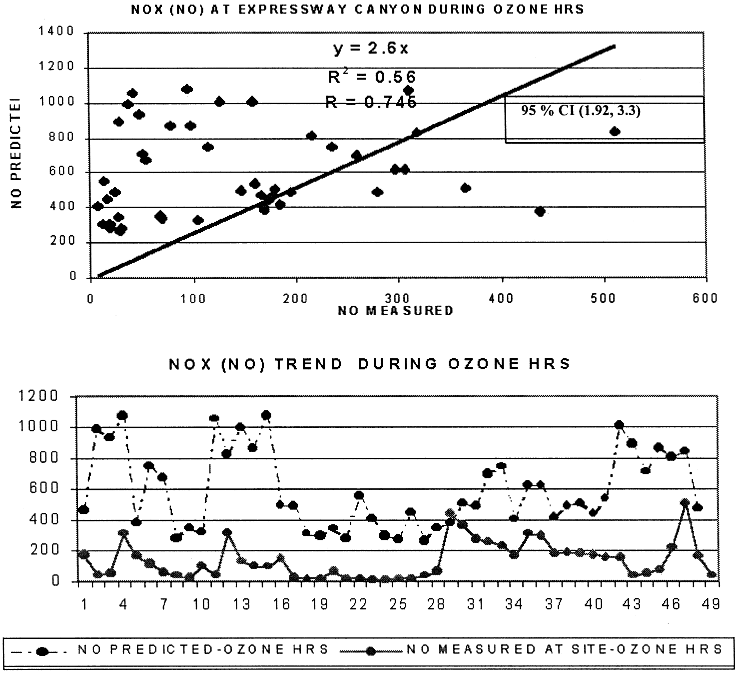

Fig. 4.

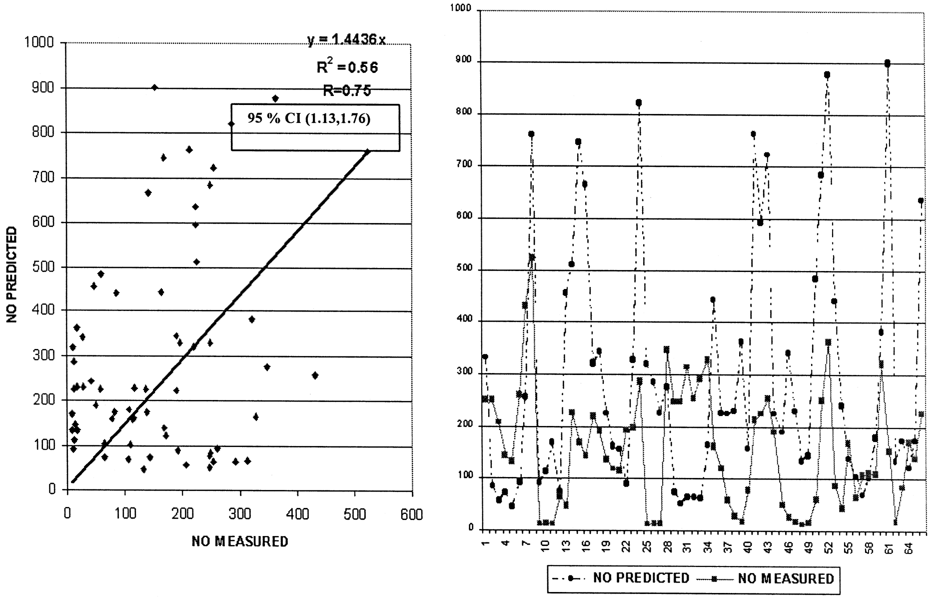

Fig. 5. NO_x (NO) at Expressway Canyon in non-ozone hours.

Photochemical oxidants are products of atmospheric reactions originating mainly from volatile organic compounds and NO_x emissions. The formation of these oxidants are a complex function of emissions and meteorological patterns. The formation of these oxidants or otherwise the concentration levels of ozone (O_3) has been widely studied at several locations (Derwent and Middleton [10]; EPA [13]; Guardani et al. [14]; Glavas [15]; Hertel et al. [16]; Luecken et al. [20]; Mendez and Trevino [21]; Seinfeld and Pandis [27]; Uwe et al. [32]; Vukovich et al. [34]; Zannetti [37]). The three primary reactions in a series of different reaction mechanisms involved are as shown below:

$$NO_2 + h\upsilon \rightarrow NO + O, \tag{3}$$

$$O + O_2 + M \rightarrow O_3 + M, \tag{4}$$

$$O_3 + NO \rightarrow NO_2 + O_2. \tag{5}$$

The most important reaction for formation of NO_2 within the street canyon is the reaction between NO and O_3 (Hertel et al. [16]). Consequently, large scale, concentrated release of NO, typical in street canyons consume O_3 (EPA [13]; Hertel et al. [16]). A comparison of the annual diurnal variation of O_3 levels at the site with a rural location and another roadside location within the study area further supports the above reasoning (Fig. 6). The difference in time lag for the rise in the ozone concentration in the morning at the main road and expressway canyon vs. the rural site is indicative of a possible photochemical reaction. The levels of O_3 at the rural site are the maximum. This is followed by the main road site which has nearly half the daily total traffic count when compared with the expressway site. Of the three sites, the peak level of ozone at the

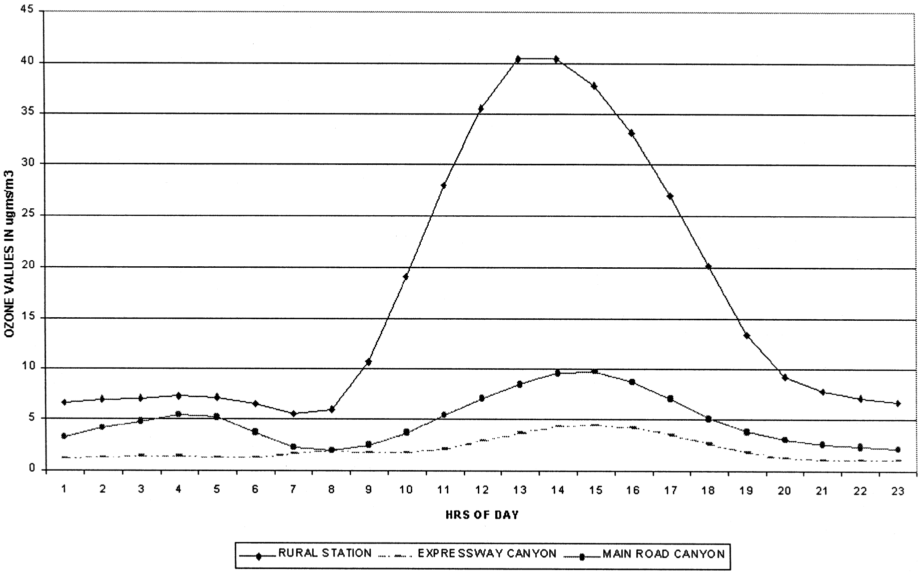

Fig. 6. Annual diurnal variation of ozone at three sites.

expressway site is the lowest but the level of emission of NO_x (NO) is the highest. This trend indicates that the ozone is being quenched at the site due to the higher rate of emission of NO. It thus establishes the importance of the reaction involving NO and O_3 within canyon sections. The resulting trend indicates that the measured values for NO_x (NO) at the canyon section on an hourly basis are affected by the photochemical transformations, which result in substantial removal of O_3 and NO. Although there could be several other mechanisms responsible for the photochemical transformations (EPA [13]; Guardani et al. [14]; Glavas [15]; Hertel et al. [16]; Luecken et al. [20]; Mendez and Trevino [21]; Seinfeld and Pandis [27]; Uwe et al. [32]; Vukovich et al. [34]; Zannetti [37]), it was not possible to include them due to the availability of only NO_x and O_3 measurements at the expressway site.

In this work, the following key photochemical reaction for ozone hours is included to account for variations in NO_x and O_3 levels with respect to traffic and meteorological data every hour:

$$\frac{d[NO]}{dt} = -k[NO][O_3], \tag{6}$$

where the value of k is $2.2 \times 10^{-12} \exp(-1430/T)$ cm^3 $molecule^{-1}$ s^{-1}, and T is the temperature in K, Temperature readings, measured at the meteorological station, are used every hour; NO and O_3 values used are hourly averages based on average 5-min measured values.

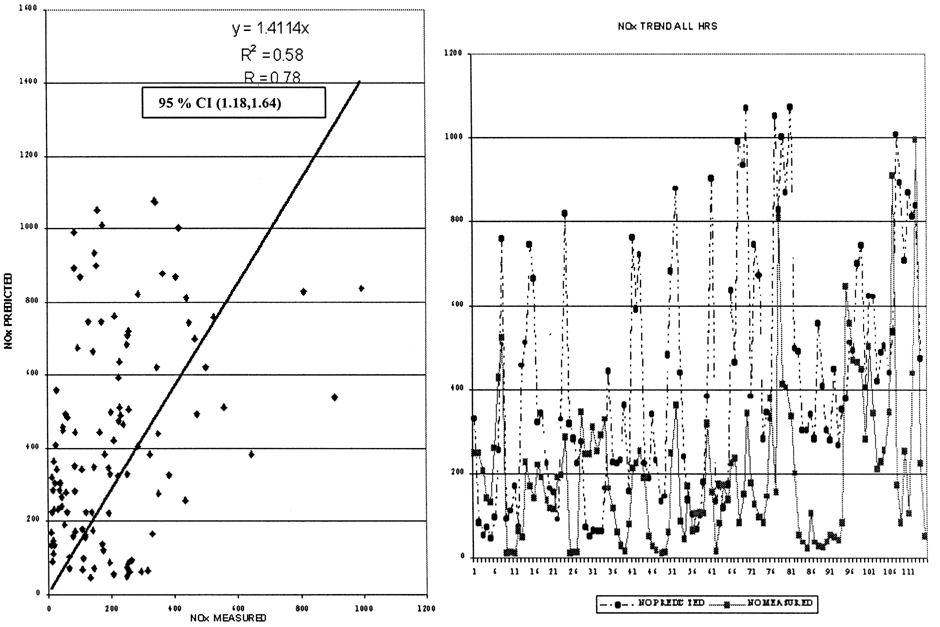

Fig. 7. NO_x for all hours (ozone hours with simplified photochemical model).

This simplified photochemical model was tested for the ozone hours only at the site. The measured NO_x (NO) readings are recomputed to account for any reduction in the concentration levels due to the ozone concentration within the canyon every hour. These recomputed values of NO_x are then used as measured values for model validation. Fig. 7 shows agreement between predicted and measured values of NO_x, if photochemical transformations are taken into account. The improved correlation is a clear indication of the validity of the model. In addition, the model parameters obtained from Fig. 5 (non-ozone hours) and Fig. 7 (all hours with ozone hours using photochemical model) match well. This is a further indication that the use of the photochemical reaction, as proposed in this work, is reasonable. It is important to note that the model-predicted NO concentrations still differ from measured values. This difference could be a consequence of not including other chemicals in the model such as hydrocarbons, RO_2 and HO_2 groups, but concentrations of these compounds have not been measured in the canyon sections.

6. Conclusions

AIRVIRO, a regional scale dispersion model was used to estimate the air quality impact of oxides of nitrogen (NO_x) from vehicular and stationary sources in Singapore. A new approach was developed to isolate the impact of mobile and nonmobile sources at urban locations. This involved the successive modeling of point, area, and mobile sources. The impact on ambient air quality within the breathing zone of the public from mobile sources at urban stations was found to be about 40%, although overall emissions from mobile sources is only 24%. This was seen to vary at different urban locations depending on their respective vicinity to traffic sources and was also seen to be affected by variations in monthly meteorological conditions. For street segments surrounded by buildings on either side, street level concentrations were simulated in a vertical plane perpendicular to the street section with the Street Canyon modeling approach. A simplified photochemical model taking in to account NO_x chemical transformations during ozone hours was used. Application of the model helped in improving the overall NO_x trend predicted at the busy expressway canyon and thus showed the importance of the reaction between NO and O_3 in canyons where large traffic volumes are expected. The effect of other photochemical mechanisms, local dilution effects, (Uwe et al. [32]) and strong scavenging effect due to urban scale transport (EPA [13]) require further study.

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