Passive Sampling of Ambient Nitrogen Dioxide at Toll Plazas in Malaysia

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Abstract

With the increasing trend of development and industrialization in Malaysia, air pollution has become an inevitable part of the process, resulting from increased vehicular activities and industrial processes. Toll operators are potentially exposed to high levels of air pollutants from working in proximity to traffic pollution sources, thereby increasing their risk of health defects associated with air pollution exposure. This study assessed the levels of Nitrogen dioxide (NO_2) toll operators are exposed to, considering the influences traffic density and meteorological factors. This is intended to serve as an indicator of the cumulative pollution emanating from the combustion process of vehicles at toll plazas. Using Passive diffusion samplers saturated with Triethanolamine (TEA), the weekly indoor and outdoor NO_2 concentrations at tollbooth were measured at Sungai Besi (SB), Kajang (KJ) and Putra-Makhota (PM) toll Plazas, to capture the varying traffic densities of 138,000, 65,000 and 24,100 vehicles/day respectively. The results showed that NO_2 concentrations increase with traffic densities, and indoor NO_2 concentrations correlated highly with outdoor NO_2 concentrations (R^2: SB = 0.767, KJ = 0.689 and PM = 0.877). The indoor/outdoor NO_2 ratio varied from 0.7 to 1.2 for all toll booths, suggesting pollution control in-efficiency, caused by technical and behavioural factors. Also, meteorological factors had no significant effect on nitrogen dioxide concentrations, contrary to previous studies, which is likely due to the far distance between tool plazas and meteorological stations. Furthermore, the NO_2 concentrations reported in this study were higher in comparison to weekly standards adopted in Germany (0.032 ppm) and previous literature. Therefore, toll operators are potentially exposed to high levels of pollution, and we advise that toll operators to wear pollution protection gears to reduce the risk of exposure, and the ventilation systems and mitigation measure (air curtain) reviewed to assess its efficiency.

1. Introduction

Air pollution in Malaysia emanates mainly from three sources, *i.e.* open burning, stationary and mobile sources, with mobile sources regarded as the highest contributor to air pollution, and accounting for approximately 70% - 75% of total air pollution for the past five (5) decades [1]. Vehicular activities such as deceleration, idling and acceleration result in the accumulation of air pollutants during travel, and studies have shown significant concentrations of air pollutants at 50 m to 200 m away from traffic interception with high traffic densities are evident, and downstream of traffic interception [2] [3].

Toll booths can be described as a worst-case scenario of air pollution exposure, as operators are exposed to a combination of pollutants including Volatile Organic Compounds (VOCs), BTEX (Benzene, toluene, ethylbenzene, and xylene), polycyclic aromatic hydrocarbons [4], ultrafine particles [5], Organic Carbon [6], and Carbon monoxide [7] and Nitrogen Dioxide [3].

Traffic congestions at toll plazas (collection of toll booths) trigger pollutant emission due to acceleration, deceleration and idle time of vehicles during toll collection [8]. Long queues at manual system (cash) tollbooths have been found to result in service delays of about 14.5 sec/vehicle, leading to vehicles emitting pollutants such as Particulate Matter (PM), Nitrogen oxide (NO_2), Ozone (O_3), Carbon monoxide (CO), Volatile Organic Compounds (VOCs) and Particulate Aromatic Hydrocarbons (PAH) which are all detrimental to human health [4] [5] [6]. Bartin *et al.* [9] also revealed that electronic (automatic) tolls experience lesser pollution than manual tolls over a short-term period, due to reduced idle time and vehicular activities.

1.1. Nitrogen Dioxide Formulation during Combustion

Nitrogen dioxide is the focus pollutant for this study, and can be defined as a colourless, odourless, irritating gas formed when oxides of Nitrogen (NOx) generated during combustion reacts with Oxygen and Hydroxide at high temperature [10] [11] and is defined by the Zeldovich equations:

$$\left.\begin{array}{l} N_2 + O \rightarrow NO + N \\ N + O_2 \rightarrow NO + O \\ N + OH \rightarrow NO + H \end{array}\right\}. \tag{1}$$
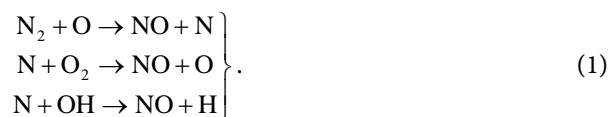
Nitric oxide (NO) then reacts readily with atmospheric ozone (O_3), to form Nitrogen dioxide (NO_2) and oxygen (O_2)

$$NO + O_3 \rightarrow NO_2 + O_2.$$
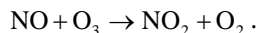
1.2. Effects of Nitrogen Dioxide Exposure

Nitrogen dioxide is known to have long and short-term effects on individuals exposed to it, due to its oxidation capacity [12]. Mücke and Wagner [13] reported that even NO_2 at low concentrations can affect respiratory tract by increasing respiratory resistance, changing pulmonary functions, decreasing defence against disease and causing morphological damage to lungs. Short-term exposures to about 100 parts per billion (ppb) were found to be harmful to rats in an experimental study [14], while long-term effects have been perceived to reduce immunity, leading to respiratory infection [15]. Using NO_2 as an indicator pollutant in this study would help us determine whether the levels of air pollution concentration present in a particular region may cause adverse health impacts, and make recommendations for pollution control, considering that NO_2 emanated from combustion is accompanied by other pollutants.

1.3. Factors that Influence NO₂ Concentration

Pollutants concentrations are mostly influenced by the source strength, intensity and meteorological factors. The strength of the emitting source determines to a large extent the concentration of the pollutant, and in this case, mobile sources (vehicles) are the primary source of NO_2 at the toll plazas. This is determined by the density of vehicles, which is directly proportional to pollution concentration, *i.e.* high traffic density implies high pollution and vice versa [16]. This was evident in Azeez *et al.* [17], where the highest concentrations of nitrogen dioxide were recorded at locations with highest traffic volume. Also, Glasius *et al.* [18] revealed that concentration of NO_2 near busy highways with high traffic was 2.9 times more than that recorded in the background areas impacted less by traffic.

Meteorological factors such as wind, precipitation, humidity and temperature, influence the dispersion, deposition, transportation and transformation of nitrogen dioxide. Wind direction defines the direction of pollutant spatial distribution, while wind speed dictates the dispersion and deposition rate [19] [20]. High wind speed tends to reduce NO_2 concentration by aiding pollutant transportation and dispersion and vice versa in the direction of up-wind [21] [22]. Precipitation reduces NO_2 concentration by enabling pollutant deposition, hence pollution is usually low during the wet (rainy) season [23] [24]. Also, Nitrogen dioxide reacts and water during the wet season to form acid rain (Nitric acid), thereby reducing atmospheric NO_2 concentrations [25] [26]. Relative humidity (the amount of water vapour in the atmosphere), which is influenced by atmospheric temperature [27] affects NO_2 dispersion and deposition, by causing atmospheric resistance. Increased humidity levels lead to reduced NO_2 concentrations and vice versa [22]. Like Nitrogen dioxide, associated pollutants such as CO and O_3 generated during combustion process are also affected by similar meteorological factors [28] [29].

1.4. Study Aim and Objectives

This study was aimed at determining the concentration of nitrogen dioxide that

tollbooth operators are being exposed to, with the specific objectives:

1) To quantify indoor and outdoor tollbooth Nitrogen dioxide concentrations, pollution control measure efficiency.

2) To determine the relationship between nitrogen dioxide concentration and meteorological conditions (Humidity, Rainfall, Wind and Temperature) and Traffic factors (Total/Lane traffic density and Toll Lane type).

3) To compare the NO_2 concentrations with known standards.

2. Methodology

2.1. Sampling Technique/Sampling Analysis

Diffusion tubes (Passive samplers) were first introduced by Palmes *et al.* (1976), and since then has been used in several studies to monitor the spatial and temporal variability of NO_2 [23] [30] [31] [32] [33].

Passive samplers operate with the principle of molecular diffusion, allowing NO_2 gas to travel through a tube to an absorbent that retains it for a period of time [34]. It provides the advantages of being low cost, easy to use, reusable, easy to store, and applicable in spatial monitoring [35].

Diffusion samplers manufactured by Passam Ltd., Switzerland [36] consist of a simple acrylic tube of dimensions (7.1×1.1 cm) that takes in gas via the principle of molecular diffusion (Figure 1(a)). The top end of the tube is tightly fitted with a black colored polythene cap that constrains a pair of stainless-steel mesh discs impregnated with 50% volume of tri-ethanolamine (TEA) and Acetone solution, while bottom end of the tube was sealed during storage and transportation to and from the sample location with a white polythene cap, which is removed during the sampling. Figure 1(a) shows the schematic of a passive sampling mechanism.

During deployment, a set of triplicate tubes were mounted vertically (impregnated mesh end hanging up) on a 5 by 2 cm spacer wooden block with the open end located below the lower surface of the spacer block, to allow free flow of air into tubes and reduce turbulence that could be caused by mounting surface. Diffusion tubes were positioned close to the breathing zone of the toll operators, to ensure sufficient levels pollutants likely inhaled by operators are captured at a measuring height not higher than 2.5 m from the ground [37].

The tubes were installed at a height of approximately 2.5 meters above ground level and left exposed for a duration of one week at each of the three (3) Toll plazas selected for sampling. Additional travel blanks were used as a control to calibrate the spectrometer. The calibration curve is presented in Figure 1(b).

2.2. Storage and Exposure Duration

The diffusion tubes were prepared for 1 - 3 days prior to exposure to atmospheric NO_2, and after collection was also stored in the refrigerator for 1 - 3 days prior to analysis. This storage timeline is generally reasonable based on previous literature [21] [38]. This was done to reduce the hassle due to logistics and
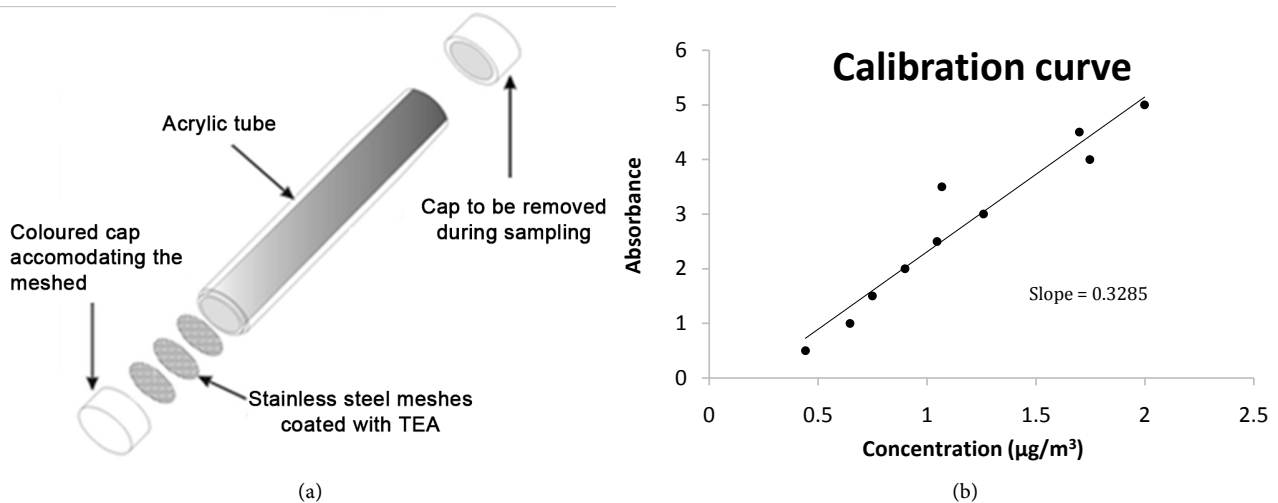
Figure 1. (a) Schematic of NOx diffusion tubes; (b) Calibration curve.

laboratory scheduling as the toll locations were at a combined distance of 57 kilometres away from the Laboratory, and it took some time to commute all three toll plazas to collect and replace diffusion tubes.

This study was conducted for a duration of one month, with pollutant concentrations measured at weekly intervals, which aligns with previous studies [23] [38]. Heal *et al.* [39] and Heal and Cape [40] also reported that weekly measurement of four times (1 week × 4) showed higher accuracy than continuous measurement for a month, as this duration leaves enough time for reagent to efficiently absorb NO_2 before the period of TEA photo-degradation [41]. Moodley *et al.* [23] likewise stipulated seven days as the optimal period it takes for the impregnated mesh to be saturation with NO_2.

2.3. Sampling Strategy

Toll Plazas in Malaysia are generally composed of manual, Touch and Go and SMART sections, where the manual toll booths are being operated by individuals and are the interest in this study. Shih *et al.* [6] revealed that the concentration of pollutants was significantly higher at manual tollbooths than automatic tollbooths, attributed to the deceleration, idling and acceleration of vehicles to make payments, and as a result. This study was focused on NO_2 sampling at the manual tollbooths.

The three Toll plazas selected for this study captured varying average daily traffic density (vehicle per day (v/d)) of High (Sungai Besi = 13,8000 v/d), Medium (Kajang = 65,000 v/d) and Low (Putra Makhota = 24,100 v/d) PLUS Express Behard (PEB). Figure 2(a) shows the map of study sites along Malaysian highway in relation to Meteorological stations, while Figure 2(b) displays the aerial view of Toll Plaza environs extracted from Google Earth and schematics of individual Toll plazas showing specific booths. Sungai Besi Toll plaza is located in an urban environment surrounded by high-rise building and other roads that can contribute to outdoor NO_2 levels, while Kajang toll plaza was located in a
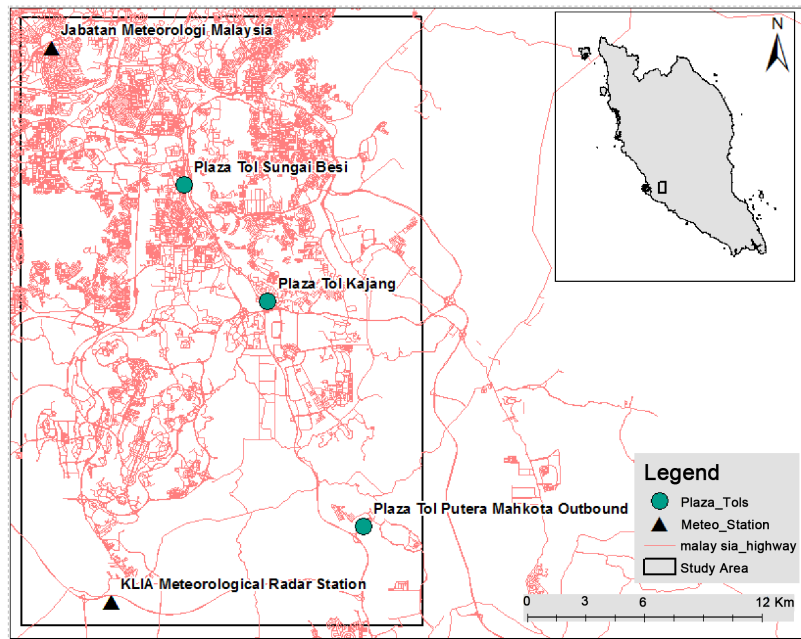
(a)
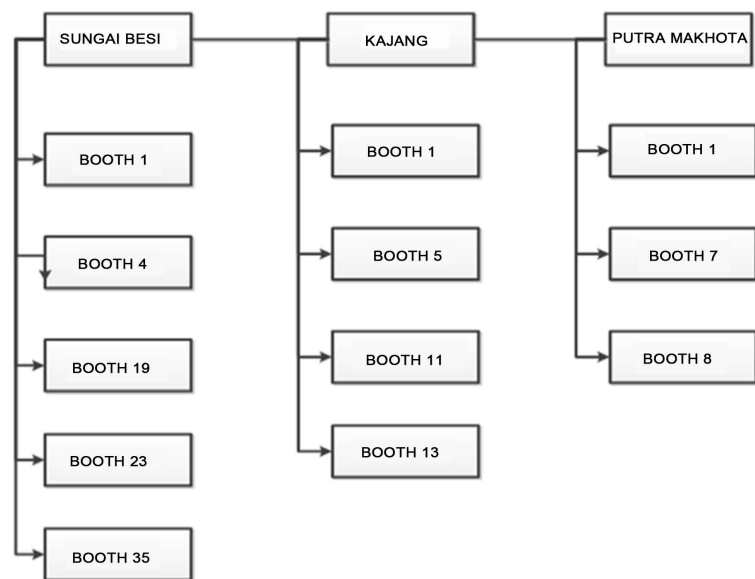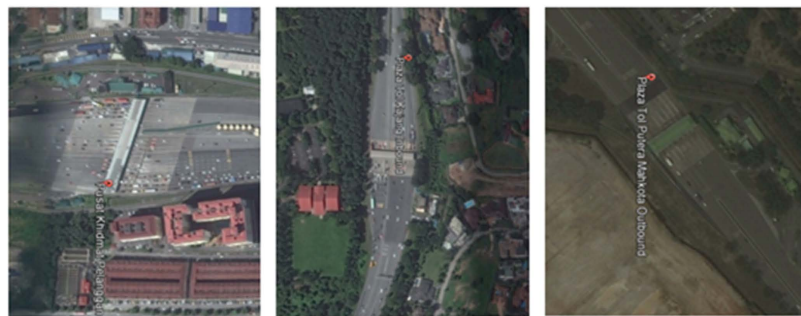
(b)

Figure 2. (a) Map of road, toll plaza locations and meteorological stations; (b) Aerial view of toll environs and schematic of specific booths sampled.

sub-urban environment surrounded by trees and Putra Makhota Toll plaza was localized in a rural environment with low traffic activities and far from other sources of pollution.

The sampling toll booths at the toll plazas were selected based on the class of vehicles that pass through them, to capture heavy and light duty vehicle passage. Heavy-duty vehicles are known to produce significantly higher nitrogen dioxide and particulate matters than carbon monoxide and contribute one-third of nitrogen oxides on highways [42]. The increased number of diesel vehicles over the last decades and use of oxidizing catalytic converters in diesel vehicles has also been identified to result in an increased ratio of NO_2/NO_x from road traffic emissions [19] [43]. Cheng *et al.* [5] also reported similar trends, where high level of pollutant NO_2 was observed on urban roads where high density of heavy-duty vehicles passes through.

2.4. Experimental Procedure to Determine NO₂ Concentration

The quantity of nitrite formed in each tube was determined using a Liquid Chromatography Triple Quadruple Mass Spectrometer System (320 LC-MS/MS), operated at a wavelength of 542 nm, using diluted N-1-napthylethyllenediaminedihydrochloride (NEDA) and sulfanilamide solutions as colour-forming reagents. Control tubes (Travel blanks) were treated similarly and used to calibrate spectrophotometer, and the average concentration of triplicates deployed each site determined. The laboratory analysis was conducted at the University of Nottingham Malaysia Campus, Selangor.

Fick's Law was used to calculate the weekly atmospheric concentration of NO_2, which states that "flux is proportional to concentration gradient". Fick's equation takes into consideration the quantity of gas absorbed over the period of time (Q), the cross-sectional area of the sampling tube (A), the time of exposure (T) and the length of the sampling tube (L) to derive NO_2 concentration (C).

$$F = -D_{12} \frac{dC}{dz} \qquad (2)$$

where: F = flux of gas across the unit area in the z direction; C = concentration of gas; z = diffusion path; D_{12} = constant of proportionality (molecular diffusion coefficient of the gas of interest). Using diffusion tube with areas $A(\pi r^2)$; and length L, the quantity of gas transferred along the diffusion tube in time T is given by

$$Q = F(\pi r^2)T . \qquad (3)$$

Substituting Equation (3) into 2;

$$Q = D_{12} \frac{(C_1 - C_0)\pi r^2 T}{L} \qquad (4)$$

where: C_1 is the concentration of pollutant absorbed by the TEA impregnated mesh after exposure, C_0 is the concentration on unexposed travel tubes, and $\frac{(C_1 - C_0)}{Z}$ is the concentration gradient (slope) presented in Figure 1(b). The

average concertation of the gas (1) at the open end of the tube over the period of exposure is:

$$C = \frac{QL}{D_{12}\pi r^2 T} \cdot$$

(5)

2.5. Traffic Count and Meteorological Data

Hourly traffic counts were obtained from the Plus Malaysia Berhad toll administration, the custodian for toll traffic records, while daily meteorological data were obtained from the department of Meteorology, Malaysia, from stations closest to the sample sites. Meteorological parameters applied in this study include Rainfall (mm), Relative Humidity (%), Wind speed (m/s) and Temperature (˚C), that have been identified from various studies to impact pollution concentration [19] [22] [23] [24].

The meteorological data were obtained from KLIA Meteorological station and Jabatan Meteorologi Malaysia (Petaling Jaya) located closest in proximity to (Putra Makhota) and (Kajang and Sungai Besi) respectively to reduce the influence of space and landscape obstructions on meteorological parameters. The distance between KLIA Meteorological station to Putra Makhota, Kajang and Sungai Besi Tolls are 13.6, 17.29 and 21.64 kilometres respectively, while the distance from Jabatan Meteorologi Malaysia (Petaling Jaya) station to similar toll plazas is 28.98, 16.92 and 9.89 kilometres respectively.

2.6. Statistical Analysis

Basic statistical analysis was undertaken in this study, first to estimate the mean of the triplicate tubes used during indoor and outdoor tollbooths sampling. Comparative analysis was applied to assess the difference between indoor and outdoor NO_2 concentrations at heavy and light duty toll booths, linear regression to relate meteorological parameters to measured NO_2 concentrations, and indoor/outdoor ratio [44] to assess the efficiency of protective booths and air-conditioning systems to reduce operators exposure to pollutants.

3. Results and Discussion

3.1. Descriptive Statistics of Toll Indoor/Outdoor NO₂ and Comparative Analysis

Statistical parameters that define the data collected from all tollbooths and Plazas are presented in Table 1, and Figure 3 displays the indoor/outdoor pollutant mean concentrations and error bars to indicate variation in sampling across the various toll plazas. Also, the traffic density difference between all three toll plazas is depicted. The weekly (one-month) average indoor and outdoor NO_2 concentrations at all three Toll plazas showed no significant difference (Table 1), however, a downward trend of pollution concentration is observed in Figure 3, corresponding with total traffic density decline from Sungai Besi to Kajang to Putra Makhota.

Table 1. Descriptive statistics.

Toll Plaza (Total Tubes)	Toll Booth	Indoor (ppm)			Outdoor (ppm)		
		Mean	SD	Range	Mean	SD	Range
SG. BESI (30)	1 (HD)	0.137	0.029	0.105 - 0.162	0.119	0.044	0.092 - 0.17
	4 (LD)	0.071	0.021	0.055 - 0.095	0.087	0.041	0.057 - 0.134
	19 (LD)	0.102	0.037	0.066 - 0.152	0.09	0.042	0.06 - 0.138
	23 (HD)	0.140	0.065	0.076 - 0.205	0.127	0.048	0.089 - 0.181
	35 (LD)	0.057	0.03	0.029 - 0.09	0.087	0.044	0.06 - 0.138
KAJANG (24)	1 (HD)	0.083	0.042	0.044 - 0.133	0.077	0.019	0.063 - 0.09
	5 (LD)	0.062	0.019	0.046 - 0.089	0.082	0.004	0.077 - 0.086
	11 (HD)	0.057	0.01	0.047 - 0.066	0.068	0.024	0.051 - 0.096
	13 (LD)	0.089	0.033	0.063 - 0.126	0.123	0.038	0.08 - 0.149
PTR. MAKHOTA (18)	1 (HD)	0.050	0.015	0.033 - 0.068	0.050	0.015	0.033 - 0.06
	7 (LD)	0.042	0.013	0.031 - 0.059	0.043	0.008	0.031 - 0.05
	8 (LD)	0.043	0.017	0.025 - 0.066	0.050	0.012	0.038 - 0.066

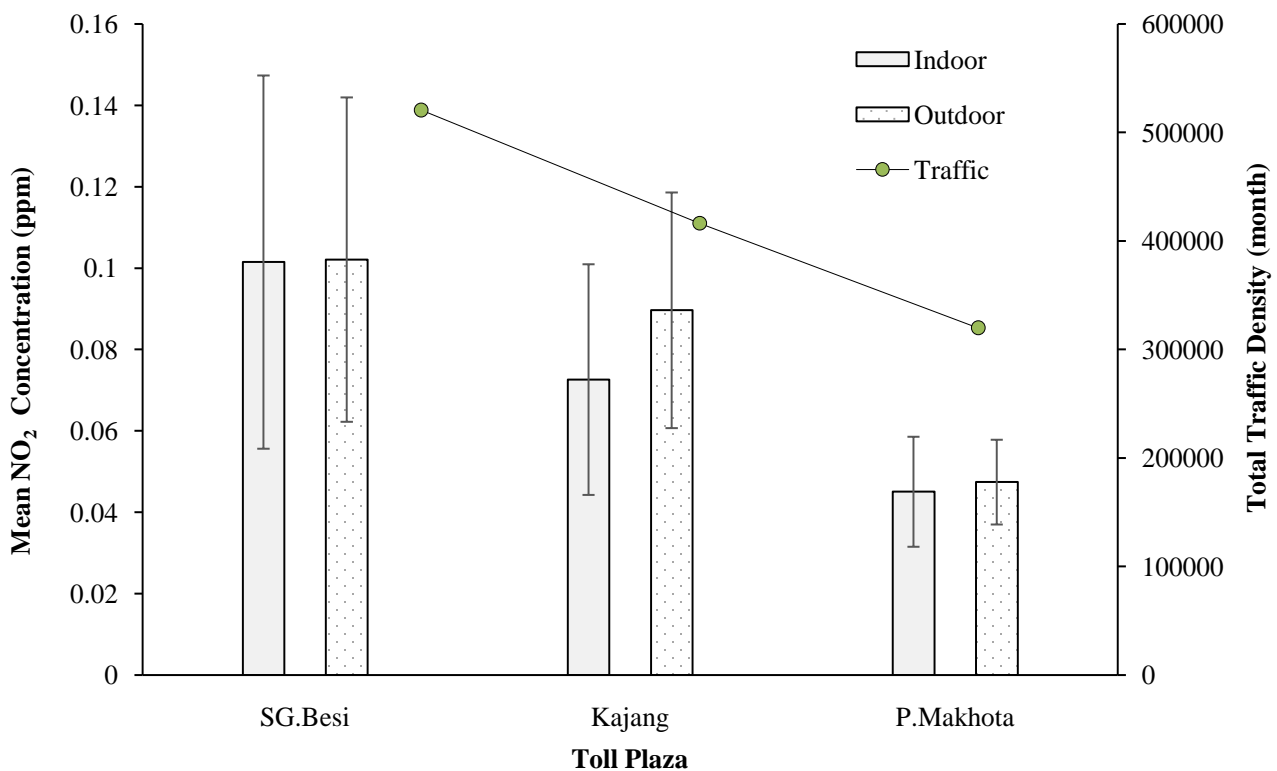HD = Heavy Duty, LD = Light Duty, SD = Standard deviation.

Figure 3. Indoor/outdoor pollutant and total traffic density.

This study was conducted during the transition monsoon period (wet season), *i.e.* September to December in Malaysia, being a period with the lowest NO_x concentrations in comparison with other periods due to high precipitations and

daily wind circulation that encourages pollutants dispersion and deposition [24] [45], implying that NO_2 concentrations could be higher during the dry season.

3.2. Indoor/Outdoor Ratio and Toll Traffic Type

The efficacy of pollution control structures, *i.e.* tollbooths structures, air curtains and ventilation systems was assessed by indoor/outdoor (I/O) pollutant concentration ratio presented in Figure 4. Variations of I/O ratio at tollbooths with different traffic types, *i.e.* heavy and light duty traffic was also accounted for and presented in Figure 4.

Indoor/outdoor nitrogen dioxide ratio aids the evaluation of the protection offered by toll booth structure and accessories [44] [46]. I/O ratio greater than one implies the presence of an indoor pollution source or reduced clean air circulation and vice versa [47]. The concentration of NO_2 in microenvironments depends on factors such as ventilation, as poor ventilation tends to reduce air circulation [38], as well as personal habits such as smoking [48]. Monn *et al.* [38] also stipulated that indoor levels of NO_2 vary from 50% to 90% of outdoor concentrations when indoor pollution sources are not present.

The ratios of indoor/outdoor NO_2 levels varied across the toll plazas, 1.05 at Sungai Besi, 0.80 at Kajang and 0.97 at Putra Makhota. Also, the comparison between heavy and light duty toll lanes I/O ratios showed insignificant differences but was greater at heavy-duty toll lanes than the light-duty lanes. The insignificant difference between in heavy and light duty toll lanes NO_2 concentrations has similarly been reported in other studies [4], and in this case can be attributed to the passage of the mixed vehicle observed during field visits, given
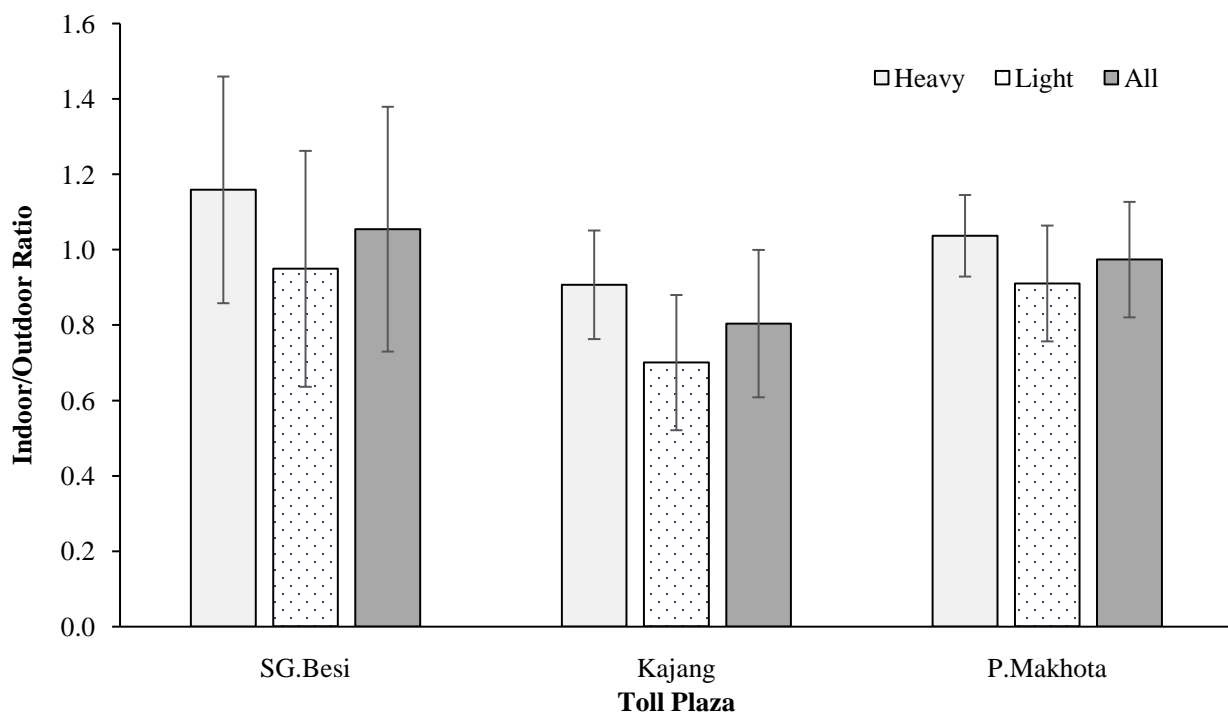
Figure 4. Indoor/outdoor NO_2 ratio of toll booths and traffic type effect.

that vehicles tend to pass through any available toll lane to save time and beat traffic congestion.

The I/O NO_2 ratio > 1 at Sungai Besi suggests the presence of an indoor pollution source, or poor operator practice, which has similarly been reported in other studies [5] [44]. This result can be explained by observatory knowledge during monitoring, as toll operators were observed to occasionally turning off air curtains and conditioners; and leave booth doors and windows open. Furthermore, Breysse *et al.* [46] argued that the use of single unit air conditioners, such as those used at the toll plazas in this study, are inefficient, given that the cooling systems draw in already contaminated air because they are located within already polluted environments.

3.3. Indoor and Outdoor NO_2 Concentration Relationship with Meteorological Factor and Traffic Density

Table 2 displays the correlation relationships between indoor, outdoor NO_2 and meteorological parameters (Wind, humidity, Temperature and Rainfall). The results showed significant positive relationship between indoor and outdoor NO_2 concentrations at all toll plazas, implying that an increase in the outdoor concentration will lead to a corresponding increase in indoor NO_2 concentration.

Table 2. Indoor and outdoor NO_2 concentration relationship with meteorological factors and traffic density.

	Wind	Humidity	Temp	Rain	Lane Density	Total Density	Indoor	Outdoor
Sungai Besi								
Indoor	−0.432	0.048	0.080	0.445	0.139	0.113	1	-
P-value	0.094	0.859	0.768	0.084	0.608	0.677	-	-
Outdoor	−0.507	−0.041	0.179	0.456	0.427	0.181	0.767	1
P-value	0.054	0.883	0.523	0.087	0.112	0.519	0.001	-
Kajang								
Indoor	0.235	−0.119	0.126	−0.481	0.375	−0.253	1	-
P-value	0.419	0.684	0.669	0.082	0.187	0.382	-	-
Outdoor	0.493	0.388	−0.363	−0.554	0.214	−0.547	0.689	1
P-value	0.103	0.212	0.246	0.062	0.500	0.065	0.013	-
Putra Makhota								
Indoor	0.811	0.811	−0.811	−0.122	−0.266	−0.729	1	-
P -value	0.001	0.001	0.001	0.705	0.402	0.007	-	-
Outdoor	0.606	0.606	−0.606	0.147	−0.289	−0.745	0.877	1
P-value	0.048	0.048	0.048	0.666	0.389	0.001	0.000	-

Correlation significant at P < 0.05, Temp = Temperature, and Rain. = Rainfall.

The impact of Lane and total traffic density was insignificant at all tolls, indicating that other sources of traffic in proximity to tolls are likely also contributing to the total measured pollutant. Evidence from several studies [2] [49] supports this suggestion, as they revealed that NO_2 generated by vehicles as far as 100 - 200 meters from sample locations can disperse and influence recorded pollution levels.

Table 2 also shows that, besides the negative relationship between wind and indoor and outdoor NO_2 concentrations that are consistent with similar studies [21] [22], the relationship between meteorological parameters and pollutant measurements were statistically insignificant. This discrepancy can be attributed to the distance between the meteorological station and pollution sampling toll plazas, and the landscape known to obstruct airflow and effective weather observation [50]. Also, variations in land use/cover have been found to impact micro-climatic conditions [51] [52], suggesting that the weather conditions at toll plazas could differ from those observed at the meteorological stations. Similar inconsistencies were disclosed by Tsai [4], where wind speed, humidity and temperature relationships with Polycyclic Aromatic Hydrocarbons (PAHs) were not statistically significant.

3.4. Comparison of the Current Study with Other Studies and World Standards

Table 3 shows an overview of other studies that were conducted over a period of one week for in indoor and outdoor environments. For consistent comparisons, the results of this study and others converted to per million (ppm). Majority of the studies reviewed revealed results comparable to outdoor NO_2 concentrations measured at Putra-Makhota and Kajang Toll Plaza, with low and medium traffic densities respectively. For example, NO_2 concentrations in a high traffic area in Abu Dhabi varied from 0.023 to 0.043 [53], while da Silva *et al.* [21], reported NO_2 concentrations ranging between 0.031 - 0.036 in Brazil, which are within the range of NO_2 concentrations measured at Putra Makhota Toll Plaza. High traffic density highway NO_2 concentration in Texas [54] was found to be lower than those reported in this study, as well as Salem *et al.* [53] and da Silva [21].

Indoor NO_2 concentrations reported for office [55] and resident [31] micro-environments were consistent with levels of theindoor NO_2 measure at Puta-Makhota toll plazas, while NO_2 concentrations at Sungai Besi and Kajang Toll plazas exceeded those reported in other studies by 3 to 4 times [20] [56]. The reason for such levels of pollution at toll plazas is possibly due to high levels of traffic density congestion which would rarely be found in residential areas.

Industrialization and urbanization have also been identified as factors that influence pollution levels [57] [58], hence developing countries are expected to be more polluted than developed ones due to ongoing development activities [15] [59].

Weekly average concentrations of indoor and outdoor Nitrogen dioxide at all three Toll Plazas were plotted against and found to exceed weekly German NO_2

Table 3. Studies showing NO_2 concentration at various environments in comparison to current study.

Location	Site	Condition	Concentration Mean ± SD (range) (ppm)	Duration	Referee
Canada	Highway		0.023 ± 0.002 (0.012 - 0.029)	1 week	[49]
Bahrain	Urban		0.019 ± 0.007 (0.007 - 0.040)	1 week	[56]
Hertfordshire and North London	Gas Cooker	Personal Exposure (Winter)	0.011 ± 0.002 (0.006 - 0.015)	1 week	[31]
		Personal Exposure (Summer)	0.015 ± 0.002 (0.001 - 0.002)		
	Electric Cookers	Personal Exposure (Winter)	0.008 ± 0.002 (0.006 - 0.011)		
		Personal Exposure (Summer)	0.013 ± 0.001 (0.001 - 0.015)		
Hong Kong	Resident and Office	Personal Exposure	0.024 ± 0.006 (0.014 - 0.039)	1 week	[55]
North, California	School Site	Road Traffic	0.021 ± 0.007 (0.001 - 0.037)	1 week	[20]
Sao-Paulo, Brazil	Street, Road and Avenues	Heavy Traffic	0.034 ± (0.031 - 0.036)	1 week	[21]
		Light Traffic	0.026 ± (0.023 - 0.028)		
El Paso, Texas	CAMS 6 (Highway) CAMS 41 (Chamizal National Memorial)	High Traffic Area	0.021 ± 0.00 (0.016 - 0.023)	1 week	[54]
			0.016 ± 0.001 (0.017 - 0.018)		
AL-Ain City, Abu Dhabi	Industrial Traffic Residential		0.022 ± 0.004 (0.005 - 0.029)	2 × 1 week	[53]
			0.032 ± 0.007 (0.023 - 0.043)		
			0.019 ± 0.004 (0.014 - 0.028)		
Wahga Town, Pakistan	Heavy Traffic, Populated Area	Outdoor	0.015 ± 0.005 (0.011 - 0.021)	1 week	[60]
Malaysia	Putra Makhota Toll Kajang Toll Sungai Besi Toll	Toll Plaza (Indoor)	0.056 ± 0.014 (0.025 - 0.068)	4 × 1 week	Current Study
			0.087 ± 0.029 (0.033 - 0.133)		
			0.102 ± 0.047 (0.029 - 0.205)		
	Putra Makhota Toll Kajang Toll Sungai Besi Toll	Toll Plaza (Outdoor)	0.041 ± 0.011 (0.031 - 0.066)		
			0.054 ± 0.011 (0.041 - 0.149)		
			0.095 ± 0.047 (0.057 - 0.181)		

standards of 0.032 ppm (Figure 5) because most NO_2 standards are defined at 1 hour, 8 hours, 1 day and 1-year intervals (Table 4). Nevertheless, besides Putra Makhota, the mean weekly indoor and outdoor NO_2 concentrations at Sungai Besi and Kajang were higher than the 24 hours NO_2 standards in Malaysia and other countries in the Asia. It is impractical to make such direct comparison due to the variation in duration; rather, this was used indicatively.

4. Conclusions, Recommendation and Study Limitation

4.1. Conclusions

This study assesses the weekly concentration of indoor and outdoor NO_2 at toll plazas, taking into account the effect of meteorological and traffic density parameters. Our findings reveal that the concentration of NO_2 at Toll booths exceeds weekly air quality standards, and were higher than those derived in other similar studies. Outdoor NO_2 concentration showed a significant positive relationship with Indoor NO_2 but was insignificantly correlated to traffic density and meteorological factors, which suggest that the NO_2 measurements at the toll plazas are likely influenced by external pollution sources and the micro-climatic conditions at the toll plazas differ from where meteorological data was acquired.
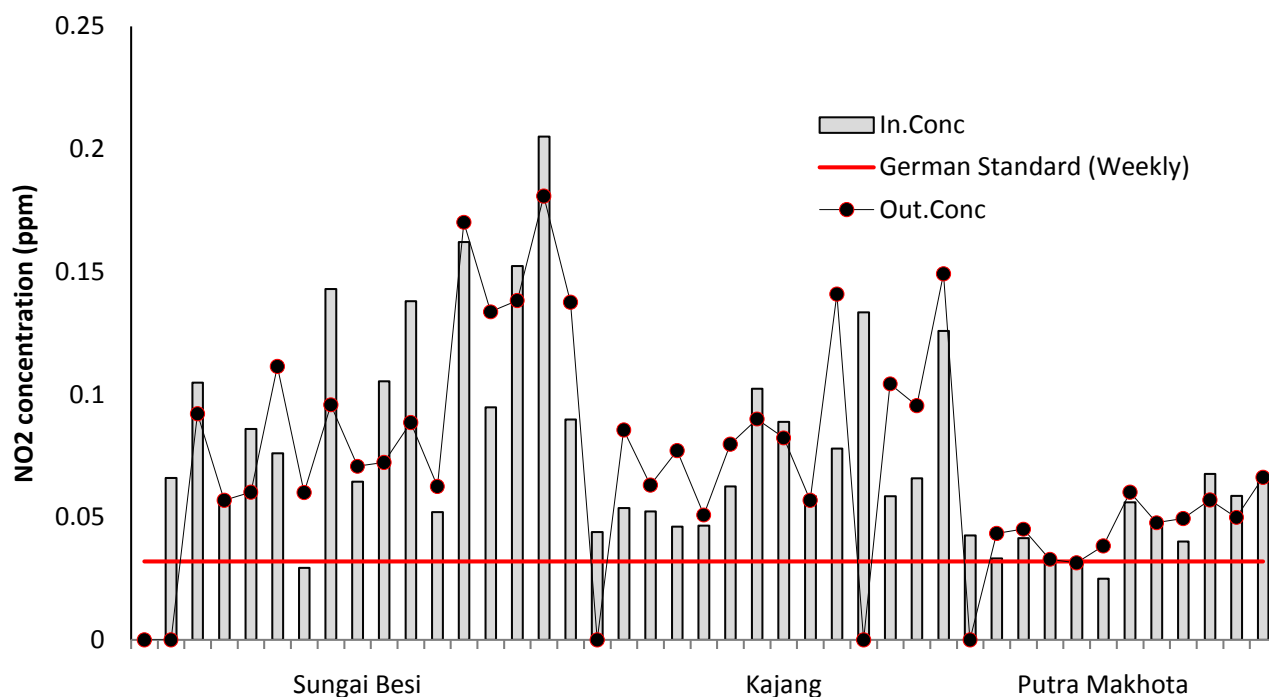
Figure 5. Comparison to German weekly standard.

Table 4. Pollution standards in Asian cities and global (ppm).

Country	1 Hour	8 Hours	24 Hours	1 Week	1 Year
Malaysia	0.170	-	0.040	-	-
China	0.128	-	0.064	-	0.043
Bangladesh	-	-	-	-	0.053
Hong Kong	0.160	-	0.080	-	0.043
India	-	-	0.043	-	0.032
Indonesia	0.213	-	0.080	-	0.053
Japan	-	-	0.060	-	-
Korea	0.150	-	0.080	-	0.050
Nepal	-	-	0.043	-	0.021
Philippines	-	-	0.080	-	-
Singapore	-	-	-	-	0.053
Sri Lanka	0.133	0.080	0.053	-	-
Thailand	0.173	-	-	-	-
Vietnam	0.106	-	-	-	0.021
Taiwan	-	-	-	-	0.051
Germany	N/A	N/A	N/A	0.032	N/A
WHO	0.106	-	-	-	0.021
EPA	0.100	-	-	-	0.053

N/A = Not Applicable to this study. Adapted from [61].

Indoor/outdoor NO$_2$ ratios that depict the efficiency of pollution control systems (*i.e.* booth structure, air conditioning systems and air curtains) were greater in heavy duty traffic lanes than the light-duty vehicle designated toll, but not significantly. The high indoor NO$_2$ concentration can be attributed to toll operator behaviour of occasionally leaving door and windows open, poor ventilation, high densities of mixed traffic fleets, and nearness to the mobile pollution source.

Indicative comparison of weekly NO$_2$ concentrations to 24 hour standards also showed that pollution levels at the toll plazas were higher, hence measures should be put in place to reduce worker's exposure and counter the potential adverse health effects.

4.2. Recommendations

The pollution risk reduction strategies recommended to reduce exposure of toll operators to NO$_2$ are presented as follows:

- Air conditioners being used to aid air circulation at toll plazas are located at the corner of the toll booths, thus are being influenced by the surrounding ambient air polluted by NO$_2$. We hereby recommend the air conditioning systems are relocated away from surrounding polluted air or redesigned to incorporate filter systems that separate pollutants from inflow air.
- Toll operators are advised to should always inspect their ventilation system to ensure it is working efficiently, and be sensitized on the importance of not tampering with pollution protection systems.
- Continuous monitoring of personal exposure during 8 hour shifts is recommended, to enable improved understanding of pollution impact, and inform improved pollution management decisions.
- Reducing the idle time of vehicles is an essential to reducing vehicular activities, hence pollution. Thus, drivers are advised to turn off their vehicles when queuing and carry the specific amount of money need for toll payment.
- Electronic toll collection systems should be encouraged to reduce idling time and operator's exposure to polluted air.

4.3. Study Limitation

Though this study clearly shows that indoor and outdoor concentrations of NO$_2$ at toll plazas were above recommended weekly standards stipulated in other regions of the world, and can likely cause health challenges, operators, however, do not spend the whole time in tollbooths. Toll operators work 8 hour shift intervals, hence would be exposed to lower levels of pollution than presented in this study. Individual exposure over time has been found to strongly correlate with indoor (home) NO$_2$ concentration, given that individuals spend more time at home than work (8 hours)/outdoors [62]. Therefore, it will be important to study the relationship between Toll Booth NO$_2$ concentration and personnel exposure at various shifts, using personal sampling devices, as well as assess long and short-term effect symptoms to make robust recommendations. The effect of meteorological factors can also be improved collecting meteorological data at the

toll plazas during the pollution measurement period, given that the micro-climatic conditions at the toll plaza and meteorological station can vary significantly due to their distance apart and variable land use/landcover.

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References

[1] Department of Environment (1996) Malaysia Environmental Quality Report. Department of Environment, Ministry of Science, Technology and Environment, Malaysia.

[2] Beckerman, B., Jerrett, M., Brook, J.R., Verma, D.K., Arain, M.A. and Finkelstein, M.M. (2008) Correlation of Nitrogen Dioxide with Other Traffic Pollutants near a Major Expressway. *Atmospheric Environment*, **42**, 275-290. https://doi.org/10.1016/j.atmosenv.2007.09.042

[3] Gilbert, N.L., Goldberg, M.S., Brook, J.R. and Jerrett, M. (2007) The Influence of Highway Traffic on Ambient Nitrogen Dioxide Concentrations beyond the Immediate Vicinity of Highways. *Atmospheric Environment*, **41**, 2670-2673. https://doi.org/10.1016/j.atmosenv.2006.12.007

[4] Tsai, P.-J., Lee, C.-C., Chen, M.-R., Shih, T.-S., Lai, C.-H. and Liou, S.-H. (2002) Predicting the Contents of BTEX and MTBE for the Three Types of Tollbooth at a Highway Toll Station via the Direct and Indirect Approaches. *Atmospheric Environment*, **36**, 5961-5969. https://doi.org/10.1016/S1352-2310(02)00768-9

[5] Cheng, Y.-H., Huang, C.-H., Huang, H.-L. and Tsai, C.-J. (2010) Concentrations of Ultrafine Particles at a Highway Toll Collection Booth and Exposure Implications for Toll Collectors. *Science of the Total Environment*, **409**, 364-369. https://doi.org/10.1016/j.scitotenv.2010.10.023

[6] Shih, T.-S., *et al.* (2008) Elemental and Organic Carbon Exposure in Highway Tollbooths: A Study of Taiwanese Toll Station Workers. *Science of the Total Environment*, **402**, 163-170. https://doi.org/10.1016/j.scitotenv.2008.04.051

[7] Diab, R., Foster, S., François, K., Martincigh, B. and Salter, L. (2005) Carbon Monoxide Levels at a Toll Plaza near Durban, South Africa. *Environmental Chemistry Letters*, **3**, 91-94. https://doi.org/10.1007/s10311-005-0111-1

[8] Currie, J. and Walker, R. (2011) Traffic Congestion and Infant Health: Evidence from E-ZPass. *American Economic Journal: Applied Economics*, **3**, 65-90. https://doi.org/10.1257/app.3.1.65

[9] Bartin, B., Mudigonda, S. and Ozbay, K. (2008) Impact of Electronic Toll Collection on Air Pollution Levels: Estimation Using Microscopic Simulation Model of Large-Scale Transportation Network. *Transportation Research Record: Journal of the Transportation Research Board*, **2083**, 105-113.

[10] Raine, R.R., Stone, C.R. and Gould, J. (1995) Modeling of Nitric Oxide Formation in Spark Ignition Engines with a Multizone Burned Gas. *Combustion and Flame*, **102**, 241-255. https://doi.org/10.1016/0010-2180(94)00268-W

[11] Finlayson-Pitts, B.J. (2000) Chemistry of the Upper and Lower Atmosphere: Theory,

Experiments, and Applications. Academic Press, San Diego, CA

[12] World Health Organization (1997) Motor Vehicle Air Pollution Public Health Impact and Control Measures. WHO, Geneva.

[13] Mücke, H. and Wagner, H. (1998) Anorganische Gase/Stickstoffdioxid. In: Wichmann, H.E., et al., Eds., *Handbuch der Umweltmedizin: Toxikologie*, *Epidemiologie*, *Hygiene*, *Belastungen*, *Wirkungen*, *Diagnostik*, *Prophylaxe*, Ecomed, Landsberg, Germany, 14.

[14] Chitano, P., Hosselet, J.J., Mapp, C.E. and Fabbri, L.M. (1995) Effect of Oxidant Air Pollutants on the Respiratory System: Insights from Experimental Animal Research. *European Respiratory Journal*, **8**, 1357-1371. https://doi.org/10.1183/09031936.95.08081357

[15] Han, X. and Naeher, L.P. (2006) A Review of Traffic-Related Air Pollution Exposure Assessment Studies in the Developing World. *Environment International*, 32, 106-120. https://doi.org/10.1016/j.envint.2005.05.020

[16] Zhang, L., Guan, Y., Leaderer, B.P. and Holford, T.R. (2013) Estimating Daily Nitrogen Dioxide Level: Exploring Traffic Effects. *Annals of Applied Statistics*, **7**, 1763-1777. https://doi.org/10.1214/13-AOAS642

[17] Azeez, K.H.A., Miura, M., Nishimura, Y. and Inokuma, S. (2005) Evaluating Transportation Impact on Environment in a Residential Area in Kuala Lumpur. *Proceedings of the Eastern Asia Society for Transportation Studies*, **5**, 1815-1826.

[18] Glasius, M., Carlsen, M.F., Hansen, T.S. and Lohse, C. (1999) Measurements of Nitrogen Dioxide on Funen Using Diffusion Tubes. *Atmospheric Environment*, **33**, 1177-1185. https://doi.org/10.1016/S1352-2310(98)00285-4

[19] McAdam, K., Steer, P. and Perrotta, K. (2011) Using Continuous Sampling to Examine the Distribution of Traffic Related Air Pollution in Proximity to a Major Road. *Atmospheric Environment*, **45**, 2080-2086. https://doi.org/10.1016/j.atmosenv.2011.01.050

[20] Singer, B.C., Hodgson, A.T., Hotchi, T. and Kim, J.J. (2004) Passive Measurement of Nitrogen Oxides to Assess Traffic-Related Pollutant Exposure for the East Bay Children's Respiratory Health Study. *Atmospheric Environment*, **38**, 393-403. https://doi.org/10.1016/j.atmosenv.2003.10.005

[21] da Silva, A.S., Cardoso, M.R., Meliefste, K. and Brunekreef, B. (2006) Use of Passive Diffusion Sampling Method for Defining NO_2 Concentrations Gradient in São Paulo, Brazil. *Environmental Health*, **5**, 19. https://doi.org/10.1186/1476-069X-5-19

[22] Elminir, H.K. (2005) Dependence of Urban Air Pollutants on Meteorology. *Science of the Total Environment*, **350**, 225-237. https://doi.org/10.1016/j.scitotenv.2005.01.043

[23] Moodley, K.G., Singh, S. and Govender, S. (2011) Passive Monitoring of Nitrogen Dioxide in Urban Air: A Case Study of Durban Metropolis, South Africa. *Journal of Environmental Management*, **92**, 2145-2150. https://doi.org/10.1016/j.jenvman.2011.03.040

[24] Mondal, R., Sen, G., Chatterjee, M., Sen, B. and Sen, S. (2000) Ground-Level Concentration of Nitrogen Oxides (NO_x) at Some Traffic Intersection Points in Calcutta. *Atmospheric Environment*, **34**, 629-633. https://doi.org/10.1016/S1352-2310(99)00216-2

[25] Wondyfraw, M. (2014) Mechanisms and Effects of Acid Rain on Environment. *Journal of Earth Science* & *Climatic Change*, **5**, 204.

[26] Driscoll, J.A. (1997) Acid Rain Demonstration: The Formation of Nitrogen Oxides

as a By-Product of High-Temperature Flames in Connection with Internal Combustion Engines. *Journal of Chemical Education*, **74**, 1424.
https://doi.org/10.1021/ed074p1424

[27] Hardwick Jones, R., Westra, S. and Sharma, A. (2010) Observed Relationships between Extreme Sub-Daily Precipitation, Surface Temperature, and Relative Humidity. *Geophysical Research Letters*, **37**, L22805.

[28] Kojić, R. and Antić, M. (2016) The Influence of Meteorological Parameters and Traffic Flows on the Concentration of Ozone (O₃) in Urban Areas in Brcko. *JTTTP—Journal of Traffic and Transport Theory and Practice*, **1**, 12.

[29] Ocak, S. and Turalioglu, F.S. (2008) Effect of Meteorology on the Atmospheric Concentrations of Traffic-Related Pollutants in Erzurum, Turkey. *Journal of International Environmental Application* & *Science*, **3**, 325-335.

[30] Vardoulakis, S., Solazzo, E. and Lumbreras, J. (2011) Intra-Urban and Street Scale Variability of BTEX, NO₂ and O₃ in Birmingham, UK: Implications for Exposure Assessment. *Atmospheric Environment*, **45**, 5069-5078.
https://doi.org/10.1016/j.atmosenv.2011.06.038

[31] Kornartit, C., Sokhi, R.S., Burton, M.A. and Ravindra, K. (2010) Activity Pattern and Personal Exposure to Nitrogen Dioxide in Indoor and Outdoor Microenvironments. *Environment International*, **36**, 36-45.
https://doi.org/10.1016/j.envint.2009.09.004

[32] Stevenson, K., Bush, T. and Mooney, D. (2001) Five Years of Nitrogen Dioxide Measurement with Diffusion Tube Samplers at over 1000 Sites in the UK. *Atmospheric Environment*, **35**, 281-287. https://doi.org/10.1016/S1352-2310(00)00171-0

[33] Campbell, G.W., Stedman, J.R. and Stevenson, K. (1994) A Survey of Nitrogen Dioxide Concentrations in the United Kingdom Using Diffusion Tubes, July-December 1991. *Atmospheric Environment*, **28**, 477-486.
https://doi.org/10.1016/1352-2310(94)90125-2

[34] Atkins, D.H.F. and Lee, D.S. (1995) Spatial and Temporal Variation of Rural Nitrogen Dioxide Concentrations across the United Kingdom. *Atmospheric Environment*, **29**, 223-239. https://doi.org/10.1016/1352-2310(94)00229-E

[35] Varshney, C.K. and Singh, A. (2003) Passive Samplers for NOₓ Monitoring: A Critical Review. *The Environmentalist*, **23**, 127-136.
https://doi.org/10.1023/A:1024883620408

[36] Chang Ho, Y., Maria, T.M. and Clifford, P.W. (2008) Passive Dosimeters for Nitrogen Dioxide in Personal/Indoor Air Sampling: A Review. *Journal of Exposure Science and Environmental Epidemiology*, **18**, 441.
https://doi.org/10.1038/jes.2008.22

[37] University of Nottingham Malaysia Campus (2011) F84M02 Techniques in Environmental Monitoring and Management, Passive Monitoring of Nitrogen Dioxide Concentration.

[38] Monn, C., Brändli, O., Schindler, C., Ackermann-Liebrich, U. and Leuenberger, P. (1998) Personal Exposure to Nitrogen Dioxide in Switzerland. *Science of the Total Environment*, **215**, 243-251. https://doi.org/10.1016/S0048-9697(98)00124-7

[39] Heal, M.R., O'donoghue, M.A. and Cape, J.N. (1999) Overestimation of Urban Nitrogen Dioxide by Passive Diffusion Tubes: A Comparative Exposure and Model Study. *Atmospheric Environment*, **33**, 513-524.

[40] Heal, M.R. and Cape, J.N. (1997) A Numerical Evaluation of Chemical Interferences in the Measurement of Ambient Nitrogen Dioxide by Passive Diffusion Samplers. *Atmospheric Environment*, **31**, 1911-1923.

https://doi.org/10.1016/S1352-2310(97)00025-3

[41] Bush, T., Smith, S., Stevenson, K. and Moorcroft, S. (2001) Validation of Nitrogen Dioxide Diffusion Tube Methodology in the UK. *Atmospheric Environment*, **35**, 289-296. https://doi.org/10.1016/S1352-2310(00)00172-2

[42] Department of Environment (2009) Malaysia Environmental Quality Report. Department of Environment, Ministry of Science, Technology and Environment, Malaysia.

[43] Keuken, M., Roemer, M. and van Den Elshout, S. (2009) Trend Analysis of Urban NO_2 Concentrations and the Importance of Direct NO_2 Emissions versus Ozone/NO_x Equilibrium. *Atmospheric Environment*, **43**, 4780-4783. https://doi.org/10.1016/j.atmosenv.2008.07.043

[44] Chen, C. and Zhao, B. (2011) Review of Relationship between Indoor and Outdoor Particles: I/O Ratio, Infiltration Factor and Penetration Factor. *Atmospheric Environment*, **45**, 275-288. https://doi.org/10.1016/j.atmosenv.2010.09.048

[45] Yassen, M.E., Jahi, J.M. and Ahmad, S. (2005) Evaluation of Long Term Trends in Oxide of Nitrogen Concentrations in the Klang Valley Region, Malaysia. *Malaysian Journal of Environmental Management*, **6**, 59-72.

[46] Breysse, P.N., *et al.* (2005) Indoor Exposures to Air Pollutants and Allergens in the Homes of Asthmatic Children in Inner-City Baltimore. *Environmental Research*, **98**, 167-176. https://doi.org/10.1016/j.envres.2004.07.018

[47] Health Canada (1989) Exposure Guidelines for Residential Indoor Air Quality: A Report of the Federal-Provincial Committee on Environmental and Occupational Health. Health Canada, Ottawa.

[48] Gallelli, G., Orlando, P., Perdelli, F. and Panatto, D. (2002) Factors Affecting Individual Exposure to NO_2 in Genoa (Northern Italy). *Science of the Total Environment*, **287**, 31-36. https://doi.org/10.1016/S0048-9697(01)00990-1

[49] Gilbert, N.L., Woodhouse, S., Stieb, D.M. and Brook, J.R. (2003) Ambient Nitrogen Dioxide and Distance from a Major Highway. *Science of the Total Environment*, **312**, 43-46. https://doi.org/10.1016/S0048-9697(03)00228-6

[50] Oke, T.R. (2007) Siting and Exposure of Meteorological Instruments at Urban Sites. In: Borrego, C. and Norman, A.-L., Eds., *Air Pollution Modeling and Its Application XVII*, Springer, New York, 615-631.

[51] Jamaludin, N., Mohammed, N.I., Khamidi, M.F. and Wahab, S.N.A. (2015) Thermal Comfort of Residential Building in Malaysia at Different Micro-climates. *Procedia—Social and Behavioral Sciences*, **170**, 613-623. https://doi.org/10.1016/j.sbspro.2015.01.063

[52] Rajagopalan, P., Chuan, L.K. and Jamei, E. (2014) Urban Heat Island and Wind Flow Characteristics of a Tropical City. *Solar Energy*, **107**, 159-170. https://doi.org/10.1016/j.solener.2014.05.042

[53] Salem, A., Soliman, A. and El-Haty, I. (2009) Determination of Nitrogen Dioxide, Sulfur Dioxide, Ozone, and Ammonia in Ambient Air Using the Passive Sampling Method Associated with Ion Chromatographic and Potentiometric Analyses. *Air Quality, Atmosphere & Health*, **2**, 133-145. https://doi.org/10.1007/s11869-009-0040-4

[54] Mukerjee, S., *et al.* (2004) Field Method Comparison between Passive Air Samplers and Continuous Monitors for VOCs and NO_2 in El Paso, Texas. *Journal of the Air & Waste Management Association*, **54**, 307-319. https://doi.org/10.1080/10473289.2004.10470903

[55] Chao, C.Y.H. and Law, A. (2000) A Study of Personal Exposure to Nitrogen Dioxide Using Passive Samplers. *Building and Environment*, **35**, 545-553. https://doi.org/10.1016/S0360-1323(99)00040-2

[56] Danish, S. and Madany, I.M. (1992) Concentrations of Nitrogen Dioxide throughout the State of Bahrain. *Environmental Pollution*, **77**, 71-78. https://doi.org/10.1016/0269-7491(92)90160-C

[57] Cho, H.S. and Choi, M.J. (2014) Effects of Compact Urban Development on Air Pollution: Empirical Evidence from Korea. *Sustainability* (*Switzerland*), **6**, 5968-5982. https://doi.org/10.3390/su6095968

[58] Fang, C., Liu, H., Li, G., Sun, D. and Miao, Z. (2015) Estimating the Impact of Urbanization on Air Quality in China Using Spatial Regression Models. *Sustainability* (*Switzerland*), **7**, 15570-15592. https://doi.org/10.3390/su71115570

[59] Bose, R.K. (1998) Automobiles and Environmental Sustainability: Issues and Options for Developing Countries. *Asian Transport Journal*, **13**, 1-16.

[60] Mehmood, T., Ali, Z., Noor, N., Sidra, S., Nasir, Z. and Colbeck, I. (2015) Measurement of NO_2 Indoor and Outdoor Concentrations in Selected Public Schools of Lahore Using Passive Sampler. *Journal of Animal and Plant Sciences*, **25**, 681-686.

[61] Schwela, D. (2006) *Urban Air Pollution in Asian Cities: Status, Challenges and Management*. Routledge, London.

[62] Valero, N., *et al.* (2009) Concentrations and Determinants of Outdoor, Indoor and Personal Nitrogen Dioxide in Pregnant Women from Two Spanish Birth Cohorts. *Environment International*, **35**, 1196-1201. https://doi.org/10.1016/j.envint.2009.08.002