Malaysian Journal of Environmental Management 6 (2005): 59 - 72

Evaluation of Long Term Trends in Oxide of Nitrogen Concentrations in the Klang Valley Region, Malaysia

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ABSTRACT

Anthropogenic emissions of NO_χ *account for a large majority of all nitrogen inputs to the environment. The major sources of man-made* NO_χ *emissions are high-temperature combustion processes such as those that occur in automobile and power plants.* NO_χ *contribute to a wide range of environmental effects directly and when combined with other precursors in acid rain and ozone.* NO_χ *react in the air to form ground-level ozone and fine particle pollution, which are associated with adverse health effects. Principally, and for a long time, transport vehicles and industrial emissions are the major sources of the pollutants emitted in the Klang Valley atmosphere. Following the increase of urbanization and industrialization, the amount of traffic has increased in the Klang Valley. Traffic is considered as the major source of* NO_χ *in the Klang Valley. Exhaust emissions from the petrol-powered motor vehicles include carbon monoxide, nitrogen oxide, hydrocarbons and particulate matter. A time series analysis of* NO_χ *monitoring data from six locations in the Klang Valley Region from 1997 to 2002 is presented. The aim of this study is to evaluate the variations and trends in* NO_χ *in the Klang Valley Region for the period 1997-2002. On an hourly and daily average basis, annual, seasonal, monthly and diurnal variations and trends in* NO_χ *concentrations are presented. The overall average daily concentration of* NO_χ *ranged from 0.03 ppm to 0.18 ppm, with maximums of about 0.32 ppm in Kuala Lumpur, with 98th percentiles in the range 0.17-0.28 ppm. It is of particular interest that all stations in 1997 showed high* NO_χ *concentrations due to haze episode. The highest level of* NO_χ *recorded in 1997 was 0.3 ppm in Kuala Lumpur. Variations in* NO_χ *were dominated by one daily peak at the morning rush hours and secondary peak in late evening. This suggests that the level of* NO_χ *during peak hour is higher and it can be attributed to the increase in traffic volume. In the Klang Valley Region, tremendous growth of motor vehicles has resulted in increasing CO and* NO_χ *emissions. Therefore, the role of source controls over emissions is stressed as a key management tool especially in relation to road transportation.*

ABSTRAK

Pelepasan NO_x *oleh kegiatan manusia menyumbang sebahagian besar nitrogen ke dalam udara. Sumber utama* NO_x *buatan manusia ialah proses pembakaran dalaman bersuhu tinggi seperti kenderaan bermotor dan stesen janakuasa.* NO_x *mendatangkan banyak kesan langsung, atau kesan tak langsung misalnya menghasilkan hujan asid dan ozon bila bercampur dengan perkusor lain.* NO_x *yang bertindak dalam udara membentuk ozon aras tanah dan partikel pencemaran yang boleh mendatangkan kesan kesihatan yang tidak diingini. Di Lembah Klang, kenderaan bermotor dan kegiatan perindustrian telah lama menjadi sumber utama pencemaran udara. Bilangan kenderaan bermotor di Lembah Klang bertambah akibat pertambahan urbanisasi dan industrialisasi. Trafik ialah punca utama pelepasan* NO_χ *di sini. Pelepasan daripada ekzos kenderaan bermotor yang digerakkan oleh petrol termasuklah karbon monoksida, nitrogen oksida, hidrokarbon dan partikel terampai. Kajian ini bertujuan menilai variasi dan tren* NO_χ *di Lembah Klang bagi tempoh 1997-2002. Analisis siri masa terhadap data pemantauan* NO_χ *pada enam lokasi telah dilakukan. Variasi dan tren kepekatan* NO_χ *mengikut jam, harian, tahunan, musiman dan bulanan dipersembahkan. Pada keseluruhannya, kepekatan purata harian* NO_χ *didapati berada sekitar 0.03 ppm hingga 0.18 ppm, dengan aras maksimum sekitar 0.32 di Kuala Lumpur, dan persentil ke 98 berada antara 0.17 ppm dengan 0.28 ppm. Suatu hal yang menarik ialah hakikat bahawa semua stesen menunjukkan aras kepekatan* NO_χ *yang tinggi pada tahun 1997 disebabkan oleh jerebu. Aras tertinggi tahun 1997 ialah 0.3 ppm yang direkodkan di Kuala Lumpur. Variasi dalam* NO_χ *dicirikan oleh satu puncak harian utama pada waktu sesak sebelah pagi, dan satu lagi puncak sekunder pada lewat petang. Ini menunjukkan aras* NO_χ *adalah tinggi pada waktu puncak, dan ini berkait rapat dengan pertambahan isipadu kenderaan. Di Lembah Klang, pertambahan kenderaan bermotor yang cepat menyebabkan pertambahan pelepasan CO dan* NO_χ *. Oleh itu, kawalan terhadap penggunaan pengangkutan jalan diketengahkan sebagai kaedah pengurusan yang utama dalam menangani bahan pencemaran ini.*

INTRODUCTION

Nitrogen oxides, the generic term for a group of highly reactive gases that contain nitrogen and oxygen in varying amounts, play a major role in the formation of ozone, PM, haze and acid rain. Nitrogen oxides (NO_χ) and sulfur oxides (SO_χ) are criteria air pollutants, emitted in large quantities from fossil-fueled electric power plants (Kasper et al. 1996). The oxides of nitrogen (NO_χ) include seven known gaseous compounds; however, only nitric oxide (NO) and nitrogen dioxide (NO_2) are emitted in significant quantities to the atmosphere (Rowe et al. 1991). Most NO_χ from combustion sources (about 95 percent) are emitted as NO; the remainder are largely NO_2 (Sloss 1998), because NO is readily converted to NO_2 in the environment. A variety of NO_χ compounds and their transformation products occur both naturally and as a result of human activities. Anthropogenic emissions of NO_χ account for a large majority of all nitrogen inputs to the environment. The major sources of man-made NO_χ emissions are high-temperature combustion processes such as those that occur in automobile and power plants. Natural sources of NO_χ are lightning, biological and stratospheric intrusion. Ammonia and other nitrogen compounds produced naturally are important in the cycling of nitrogen through the ecosystem. Atmospheric transformation of NO_χ can lead to the formation of ozone and nitrogen-bearing particles (e.g., nitrates and nitric acid). Nitrogen oxides contribute to a wide range of effects on public welfare and the environment, including global warming and stratospheric ozone depletion (U.S Environmental Protection Agency 1998). Nitrogen alone or in acid rain, also can acidify soils and surface waters. Acidification of soils causes the loss of essential plant nutrients and increased levels of soluble aluminum that are toxic to plants. Acidification of surface waters creates conditions of low pH and levels of aluminum that are toxic to fish and other aquatic organisms. NO_χ also contribute to visibility impairment.

DATA AND METHODS

This paper reports on measurements of NO_χ performed over several years at six stations within the Klang Valley Region, namely; Country Heights (Kajang), Victoria Institution (Kuala Lumpur), Sekolah Rendah Kebangsaan (Petaling Jaya), Jabatan Bekalan Air Daerah (Gombak), Shah Alam and Sekolah Menengah Perempuan Raja Zarina (Klang). The measuring project has been conducted by Alam Sekitar Malaysia Sdn. Bhd. (ASMA) which was started in mid 1996 and continues to date. We have analyzed the NO_χ hourly and daily data from 1997 to 2002 provided by the DOE and ASMA for six representative locations in the Klang Valley Region.

The first consideration in determining whether there is a trend in the air quality measurements is to select the parameter, or parameters, of interest. Useful parameters for example, are the annual average and certain specified percentiles such as the 50th and 98th percentiles.

Techniques that measure the statistical significance of trends usually involve correlation of concentrations in the air of the pollutant with the sequence in which they are observed. Since the time interval between observations is not considered, missing observations can be ignored (WHO 1980). In this respect, a technique often used is the Daniel's test for trends using the Spearman rank correlation coefficient. To utilize this procedure, at least 4 time periods should be available. Given time period Xi, …Xn and their corresponding value (e.g. yearly average 1,…yearly average n) ranked from the lowest to the highest, the test statistic is calculated as the rank-correlation coefficient. The Daniel's test will be used because it is a powerful test designed specifically for detecting the trend in any time series data (WHO 1984). A description of this test is given as follows: the hypothesis H_0: The time series has no trend. The alternative hypothesis H_1: The time series has a trend (upward or downward). The Daniel's test statistic is:

$$r_s = 1 - \left[6 \sum^{N} i = 1 di^2 \right] / \left[N^3 - N \right] (1)$$

where N is sample size, *di* is the difference between the *x* variable (time starting with period one through period *N*) and the *y* variable ranked by measured concentration for ith observations.

The absolute value of r_s (the coefficient of rank correlation), is compared with a critical value Wp in a statistical table of Spearman rank correlation coefficients. If $|r_s| >$ Wp then a trend is declared significant. A negative value of r_s indicates a downward trend. Then decision rule is: Reject H_0. Note that if r_s is –ve then the trend is downward. The detailed analysis is discussed below.

RESULTS AND DISCUSSIONS

Statistical Characteristics of NO_χ

Summaries of the daily statistics of the NO_χ measurements for the Klang Valley Region are given in Tables 1a-1f, respectively. The overall average daily concentration of NO_χ ranged from 0.03 ppm to 0.18 ppm, with maximums of about 0.32 ppm in 1997 at Kuala Lumpur, with 98th percentiles in the range 0.06-0.28 ppm. While the low NO_χ concentrations were observed at Gombak (0.03-0.04 ppm) and Kajang

Table 1a. Daily concentrations of NO_χ (ppm) in Kajang, 1997-2002

		Percentile							Statistic				
Year	NO	10	30	50	70	90	95	98	Max	Min	Mean	Mode	SD
1997	345	0.02	0.04	0.05	0.06	0.08	0.09	0.10	0.11	0.01	0.05	0.03	0.02
1998	357	0.02	0.04	0.05	0.06	0.07	0.08	0.09	0.13	0.01	0.05	0.04	0.02
1999	365	0.03	0.04	0.05	0.06	0.07	0.08	0.09	0.10	0.01	0.05	0.05	0.02
2000	364	0.03	0.04	0.05	0.06	0.07	0.08	0.09	0.11	0.01	0.04	0.04	0.02
2001	365	0.03	0.04	0.05	0.06	0.07	0.07	0.09	0.11	0.01	0.05	0.05	0.02
2002	365	0.02	0.02	0.04	0.05	0.07	0.07	0.08	0.10	0.01	0.04	0.02	0.02

Table 1b. Daily concentrations of NO_χ (ppm) in Kuala Lumpur, 1997-2002

		Percentile							Statistic				
Year	NO	10	30	50	70	90	95	98	Max	Min	Mean	Mode	SD
1997	313	0.13	0.16	0.18	0.21	0.24	0.26	0.28	0.32	0.05	0.18	0.14	0.04
1998	360	0.08	0.11	0.14	0.16	0.19	0.21	0.23	0.28	0.03	0.14	0.17	0.04
1999	363	0.07	0.10	0.12	0.15	0.17	0.19	0.21	0.25	0.01	0.12	0.10	0.04
2000	366	0.07	0.09	0.11	0.12	0.15	0.16	0.17	0.21	0.03	0.11	0.11	0.03
2001	365	0.05	0.08	0.10	0.11	0.14	0.15	0.17	0.19	0.02	0.09	0.09	0.03
2002	365	0.08	0.10	0.11	0.12	0.14	0.15	0.17	0.24	0.04	0.11	0.11	0.02

Table 1c. Daily concentrations of NO_χ (ppm) in Petaling Jaya, 1997-2002

		Percentile							Statistic				
Year	NO	10	30	50	70	90	95	98	Max	Min	Mean	Mode	SD
1997	365	0.07	0.09	0.10	0.11	0.13	0.15	0.17	0.18	0.02	0.10	0.10	0.03
1998	361	0.06	0.08	0.09	0.11	0.13	0.14	0.15	0.18	0.03	0.09	0.07	0.02
1999	365	0.06	0.08	0.09	0.10	0.12	0.13	0.15	0.20	0.02	0.09	0.08	0.02
2000	352	0.06	0.08	0.10	0.12	0.16	0.18	0.20	0.22	0.02	0.10	0.09	0.04
2001	365	0.06	0.07	0.08	0.10	0.12	0.13	0.14	0.15	0.03	0.09	0.08	0.02
2002	355	0.06	0.07	0.08	0.09	0.11	0.12	0.13	0.17	0.01	0.08	0.08	0.02

Table 1d. Daily concentrations of NO_χ (ppm) in Gombak, 1997-2002

		Percentile							Statistic				
Year	NO	10	30	50	70	90	95	98	Max	Min	Mean	Mode	SD
1997	361	0.02	0.03	0.03	0.04	0.05	0.06	0.06	0.09	0.01	0.04	0.03	0.01
1998	361	0.02	0.03	0.03	0.04	0.04	0.04	0.05	0.06	0.00	0.03	0.03	0.01
1999	363	0.02	0.03	0.03	0.04	0.04	0.05	0.05	0.06	0.01	0.03	0.03	0.01
2000	366	0.02	0.03	0.03	0.04	0.05	0.05	0.06	0.07	0.01	0.03	0.03	0.01
2001	356	0.02	0.03	0.04	0.04	0.05	0.05	0.06	0.07	0.02	0.04	0.04	0.01
2002	356	0.03	0.04	0.04	0.05	0.06	0.06	0.07	0.08	0.01	0.04	0.05	0.01

Table 1e. Daily concentrations of NO_χ (ppm) in Shah Alam, 1997-2002

		Percentile							Statistic				
Year	NO	10	30	50	70	90	95	98	Max	Min	Mean	Mode	SD
1997	191	0.03	0.04	0.04	0.05	0.07	0.07	0.08	0.10	0.01	0.04	0.04	0.01
1998	365	0.01	0.02	0.03	0.04	0.06	0.06	0.07	0.09	0.01	0.03	0.03	0.02
1999	365	0.01	0.02	0.03	0.04	0.05	0.06	0.07	0.09	0.00	0.03	0.02	0.01
2000	366	0.01	0.03	0.03	0.04	0.05	0.06	0.07	0.08	0.01	0.03	0.03	0.01
2001	358	0.01	0.03	0.03	0.04	0.06	0.06	0.07	0.09	0.01	0.03	0.04	0.01
2002	357	0.02	0.03	0.04	0.04	0.05	0.06	0.06	0.07	0.01	0.04	0.03	0.01

Table 1f. Daily concentrations of NO_χ (ppm) in Klang, 1997- 2002

		Percentile							Statistic				
Year	NO	10	30	50	70	90	95	98	Max	Min	Mean	Mode	SD
1997	345	0.02	0.04	0.05	0.06	0.08	0.09	0.10	0.11	0.01	0.05	0.03	0.02
1998	357	0.02	0.04	0.05	0.06	0.07	0.08	0.09	0.13	0.01	0.05	0.04	0.02
1999	365	0.03	0.04	0.05	0.06	0.07	0.08	0.09	0.10	0.01	0.05	0.05	0.02
2000	364	0.03	0.04	0.05	0.06	0.07	0.08	0.09	0.11	0.01	0.05	0.04	0.02
2001	365	0.02	0.04	0.05	0.06	0.07	0.08	0.09	0.11	0.01	0.05	0.05	0.02
2002	365	0.03	0.04	0.05	0.06	0.07	0.08	0.09	0.10	0.01	0.05	0.06	0.02

(0.04-0.05 ppm), respectively. The low NO_χ levels indicate little immediate influence of urban activities. At all station the unusually high concentrations were observed in 1997 due to serious haze episode.

Annual Variation and Trend of NO_χ Concentrations

Figures 1a-f and Table 2 show annual mean (50th percentile and 98th percentile) of NO_χ concentrations over the Klang Valley Region for 1998-2002 (excluding the 1997 haze episodes year which effected the annual trend), which have changed little over the period. It is of particular interest that three stations (Kuala Lumpur, Shah Alam and Klang) in 1998 showed high NO_χ concentrations. NO_χ levels present a clear upward trend at all stations except Kuala Lumpur. The NO_χ decrease reaches about -0.003 ppm in Kuala Lumpur over a 5-year period. The decreases in NO_χ emissions Kuala Lumpur over the 1998-2002 for NO_χ are attributed to traffic and industrial source emission reductions. Whereas NO_χ concentrations in Petaling Jaya, Gombak, Shah Alam and Klang showed small increases over 1998-2002.

Petaling Jaya presents a clear upward trend for NO_χ, indicating the large influence of industrial activities and the nearby highway. The results show increasing trends over 1998-2002 at Kajang and Gombak stations which is significant at 0.05 level. Other stations like Petaling Jaya, Shah Alam and Klang do not present any significant trend in NO_χ concentrations during 1998-2002. Analysis of the 50th and 98th percentiles

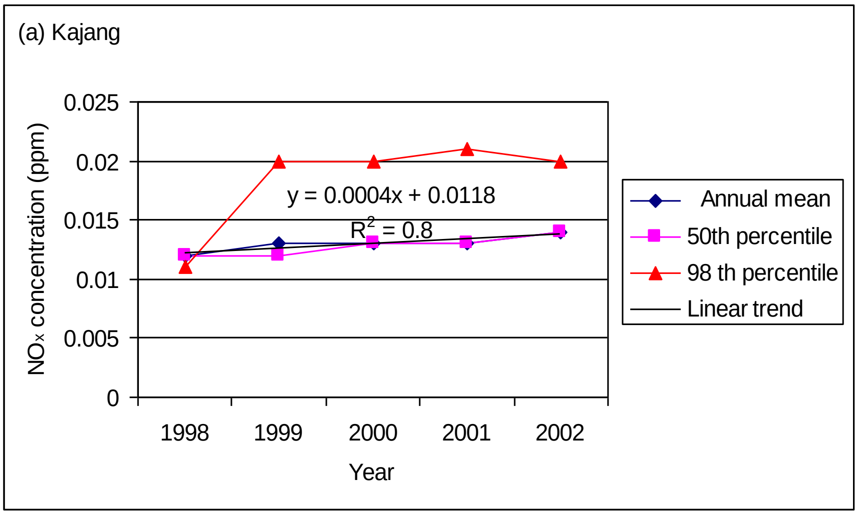

Figure 1a. Annual mean, 50th percentile and 98th percentile of NO_χ concentrations in Kajang, 1998-2002

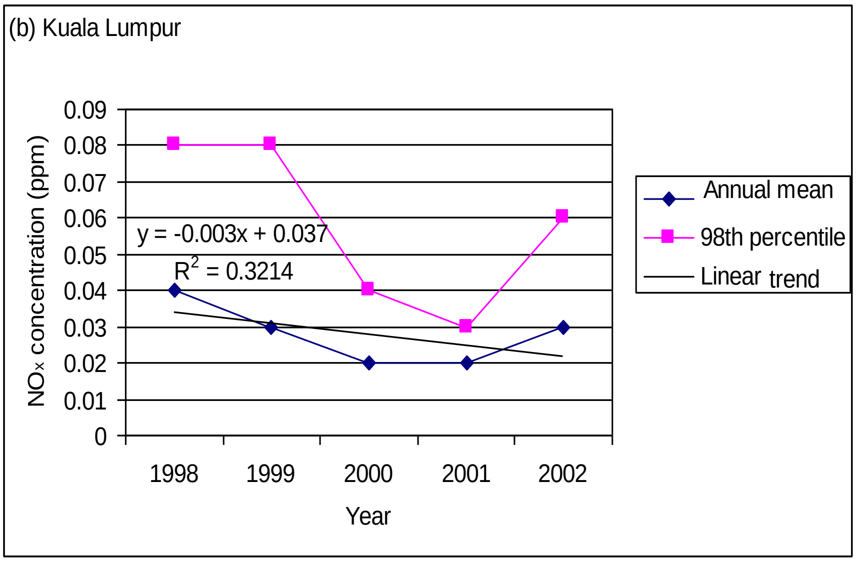

Figure 1b. Annual mean and 98th percentile of NO_χ concentrations in Kuala Lumpur, 1998-2002

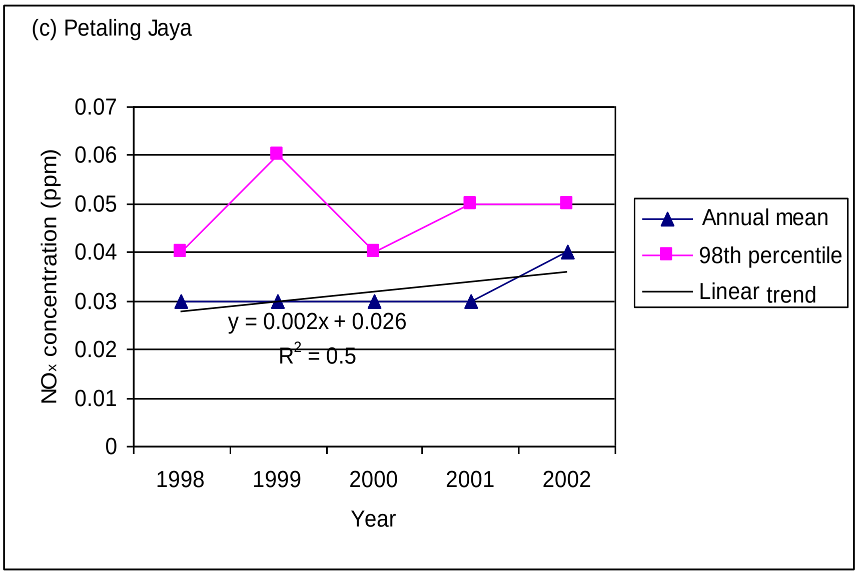

Figure 1c. Annual mean and 98th percentile of NO_χ concentrations in Petaling Jaya, 1998-2002

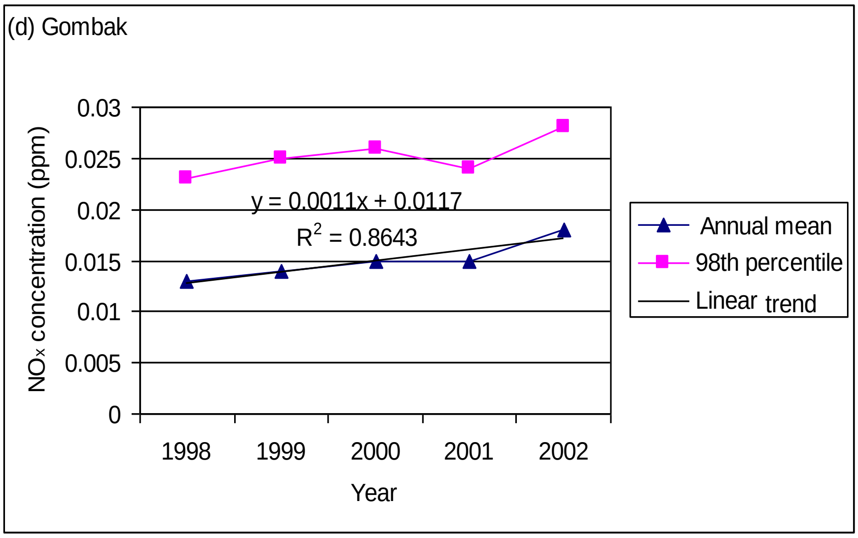

Figure 1d. Annual mean and 98th percentile) of NO$_\chi$ concentrations in Gombak, 1998-2002

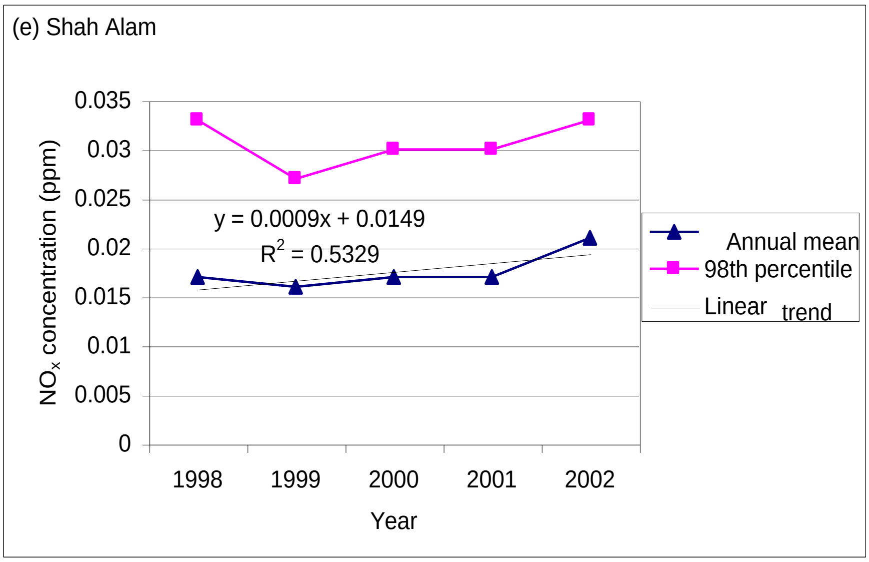

Figure 1e. Annual mean and 98th percentile of NO$_\chi$ concentrations in Shah Alam, 1998-2002

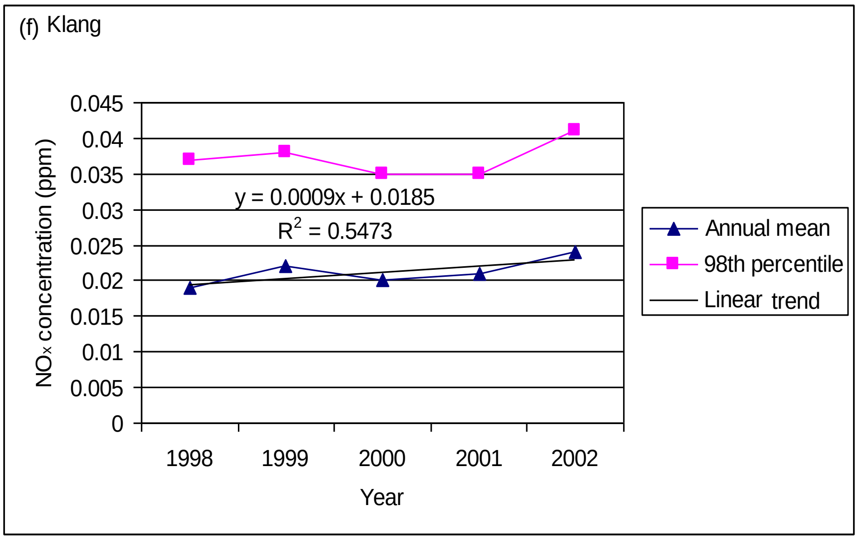

Figure 1f. Annual mean and 98th percentile of NO_{χ} concentrations in Klang,1998-2002

Table 2. Linear regression and slope of NO_{χ} annual mean concentrations (ppm) in the Klang Valley Region for 1998-2002

Station	Constant	Slope	r^2	Sig.
Kajang	0.012	0.0004	0.80	0.041
Kuala Lumpur	0.037	-0.003	0.32	0.319
Petaling Jaya	0.026	0.002	0.50	0.181
Gombak	0.012	0.001	0.86	0.022
Shah Alam	0.015	0.001	0.53	0.161
Klang	0.018	0.001	0.55	0.153

of annual NO_{χ} levels results in small positive or negative trends at some stations, and they were statistically not significant. For example, the values of r_s for Kuala Lumpur were -0.7 and -0.6 for 50th and 98th percentiles respectively, which are smaller than the critical value of Wp = 0.90 to indicate that a series has a trend. Since $r_s < Wp$, we may conclude that the downward trends in the study area were not significant at 0.05 level of significance.

Seasonal Variation and Trend of NO_{χ} Concentrations

Seasonal variations of NO_{χ} concentrations in the Klang Valley Region for 1997-2002 are shown in Figure 2. The NO_{χ} concentrations were highest during southwest monsoon season at Kuala Lumpur (0.13 ppm) and lowest during northeast monsoon at Kajang and Gombak (0.013 ppm) and during transitional monsoon (October-November) at Kajang (0.013 ppm). During transitional monsoon (April-May) higher and more regular levels of NO_{χ} are observed at Kuala Lumpur, Petaling Jaya and Shah Alam stations. The seasonal difference in NO_{χ} levels is found to be small. The highest concentrations occurred during southwest monsoon followed by transitional monsoon (April-May), northeast monsoon and transitional monsoon (October-November) seasons. The monthly mean concentrations coincide well with seasonal cycle with higher values occurring during the month of dry seasons and lower concentrations during rainy seasons. The seasonal means of NO_{χ} concentrations suggest that some reduction in NO_{χ} levels values occurred during the transitional monsoon (October-November) and northeast monsoon seasons, perhaps due to rain that washed out the NO_{χ}.

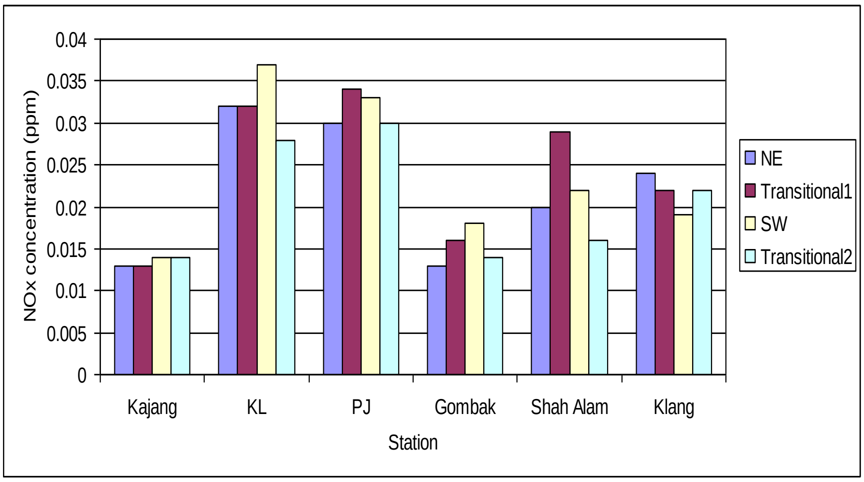

Figure 2. Seasonal variation of NO_{χ} concentrations (ppm) in the Klang Valley Region, 1997-2002.

Monthly Variation and Trend of NO_{χ} Concentrations

Figure 3 shows monthly mean of NO_{χ} concentration in the Klang Valley Region for 1997-2002. There was an increase in the average levels of NO_{χ} during July and September, after the low values during April and May. From September to December the monthly NO_{χ} concentrations showed decreasing trends. Generally, NO_{χ} concentrations were relatively high during dry months but low during rainy months which coincide with the high amounts of precipitation received in November and December. The general trend of decreasing NO_{χ} concentrations seems to be well matched inversely with the monthly variability in cloud cover and rainfall.

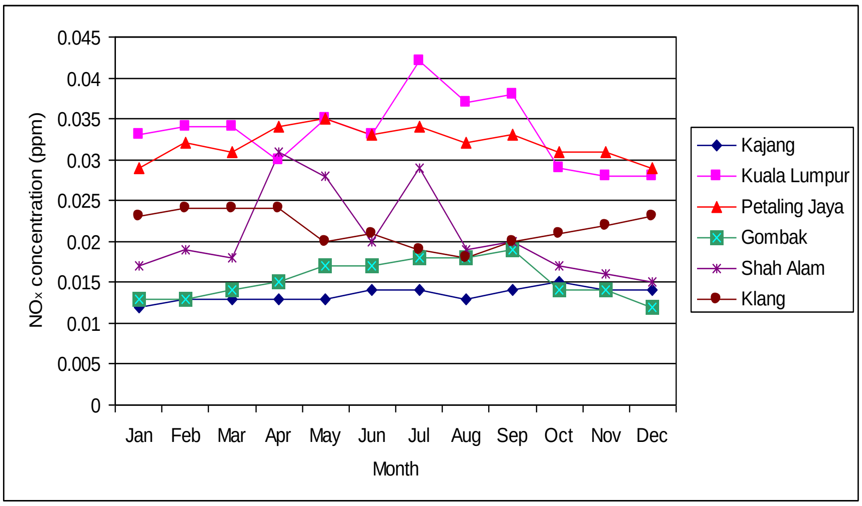

Figure 3. Monthly variation of NO_χ concentrations (ppm) in the Klang Valley Region, 1997-2002

Diurnal Variation of NO_χ Concentrations

Figures 4a and 4b show the diurnal pattern of NO_χ for 1997 and 2002, respectively. Total NO_χ concentration had a pronounced diurnal maximum at 09:00 a.m. Trends in NO_χ are dominated by one daily peak during the morning rush hours and a secondary peak in late evening. The highest level of NO_χ recorded during the morning was 0.3 ppm in 1997 at Kuala Lumpur station. This suggests that the level of NO_χ during peak hour is higher and it can be attributed to the increase in traffic volume.

It is not surprising to find that the highest concentrations are observed in Kuala Lumpur, where the measurement site accommodated high daily flow of vehicles, giving rise to higher concentrations of NO_χ. Furthermore, Kuala Lumpur is in a busy commercial area of the Klang Valley Region where traffic forms an important source of NO_χ emissions. NO_χ concentrations were relatively much lower in Gombak and Kajang in comparison with the large cities of Kuala Lumpur and Petaling Jaya. Nitrogen oxides concentration in the Klang Valley Region is generally similar to that for NO_2 reflecting the fact that population density, road traffic and industrial activities are closely related contributors to pollution (Yasser 2004).

It was also observed that concentrations in the morning peak were higher compared to the evening peak. This observation conforms to normal traffic behaviour. Sham (1987) reported that during the morning when traffic is building up, air pollution concentration would increase quite substantially in the city area. The dispersion of pollutants during this period is relatively restricted particularly due to low wind speed. In the afternoon, as convection becomes more vigorous, the pollutants, which have been blanketing the city during the morning, will then be dispersed causing the city air to clear somewhat.

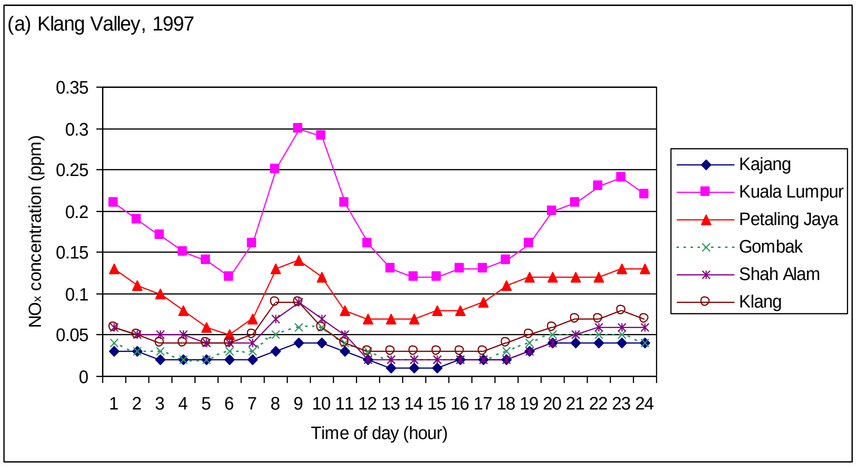

Figure 4a. Diurnal variation of hourly NO_χ concentrations (ppm) in the Klang Valley Region, 1997.

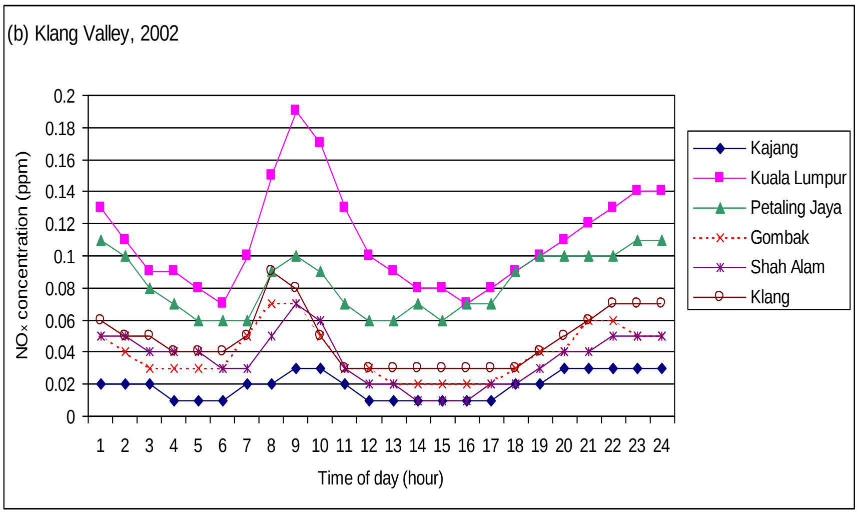

Figure 4b. Diurnal variation of hourly NO_χ concentrations (ppm) in the Klang Valley Region, 2002.

CONCLUSION

The results show that the highest concentrations are observed in Kuala Lumpur, where the measurement site accommodated higher daily flow of vehicles, giving rise to higher concentrations of NO_χ. Furthermore, Kuala Lumpur is located in the busiest area of the Klang Valley Region where traffic forms an important source of NO_χ emissions. NO_χ concentrations were very low in Gombak and Kajang in comparison with the large cities of Kuala Lumpur and Petaling Jaya. It is of particular interest that all stations in 1997 showed high NO_χ concentrations due to haze episode. NO_χ levels do present a clear trend at most of the stations. Diurnal trends in NO_χ are dominated by one daily peak at the morning rush hours and a secondary peak in late evening. Therefore, as a management tool it is suggested that emissions, especially in relation to road transport, should be controlled at source

ACKNOWLEDGEMENTS

Sincere thanks and gratitude are expressed to Alam Sekitar Malaysia Sdn. Bhd. (ASMA) and the Department of Environment (DOE) for providing valuable data used for this paper.

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