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Variability of NO_x and NO_2 concentrations observed at pedestrian level in the city centre of a medium sized urban area

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Abstract

NO_x and NO_2 concentrations were measured at different locations in a city centre of an urban zone (Population 450 000) in order to study the variation of the outdoor exposure at pedestrian level. These measurements were carried out to understand the influence of traffic emissions at each measured site. The observations were done during four weeks in winter, including several days with high pollution levels. The results at different locations have been used to analyse criteria recommended for locating observation sites in a monitoring network. No large differences in background pollution averaged over several weeks have been found throughout the city centre, even during pollution peaks. Measurements were also carried out inside one street canyon. The contribution of the street traffic to the $NO = NO_x - NO_2$ concentrations observed at side-walk has been found important, i.e., several times the background level. On the other hand, the majority of observed NO_2 pollution is due to the contribution of background pollution within the street. The pollutant excess at pedestrian level is strongly correlated to the street traffic emission and to the atmospheric turbulence observed at roof level. Application of a box model to the street data demonstrates that such models can be useful to estimate the pollutant accumulation within the street.

Keywords: NO_x; Traffic emission; Urban pollution; Street canyon

1. Introduction

Exposure to traffic emissions is believed to result in significant risk to human health. To assess this exposure it is necessary to know 'how much' and 'when' people breathe the pollutants. In urban areas, time-varying traffic activities result in differences between short term peaks and long term means. Source emission patterns result in spatial variabilities within and between neighbourhoods (Moon, 2001). In order to assess the exposure variations, observations were carried out at several locations in a city, and the measurements used to analyse spatial and short term pollution variabilities.

In the present study, measurement were obtained with the devices and the methodology used in air quality monitoring networks. That makes it possible to translate results from this study to other cities and also to interpret existing data obtained by such networks.

In France and in Europe, several criteria are used to check the representativeness of observations obtained at a particular site (LCSQA, 1999; EUROAIRNET, 1999); this results in a classification of the monitoring stations. The present study was focused on the assessment of traffic exposure in an urban area, so only two kinds of site were used, 'background' and 'roadside' locations. The measurements were interpreted with a view to providing some insights into the representativeness of such monitoring stations.

The measurements are described in the first section. Next, observations obtained at two background stations

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in the city centre were compared and analysed as a function of the wind direction. The short term variability is compared to the differences observed over longer periods. In the last section, information on traffic-related contributions versus 'far-travelled' contributions to pollutant concentrations observed within a street are provided.

2. Geographical layout and measurement descriptions

The pollutant observations were carried out during one month in winter and in a medium sized urban area, the Rouen City, in the west of France. The city has a population of 450 000 inhabitants. Fig. 1 shows the city layout and indicates heaviest traffic lanes. At 10 km SW from the city centre, there is an important industrial zone and at 2 km SE a waste incinerator. Total NO_x emissions are estimated as 7949 ton yr^{-1}, of which traffic accounts for 46%, industry 47% and heating a mere 7%. The air pollution is thus equally from traffic and industry.

During four weeks (19 Jannuary–16 February 1998), NO_x concentrations were measured at different locations shown in Fig. 2. Average values were calculated every 15 min and the sampling was performed at 2 m height. Two background sites were used: one is located in the city centre, the other is in the residential neighbourhood of the city, 2.5 km far from the 'Hachette' site and in the court of the "Carrel" school. Two other stations were installed on the kerb of busy streets, "rue Crevier" and "bd de la Marne", located in the vicinity of the Hachette School, as shown in Fig. 2. All results presented here were obtained during working days. Traffic volumes were monitored at the two street sites, "bd de la Marne" which is a large busy avenue, and "rue Crevier", which is a street canyon. Meteorological observations were performed at a station placed at roof level and close to the Hachette school (see Fig. 2). All wind directions were observed during the measurement campaign. The monitoring period included several days during which there were calm episodes and the meteorological conditions were not favourable to pollutant dispersion.

3. Results obtained at background stations

In France, two criteria are used to check the representativeness of an urban background station (LCSQA 1999). First, the annual average value of the NO/NO_2 ratio must be less than 1.5 in ppb/ppb. Secondly, the sampling must be carried out at a minimum distance from a busy road, this distance being a function of the traffic intensity. The two sites chosen in the present study, 'Carrel' and 'Hachette', satisfy these criteria. However, their different locations in the city, as shown in Fig. 1, may suggest differences in the observations.

Figs. 3 and 4 show the NO_x and NO_2 concentrations measured at 'Carrel' plotted as a function of the 'Hachette' values. Considerable variability is seen between paired values. This can be explained by the wind direction, as discussed below, or by the atmospheric turbulence and the instantaneous mixing which are not homogeneous in the lower part of the urban boundary layer when observed over short time. However, the shape of the scatter plot is oblong suggesting a relationship between the average values. We applied a linear least square regression to both NO_x and NO_2 series and the results are given in Table 1. The two correlation coefficients are high and the two slopes are

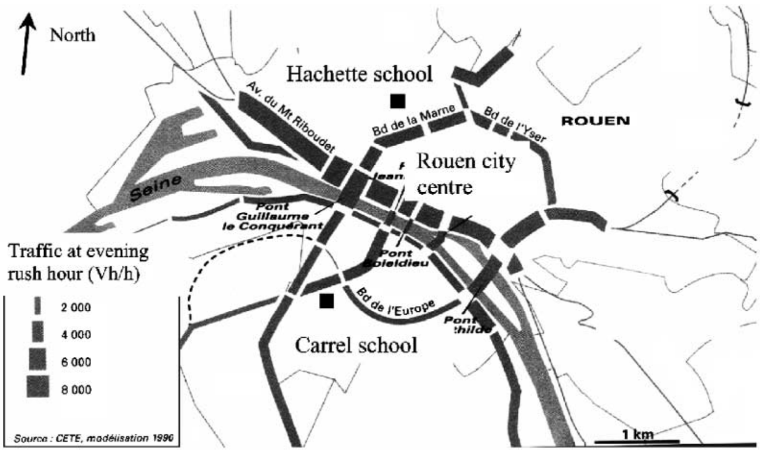

Fig. 1. Study are and main traffic flows: the square dots show the location of the background monitoring sites.

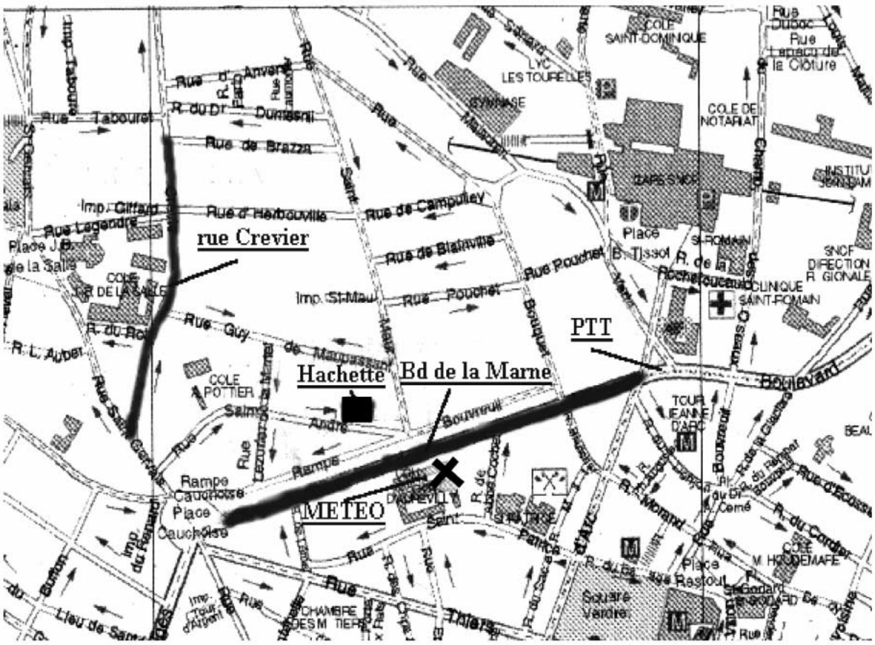

Fig. 2. Detail of the city centre, the circles indicate the location of the street sites.

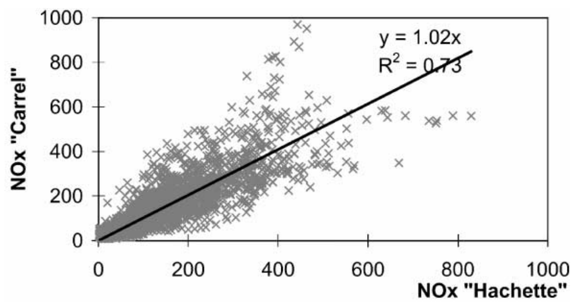

Fig. 3. Scatter plot for NO_x observations ($mg\,m^{-3}$) obtained at the two background sites.

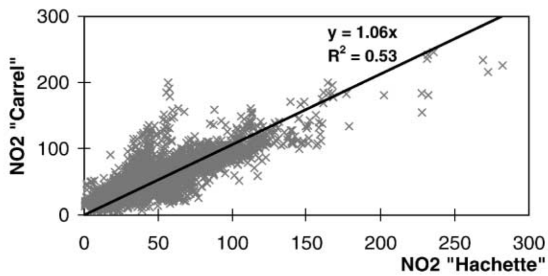

Fig. 4. Scatter plot for NO_2 observations ($mg\,m^{-3}$) obtained at the two background sites.

close to one. These indicate a linear correlation for the NO_x and NO_2 pollutants when they are observed over a long period, one month in the present work.

Mean values of NO_x or NO_2 are reported in Figs. 5 and 6 as a function of the wind sectors, and the corresponding mean wind speeds are given in Fig. 7 (wind direction is equal to 0° and 180° for wind blowing from north and south, respectively). Comparison of Figs. 5 and 6 with Fig. 7 shows that the concentration is a function of the inverse of wind speed, higher pollution level corresponding to low wind speed. Careful examination of Figs. 5 and 6 shows that "Carrel" observations are slightly greater for winds from the North (sectors 45° and 360°), while the opposite is true for "Hachette", where concentrations are greater for winds from the SW (sector 225°). The explanation is suggested by the geographical location of the two sites. As we can see in Fig. 1, the "Carrel" station is located to the SW of the urban zone and is thus downwind of the main emission sources for wind blowing from north, the opposite of "Hachette" site. However for all wind sectors, the differences remain low and would become less if averaged over longer period (e.g. 1 yr). Values averaged over all wind sectors are given in Table 1, and the relative difference is 5%.

We have to remember there is an important industrial area to the SW. As a result, we would expect differences between "Carrel" and "Hachette" observations for

Table 1
Statistic of the observations obtained at the two background sites "Hachette" and "Carrel" during one month in winter. Parameters of the linear least squares regression between the two site observations; mean and r.m.s. values for both sites; mean and r.m.s. values for the difference

	Parameters of the linear least squares regression (Figs. 3 and 4)		Mean (all wind sectors in Figs. 5 and 6)		R.m.s. (all wind sectors in Figs. 8 and 9)		Difference (Hach–Carrel) Distribution			
	Slope	Correlation coefficient R_2	Hach	Carrel	Hach.	Carrel	Mean	R.m.s.	Kurtosis	Asymetry
NO_x (μg m^{-3})	1.02	0.73	119	125	113	121	9.6	61	10.5	1.5
NO_2 (μg m^{-3})	1.06	0.53	51	59	33	31	8.9	19.8	3.6	0.8

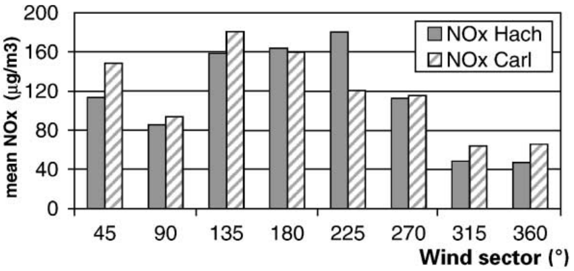

Fig. 5. Mean NO_x values for each wind sector; averaged value for all sectors: Hach = 119 and Carl = 125 in mg m^{-3} (labels are midpoints of sectors).

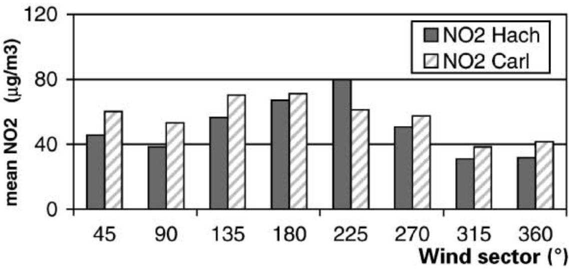

Fig. 6. Mean NO_2 values for each wind sector; averaged value for all sectors, Hach = 51 and Carl = 59 in mg m^{-3} (labels are midpoints of sectors).

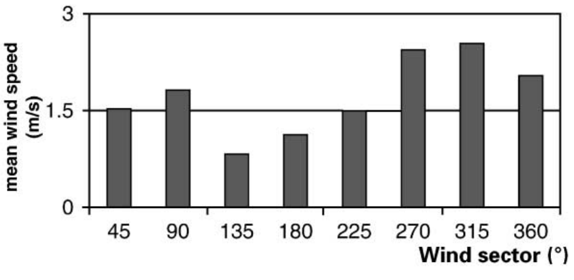

Fig. 7. Mean wind speed values for each wind sector; averaged value for all sectors: 1.5 m s^{-1}.

winds blowing from the industrial area toward the city (see wind sectors 180° and 225° in Figs. 5 and 6). That is not the case, that to probably because the industrial area is too far away (10 km) to have a noticeable impact on city centre air quality. This implies NO_x background pollution in the city centre is mainly due to local traffic emissions.

For each wind sector, we have also calculated the root mean square (r.m.s.) values; they are presented in Figs. 8 and 9 in a dimensionless form, as r.m.s./mean ratios. Corresponding ratios for wind speed are given in Fig. 10. In each case, the ratio is often greater than 0.5. For NO_x, it is some times greater than 1 and always greater than the one of NO_2, as it was also observed in other study (Moon, 2001). This reflects short time variability, as also shown in Figs. 3 and 4. However for each wind direction, the difference between the two r.m.s. values is small. Values averaged over all wind sectors are given in Table 1. From Figs. 8–10, we can see the shapes of pollutants and wind speed diagrams are similar, suggesting a relationship between pollutant concentration and wind turbulence intensity at the two sites.

The distribution of the differences between paired observations has also been analysed. The mean and root mean square values of this distribution are given in Table 1. The r.m.s. values are much greater than the mean, 6 times greater for NO_x and two times for NO_2. For NO_x, the distribution is not normal, the Kurtosis parameters given in Table 1 indicate a strong deviation from the normal distribution.

In general, therefore, the results in Figs. 3–10 show that the spatial variability over short time period (15 min) is great and the pollutant mixing at roof level is not sufficient to homogenise the traffic emissions throughout the city centre. However, over longer periods, such as the month in the present study, spatial variability is limited and no large differences are seen between background pollution concentrations in the city centre and in the suburb 2.5 km away.

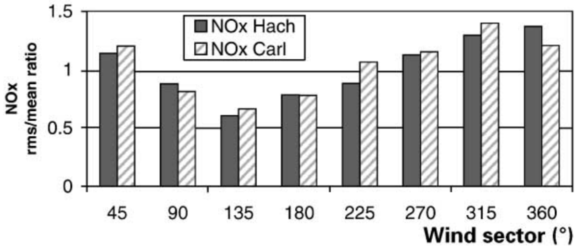

Fig. 8. NO_x rms/mean ratio for each wind sector; Averaged r.m.s value for all sectors, Hach = 113 and Carl = 121 in mg m^{-3}.

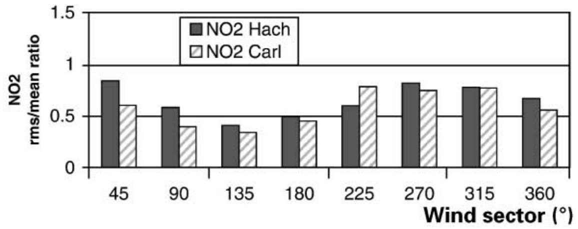

Fig. 9. NO_x r.m.s/mean ratio for each wind sector; Averaged r.m.s value for all sectors, Hach = 33 and Carl = 31 in mg m^{-3}.

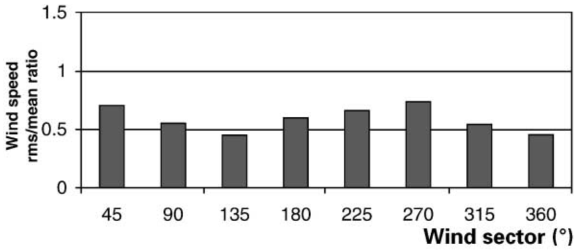

Fig. 10. Wind speed r.m.s/mean ratio for each wind sector; Averaged r.m.s value for all sectors: 1.1 m s^{-1}.

Mean concentration of the background NO_x concentration can be estimated in the city centre. From the annual emissions, we determine a theoretical value of the traffic and heating contribution. This has been proposed by Derwent et al. for London (Derwent et al., 1995), using the relation

$$C_{\text{bgd}}^{\text{traffic,heat}}(\mu\text{g m}^{-3}) = \sqrt{\frac{2}{\Pi}} \frac{1}{U_{\text{wind}} a(s-1)} (R_{\text{eff}})^{1-S}\, q,$$

which is an application of the Gaussian plume model to ground source area. In the above relation, q is total emission in $\mu\text{g m}^{-2}\text{s}^{-1}$ and R_{eff} is the effective radius of the city. The coefficients a and s are parameters giving the variation of the vertical dispersion parameter with distance x from the source, sigma $z = ax^s$ ($a = 0.05$ km and $s = 0.68$). In the case of Rouen, we have defined the effective radius of the city as the area inside which 85% of the population reside. This area measures 50 km^2 and

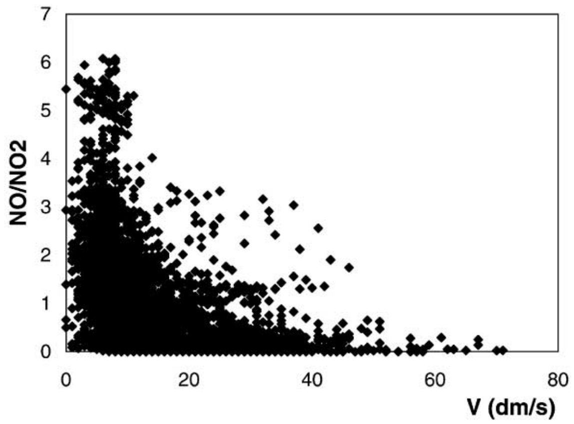

Fig. 11. NO/NO_2 ratio as a function of wind speed, observation at Hachette site.

thus R_{eff} is equal to 4 km. Application of the previous relation gives $C_{\text{bgd}}^{\text{traffic,heat}} = 39q$. With the NO_x emission inventory given above, $q_{\text{traffic+heat}}$. is equal to 2.67, so we find $C_{\text{bgd}}^{\text{traffic,heat}} = 104\ \mu\text{g m}^{-3}$. During the measurement campaign, the rural background concentration was $C_{\text{bgd}}^{\text{rural}} = 25\ \mu\text{g m}^{-3}$. So, the theoretical value of the city background concentration is $C_{\text{bgd}}^{\text{city}} = C_{\text{bgd}}^{\text{traffic,heat}} + C_{\text{bgd}}^{\text{rural}} = 129\ \mu\text{g m}^{-3}$. This value is in close agreement with the mean values shown in Table 1.

As Fig. 11 shows, the NO/NO_2 ratio is dependent on the meteorological conditions. Periods of low wind speed periods correspond to meteorological conditions which are not favourable to pollutant dispersion, and they tend to coincide with stable atmospheric conditions. Consequently NO concentrations increase over the urban zone; the production of NO_2, however, is limited by ozone availability, which is rather low in winter (Harrison, 1996). The average value of the ratio shown in Fig. 11 is 1.8 (in ppb/ppb). We can see that, under meteorological conditions which are not favourable to pollutant dispersion, marked variations of the ratio NO/NO_2 occur and values may be greater than 1.5 (in ppb/ppb).

4. Results obtained along the kerb of two busy streets

Average traffic flow on 'Boulevard de la Marne', a large busy avenue, is estimated as 30 000 Vh day^{-1} and in 'Crevier' street canyon 8000 Vh day^{-1}. The diurnal variations of traffic are typical of busy streets and are similar to observations reported elsewhere (Qin and Chan, 1993; Kemp, 1996; Gram, 1996). 'Crevier' street has an aspect ratio, defined by the height to the width, H/D, equal to 1.3. It thus has the characteristics of a street canyon and accumulation effects are often observed.

In cities, three criteria are generally used in France to test if a location is exposed to a close emission source: (1) the annual average value of the NO/NO_2 ratio must be greater than 2 in ppb/ppb; (2) there is a traffic emission source at less than 5 m and (3) accumulation effects may be expected by looking on basis of the aspect ratio H/D value. It is obvious that kerb stations located in 'Bd de la Marne' and 'rue Crevier' satisfy these criteria.

In order to calculate the contribution of the street traffic pollution to the pollution observed on the kerb, a background correction was applied. The background concentration was subtracted from the one measured on the kerb. We have analysed both NO and NO_2 contributions. The results for 'rue Crevier' are shown in Figs. 12 and 13, in which the relative contribution of the street traffic emission $C\% = (C_{kerb} - C_{bgd})/C_{bgd}$ have been plotted as a function of the traffic volume (counted during 15 min). The $C\%$ value averaged over one month is equal to 2.8 for NO and 0.6 for NO_2. Thus, street traffic is seen to be the main contribution to NO pollution, account for 74% of observed concentrations.

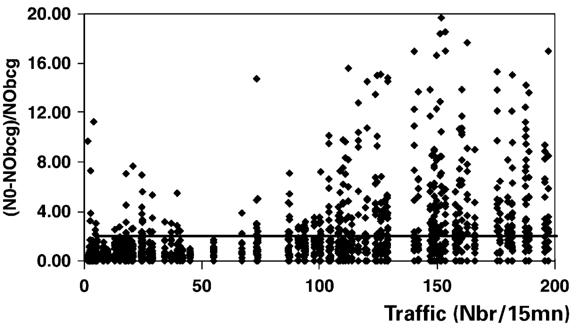

Fig. 12. Relative contribution $C_{NO}\%$ of the street traffic emission to the pollution observed on the kerb: $C_{NO}\% = (-NO - NO_{bcg})/NO_{bcg}$, the bold line represent the averaged value of $C_{NO}\%$.

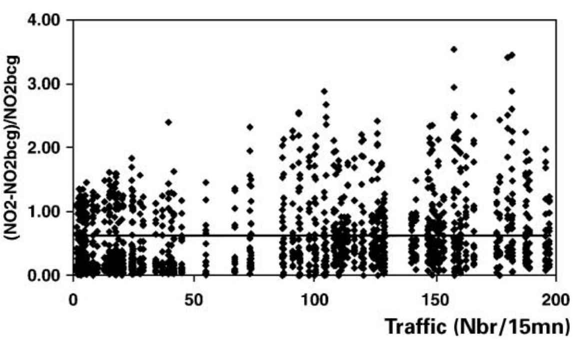

Fig. 13. relative contribution $C_{NO_2}\%$ of the street traffic emission to the pollution observed on the kerb: $C_{NO_2}\% = (NO_2 - NO_{2bcg})/NO_{2bcg}$, the bold line represent the averaged value of $C_{NO_2}\%$.

For NO_2, the traffic contribution is less (37%), suggesting that the majority of NO_2 is produced outside the 'Crevier' street.

During the measurement campaign, we had the opportunity to monitor the CO concentration at the 'bd de la Marne' site and at the Hachette school. The results for 'bd de la Marne' are plotted in Fig. 14 as a function of NO_x concentrations (a background correction has again been applied to both CO and NO_x concentrations). These results show that CO observations are correlated with those of NO_x. The average CO/NO_x ratio, which is given by the slope of the straight line drawn on Fig. 14, is equal to 12. The same measurements were also carried out at the PTT intersection, at one end of the boulevard (see Fig. 2), where we found an average CO/NO_x ratio of 18. Under assumptions discussed below, a relation can be established between the CO/NO_x ratio and the traffic emission rates.

Table 2 gives the emission rates, in g km^{-1}, for two urban speeds (INRETS, 1995; Joumard, 1995). Each value is calculated using the relations $e^{CO} = \sum_j \alpha_j e_j^{CO}$ and $e^{NO_x} = \sum_j \alpha_j e_j^{NO_x}$, in which α is the contribution of each vehicle type or each motor-fuel type.

The apportionment between trucks and light vehicles is not known with accuracy in this part of the city, but it can be assumed to be the same at the two sites. On the other hand, the traffic speed is greater in 'Bd de la Marne'. The average theoretical ratios may thus be approximated as about 15 for a fluid traffic and 29 for dense traffic. These theoretical values are high and are close to those observed, 12 in 'Bd de la Marne' and 18 at the 'PTT' intersection. This ratio provides a fingerprint of traffic emissions in the area. Factors for other source sectors are much smaller. For space heating, for example (Satehelin, 1997), the CO/NO_x ratio is one for both natural gas and oil combustion.

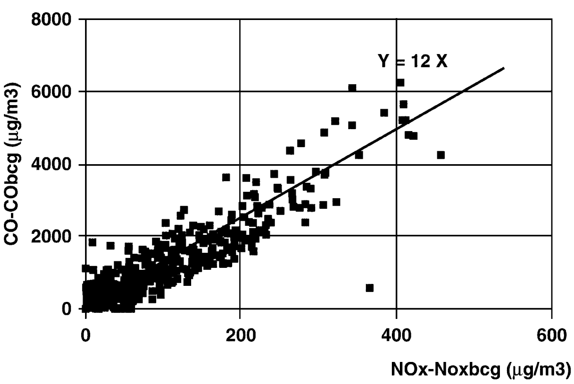

Fig. 14. CO as a function of NO_x, values observed on the kerb of the 'bd de la Marne', background concentration (Hachette site) has been subtracted.

Table 2
Traffic emission factors, VL is for light vehicles and T for trucks

Emission rate ($g\,km^{-1}$)	Low speed busy traffic VL + T	High speed fluid traffic VL + T	Low speed busy traffic VL alone	High speed fluid traffic VL alone
e^{CO}	28.5	14.0	27.4	12.9
e^{NO_x} [a]	1.2	1.2	0.8	0.7
e^{CO}/e^{NO_x}	23.7	11.7	34.2	18.4

[a] e^{NO_x} represent the true emission factor calculated from the NO_2 equivalent emission factor $e^{NO_x,\,equiv}$, $e^{NO_x}, = e^{NO_x,\,equiv}$ *MnO/MnO$_2$.

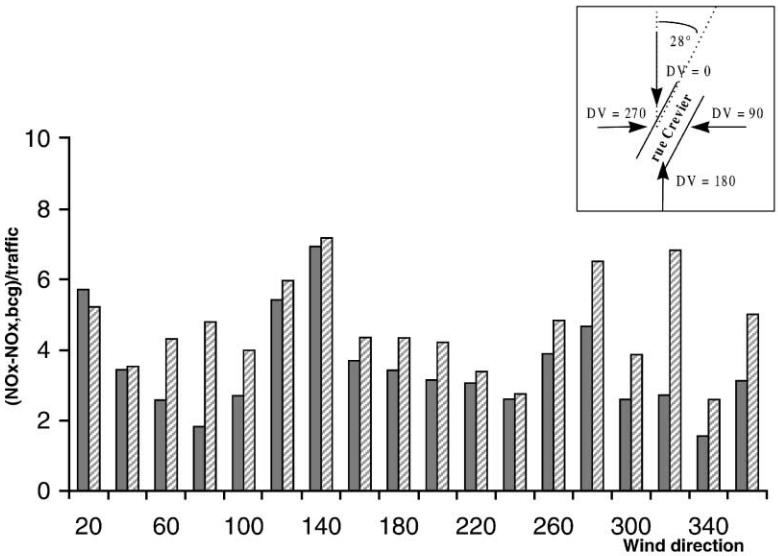

Fig. 15. Mean ratios of the street traffic pollution to the traffic volume, as a function of the wind direction, symbols are for all U_{wind} wind speed, dashed for $U_{wind} < 1\,m\,s^{-1}$.

Fig. 15 shows the ratio (NO_x–$NO_{x,bcg}$)/traffic volume as a function of the wind direction, which is perpendicular to the street axis for directions equal to 120° or 300°; sampling was downwind for 300°. For each wind direction, we have observed a large scatter in the data, since the r.m.s. value (not shown here) is some times 10 times greater than the mean value reported in Fig. 15. This is explained by the variability of the atmospheric conditions during the observation period. For all wind speeds observed during the campaign, we can see that the maximum concentrations are found for wind directions perpendicular to the street axis. This is a well known feature of the pollution pattern in street canyons (Berkowitcz, 1997; Rafailidis, 1997). However in Fig. 15, we also report values for wind speeds less than $1\,m\,s^{-1}$. In this case, we can see that the differences are not so pronounced. For example, values for wind directions between 60° and 100° increased 2–3 times; for wind directions at 120° and 140°, there is little difference between the two wind speeds. At low wind speed, therefore we do not find such a strong influence of wind direction on pollution levels observed at either side of the street canyon.

In Fig. 16, the same index, (NO_x–$NO_{x,bcg}$)/traffic, is reported as a function of the traffic volume. For each traffic volume, there exists a large scatter in the data, since the r.m.s. value may be ten times greater than the mean value. For low traffic ($<40\,Vh\,15\,min^{-1}$), the mean and r.m.s. values are high, these values must be interpreted with caution since they were calculated on the basis of a small number of vehicles. For traffic volume greater than $40\,Vh15\,min^{-1}$, the average values do not depend on traffic volume and are close to two (in $\mu g\,m^{-3} Vh\,15\,min^{-1}$).

The results shown in Figs. 15 and 16 suggest that pollutant accumulations in the street canyon can be estimated using a well mixed box model. In this approach, pollutant accumulations are driven by the balance between NO_x emissions and exchanges with the outside environment, so the relation between the

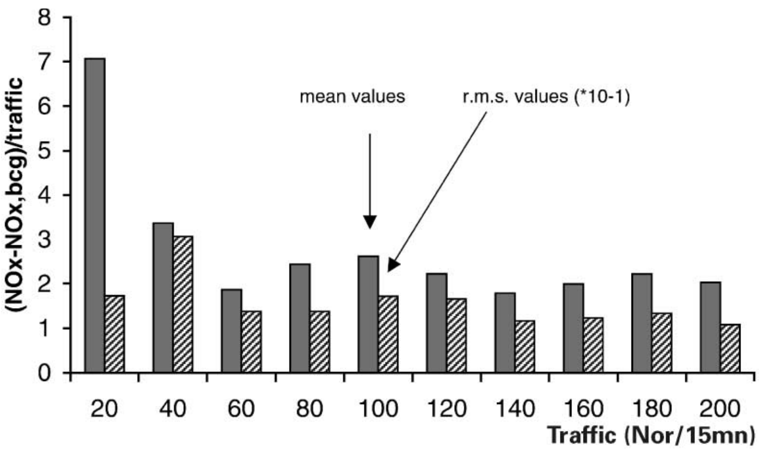

Fig. 16. Ratio of the street traffic pollution to the traffic volume, as a function of the traffic volume, plain symbols are for the mean and dashed for the r.m.s. values.

emission E_{NO_X} and the NO_x concentration is

$$\frac{dNO_x(t)}{dt} = -\frac{(NO_x(t) - NO_{x,bcg})}{\tau} - \frac{U_{wind,\parallel}}{L}(NO_x(t) - NO_{x,bcg}) + E_{NO_x}(t),$$

where $U_{Wind,\parallel}$ is wind speed parallel to the street axis, L the length of the street, and τ the characteristic time scale of the exchange at roof level. Here, it is assumed that the pollution which is brought into the street by the wind is equal to the background pollution. Under the steady state approximation (Palmgren, 1996), the above relation becomes

$$(NO_x(t) - NO_{x,bcg}) = \tau_{ech} E_{NO_x}(t)$$

with

$$\frac{1}{\tau_{ech}} = \frac{1}{\tau} + \frac{U_{Wind,\parallel}}{L}$$

and

$$E_{NO_x}(t) = e^{NO_x}(\mathrm{g\,km^{-1}})10^3 N_{trafic}(\mathrm{veh\,s^{-1}})/(HW),$$

τ_{ech} being the global characteristic exchange time. From Table 2, the mean e^{NO_x} value across all traffic conditions can be calculated as 0.975 g km^{-1}. The application of the previous relations to the data obtained during the campaign gives an average value τ_{ech} equal to 60 s. thus the magnitude of the exchange time is about 1 min in the case of 'rue Crevier'. We can see this time scale is much shorter than the characteristic time scale of the emission rate E_{NO_X} within the street (about 15 min at rush hours)

In reality, the τ time scale is a function of the meteorological conditions. In recent work, Soulhac and Perkins (1998) has shown that τ can be determined by the relation

$$\frac{1}{\tau} = \frac{\sigma_w}{\sqrt{2\Pi}} \times \frac{1}{H},$$

where σ_w is the variance of vertical wind speed at roof level. This σ_w parameter was not available in the present work. We have assumed it to be given by the relation $\sigma_w = a_1 + a_2\, U_{wind,\perp}$, where $U_{wind,\perp}$ is the wind speed component perpendicular to street direction, and a_1 and a_2 are constants. With this relationship, both mixing produced by the atmospheric turbulence and induced by the traffic inside the street are taken into account. The parameters a_1 and a_2 have been determined by a least square fitting minimisation of the model errors, performed on the NO_x data observed at the 'rue Crevier' station. The results are $a_1 = 0.27$ (m s^{-1}) and $a_2 = 0.16$. In order to assess the model reliability, we have calculated a number of performance indices, suggested in other works (Olesen, 1994). We find the fractional bias FB = 3.14E-02, the normalized mean square error NMSE = 0.167 and the correlation error COR = 0.870. It is not easy to interpret such statistical indices, but a comparison with other studies (Olesen, 1995; Kukkonen, 2000; Buckland, 1998) suggests a rather low discrepancies between the model predictions and the present observations. This is encouraging for the use of the box model approach. However, this attempt must not be considered as a validation of the box model since the performance indices were calculated on the same data as those used to determine the model parameters a_1 and a_2. Further studies are needed to validate the present box model approach.

5. Conclusions

NO_x measurements were carried out at different locations in the centre of a medium sized city during four weeks in winter. All wind directions were observed, the study period included several days of low wind speeds, when the meteorological conditions were not favourable for pollutant dispersion.

From these observations, it is concluded that:

- The spatial variability over short time periods, 15 min, is great and the pollutant mixing at roof level is not sufficient to homogenise the traffic emission throughout the city centre. However over longer periods (one month in this study), this variability is not observed and no large differences between the background concentrations in the city centre and those observed in the suburb 2.5 km away have been detected.
- In a street canyon, we have found that most NO pollution is due to traffic within the street, for NO_2, it is the opposite, the main contribution coming from outside the street.
- At low wind speed ($<1\,m\,s^{-1}$), the wind direction effect on pollution measured on the kerb is not always observed.
- The difference ($NO_x - NO_{x,bcg}$) represents the contribution from the street traffic emissions. This value, divided by the traffic volume, is seen to be independent of the traffic volume value itself. This argues in favour of using a simple approach, such as a box model, in order to calculate pollutant accumulation within the street canyon.

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