On the determination of the 99.8 percentile of NO_2 concentrations required by the directive 99/30/EC: A case study

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1 Introduction

The daughter directive 99/30/EC requires the determination of the 99.8 percentile of NO_2 concentrations. From the modellers point of view such high percentiles are more difficult to calculate because they depend on a very limited number of situations (e.g. specific meteorological conditions, unusual emissions, background concentrations), which are not predictable with the same accuracy as for instance annual mean values.
The main source of NO_2 is road traffic, which further complicates the matter, because of daily and seasonal variations (e.g. holiday and weekend traffic) of traffic flow. The motivation of this study is to test a simple methodology to compute the 99.8 percentile of NO_2 against field measurements near a motorway south of the city of Vienna (Austria).
The modelling approach consists of the Lagrangian dispersion model GRAL (=Graz Lagrangian Model), an empirical relationship for the NO conversion into NO_2, and another empirical relationship between the 98 percentile and 99.8 percentile of NO_2. Emphasise was laid that the whole methodology is applicable for regulatory purposes.

2 Data base

The Institute for Analytical Chemistry at the Technical University Vienna (IAC) has performed a measurement program of various chemical compounds in the vicinity of a major motorway south of Vienna from August 1998 to September 1999 (Kalina et al., 2000). A map of the position of the four monitoring stations (W1, E1, E2, and E3; sampling height was 4 m above ground level) relative to the motorway is given in Figure 1. Also shown is the mean wind direction distribution as observed during the measurement campaign. Winds from the sectors northwest and southeast were dominant. The mean wind speed during the observational period was 3.1 m s^{-1} 10 m above ground level at the monitoring station E3. The distances of the monitoring stations from the centre line of the motorway were 225 m (W1), 61 m (E1), 391 m (E2), and 900 m (E3).

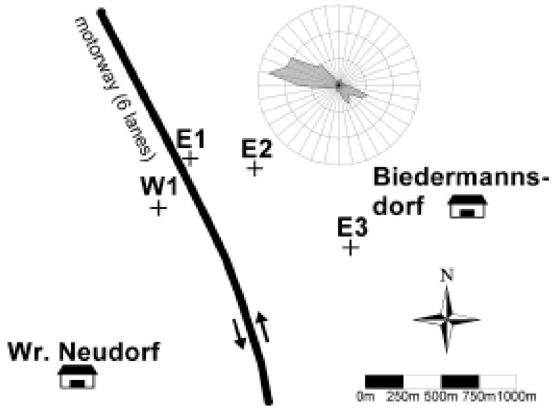

Figure 1 Map of the motorway and the position of the monitoring stations during the field experiment of the IAC, Vienna.

3 Modelling approach and results

The main difference of the Lagrangian particle model GRAL compared to other Lagrangian models is the treatment of the horizontal dispersion. In low wind conditions (regardless the stability), a negative lobe in the Lagrangian autocorrelation functions for the horizontal wind component fluctuations is assumed. This assumption is based on observed corresponding Eulerian autocorrelation functions, which do also show negative lobes in such conditions (Oettl et al., 2001a). To achieve such an autocorrelation function, a Markov Chain – Monte Carlo model as proposed by Wang and Stock (1992) is used for the horizontal components of the wind fluctuations. For the vertical dispersion the model of Franzese et al. (1999) is used.

The model has been tested against measurements taken by the Finish Meteorological Institute near a major road in Helsinki (Oettl et al., 2001b), and it was shown that it performs better in low wind conditions, and when the wind direction is close to the road orientation, compared to a Gaussian model.

The chemical transformation of NO into NO_2 was treated via an empirical relationship (Romberg et al., 1996).

$$NO_2 = NO_x \cdot \left\{ \frac{103}{NO_x + 130} + 0.005 \right\}, \text{ annual mean} \tag{1}$$

$$NO_2 = NO_x \cdot \left\{ \frac{111}{NO_x + 119} + 0.039 \right\}, \text{ 98 percentile} \tag{2}$$

The concentrations in eq. (1) – (2) need to be in units of microgram per cubic meter. Note, that neither information on the background concentrations of NO, NO_2, and O_3 nor information on the radiation is needed, which facilitates the application of eq. (1)-(2) in modelling studies for regulatory purposes. On the other hand, those quantities play an important role in the chemical transformation of NO into NO_2. Hence, eq. (1) – (2), which were derived from a numerical best fit to measured concentrations, may only give good results for regions, where similar conditions as in the field measurements, upon which eq. (1) – (2) are based, prevail.

Background concentrations were obtained as follows: For westerly winds W1, and for easterly winds and for winds parallel to the street E3 were taken as background stations. In this way a time-series for the background concentrations was obtained from which an annual mean and the 98 percentile were calculated. These read 38 $\mu g\ m^{-3}$ (annual mean NO_x) and 197 $\mu g\ m^{-3}$ (98 percentile NO_x). In environmental assessment studies background concentrations are rather poorly known in most cases, because existing monitoring stations (if there are any) are seldom placed in a way to allow for an approach as mentioned above, and hence leading to enormous uncertainties in the total concentration.

The total concentrations were obtained by adding the background concentrations to the computed concentrations. It is usual to take the annual background concentration and add it to the calculated annual mean, to have an estimate of the total impact on the air quality. This may lead to errors when applied also to the 98 percentile, since this would assume a perfect correlation between the background concentrations and the ones caused by the traffic, which is not likely to occur. On the other hand – especially the NO_x concentrations – will not be totally uncorrelated with the background concentration, since it is mainly caused by traffic in the surroundings. In the following, a low correlation between background concentrations and the concentrations generated by the traffic on the motorway is assumed and the simple addition theorem suggested by the German guideline TA-Luft (1999) is applied, although it was derived for elevated point sources only.

As it is not possible at the moment to compute a time-series of one year with the GRAL model on a PC, it is useful to establish some classification scheme for the various meteorological conditions. Here, we consider the wind speed and direction, and the stability class, which was derived from the observed wind speed and the cloud cover (Kalina et al. 2000). To limit the number of meteorological conditions in the simulations, only three stability classes were distinguished (stable, neutral, and unstable). Further, 36 classes of wind direction, and 7 classes of wind speed were used (0-1, 1-2, 2-3, 3-4, 4-6, 6-8, and >8 m s^{-1}), leading to 391 different meteorological situations weighted by their frequency.

Emission factors for NO_x were calculated according to the Austrian handbook of emission factors (Version 1.1A 1997). These depend on the vehicle speed, inclination of the street, year, the type of vehicle, and the type of street. The emission factors for light-duty vehicles and heavy-duty vehicles are 0.42 g km^{-1} and 7.5 g km^{-1} respectively. Uncertainty analysis of the emission factors provided by the Austrian handbook of emission factors showed an accuracy of about ±30 % (Sturm et al. 1998). Recent tunnel studies brought evidence, that the NO_x emission factors for light-duty vehicles are captured satisfactory, but the ones for heavy-duty vehicles are heavily underestimated. Such tunnel studies were performed in Germany, Switzerland, and Austria – naturally, we rely on the most recent one performed in Austria (Rodler 2000, Sturm et al. 2001), because the differences in the car fleet, year, and even vehicle speed compare best with the ones observed at the motorway near Vienna. Taking these results into account, the average emission factor per unit length (light-duty and heavy duty vehicles) for the year 1998 reads 7.53 kg km^{-1} h^{-1}. It is based on hourly traffic counts during the observational period resulting in a mean traffic flow of 113 400 vehicles a day (both directions), and a fraction of heavy-duty vehicles of 10.2% (Kalina et al. 2000). Although we make use of the corrected emission factor, there will still be some uncertainty, which in fact can not be assessed by now.
Lohmeyer et al. (2000) derived a linear relationship between the 98 percentile and 99.8 percentile of NO_2 from measurements. Figure 2 shows the same but for Austrian air quality stations only. There is a rather good correlation of 0.93. Here, we made use of this relationship to calculate the 99.8 percentile of NO_2 as demanded by the daughter directive 99/30/EC.

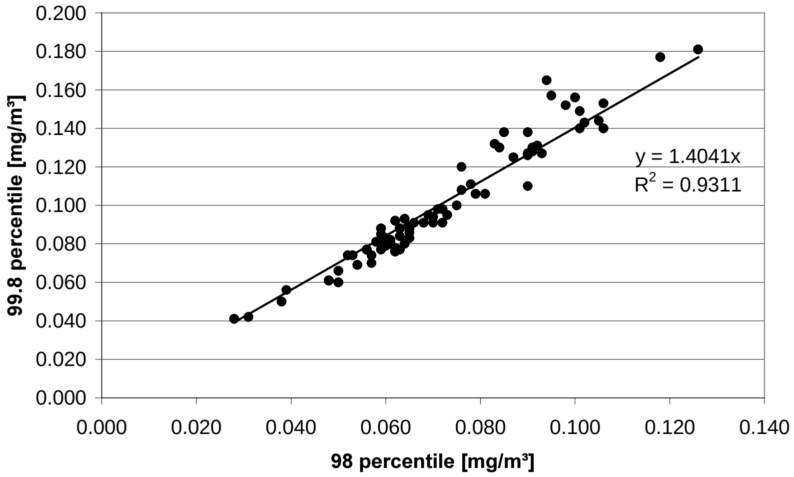

Figure 2 Relationship between the 98 percentile and the 99.8 percentile of NO_2 as observed by Austrian air quality stations.

The computed concentrations are listed in Table 1. There is a rather good agreement for all NO_x concentrations except at site E1, were the concentrations are overestimated by +36% (annual mean) and +51% (98 percentile). This may be an effect of the dam and the wall on both sides of the street, which reach a height of 6 m above ground level. The influence of the street geometry on the dispersion process can not be handled properly within Lagrangian dispersion models, since they may change the flow pattern in the vicinity of the road. Due to the non-linear relationship between the NO_x and NO_2 concentrations (eq. (1)-(2)), the error gets much smaller for the NO_2 concentrations. A very good agreement can be found at all sites except at site E3, where the annual mean is overestimated by +43%. Because, the NO_x annual mean is simulated with an accuracy of +20%, it can be concluded, that eq. (1) is not suitable at this site. Finally, the 99.8 percentile of NO_2 was simulated with the relationship shown in Figure 2. Again, a good agreement was found between measured and calculated concentrations, with the maximum deviation at site E1, where the concentrations were overestimated by +25%.

Table 1 Summary of modelled and observed NOx and NO_2 concentrations.

		W1 (225 m)	E1 (61 m)	E2 (391 m)	E3 (900 m)
Total annual mean NOx [µg m^{-3}]	Observed	63	97	67	50
	Predicted	61	132	75	60
Total 98 percentile NOx [µg m^{-3}]	Observed	327	374	342	288
	Predicted	340	564	331	284
Total annual mean NO_2 [µg m^{-3}]	Observed	32	42	32	23
	Predicted	33	53	38	33
Total 98 percentile NO_2 [µg m^{-3}]	Observed	88	92	96	76
	Predicted	95	114	95	89
Total 99.8 percentile NO_2 [µg m^{-3}]	Observed	115	128	141	109
	Predicted	133	160	133	125

4 Conclusions

The model results with the Lagrangian dispersion model GRAL for the spatial concentration distribution of the annual mean, the 98 percentile and the 99.8 percentile of NO_2 are found to be in good agreement with the observed values near a major highway south of Vienna, except at the closest site to the highway (distance = 61 m), where the model / method overestimated concentrations. In our opinion, this is a result of the complex geometry of the motorway rather than a failure of the model to simulate the dispersion process. The meteorological situations leading to high concentrations in the vicinity of the motorway are low wind speeds, combined with a stable stratification, and wind directions almost parallel to the street. In such conditions it might happen, that almost no vertical eddies develop at the upwind and downwind side of the noise barriers, resulting in a low vertical pollutant flux. Hence, the pollutants will remain in a mixed layer at street level, and are transported along the street. Whenever the street orientation changes to a certain extend, vertical eddies will appear more intensive and lead to relatively high concentrations near the street at those locations. However, such processes are difficult to handle in Gaussian as well as in Lagrangian dispersion models. The usual way to treat the effect of noise barriers in Gaussian and Lagrangian models is to increase the initial vertical plume spread. This approach would lead to errors in the spatial concentration distribution for the situations described above.

The usage of the NO-NO_2 conversion according to Romberg et al. (1996), and the linear relationship between the 98 and the 99.8 percentile of NO_2 according to Lohmeyer et al. (2000) were found to give good results in the frame of environmental assessment studies. It is remarkable, that using the annual daily mean traffic as input for the calculation of high percentiles led to reasonable results. This parameter is usually available with a relatively high accuracy. However, care has to be taken to generalise the good agreement here to other types of streets owing a different daily and annual variation of traffic.

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