

A new simplified NO/NO₂ conversion model under consideration of direct NO₂-emissions

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Abstract

Although many German monitoring sites report declines of NO_x concentrations, NO₂-concentrations actually stagnate or even increase quite often. Various analyses have identified the altered compositions of nitrogen oxides (NO₂/NO_x-ratio) emitted by motor vehicles (resulting in an increase of primary NO₂-emissions) as well as the chemical environmental conditions (mainly ground level ozone) as the main causes. The chemical conversion of NO to NO₂ is often parameterized in dispersion calculations of exhaust emissions. A widely applied conversion model is the so-called Romberg approach from 1996. However, the Romberg approach has to be re-evaluated to accommodate the above-mentioned conditions. This article presents an adjustment to the Romberg approach in accordance with the measured data from 2000 to 2006, taking into consideration substantially higher NO₂/NO_x-ratios especially for higher NO_x-concentrations. Model calculations with OSPM (Operational Street Pollution Model) including its internal chemistry module are able to reproduce very well the trends in the measured annual NO₂-concentrations over a 10 year period. The relevant parameters for variations between the years are the NO_x-emissions, primary NO₂-emissions, ozone concentrations, wind conditions, and background concentrations. A simplified chemistry model based on annual mean NO_x- and NO₂-concentrations, and background ozone concentrations, as well as primary NO₂-emissions is presented as a better method than the updated Romberg approach. This model simulates the annual mean NO₂-concentrations much more accurately than the conventional and the updated Romberg approaches.

Zusammenfassung

An vielen deutschen Messstationen wird zwar ein Rückgang der NO_x-Konzentrationen, aber häufig eine Stagnation der NO₂-Konzentrationen oder gar deren Anstieg beobachtet. Verschiedene Untersuchungen identifizierten als Hauptursachen eine geänderte Zusammensetzung der seitens der Kraftfahrzeuge emittierten Stickoxide (NO₂/NO_x-Verhältnis) und die daraus folgende Erhöhung der primären NO₂-Emissionen des Kfz-Verkehrs sowie die chemischen Umgebungsbedingungen (insbesondere bodennahes Ozon). Die chemische Umwandlung von NO in NO₂ wird innerhalb von Ausbreitungsrechnungen für Autoabgase oft parametrisiert. Ein häufig angewendetes Konversionsmodell stellt hierbei die so genannte Romberg-Formel aus dem Jahr 1996 dar. Unter den o. g. veränderten Randbedingungen steht die Romberg-Formel derzeit auf dem Prüfstand. Eine Anpassung der Romberg-Formel an Messdaten aus den Jahren 2000 bis 2006 wird aufgezeigt. Diese zeigt insbesondere bei hohen NO_x-Konzentrationen deutlich höhere NO/NO₂-Umwandlungsraten. Modellrechnungen mit OSPM inkl. nachgeschalteten Chemiemodell können die Trends der NO₂-Jahresmittelwerte über einen Zeitraum von 10 Jahren gut reproduzieren. Die relevanten Einflussgrößen auf die Variationen zwischen den Jahren sind die NO_x-Emissionen, die primären NO₂-Emissionen, die Ozonkonzentrationen, die Windverhältnisse und die Hintergrundbelastungen. Als möglichen Ersatz für die Romberg-Formel bzw. dessen Aktualisierung wird ein vereinfachtes Chemiemodell auf Basis der Jahresmittelwerte von NO_x-, NO₂- und Ozon-Hintergrundbelastung sowie primärer NO₂-Emission vorgestellt. Dieses kann die Trends in den NO₂-Jahresmittelwerten deutlich besser abbilden als der Romberg- bzw. der aktualisierte Rombergansatz.

1 Introduction

The formation of NO₂ from NO is a complex photochemical process. It is mainly depending on the total

amount of available NO_x and ozone. The ambient NO₂-concentration is also depending on the primarily emitted NO₂. Besides NO-emission, ozone concentrations as well as primary NO₂-emission are changing from year to year.

To date, the empirical conversion model of ROMBERG et al. (1996) is often used for dispersion calculations.

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It describes the conversion according to a $[NO_2] = f([NO_x])$ equation with

$$[NO_2] = \frac{A \cdot [NO_x]}{[NO_x] + B} + C \cdot [NO_x] \quad (1.1)$$

where A, B, and C are regression parameters. Two sets of parameters exist for both the annual average value as well as the 98-percentile value.

The results of this model are satisfactory, however, they cannot describe the tendency to stagnating or slightly increasing NO₂-concentrations, which has been observed by data from roadside monitoring stations over the last years (BÄCHLIN et al., 2006). This is due to changes in ozone values compared to before 1996, when the model was calibrated, as well as increased primary NO₂-emissions following increases in installations of after-treatment systems in automobile exhausts (Oxidation catalysts).

2 Updating the Romberg approach

ROMBERG et al.'s (1996) approach was recalibrated to current data within a research project for the regional environmental office of North Rhine-Westphalia. Based on the approach described above (Equation 1.1), the coefficients for A, B, and C were determined according to the new data sets. They were adjusted to capture the annual average values, the 98-percentile values, and the 19th-highest hourly values.

The data were evaluated for the years 2004, 2005, and 2006 in BÄCHLIN and BÖSINGER (2008) accordingly to ROMBERG et al. (1996) and graphically displayed. The regression functions were computed by using the least square method.

The constants for Equation (1.1) are listed in Tab. 1 for the given period of 2004 to 2006 along with the parameters from the conventional approach. The results are shown in Fig. 1. Next to the values for the conventional method the new regression results are plotted together with the observed data. The regression parameters are those given in Tab. 1. Using the new approach, higher NO₂/NO_x ratios are obtained for NO_x-values smaller than approx. 36 µg/m³. Marginally smaller NO₂/NO_x ratios are obtained for NO_x-values from approx. 36 µg/m³ to approx. 140 µg NO_x/m³. If the NO_x-value is greater than 140 µg/m³, the new approach yields noticeably higher NO₂/NO_x ratios. Several of the roadside monitoring sites are located in this range.

Fig. 2 shows the observed NO₂-concentrations depending on NO_x, as well as the two regression curves. It can be clearly seen that the approach by ROMBERG et al. (1996) results in too low NO₂-values for high NO_x-values. According to the current approach, a NO_x-value of approx. 81 µg NO_x/m³ yielded a NO₂-value around the limit of 40 µg/m³. Using to the new approach, slightly lower NO₂/NO_x ratios are predicted in this range. The annual average value of 40 µg NO₂/m³

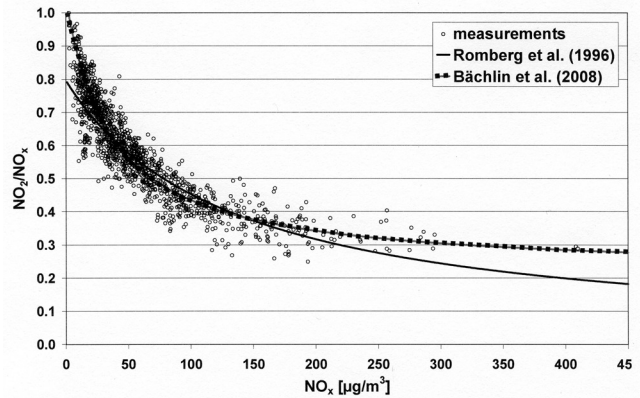


Figure 1: Ratios of NO₂/NO_x annual mean concentrations and all observed stations for the years 2004–2006. Source: BÄCHLIN and BÖSINGER (2008)

Table 1: Parameters for the regression of NO₂/NO_x ratios according to Equation (1.1) (Data are from BÄCHLIN and BÖSINGER, 2008). Units for NO₂ and NO_x concentrations are in µg (NO₂)/m³.

| Statistical concentration parameters | parameter of function | | |
|--------------------------------------|-----------------------|-----|-------|
| | A | B | C |
| Romberg et al. (1996) | | | |
| annual average | 103 | 130 | 0.005 |
| 98-percentile | 111 | 119 | 0.039 |
| Bächlin et al. (2008) | | | |
| annual average | 29 | 35 | 0.217 |
| 98-percentile | 40 | 20 | 0.170 |
| 19. highest hourly value | 43 | 10 | 0.151 |

would be reached by an annual mean NO_x-concentration of approx. 88 µg/m³ with the new approach. If annual average NO₂-values are calculated using these functions, the results are associated with a certain degree of uncertainty.

The measured data, the new approach, and the 95 %-interval for forecasts are given in Fig. 3. The 95 %-interval for forecasts is between approx. 65 and 112 µg NO_x/m³ for the limit of 40 µg NO₂/m³. This range shows the uncertainty connected with this approach. The uncertainty is estimated based on the scatter in the experimental data. Hence, for values greater than 112 µg NO_x/m³, it can be assumed with a certainty of 95 % that the limit is exceeded.

This conversion model is calibrated to the time range of 2004 to 2006 as described. Forecasts for which the relevant boundary conditions (i.e. ozone concentrations, radiation, primary NO₂-emissions) change with respect to the current situation are uncertain or their influences cannot be explicitly evaluated.

3 OSPM chemistry model

More realistic conversions can be obtained if a chemistry model is applied. A simplified relation is often used to describe the chemical conversion in the equilibrium

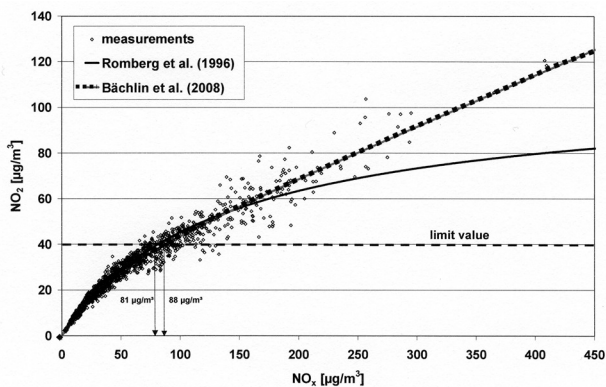


Figure 2: Measured annual mean NO₂-concentrations as a function of NO_x, and both regression curves. Source: BÄCHLIN and BÖSINGER (2008).

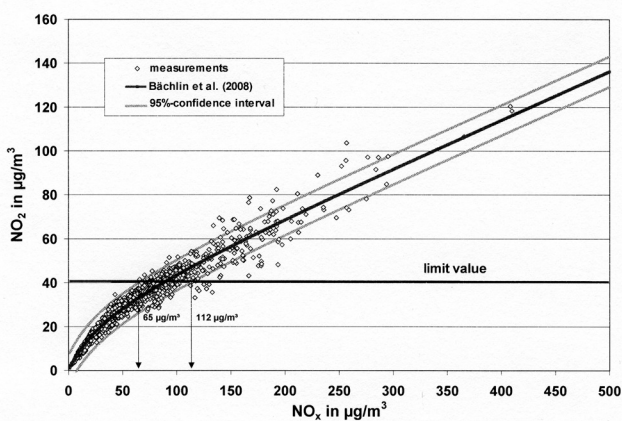
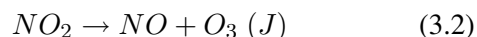
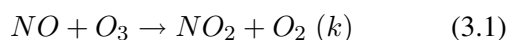


Figure 3: Measured annual mean NO₂-concentrations as a function of annual mean NO_x-concentrations, and the new regression curve. The 95 %-confidence interval is additionally highlighted. Source: BÄCHLIN and BÖSINGER (2008).

state of NO₂, NO, and ozone. The assumptions leading to the simplifications are:

- An equilibrium in the system NO₂-NO-O₃ is quickly obtained
- The reaction with volatile organic compounds (VOC) is not considered
- Mixing processes and sinks (advection, turbulent diffusion, influence of boundary conditions, deposition) are parameterized via the mixing time
- The contribution of traffic emissions is estimated by the difference of the measured NO_x-concentration at two air quality monitoring sites or calculated via a dispersion model
- The influence of the wind direction is not considered.

The considered reactions are:



J (s⁻¹) is the photolytic frequency of NO₂, k (ppb⁻¹ s⁻¹) is the reaction coefficient.

In the atmosphere, equilibrium between the three reacting gases (NO, NO₂, O₃) is quickly obtained. The photochemical equilibrium describes the condition, in which all three time derivatives (left sides of Equations 3.3, 3.4, 3.5) identically yield zero. This means that production and loss of each gas are equal and the sum is zero. When these reactions are considered independently, the differential equations describing the decomposition of NO₂, NO, and O₃ are defined as:

$$\frac{d[NO_2]}{dt} = k \cdot [NO] \cdot [O_3] - J \cdot [NO_2] + \frac{[NO_2]_V}{\tau} + \frac{[NO_2]_B - [NO_2]}{\tau} \quad (3.3)$$

$$\frac{d[NO]}{dt} = -k \cdot [NO] \cdot [O_3] + J \cdot [NO_2] + \frac{[NO]_V}{\tau} + \frac{[NO]_B - [NO]}{\tau} \quad (3.4)$$

$$\frac{d[O_3]}{dt} = -k \cdot [NO] \cdot [O_3] + J \cdot [NO_2] + \frac{[O_3]_B - [O_3]}{\tau} \quad (3.5)$$

The first two terms on the right side describe the chemical transformation by thermal and photochemical reactions. The next term in Equations 3.3 and 3.4 describes the effect of emissions (index V , no direct emissions for ozone). The last term describes the mixing as a function of concentration differences between the background station (index B) and the point, at which the concentration should be calculated, in this case the traffic station: if the concentration is higher at the traffic station than at the background station, this term is negative and the concentration decreases by the addition of air with a lower concentration in the assumed volume. The mixing is governed by the time constant τ , the residence time. The two terms $[NO]_V$ and $[NO_2]_V$ are the amount being produced by traffic emissions as an increase of concentration above the background concentration. $[NO_x]_V$ and $[NO]_V$ are calculated from the difference in NO_x between the traffic station and the background station as

$$[NO_2]_V = p \cdot ([NO_x] - [NO_x]_B) \quad (3.6)$$

$$[NO]_V = ([NO] - [NO]_B) \quad (3.7)$$

with $p = NO_2/NO_x$ being the fraction of NO₂ in the direct traffic emissions (will be discussed below) and

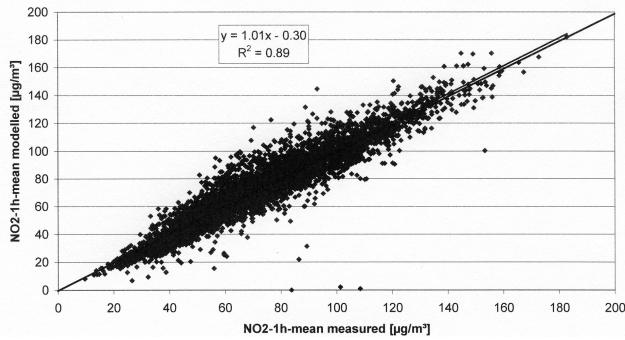


Figure 4: Comparison of hourly mean NO₂-concentrations, calculated with the OSPM chemistry model from observed NO_x data, and values from the measurement site Corneliusstraße in Duesseldorf for the year 2006.

[NO_x] as the concentration, which is estimated at the traffic station by measurements or a dispersion model. The unknown variables are [NO], [NO₂], [O₃]. Assuming that the equilibrium is quickly reached, the three differential equations (Equation 3.3 through 3.5) turn into three algebraic conditional equations. Solving them yields the analytical equation for the concentration:

$$[NO_2] = 0.5 \left(B - \sqrt{B^2 - 4 \left([NO_x][NO_2]_O + [NO_2]_n/k\tau \right)} \right) \quad (3.8)$$

With the variables

$$\begin{aligned} [NO_2]_n &= [NO_2]_V + [NO_2]_B \\ [NO_2]_O &= [NO_2]_n + [O_3]_B \\ B &= [NO_x] + [NO_2]_O + \frac{1}{k} \left(J + \frac{1}{\tau} \right) \end{aligned}$$

This equation is used in the dispersion model OSPM to calculate the NO₂ conversion with an hourly resolution (PALMGREN et al., 2007; BERKOWICZ, 2000). The parameter is calculated from meteorological values (e.g. wind speed and turbulence) as well as the road geometry (height of the street canyon) and represents the typical residence time the pollution is trapped inside the street canyon and is available for chemical reactions (see BERKOWICZ et al., 1997). Typical values for are in the range of 80s to 150s.

The average hourly NO₂-values calculated via the OSPM chemistry model (considering hourly data of primary NO₂ emissions, ozone concentrations, global radiation, temperature, and wind speed) match the observed data in 2006 from the station Corneliusstraße in Düsseldorf (see Fig. 4) very well. Comparable results have been obtained at other stations and for other years. Therefore, the OSPM chemistry model is well suited to describe the formation of NO₂, while considering the

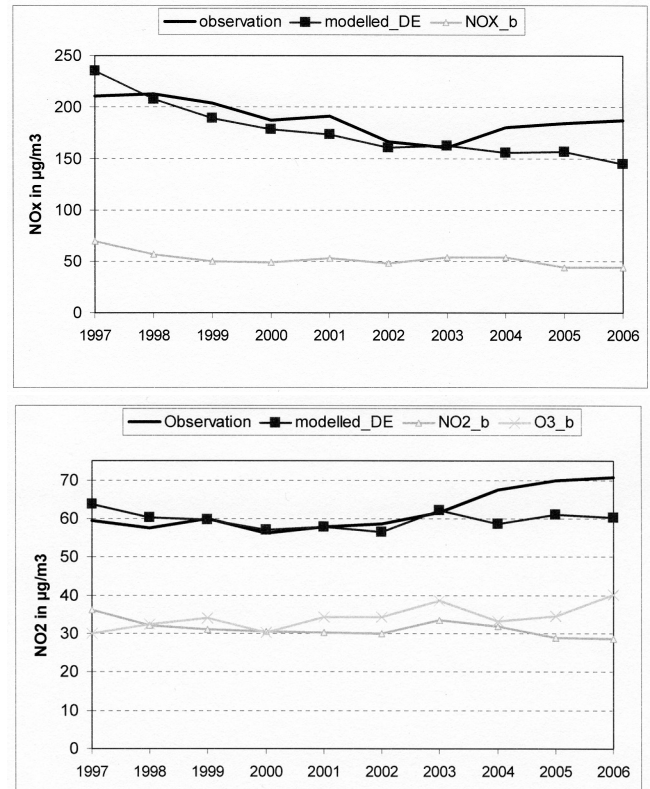


Figure 5: Top: Comparison of annual mean NO_x-concentrations, calculated with OSPM, with observed data from the measurement site Corneliusstraße in Duesseldorf for the years 1997–2006. NO_{x_b} = NO_x background concentration. Bottom: Comparison of annual mean NO₂-concentrations, calculated with OSPM, with observed data from the measurement site Corneliusstraße in Duesseldorf for the years 1997–2006. NO_{2_b} = NO₂ background concentration, O_{3_b} = ozone background concentration

relevant influences in the NO-NO₂-O₃ cycle, including the primary NO₂-emissions.

Calculations were also run with OSPM for the station Corneliusstraße in Düsseldorf (DÜRING et al., 2008) for the time range of 1997 to 2006. The results of the corresponding annual mean NO_x and NO₂-concentrations are given in Fig. 5. It can be noted that the OSPM model can reproduce these values very well for the time range of 1997 to 2003. Even though NO_x emissions decreased (not depicted here) due to the modernization of the motor fleet and, as a consequence, annual mean street canyon NO_x-concentrations dropped by approx. 25 % between 1998 and 2003, the annual mean street canyon NO₂-concentrations stagnates around a value of 60 µg/m³. The model calculations show that the decreasing NO_x emissions were compensated by an increase in ozone concentrations and primary NO₂ emissions. The increase in the NO₂/NO_x ratio in the traffic emissions is shown as an example in Fig. 6.

The increase in street canyon annual average NO_x-values after 2003 observed in the data cannot be reproduced by emission calculations based on HBEFA

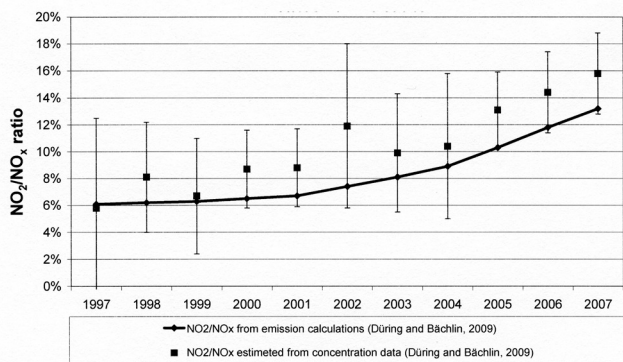


Figure 6: Comparison of annual mean NO_x-concentrations, calculated with OSPM, with observed data from the measurement site Corneliusstraße in Duesseldorf for the years 1997–2006. NO_{x-b} = NO_x background concentration.

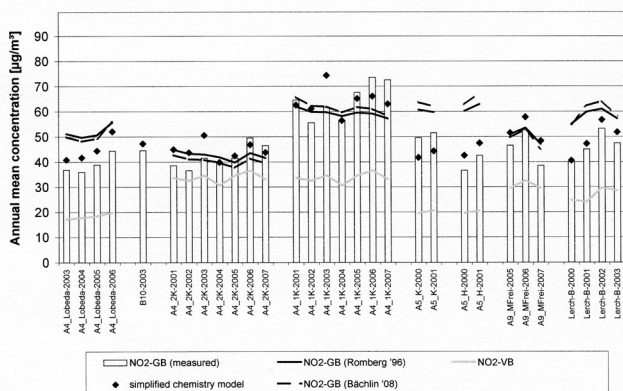


Figure 7: Annual mean NO₂-concentration from federal highway measurement sites, including values estimated from different conversion models. VB = background concentration; GB = total concentration. Bächlin '08 = BÄCHLIN and BÖSINGER (2008).

2.1 (Manual of Emission Factors, see: www.hbefa.net). Various sensitive calculations also show that the remarkable increase in street canyon annual mean NO₂-concentrations after 2003 (7 µg/m³ to 10 µg/m³ increase in comparison to 2003) can not be explained by an increase of the NO₂/NO_x ratio emissions, than by an increase in the total NO_x emissions (keeping the NO₂/NO_x ratio constant). The reason for this discrepancy could possibly be found in inaccuracies during the emission determination (percentage of diesel-cars, amount of traffic, etc.).

4 Simplified chemistry model based on annual average values

Strictly speaking, the above equations of the chemistry model can only be used in time series calculations, because the parameters J and k are dependent on meteorological parameters. Based on research projects by BASt (Bundesanstalt für Straßenwesen) (DÜRING et al., 2009)

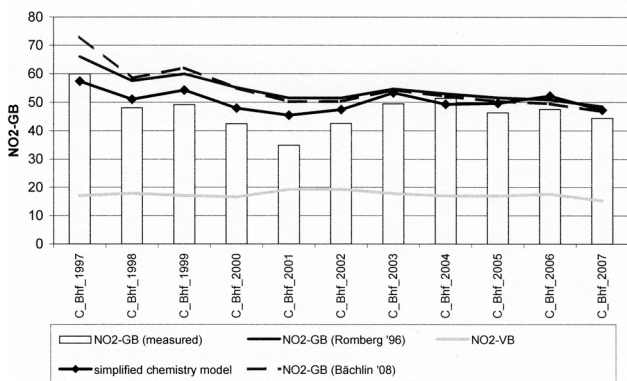
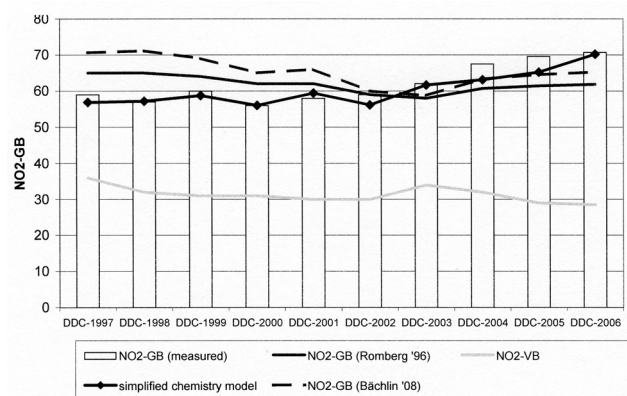


Figure 8: Top: Annual mean NO₂-concentration in µg/m³ from the measurement site Corneliusstrasse (Duesseldorf), including values estimated from different conversion models. Bottom: Annual mean NO₂-concentration in µg/m³ from the measurement site Bahnhofstraße (Cottbus), including values estimated from different conversion models. VB = background concentration; GB = total concentration. Bächlin '08 = BÄCHLIN and BÖSINGER (2008).

as well as the Landesumweltamt Brandenburg (DÜRING and BÄCHLIN, 2009), these equations can also be applied for annual mean concentrations using the following parameters:

$$J = 0.0045 \text{ s}^{-1}$$

$$k = 0.00039 \text{ (ppb s)}^{-1}$$

$$\tau = 100 \text{ s (street canyons) or } 40 \text{ s (free dispersion)}$$

The input data are the same as for the Romberg approach:

NO_x – annual mean at traffic station (in the model=calculated value)

NO_x – annual mean at background station

NO₂ – annual mean at background station

as well as two additional values:

Ozone – annual mean at background station

p = share of primary NO₂ emissions for all NO_x emissions.

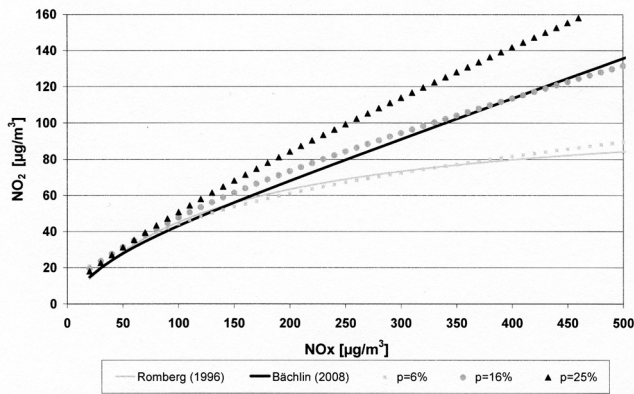


Figure 9: Dependence of annual mean NO₂- on annual mean NO_x- concentration, estimated with different models. Description see text. Bächlin (2008) = BÄCHLIN and BÖSINGER (2008).

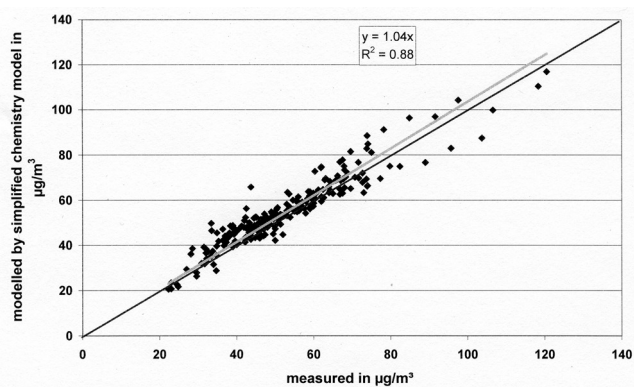


Figure 10: Comparison of results for annual mean NO₂- concentration with the simplified chemistry model and measurement data.

It has not been possible, to date, to compute the primary NO₂ emissions with the current HBEFA 2.1. Therefore typical values listed in Tab. 2 (from literature and own research data) were assigned equally to all stations. As soon as the new manual of emission factors is available, these shares can be calculated, specifically for every individual road.

The conversion calculations were performed with these values for observation data along rural and city roads. The NO_x concentration, the background NO₂ concentration, and the ozone concentration were set as annual average values for each individual traffic station and the corresponding representative background station. The results are given in Fig. 7 for monitoring sites along federal highways. Fig. 8 shows the data from two city roads as an example.

Table 2: Shares of primary NO₂ emissions for overall NO_x emissions.

| Year | Share of NO ₂ for all NO _x emissionens [%] |
|------|--|
| 2000 | 8 |
| 2001 | 9 |
| 2002 | 11 |
| 2003 | 12 |
| 2004 | 14 |
| 2005 | 15 |
| 2006 | 17 |
| 2007 | 18 |

5 Conclusions

The following can be observed:

- The approach by ROMBERG et al. (1996) is often conservative for existing data up to the year 2003; however, it cannot reproduce the long term trends (e.g. A4_1k and A4_2k in Fig. 7).
- The approach by BÄCHLIN and BÖSINGER (2008) reproduces the existing measured data better than the approach by Romberg (Fig. 1 and Fig. 2), but it cannot reproduce the long term tendencies very well, either (Fig. 7 and Fig. 8).
- The simplified chemistry model that was introduced here can best reproduce the tendencies and the absolute values.

The reason for the good fit for trends in the NO₂/NO_x ratios is mainly due to the explicit incorporation of the share of primary NO₂ within all NO_x emissions. Fig. 9 shows the annual mean NO₂-concentration depending on the annual mean NO_x-concentration for the simplified chemistry model at a share of 6 %, 16 % and 25 % primary NO₂, as well as the parameterization by ROMBERG et al. (1996) and BÄCHLIN and BÖSINGER (2008), for comparison. It can be seen that the Romberg-parameterization is set at p = 6 %. This was to be expected because the calibration was performed before 1996 and p was about 5 % to 10 % at that time. The parameterization by BÄCHLIN and BÖSINGER (2008) can be reproduced with a p of 16 %. The calibration for this model was done on data from 2003 to 2006, and the average NO₂ emission share then was between 12 % and 17 % (Tab. 2).

A further increase of NO₂ shares is to be expected over the next years (e.g. IFEU, 2007) leading to a further increase of the NO₂/NO_x ratios. This also means: the higher the proportion of primary NO₂ emission becomes, the more NO_x emissions have to be reduced in order to comply with the limit value of 40 µg/m³.

Fig. 10 shows the comparison between annual mean NO₂-values concentrations, calculated via the simplified chemical model, and measurements from more than 30

German monitoring sites (highways and city roads) between 1997 and 2006. It can be noted that this approach reproduces the observed data very well ($r^2 = 0.88$, the slope of the curve is approx. 1). Therefore, this model is better suited to calculate the NO-NO₂ conversion than the parameterization by ROMBERG et al. (1996) and BÄCHLIN and BÖSINGER (2008), by explicitly including primary NO₂ emissions and ozone concentrations.

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