HIGH NO_2 CONCENTRATIONS IN URBAN STREETS - CAUSES AND IMPACT OF MEASURES

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ABSTRACT

The ambient air quality limit value for NO_2 for the year 2010 is currently exceeded in many busy streets in Europe. Despite the reductions of NO_x emissions, the concentrations of NO_2 have hardly decreased in the past years; increases have been observed at some traffic sites. Causes for the development of NO_2 concentrations in streets in Germany are primarily the increase of primary NO_2 emissions from modern diesel passenger cars and in addition their increased mileage share. NO_2 formation by atmospheric chemical reactions of emitted NO still has a considerable share on NO_2 concentrations in streets, though.

Results of new projects in Germany give a clearer picture of the reasons for high NO_2 concentrations in streets and the effectiveness of reduction measures. The contribution of primary NO_2 emissions and atmospheric chemical NO_2 formation to NO_2 air pollution was analysed with several methods for a busy major street in the city of Stuttgart. The impact of more stringent exhaust emission limits on future air quality was calculated by using a chemical box model. At the analysed location, the future NO_2 limit value can only be met with a considerable reduction of primary NO_2 exhaust emissions from vehicles in addition with reduction of atmospheric chemical NO_2 formation in the street and in the urban background.

Keywords: *NO_2, diesel cars, primary NO_2, atmospheric chemistry, ozone, modelling.*

1. INTRODUCTION

1.1. Development of NO_x emissions, NO_x concentrations and NO_2 concentrations

In recent years, nitrogen oxide emissions (NO_x = NO + NO_2) from vehicles in Germany have decreased considerably. This has also led to a decrease in the nitrogen oxide concentrations measured at many traffic sites in Germany (Figure 1).

The European ambient air quality legislation for the protection of human health, however, does not limit the sum of nitrogen oxides (NO_x), but the nitrogen dioxide (NO_2) concentration. According to EU directive 1999/30/EG, the annual mean limit value of 40 µg/m³ has to be complied with from 2010. Under certain conditions, it will also be possible to request for an extension of the deadline for compliance with the limit values "until" 2015 [BMU 2008].

The decrease in nitrogen oxide emissions and concentrations is not yet reflected in the nitrogen dioxide concentrations (NO_2). These have only slightly decreased in recent years and at some sites, the concentrations have even increased (Figure 1 right; see also [Rabl 2005]).

The annual mean NO_2 limit value is therefore currently exceeded at many traffic sites. In order to achieve a compliance by 2010, efficient measures for a reduction of the pollution have to be developed and implemented. This requires a detailed knowledge of the reasons underlying the unexpected NO_2 development in recent years.

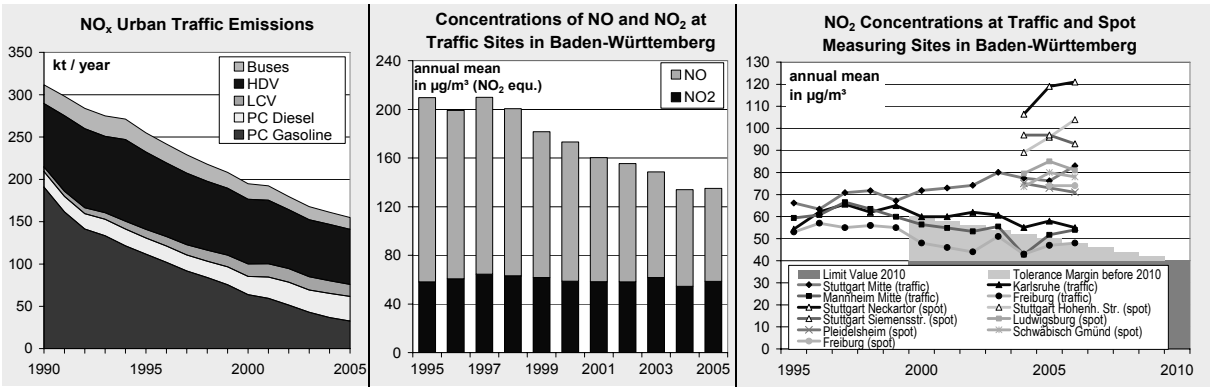

Figure 1: Urban NO_x emissions, NO_x concentrations, NO_2 concentrations

1.2. Causes of the NO_2 concentrations in highly polluted streets

The main cause for high NO_2 concentrations measured in urban streets are the NO_x emissions of traffic. On the one hand, emissions of the entire urban and also regional traffic contribute to a general concentration level, the "urban background". On the other hand, local traffic emissions further contribute to the concentration, especially in streets with heavy traffic ("additional local pollution").

Different studies (e.g. Carslaw 2005], [IFEU 2006]) have shown, that the additional local NO_2 pollution has two main causes:

1. NO_2 is directly emitted by vehicles in the street ("primary NO_2"). In the last years, directly emitted NO_2 from diesel vehicles has increased due to modern exhaust aftertreatment technologies.
2. NO_2 is formed out of locally emitted NO by atmospheric chemical reactions, mainly with ozone.

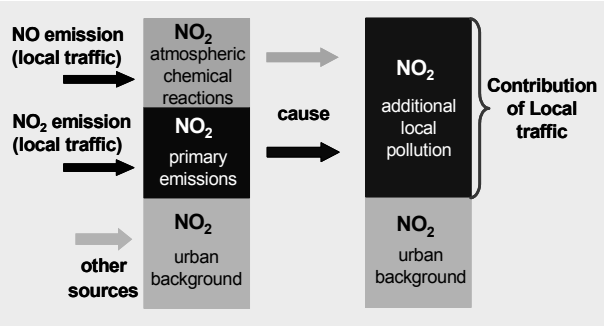

Figure 2: Causes of NO_2 concentrations in urban streets (schematic)

The share of the different sources on the total NO_2 concentration varies, depending on the traffic situation (e.g. traffic volume, share of diesel vehicles); local ventilation as well as meteorological parameters (e.g. conditions for diffusion, available ozone). A detailed analysis of the share of different sources is therefore an important basis for the identification of efficient measures to reduce NO_2 concentrations in the future. Such an analysis has been undertaken for the highly polluted traffic measurement site Stuttgart-Mitte.

In order to determine the share of different sources on the NO_2 concentration, the following methodological approaches have been applied:

- Statistical analyses of air quality measurement data, incl. ozone concentrations, at the traffic site,
- Chemical box modelling, considering NO and NO_2 emissions from local traffic, background concentrations and a set of atmospheric chemical reactions.

An important basis for this analysis has been the derivation of emission factors for primary NO_2 in urban traffic situations from comprehensive exhaust emission measurements. These emission factors together with data on the local traffic composition have been used to calculate the NO_x and NO_2 emissions at the measurement site as an input parameter for the air quality modelling.

2. ESTIMATE OF THE CONTRIBUTION OF DIFFERENT SOURCES TO NO_2 CONCENTRATIONS BASED ON AIR QUALITY MEASUREMENT DATA

This estimation of the contribution of different sources to the NO_2 concentration at Stuttgart-Mitte traffic site is based on the measured ozone concentration at the site, making the assumption, that the difference in ozone concentration between the urban background and the traffic site is due to the atmospheric chemical reaction of ozone with locally emitted NO ($NO + O_3 \Leftrightarrow NO_2 + O_2$). The difference in the ozone concentration in the street and in the urban background thus allows for an estimate of the rate of ozone depletion and the amount of NO_2 formed during this process. The remaining additional local NO_2 pollution is in small part due to other atmospheric chemical reactions (e.g. with radicals), but mainly due to primary NO_2 from direct exhaust emissions.

Ozone concentrations have been measured by LUBW[1] at Stuttgart-Mitte for this research project since August 2006 and have been analysed for the period of one year. During the entire analysed period, the ozone concentration at the traffic measurement site has been lower compared to the urban background concentration (Figure 2, left). At the traffic site, the ozone concentration level has been about 30-40% lower in summer and 50-60% lower in winter (with clearly reduced ozone availability) (Figure 2, right).

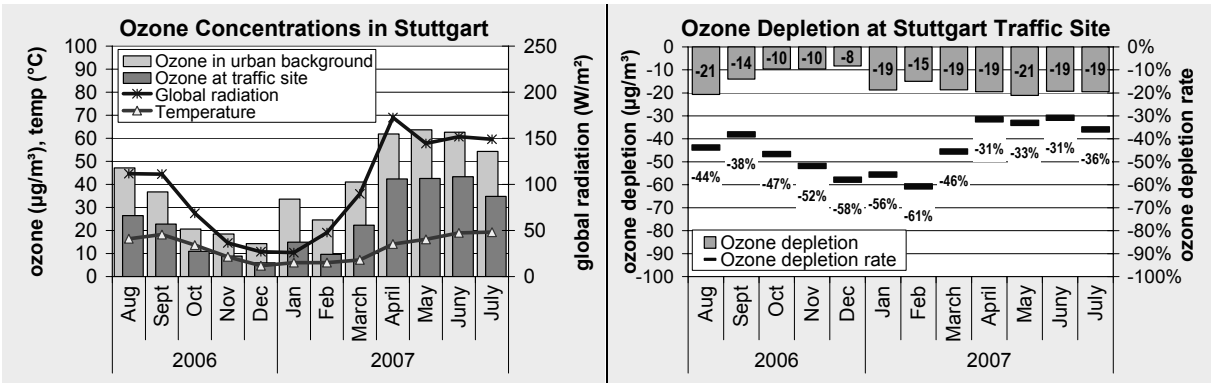

Figure 3: Ozone concentrations in Stuttgart (left), ozone depletion at Stuttgart traffic site (right) on monthly average

Figure 4 shows an exemplary daily development of the different contributions in summer. The urban background (overall city traffic) as well as primary NO_2 increases with the morning traffic. Ozone depletion is not yet relevant, since ozone availability is low at this time of day. In the course of the day, the urban background decreases despite traffic emissions remaining on a high level, because the conditions for diffusion improve (dissolution of the nightly inversion in the Stuttgart valley). Simultaneously, the contribution of NO_2 from ozone depletion increases due to the increase in ozone availability (inflow from the regional background or higher atmospheric layers; inner city ozone formation (in the background) by solar radiation).

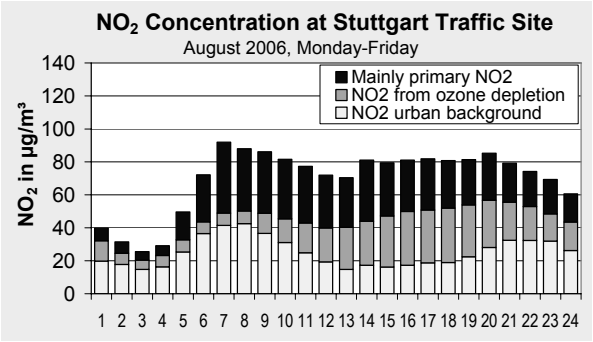

Figure 4: Composition of NO_2 Concentrations in diurnal variation (exemplary)

[1] Landesanstalt für Umwelt, Messungen und Naturschutz Baden-Württemberg

Over the entire analysed period of one year, the total NO_2 concentration has been about 15-30% higher in winter compared to the warmer season (Figure 5). In the cold season, the urban background with a share of 40-52% made the highest contribution to the NO_2 concentration. Ozone depletion at the site was responsible for up to 30% of the NO_2 concentration in summer and partly less than 10% in winter. The remaining additional local NO_2 pollution, which is mainly due to primary NO_2, had a share between 24 and 45%.

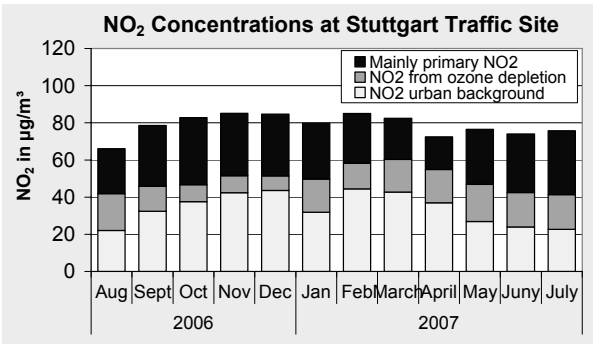

Figure 5: Composition of NO_2 Concentrations on monthly average

3. PRIMARY NO_2 EMISSIONS IN URBAN ROAD TRAFFIC

3.1. Shares of primary NO_2 in NO_x exhaust emissions of urban road traffic

The emission legislation for road vehicles defines a limit value for the sum of nitrogen oxides (NO_x). Emission measurements therefore mainly determine total NO_x emissions and only few data on NO_2 emissions are thus available. Only in recent years, specific measurements of NO_2 emissions have been undertaken by European laboratories (e.g. [TNO 2007], [EMPA 2007], [Millbrook 2006]).

An analysis of such recent measurements by IFEU shows that NO_x emissions of gasoline cars only have a small share of primary NO_2 and therefore mainly consist of NO. For diesel cars, the share of NO_2 on NO_x emissions is already higher than assumed in many diffusion models (5%) for old cars and has increased even more especially from Euro 3 (Figure 6). This increase in the share of NO_2 on total NO_x emissions is among others attributed to the introduction of oxidation catalytic converters. These are used in diesel vehicles in order to reduce hydrocarbon and carbon monoxide emissions, but as a side effect also oxidise NO to NO_2. The highest NO_2/NO_x ratio has been measured for Euro-4 diesel cars with particulate filters.

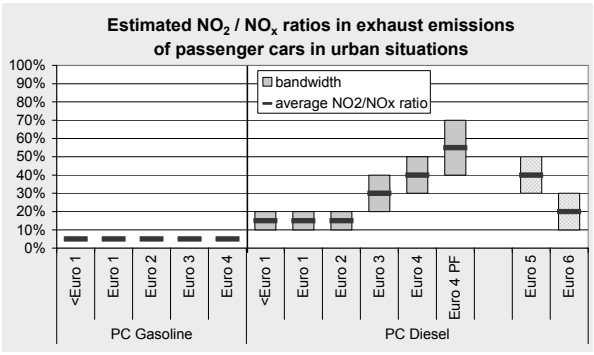

Figure 6: NO_2 share in urban NO_x emissions of passenger cars

Only few measurements are available for future emission reduction technologies for passenger cars. The available measurements, however, indicate that the use of different coating materials in oxidation catalytic converters and particulate filters as well as the use of SCR systems ("selective catalytic reduction" to reduce nitrogen oxide emissions) will lead to a decrease in the NO_2/NO_x ratio in direct exhaust emissions in the future.

Trucks and buses without exhaust treatment technologies show low NO_2/NO_x ratios. Therefore, with decreasing NO_x emissions due to more strict emission limits, also NO_2 emissions have decreased. However, for buses with particulate filter systems, specific NO_2 emissions can increase by a factor 3-6 with some systems. Such systems actively generate NO_2 in order to regenerate the filter.

The derived NO_2/NO_x emission ratios have been linked to the NO_x emission factors of the emission model TREMOD [IFEU 2006] in order to derive NO_2 emission factors (Figure 7).

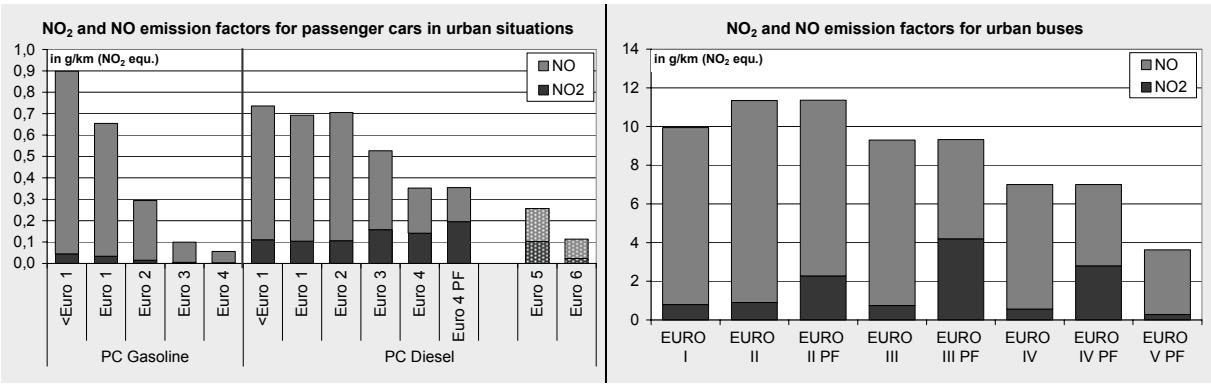

Figure 7: Urban NO_x and NO_2 emission factors of passenger cars and buses

3.2. NO_x and NO_2 Emissions of Local Traffic at Stuttgart-Mitte

A detailed modelling of local NO_x and NO_2 emissions has been undertaken. The results can afterwards be used to calculate the NO_2 concentration at „Stuttgart-Mitte“ with a chemical box model. A special characteristic of the measurement station is its position close to a highly frequented bus stop and the accordingly high share of buses on the local traffic. The local emission calculation considers the following parameters:

- Intraday traffic volume and composition based on traffic counting data,
- Diesel share on the passenger car fleet of Baden-Württemberg,
- Number of buses per day,
- Technical equipment of buses (Euro stage, CRT®-System).

In 2005, gasoline and diesel passenger cars had a share of just over 20% on the local NO_x emissions, trucks and buses each contributed about 25% to the NO_x emissions. NO_2 emissions, in contrast, have been dominated by diesel cars which contributed 40% and buses with 35%. The high contribution of buses to the NO_2 emissions is due to the high share of older Euro II/III buses with retrofitted particulate filter systems. Gasoline cars and trucks made only a small contribution to the primary NO_2 emissions due to their low share of NO_2 in the exhaust emissions.

Between 2005 and 2020, NO_x emissions of the local traffic at Stuttgart-Mitte are expected to decrease considerably. The NO_2 emissions in 2010, however, have been calculated to be higher as in 2005. Not until 2015 will the NO_2 emissions again reach the level of 2005 due to emission reductions of diesel cars with the introduction of Euro5/6.

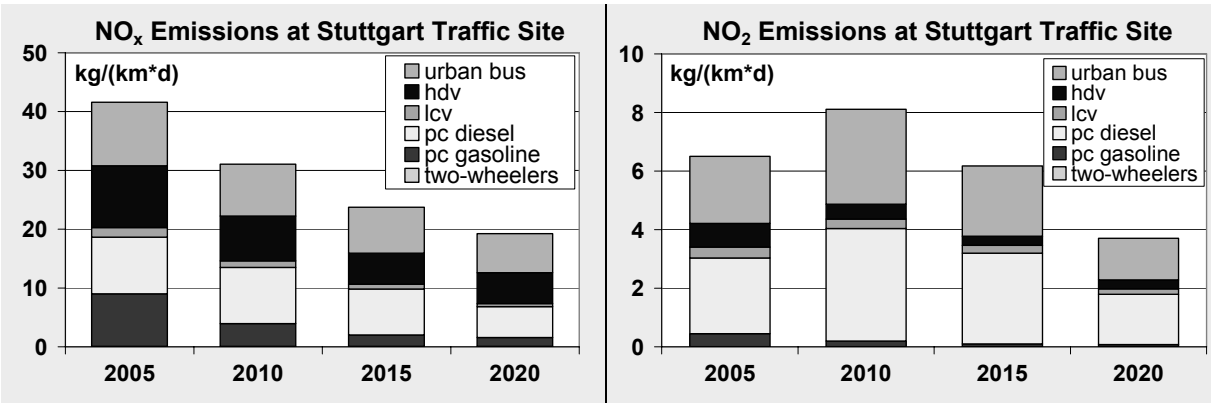

Figure 8: NO_x and NO_2 emissions 2005-2020 at Stuttgart traffic site

4. MODELLING OF NO_2 CONCENTRATIONS WITH A CHEMICAL BOX MODEL

4.1. AVISO's Chemical Box Model

A detailed analysis of the NO_2 concentration at the measurement site Stuttgart-Mitte has been undertaken with a chemical box model developed by AVISO (see also companion paper [Niederau 2008] "Model based source estimation of roadside concentration of NO_2 at Stuttgart, Germany"). This box model calculates the atmospheric diffusion and the chemical conversion of inflow substances (traffic emissions, background concentration) under the influence of solar radiation and wind speed. Besides the NO_x emissions of traffic also emissions of primary NO_2 in the exhaust of vehicles are taken into account.

On the inflow side of the box, the intraday NO_2, NO and O_3 background concentration as well as wind speed is given (Figure 9, left). The model uses values measured in the urban background. At the bottom side of the box, the hourly NO_x and NO_2 emissions of traffic are fed into the model. The upper side of the box is permeable for solar radiation, but not for substances. Inside the box, the atmospheric chemical conversion of 59 substances is described (reaction model RADM2 [RADM2]). As a result of the modelling, the hourly development of the NO, NO_2 and ozone concentration at the outflow side of the model is calculated.

The box model allows not only for a variation of different parameters, but also to switch on or off certain parameters or processes. Thus, concentrations can be modelled under the assumption that no atmospheric chemical conversion is taking place. Furthermore it can be modelled how much lower the NO_2 concentration would be without any emissions of primary NO_2, i.e. if all NO_x emissions would be NO. The box model thus allows for an analysis of the influence of different causes to the total NO_2 concentration as well as the impact of different measures.

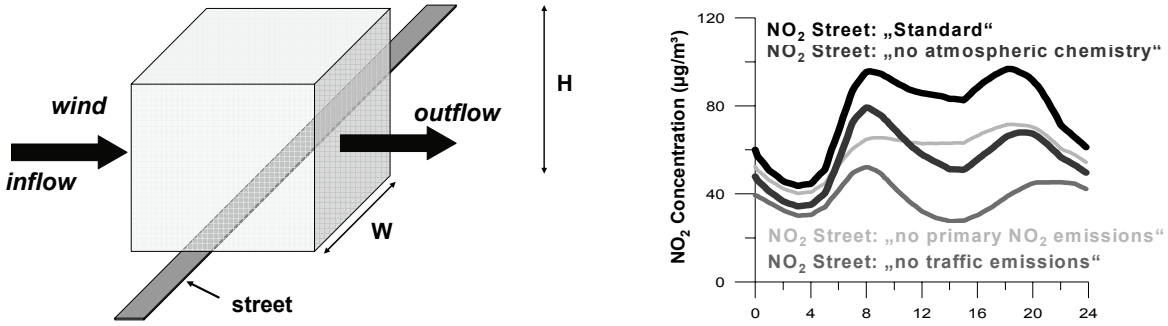

Figure 9: Schematic presentation of the chemical box model (left), typical intraday development of modelled NO_2 concentrations with variations of certain processes (right)

4.2. Results of chemical box modelling for 2005

a) Composition of the NO_2 concentration (causes, contribution of vehicle categories)

For 2005, the chemical box model has calculated a total NO_2 concentration of 73 µg/m³ for Stuttgart-Mitte. 34 µg/m³ can be attributed to the background concentration.

Assuming an inert diffusion of primary NO_2 from the exhaust, a contribution of 19 µg/m³ from primary NO_2 has been calculated (Figure 10, left). The actual contribution of primary NO_2 will be slightly lower, due to a small reduction of primary NO_2 by photolysis and other processes. The contribution of primary NO_2 to total NO_2 is therefore about as high as the contribution of the atmospheric chemical conversion of locally emitted NO (Figure 10, left).

In order to answer the question of how the NO_2 concentration would develop if traffic exhaust emissions of NO_x would consist entirely of NO (NO_x emissions = 100% NO), an additional scenario without primary NO_2 emissions has been calculated with the box model. In this

scenario the contribution of primary NO_2 is eliminated, but at the same time the NO_2 contribution from atmospheric chemical reactions increases, since more NO is emitted in comparison with the standard scenario.

Without primary NO_2 emissions the modelled annual mean NO_2 concentration in 2005 would have been about 57 µg/m³, thus 16 µg/m³ lower (see white box of avoidable NO_2 in Figure 10, right). This reduction in the NO_2 concentration is lower than the contribution of primary NO_2 of 19 µg/m³ (assuming an inert diffusion) in the standard scenario due to an increase of NO_2 formation by atmospheric chemical processes to 23 µg/m³.

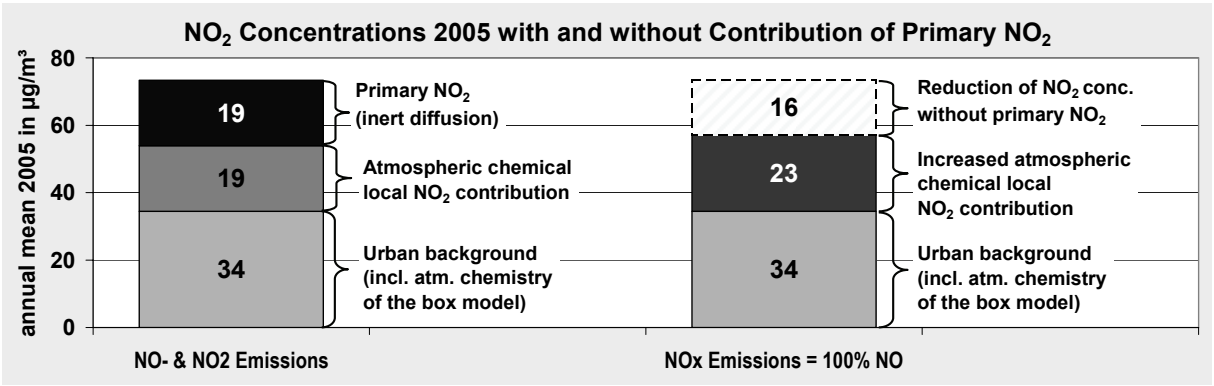

Figure 10: Sources of the total NO_2 concentration with the contribution of primary NO_2 (left), avoidable NO_2 concentration without direct NO_2 emissions (right)

The share of different polluters (vehicle types, other sources) on the total concentration can be estimated based on the sources of the NO_2 concentration as derived above (primary, atmospheric chemistry, background) and the emission shares.

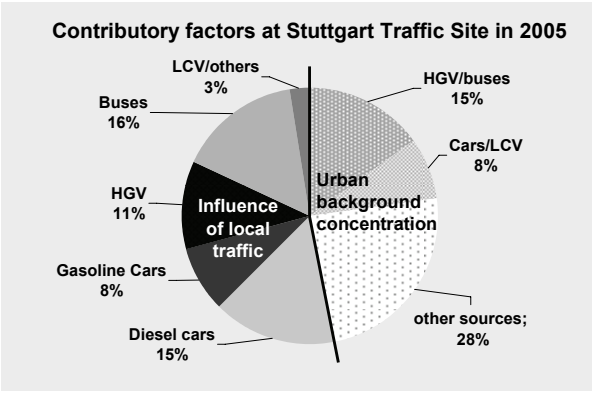

Figure 11: Contribution of vehicle categories to NO_2 concentrations in 2005

The contribution of primary NO_2 to the NO_2 concentration has been allocated to vehicle types according to the shares on NO_2 emissions. The remaining additional local NO_2 pollution is due to atmospheric chemical conversion of emitted NO and has been allocated to vehicle types according to their shares on NO emissions. The allocation of the urban background NO_2 concentration can be undertaken according to the shares of vehicles on total NO_x emissions in the city area.

Accordingly, local traffic contributes about 53% to the NO_2 concentration. The largest single polluters are diesel cars with 15% and buses with 16%. Trucks (despite lower NO_2 emissions, but with high NO emissions) still contribute 11%. About half of the urban background concentration in 2005 is due to traffic emissions. Traffic is thus responsible for about 3/4 of the total NO_2 concentration at the traffic measurement site Stuttgart-Mitte.

b) Relevance of further atmospheric chemical processes besides ozone chemistry

Simplified estimates of the share of different sources on the NO_2 concentration (analysis of ozone measurements see above, simplified modelling [Carslaw 2005]) use a simplified conservation-of-mass equitation. This is based on the assumption that the additional local NO_2 pollution can be – besides primary NO_2 emissions – fully described by ozone depletion, thus that the influence of other atmospheric chemical reactions on NO_x concentration and

NO_2 formation can be neglected. Such methodologies use a constant sum of oxidants ($[O_x] = [NO_2] + [O_3]$) and do not take into account any degradation of nitrogen oxides.

The box model takes into account a range of other atmospheric chemical processes besides the NO_x-Ozone reaction equilibrium, including degradation processes. The modelling, however, can also be undertaken considering only ozone chemistry. The box model can therefore be used to assess the influence of other chemical processes and thus determine the accuracy of a simplified methodology which neglects them.

The left bar in Figure 12, left, represents the modelled NO and NO_2 concentrations with complete atmospheric chemistry, the right bar is the result of the simplified calculation with the NO_x-ozone reactions only. The modelled annual mean NO_2 concentration for 2005 is about 3.1 µg/m³ ***higher*** with complete chemistry than with simplified chemistry. The NO concentration, in contrast, is about 5.1 µg/m³ (in NO_2 eq.) ***lower***. Altogether the overall NO_x concentration is about 2 µg/m³ lower if complete chemistry is taken into account compared to the simplified calculation. According to this modelling, the NO_2 formation by other than ozone related processes has a share of about 20% on the total NO_2 contribution of atmospheric chemical reactions. If only NO_2 formation from ozone depletion is taken into account, the contribution of atmospheric chemical reactions to the NO_2 concentration is underestimated.

The contribution of other atmospheric chemical reactions at least partly explains why the NO_2 shares in exhaust NO_x emissions, which have been recalculated for Stuttgart traffic site from ozone measurements or simplified modelling, are significantly higher than can be explained based on the current NO_x and NO_2 emission calculations for the site (Figure 12, right).

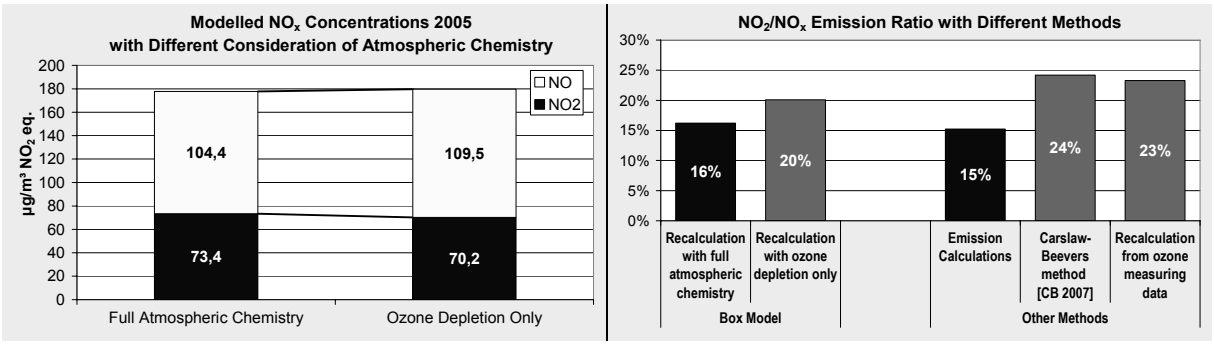

Figure 12: NO and NO_2 box modelling with complete atmospheric chemistry and with ozone chemistry only (left); Recalculated NO_2/NO_x emission ratio with complete atmospheric chemistry and with ozone chemistry only (right)

4.3. Future development of NO_2 at Stuttgart-Mitte

The NO_x concentration as calculated with the box model decreases between 2005 and 2020 by 52% due to a decrease of NO_x emissions (see Figure 8) as well as a decrease in the urban background concentration. The NO_2 concentration, however, which is relevant for the compliance with ambient air quality standards, is calculated to decrease by only 32%, i.e. from 73 to 50 µg/m³ in the same period (Figure 13). According to this scenario, the NO_2 air quality standard of 40 µg/m³, which becomes effective in 2010, will thus still be exceeded in 2020.

A more detailed analysis shows that the NO_2 contribution from the urban background and from atmospheric chemistry continuously decreases until 2020. However, the contribution of primary NO_2 at first increases until 2010. The contribution of primary NO_2 decreases only after 2010 due to the decrease of direct NO_2 emissions (see Figure 8, right). In 2010 and 2015, primary NO_2 contributes more to the total NO_2 concentration than NO from local traffic emissions which is converted into NO_2. This relation is finally reversed in 2020.

In order to analyse the effect of a reduction of primary NO_2 emissions in traffic exhaust, it has been assumed for the modelling, that NO_x emissions of local traffic would consist only of NO, i.e. no primary NO_2 is emitted. This would lead to a 24 µg/m³ reduction of the contribution of primary NO_2 to the total NO_2 concentration in 2010. The NO_2 contribution from atmospheric chemistry, however, would increase by about 5 µg/m³ compared to reference scenario (with primary NO_2) due to the higher NO emissions. The overall reduction effect would thus be only 19 µg/m³ (Figure 13, right). Accordingly, the NO_2 air quality limit would not be complied with in 2010 in the trend scenario, even if primary NO_2 emissions would be fully eliminated.

In order to comply with the 2010 air quality limit at Stuttgart-Mitte, a further reduction of NO_x emissions in the urban background (e.g. by introducing Euro VI trucks, other NEC measures) as well as at Stuttgart-Mitte (Trucks Euro VI, low emission zone) will be necessary

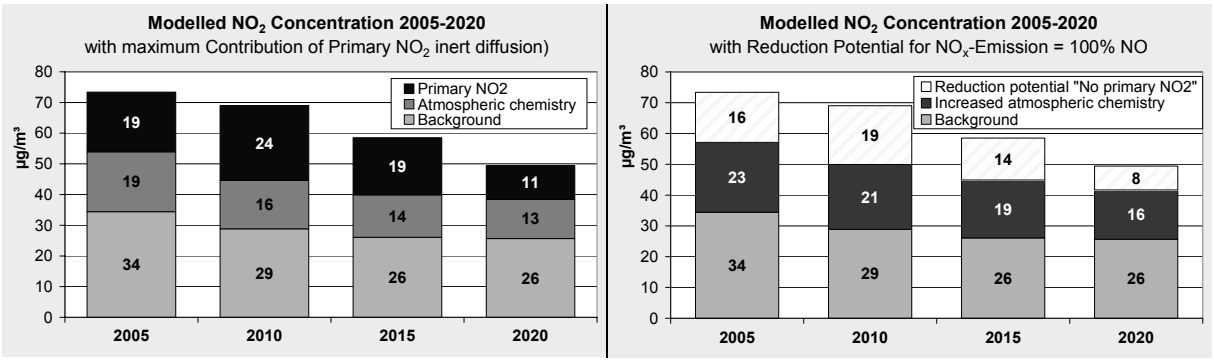

Figure 13: Modelled NO_2 concentrations 2005-2020 for Stuttgart traffic site

5. CONCLUSIONS

Based on an extensive analysis of emission data, it has been shown that the NO_2 emissions in the exhaust of diesel cars have increased in recent years. This is basically due to the fact that these vehicles have been equipped with oxidation catalytic converters. Also particulate filters for buses which use NO_2 for regeneration, can lead to an increase in NO_2 emissions.

The impact of this change in exhaust composition has been exemplified for the traffic measurement site Stuttgart-Mitte. On the emission side, an increase in primary NO_2 emissions until 2010 has been calculated due to increasing numbers of modern diesel cars and buses with exhaust after treatment. NO_2 emissions decrease only after 2010 because of the introduction of Euro-5 and Euro-6 vehicles.

The NO_2 concentration calculated with a chemical box model for Stuttgart-Mitte consists of an urban background concentration, NO_2 from atmospheric chemistry as well as primary NO_2. The development of traffic emissions leads to an increase in the contribution of primary NO_2 to the overall NO_2 concentration between 2005 and 2010, the urban background concentration and NO_2 from atmospheric chemistry show a slight decrease. This leads to a 40% contribution of primary NO_2 to the overall concentration in 2010. The overall NO_2 concentration therefore decreases only slightly between 2005 and 2010. According to the calculations, without additional measures the NO_2 air quality standard for 2010 is not even reached in 2020.

The analysis shows that the reduction of primary NO_2 emissions as well as the reduction of the NO emissions of local traffic and a reduction of the urban background concentration can make a relevant contribution to an improvement of air quality. The shares of different causes need to be analysed site-specific in order to take into account the influence of the local traffic composition and volume and also the influence of other sources.

6. ACKNOWLEDGEMENT

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