THE IMPACT OF INCREASED PRIMARY NO_2 VEHICLE EMISSIONS ON URBAN NO_2 CONCENTRATIONS

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ABSTRACT

In the next years traffic NO_x emissions will decrease strongly, but in contrast, due to exhaust gas after treatment systems, traffic NO_2 emissions will increase. To calculate urban NO_2 concentrations and to define air quality management zones, the transformation from NO to NO_2 must be considered. Therefore reliable methods to calculate the NO-NO_2-formation dependent on the primary NO_2 emissions are required. A new simple method is presented in this work. This function is linked to a high resolution dispersion model to calculate the effect of the changing emission situation on the urban NO_2 concentration in the city of Klagenfurt. Trend scenarios for the years 2010, 2012 and 2020 are illustrated. The results of the study show that the very stringent threshold values in 2010 and 2012 will be exceeded extensively in the urban area.

Keywords: *NO_2-conversion, primary NO emissions, NO-NO_2-model*

1. INTRODUCTION

In many regions in Austria the actual air quality threshold values for nitrogen dioxide (NO_2) are exceeded. Recent investigations show that traffic emissions are the major source for urban NO_2 concentrations. During the combustion mainly nitrogen monoxide (NO) is generated which is consequently oxidised in the atmosphere to NO_2:

$$NO + O_3 \rightarrow NO_2 + O_2 \qquad (1)$$

$$NO + RO_2 \rightarrow NO_2 + RO \qquad (2)$$

$$NO + HO_2 \rightarrow NO_2 + OH \qquad (3)$$

Under the effect of solar radiation ($\lambda < 420$ nm) NO_2 dissociates into NO and atomic oxygen, which will react immediately to ozone (O_3):

$$NO_2 + h\nu \rightarrow NO + O(^3P) \qquad (4)$$

$$O(^3P) + O_2 \rightarrow O_3 \qquad (5)$$

With adequate chemical models it is possible to describe the complex reactions in the atmosphere and to calculate the concentrations of NO and NO_2. Therefore many parameters are needed (e.g. solar radiation, temperature, concentrations of HC and VOC) which are usually not known. In addition such complex models require calculations on time series (episodes) and do not allow the use of simple statistical approaches. In practice (e.g. in the framework of environmental assessment studies) simple empirical approaches like that of Romberg [1] have been approved. With this function the annual mean NO_2 concentration can be determined dependent on the total NO_x concentrations. In **Figure 1** this function is compared with measurements of annual mean values at Austrian monitoring stations during the last years. Older measurements were overestimated by the Romberg function, but recently an increase of the NO_2/NO_x ratio could be observed, especially at roadside stations.

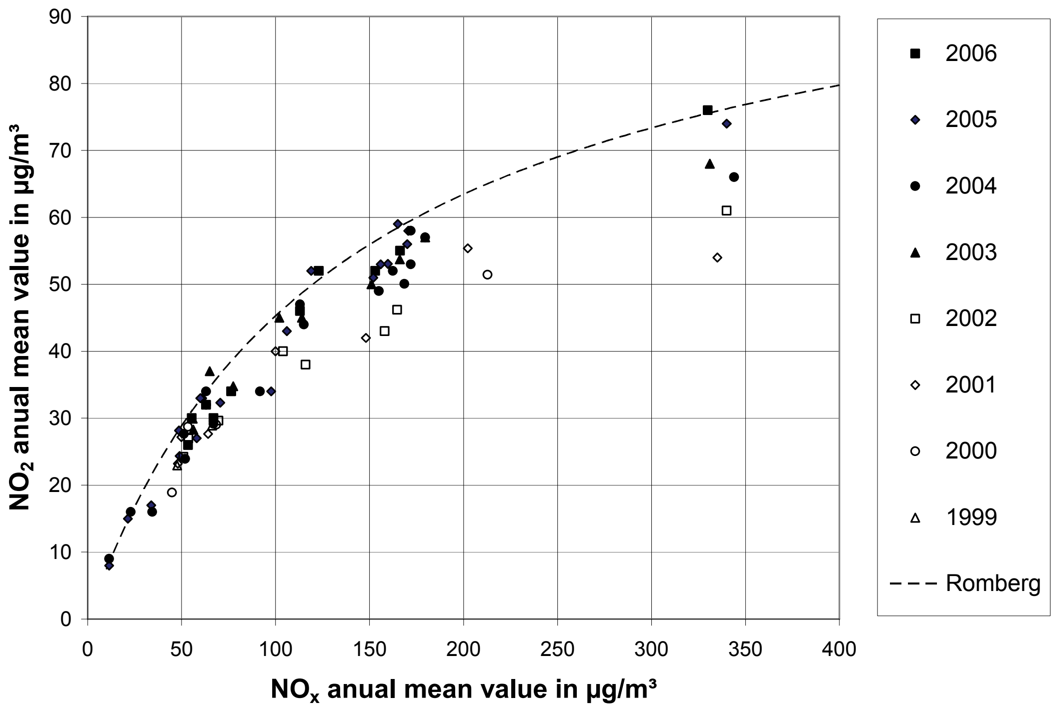

Figure 1: Comparison of the Romberg function with annual mean measurements

Predictions indicate that this trend will be continued and the average fleet NO_2/NO_x emission ratios of between 30-40% will be realistic for the year 2020. For these changed conditions the Romberg function can not be applied with the actual parameters, because the projected NO_2 concentrations will be underestimated, especially at roadside locations. For future scenarios it is necessary to consider the rising primary NO_2 tailpipe emissions in the applied models. Therefore, a novel method is presented to estimate the effect of the rising primary NO_2 emissions on the urban NO_2 concentrations [2].

2. METHODICAL APPROACH

Long-term measurements of monitoring stations in different locations in Austria were used to analyse the evolution of the annual NO_2/NO_x ratio of the past years. First, the NO_2 and NO fleet average traffic emissions were calculated retrospectively with the model GLOBEMI [3]. In addition, the share of traffic emissions to the annual mean NO_x concentrations was estimated from dispersion calculations using the GRAMM/GRAL model [4]. The total annual mean NO_2 air quality concentrations are split into traffic and all other sources (background):

$$NO_{2_{total}} = NO_{2_{traffic}} + NO_{2_{background}} \tag{6}$$

For NO_x emissions from all other sources except from road traffic, the Romberg function is applied using the standard parameters:

$$NO_{2_{background}} = NO_{x_{background}} \cdot \left(\frac{103}{130 + NO_{x_{total}}} + 0{,}005 \right) \tag{7}$$

The photo dissociation of the primary emitted NO_2 is neglected and the emitted NO_2 is treated as an inert pollutant:

$$NO_{2_{traffic}} = NO_{2_{primary}} + NO_{2_{conversion}} \tag{8}$$

For the share of the remaining NO traffic emissions a new exponential conversion function was found:

$$NO = NO_{x_{traffic}} - NO_{2_{primary}} \tag{9}$$

$$NO_{2_{conversion}} = NO \cdot \frac{0{,}72}{e^{\left(\frac{NO_{x_{total}}}{126}\right)}} \tag{10}$$

In **Figure 2** this new conversion function is compared with data derived from measurements. Both the conversion functions, the Romberg function for the background NO_2 concentration and the new exponential function for the traffic induced NO_2 concentrations, are related to the total NO_x concentration. The comparison of the new method with the Romberg function is shown in **Figure 3** for different primary NO_2 emission proportions. In the example 30 µg/m³ NO_x background are assumed. With a share of around 20% primary NO_2 emissions the new method is consistent with the Romberg function. For increasing NO_2/NO_x ratios the total conversion increases, especially at roadside locations. If the traffic emissions approach zero, the conversion is in accordance with the Romberg function, and for very high NO_x concentrations the conversion fits to the primary NO_2/NO_x ratio.

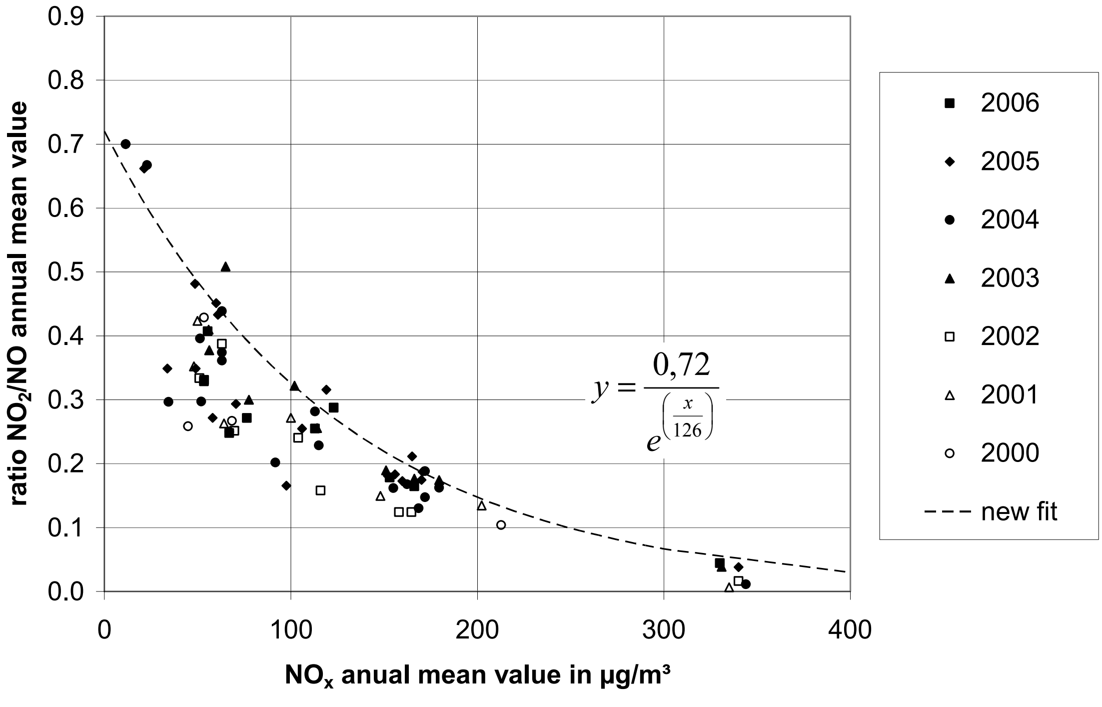

Figure 2: NO_2/NO ratio related to the total NO_x concentration

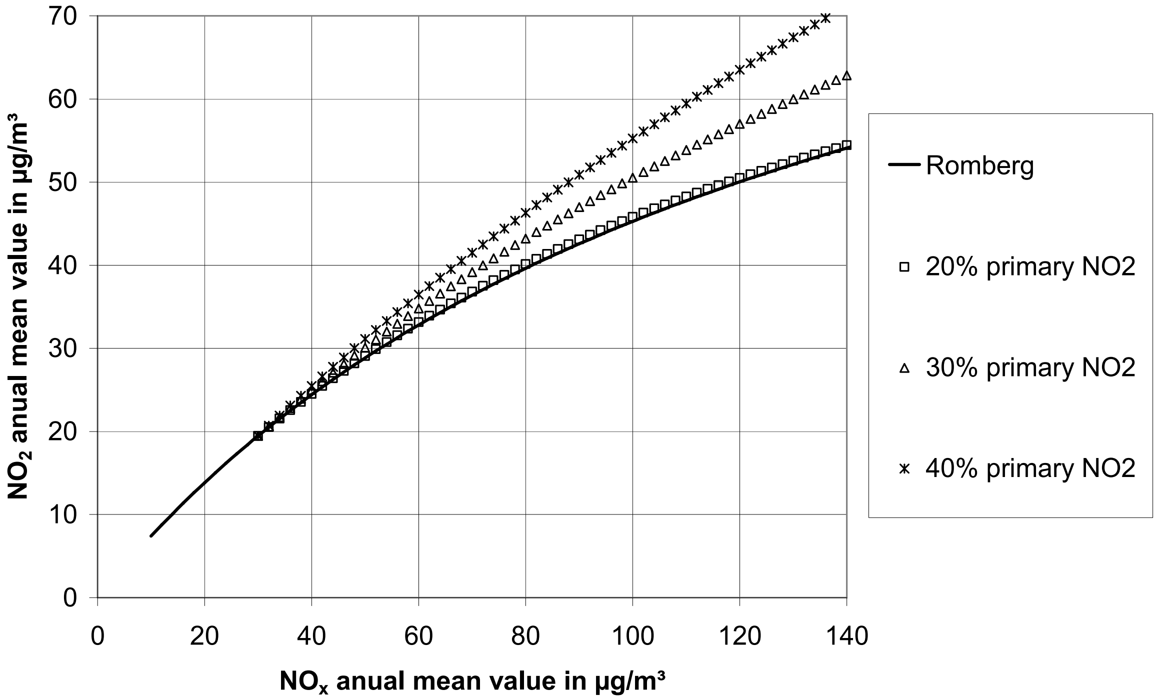

Figure 3: Comparison of the new method with the Romberg function for different primary NO_2 ratios

3. VALIDATION

In **Figure 4** the measured NO_2 concentrations are correlated with the calculated values using the Romberg function. The function is overestimating the older values and fits well to the actual NO_2 concentrations. It is expected, that this function will underestimate the NO_2 conversion in the future, because of rising primary NO_2 emissions. In **Figure 5** it is shown, that the new method accounts for these rising NO_2 emissions. In addition the correlation of the new method is slightly better than with the Romberg function.

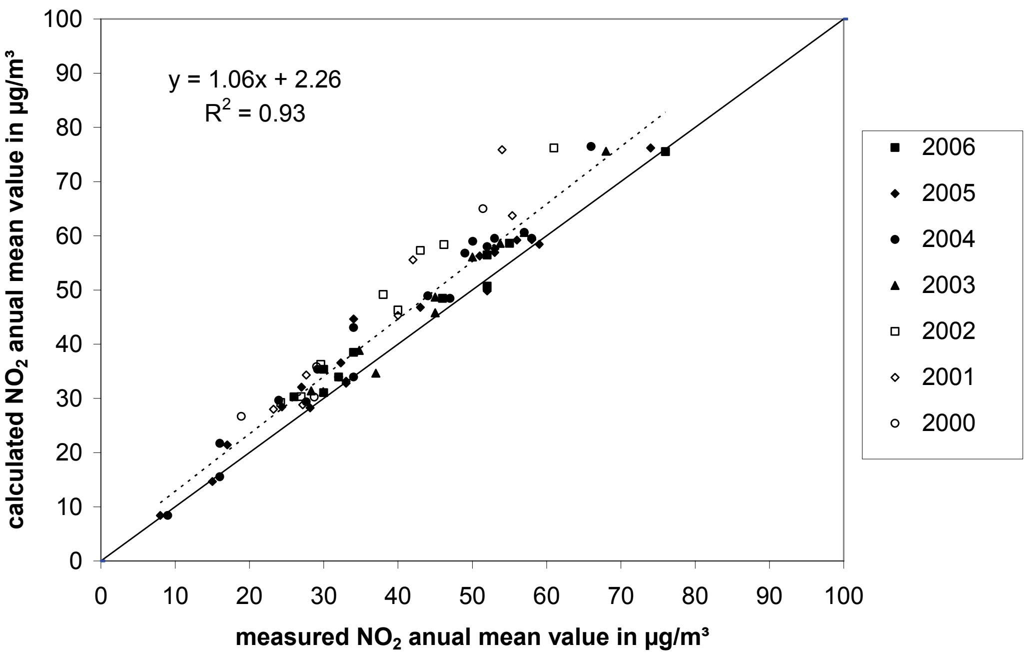

Figure 4: Scatter plot for the Romberg function

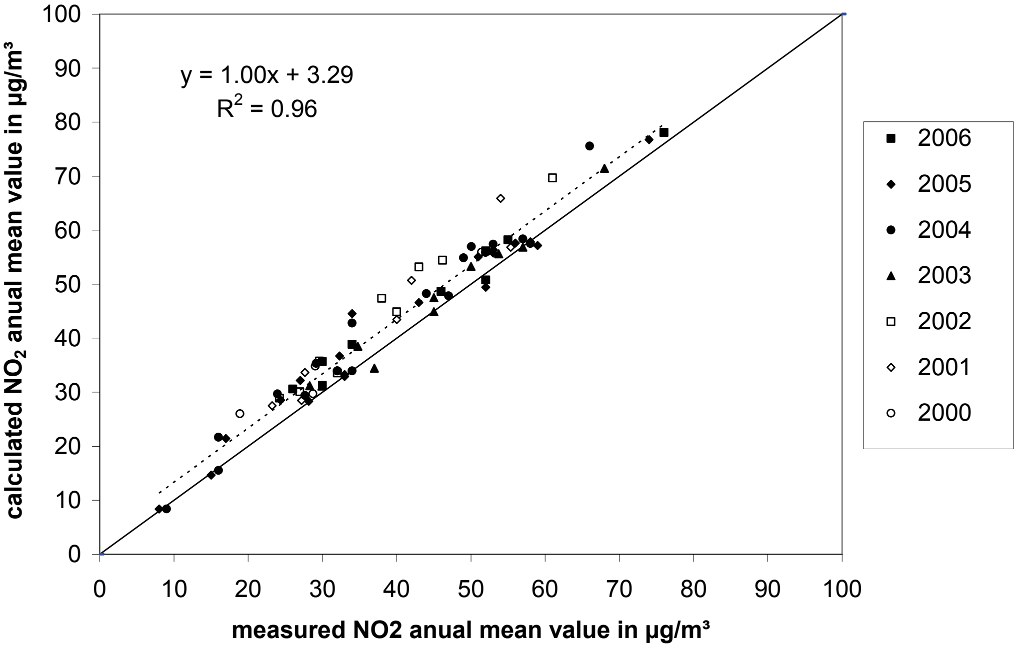

Figure 5: Scatter plot fort new method with the exponential conversion function

4. TREND SCENARIOS FOR NO_2

As an example the new method is applied to the city of Klagenfurt. In a first step the spatial distribution of the annual NO_x concentrations was calculated for the base case in the year 2005. Therefore, traffic emissions were calculated with the model NEMO [5] and the emission inventory of domestic heating and industry was implemented in the simulations. The GRAMM/GRAL model system was used to calculate the spatial distribution of the NO_x concentrations. For the NO-NO_2-convertion in the base case the Romberg function was applied.

For the trend scenarios a linear traffic increase of 2,5% per year was assumed and for the calculation of the traffic emissions the Austrian standard fleet composition for urban traffic was applied. Although the traffic volume increases about 40% until the year 2020 the total NO_x-emissions will be reduced by some 50% (**Figure 6**). But due to exhaust gas after treatment systems the absolute NO_2-emissions will rise in the next years and will not fall below the actual level before 2015. Then they will decrease consistently until the year 2020.

With the new method it is possible to calculate the effects of the changing emission situation on the annual NO_2 concentrations in the city of Klagenfurt. The traffic NO_x concentrations of the base case were scaled to the total NO_x traffic emissions. NO_x emissions of domestic heating and industry and the NO_x background concentration were assumed to remain constant. The new conversion function was the applied.

While the NO_2 concentrations at urban background locations are predicted to stay at the same level, the NO_2 concentrations near main roads or highways will increase in the next years (**Figure 7**). In **Figure 8** the modelled trend scenarios for the NO_2 annual mean values are illustrated. The air quality nonattainment zone is framed with a thicker line. At present the NO_2 air quality standard is exceeded only near main roads and the highway. Due to rising NO_2 concentrations and more stringent threshold values in 2010 and 2012, the nonattainment zone is growing strongly until 2012, when a larger proportion of the urban area is forecast to be affected. After 2012 the reduction of the NO_x and the primary NO_2 emissions is predicted to a decrease in NO_2 concentrations. In 2020 the nonattainment zone will predominantly only include areas near main roads or highways.

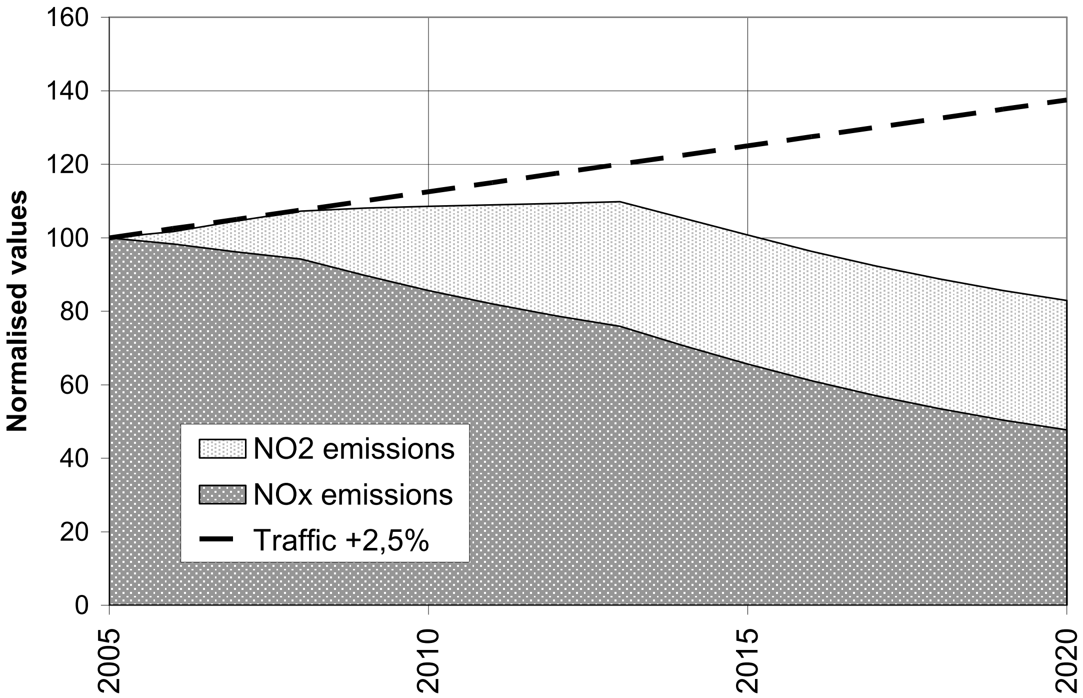

Figure 6: Modelled NO_x-and NO_2-emissions

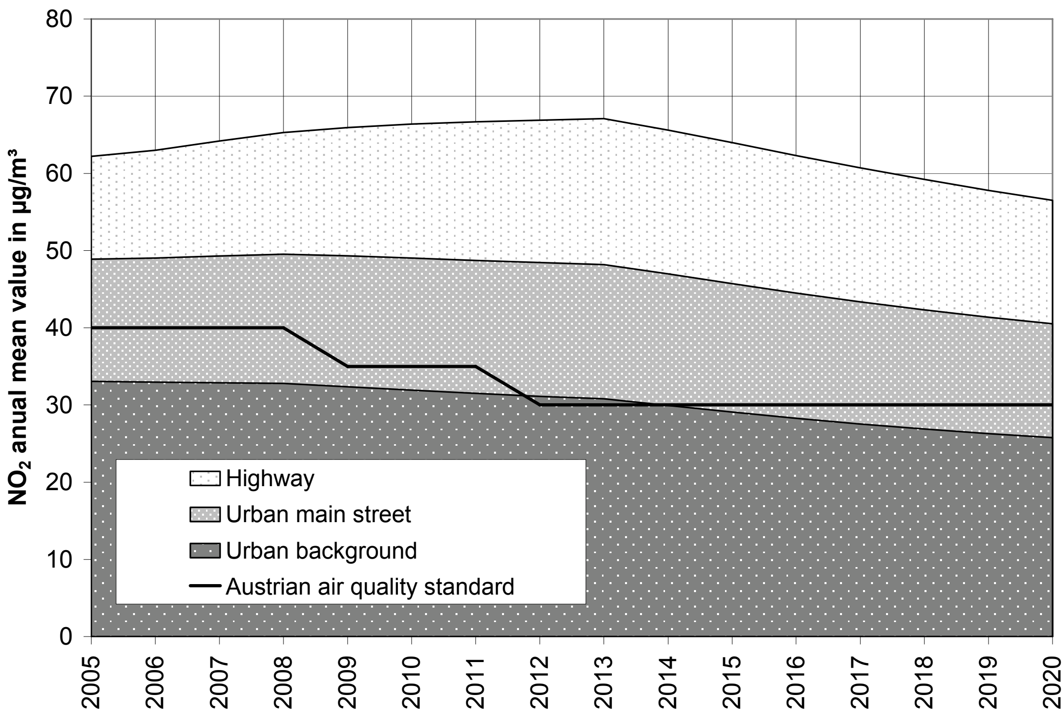

Figure 7: Comparison of the modelled NO_2 concentrations with the Austrian air quality standard

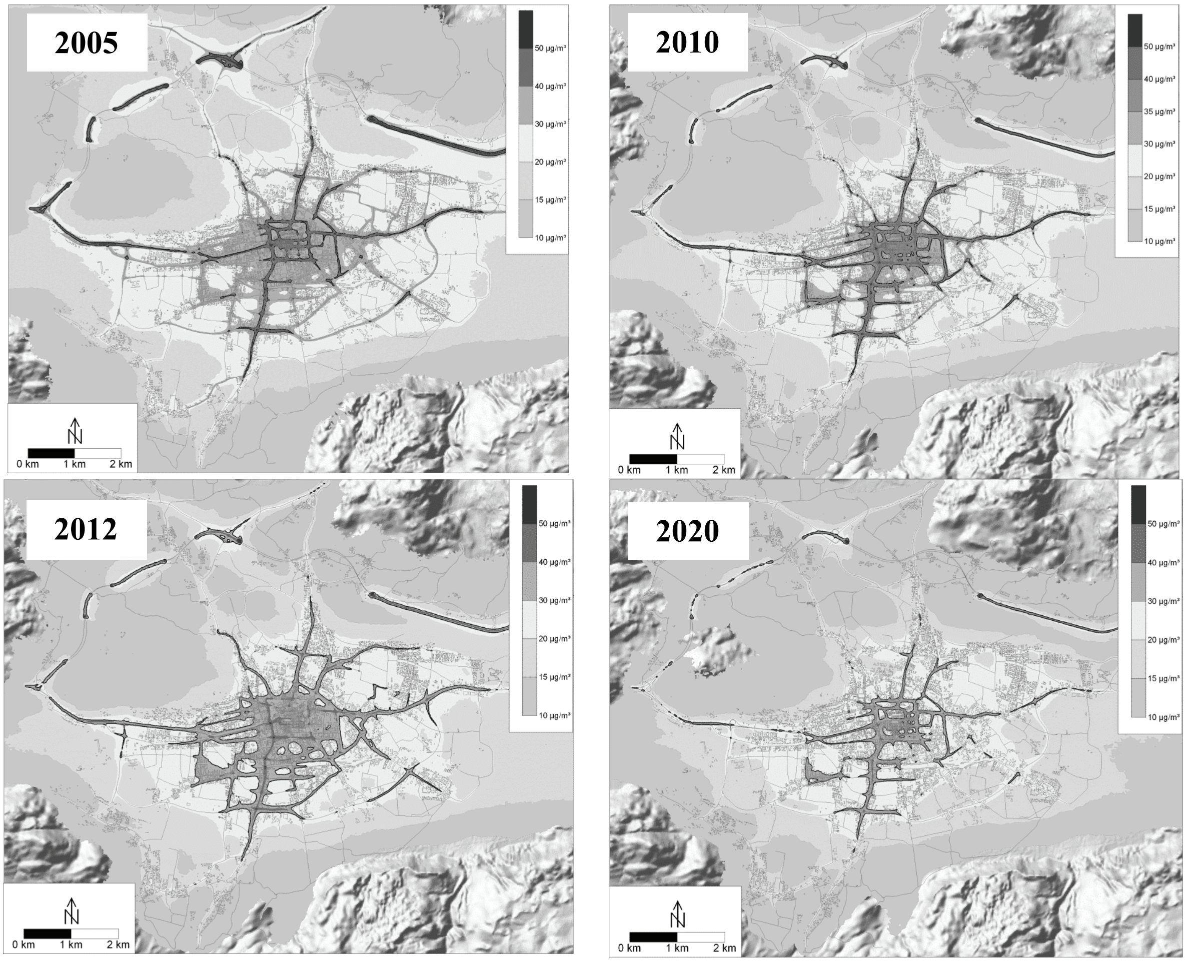

Figure 8: Modelled trend scenarios for the NO2 annual mean value in the city of Klagenfurt

5. CONCLUSIONS

In recent investigations it is shown that traffic NO_x emissions are forecast to decrease strongly in the forth coming years, but primary NO_2 emissions are predicted to increase. Existing statistical models for the prediction of the NO_2-NO-conversion neglect this aspect. In order to take the primary NO_2 tailpipe emissions into account for future scenarios a new simple approach has been developed. While the NO_2 concentrations at urban background locations are predicted to stay at the same level, the NO_2 concentrations near main roads or highways are forecast to increase in the next years. When considered together with the lowered Austrian NO_2 threshold values in the near future, this is likely to lead to expanding nonattainment zones. Especially in the year 2012 it is expected that many urban regions will have problems to meet the future threshold values. Based on this new NO-NO_2-model it is possible to consider changing NO/NO_2 emission shares in scenario calculations in order to evaluate the effects of NO_x emission reduction strategies on NO_2 air quality.

6. REFERENCES

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