UPDATE OF THE ROMBERG-APPROACH AND SIMPLIFIED NO / NO$_2$ CONVERSION MODEL UNDER CONSIDERATION OF DIRECT NO$_2$-EMISSIONS

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Outline

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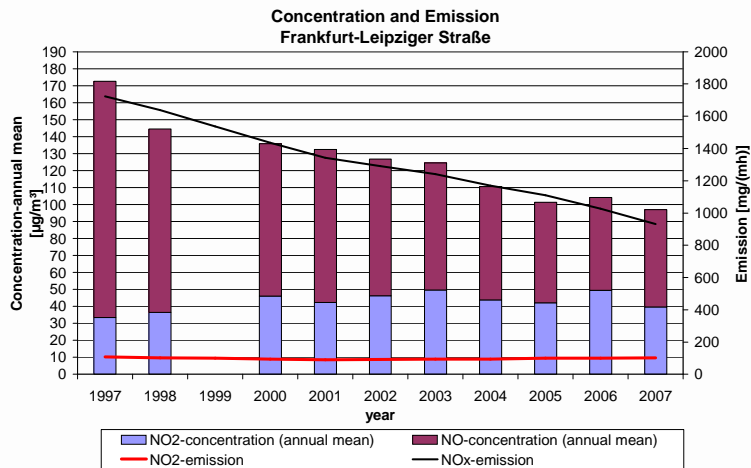
6. Conclusions

Motivation

NO$_2$-limit value: 40µg/m³ (annual mean)

Air quality measurements at street stations show often reduction in NO$_x$ levels, but no significant reduction in NO$_2$

– NO$_x$ = NO + NO$_2$ [ppb]

– NO + O$_3$ => NO$_2$ +O$_2$
... ... more chemistry ... ...

Motivation
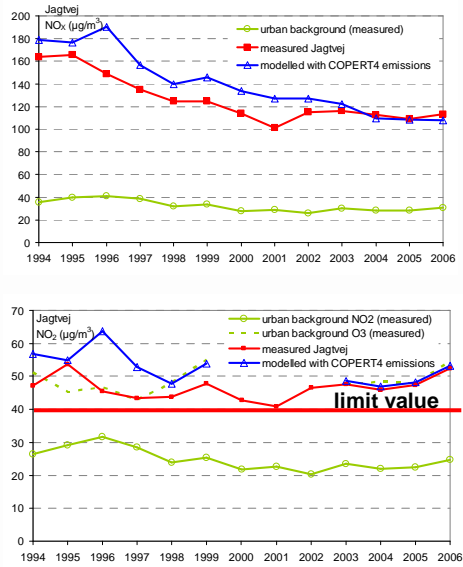
According to the EU directive the EPA in co-operation with local authorities has to prepare action plans to assure compliance by 2010

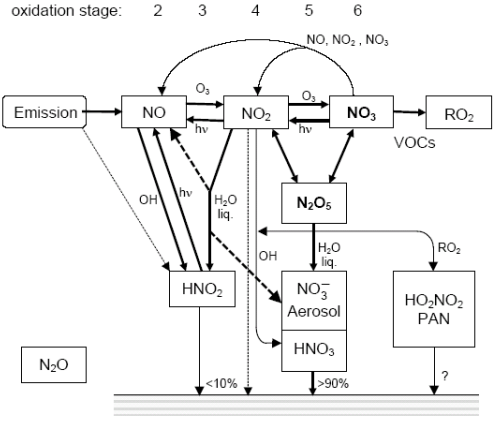Concentration and Emission
Frankfurt-Leipziger Straße

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NO$_x$- chemistry

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Formation of NO$_2$ from NO is a complex photochemical process.

Mainly depending on the total amount of available NO$_x$ and ozone.

NO$_2$ concentration is also depending on the primarily emitted NO$_2$.

Besides NO-emission, ozone concentrations as well as primary NO$_2$ emission are changing from year to year.


(Platt U., 2008)

Updating the Romberg approach

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$$[NO_2] = \frac{A \cdot [NO_x]}{[NO_x] + B} + C \cdot [NO_x]$$

The new constants for the given period of 2004 to 2006 (German measurement sites) along with the earlier parameters from 1996:

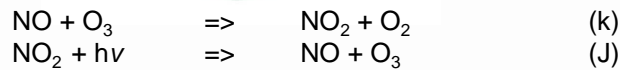| Statistical concentration parameters	parameter of function		
Romberg et al. (1996)	**A**	**B**	**C**
annual average	103	130	0.005
98-percentile	111	119	0.039
Bächlin et al. (2008)	**A**	**B**	**C**
annual average	29	35	0.217
98-percentile	40	20	0.170
19. highest hourly value	43	10	0.151

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Comparison old / new
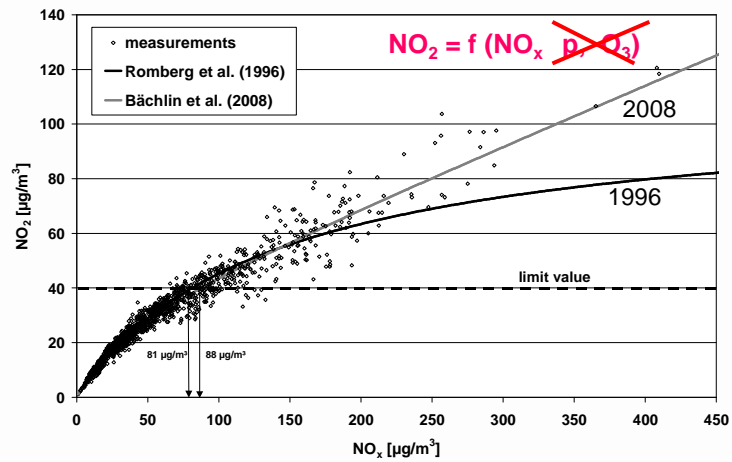
The approach by Romberg et al. (1996) results in too low NO_2-values for high NO_x-values. The annual average value of 40 µg NO_2/m^3 would be reached by an annual mean NO_x-concentration of approx. 88 µg/m³ with the new approach.

OSPM Chemistry Model

More realistic conversions can be obtained using a chemistry model. A simplified relation scheme is often used to describe the chemical conversion in the equilibrium state of NO_2, NO, and ozone.
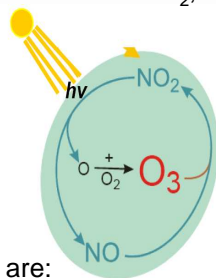
The considered reactions are:

$$NO + O_3 \Rightarrow NO_2 + O_2 \quad (k)$$
$$NO_2 + h\nu \Rightarrow NO + O_3 \quad (J)$$

k (ppb^{-1} s^{-1}) is the reaction coefficient
J (s^{-1}) is the photolytic frequency of NO_2,

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OSPM Chemistry Model

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Assuming that the **equilibrium is quickly reached**, the differential equations have an **analytical solution** for the concentration:

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

With the variables :

$$B = [NO_x] + [NO_2]_O + \frac{1}{k}\left(J + \frac{1}{\tau}\right)$$

$$[NO_2]_O = [NO_2]_n + [O_3]_B \qquad [NO_2]_n = [NO_2]_V + [NO_2]_B$$

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. $[NOx]_V$ is calculated from the difference in NOx between the traffic station and the background station as

$$[NO_2]_V = p\left([NO_x] - [NO_x]_B\right)$$

$$[NO]_V = \left([NO] - [NO]_B\right)$$

$NO_2 = f (NO_x , p, O_{3\,B},$
**$NO_{x\,B}, NO_{2\,B}$**
$k, J, \tau)$

with p = NO_2/NOx being the fraction of NO_2 in the direct traffic emissions and $[NO_x]$ the concentration, which is estimated at the traffic station by measurements or dispersion model.

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Used in OSPM for hourly time series.
The parameter τ 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 τ are in the range of 80s to 150s.

NO_2 modelled
(based on measured NO_x !)

$y = 1.01x - 0.30$
$R^2 = 0.89$
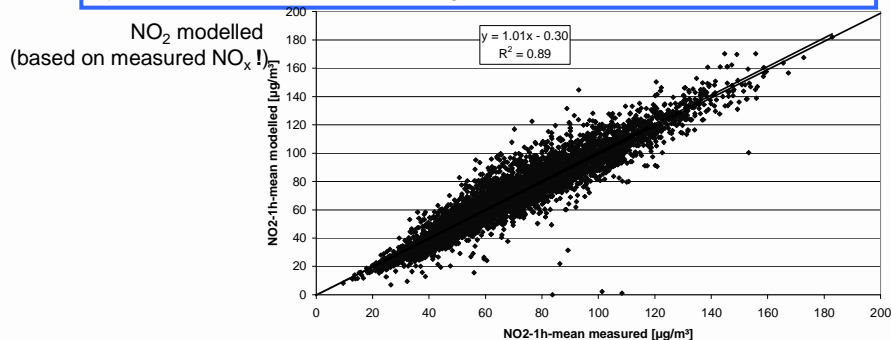
Comparison of hourly mean NO_2-concentrations, calculated with the OSPM chemistry model from observed NOx data, and values from the measurement site Corneliusstraße in Duesseldorf for the year 2006.

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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.

However, based on several research projects, these equations can **also be applied for annual mean** concentrations using the following parameters:

J = 0.0045 s^{-1}
k = 0.00039 m^3 $(ppb\ s)^{-1}$
τ = 100 s (street canyons) or 40 s (free dispersion)

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

NOx- annual mean at traffic station (observed value or estimated by dispersion model)
NOx- annual mean at background station
NO_2- annual mean at background station

$$NO_2 = f\ (NO_x,\ p,\ O_{3\,B},\ NO_{x\,B},\ NO_{2\,B},\ k,\ J,\ \tau)$$

as well as two additional values:
Ozone- annual mean at background station
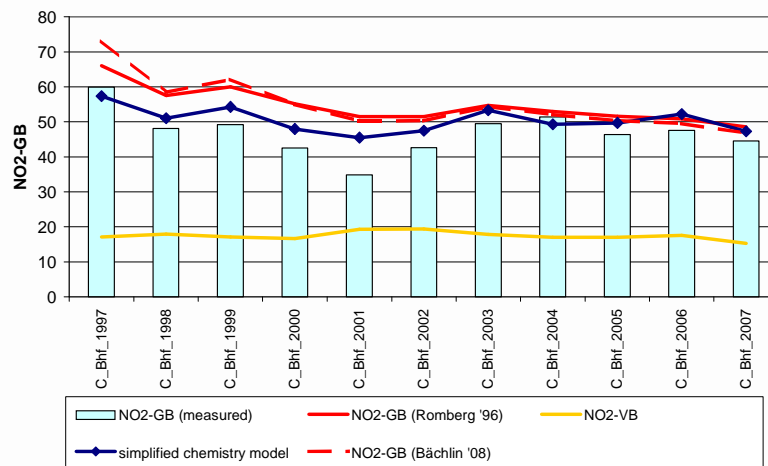p = share of primary NO_2 emissions for all NOx emissions.

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Düsseldorf-Corneliusstraße
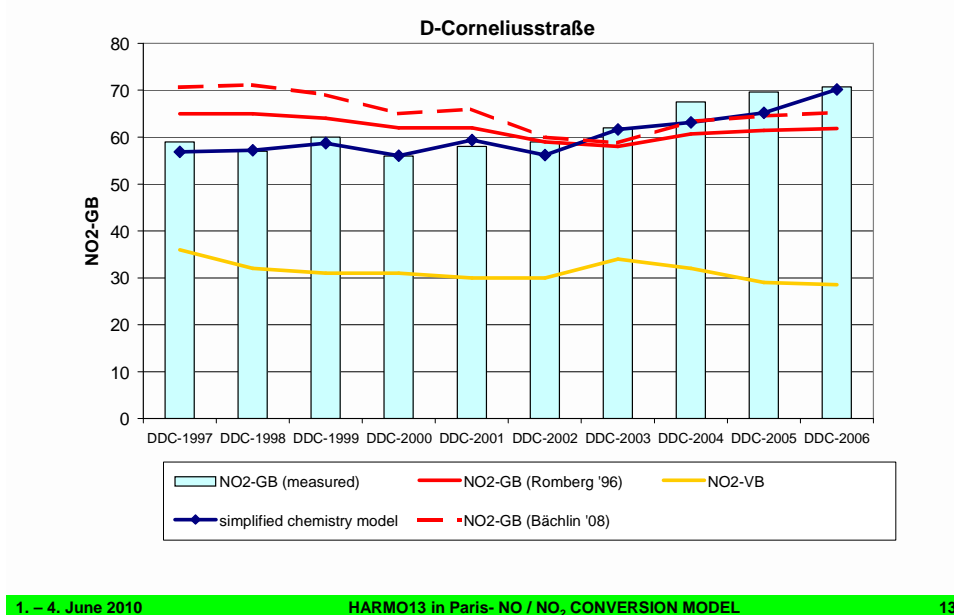
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D-Corneliusstraße

NO2-GB (measured) — NO2-GB (Romberg '96) — NO2-VB
simplified chemistry model — - NO2-GB (Bächlin '08)

**Comparision (Data 1997 - 2007;
30 German measurement sites)**

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Romberg (1996)

Bächlin (2008)

Simplified
chemistry
model
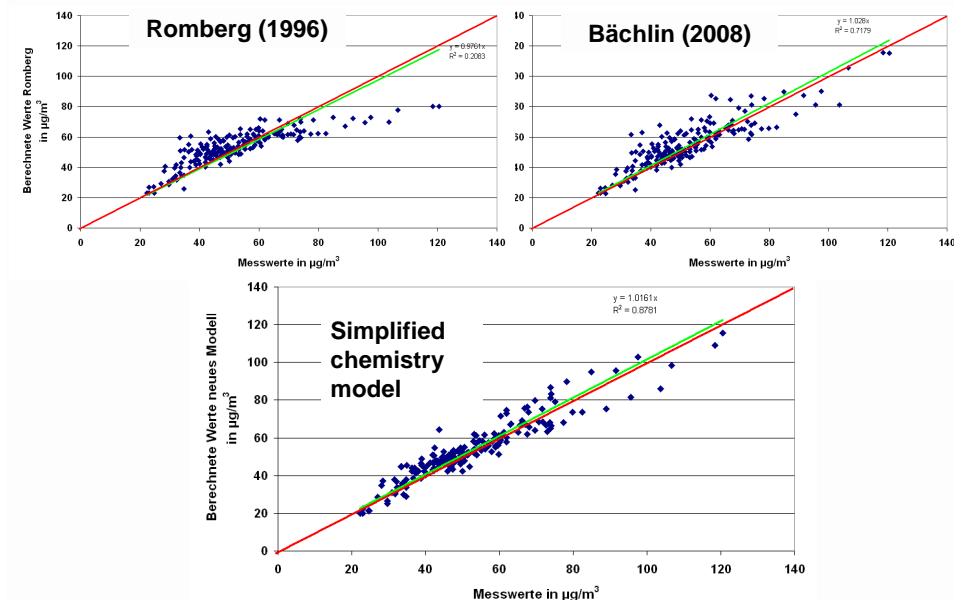
Conclusions

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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.

The approach by Bächlin and Bösinger (2008) reproduces the existing measured data better than the approach by Romberg, but it cannot reproduce the long term tendencies very well, either.

The simplified chemistry model can best reproduce the tendencies and the absolute values. The main reason for the good fit for trends in the NO_2/NOx ratios is the explicit incorporation of the share of primary NO_2 emissions.

➔ Better NO_2-prognosis possible, especially if primary NO_2-emission changes (e.g. change of the vehicle fleet mix due to environmental zone, future fleet trend etc.)

Acknowledgements

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