

# **Site specific projections of NO<sub>x</sub> and NO<sub>2</sub> concentrations for the UK**

A report produced for The Department of the Environment, Transport and the Regions, The National Assembly for Wales, The Scottish Executive and The Department of the Environment for Northern Ireland

John R Stedman

September 1999

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# Executive Summary

The 1997 UK National Air Quality Strategy (NAQS) gives the following provisional objectives for nitrogen dioxide (NO<sub>2</sub>) to be achieved by the end of 2005:

- Annual mean: The annual mean must not exceed 21 ppb.
- Hourly mean: The hourly mean must not exceed 150 ppb.

The recently published consultation document, The Air Quality Strategy for England, Scotland, Wales and Northern Ireland proposes the retention of the provisional annual mean objective and the replacement of the hourly mean objective by the following:

- Hourly mean: 104.6 ppb, not to be exceeded more than 18 times a year.

The annual mean objective is likely to be the most stringent of the objectives, particularly at the roadside.

Site specific projections of annual mean concentrations for NO<sub>2</sub> and NO<sub>x</sub> for all years between 1990 and 2005 are presented here by projecting both forwards and backwards from 1996 and 1997 monitoring data. Projecting backwards as well as forwards provides an indication of the reliability of the prediction methods that have been used in the review of the National Air Quality Strategy. These site specific projections have the advantage that they do not have the additional uncertainty associated with mapping methods.

The site specific projections presented in this report clearly illustrate the impact of emissions reductions on ambient NO<sub>x</sub> concentrations and the correspondingly smaller reduction in annual mean NO<sub>2</sub>.

Overall, the backwards projections at urban background sites show good agreement with the measurements, which gives confidence in the conclusions reached in the review of the NAQS. These conclusions were that inner London background sites are likely to be at risk of exceeding 21 ppb in 2005 but all other background sites should have concentrations lower than 21 ppb.

Measured annual mean concentrations of both NO<sub>x</sub> and NO<sub>2</sub> were generally higher in 1997 than in 1996, particularly set against the expected decline in emissions. This was presumably caused by unusually poor dispersion of primary pollutants during 1997 due to the meteorological conditions. Backwards projections based on these two years therefore provide reasonable estimates of the range of mean values in earlier years. Measured concentration in 1998 were generally lower than would be expected due to the falling trend in emissions, again presumably due to the prevailing meteorological conditions which lead to unusually efficient dispersion of pollutants.

The backwards projections at roadside sites generally show reasonable agreement with the limited amount of monitoring data that are available. This confirms the conclusions in the review

of the NAQS that many of the roadside sites included in the analysis are likely to be at risk of exceeding the 21 ppb objective in 2005.



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# 1 Introduction

The 1997 UK National Air Quality Strategy (NAQS) gives the following provisional objectives for nitrogen dioxide (NO<sub>2</sub>) to be achieved by the end of 2005 (DoE *et al*, 1997):

- Annual mean: The annual mean must not exceed 21 ppb.
- Hourly mean: The hourly mean must not exceed 150 ppb.

The recently published consultation document, The Air Quality Strategy for England, Scotland, Wales and Northern Ireland (DETR *et al*, 1999b) proposes the retention of the provisional annual mean objective and the replacement of the hourly mean objective by the following:

- Hourly mean: 104.6 ppb, not to be exceeded more than 18 times a year.

The annual mean objective is likely to be the most stringent of the objectives, particularly at the roadside.

Concentrations of both NO<sub>2</sub> and oxides of nitrogen (NO<sub>x</sub>, the sum of NO and NO<sub>2</sub>) are currently monitored at a wide range of automatic monitoring sites around the UK (Broughton *et al*, 1998). Annual mean NO<sub>2</sub> concentrations in 1998 were higher than 21 ppb at about half of the UK national monitoring sites. Emissions of NO<sub>x</sub> from road traffic sources make an important contribution to urban background NO<sub>2</sub> concentrations and will clearly dominate concentrations at the roadside. National measures are likely to deliver significant reductions in road traffic NO<sub>x</sub> emissions between 1996 and 2005 (Murrells, *pers comm*). It is likely, however, that there will still be exceedances of the annual mean objective in 2005 if national measures are considered in isolation, particularly at the roadside. It was envisaged that this 'policy gap' would be addressed by implementing local air quality management to reduce concentrations in locations that are at risk of exceeding the objective.

Maps of estimated annual mean NO<sub>2</sub> concentrations for 2005 in both urban background and roadside locations have recently been prepared as part of the review of the NAQS (DETR *et al*, 1999a DETR *et al*, 1999b, Stedman *et al*, 1998). These maps indicate that current national policies are likely to result in annual mean urban background NO<sub>2</sub> concentrations in 2005 of below 21 ppb in all areas except inner London. Roadside NO<sub>2</sub> concentrations in urban areas in 2005 are expected to be significantly higher, with about 10% of the total number of UK urban major road links likely to have concentrations higher than the annual mean objective of 21 ppb. The majority of these links are expected to be in the Greater London area. The remainder are generally confined to the most heavily trafficked roads in other big cities. The review concluded that on the evidence available, achieving the annual mean NO<sub>2</sub> objective at the roadside is likely to be very challenging in London and may also be difficult in some other major conurbations. For this reason and because of the uncertainties associated with the NO<sub>2</sub> modelling work carried out for the review, the Government decided that the annual mean nitrogen dioxide objective should remain as a provisional objective at this stage, to be reviewed in two years time.



Site specific projections of annual mean NO<sub>2</sub> concentrations in 2005 for the review of the NAQS were also calculated by Stedman *et al* (1998) based on 1996 and 1997 monitoring results. These site specific projections have the advantage that they do not have the additional uncertainty associated with the mapping methods. The site specific projections were found to be generally consistent with those derived from mapping methods.

Site specific projections for NO<sub>2</sub> and NO<sub>x</sub> for all years between 1990 and 2005 are presented here by projecting both forwards and backwards in time from 1996 and 1997 monitoring data. Projecting backwards as well as forwards provides an indication of the reliability of the prediction methods that have been used in the review of the NAQS. The inter-year changes in measured concentrations are also put into the context of changes in emissions, enabling the identification of years with unusually efficient or poor dispersion of primary pollutants. The projections also clearly illustrate the impact of emissions reductions on ambient NO<sub>x</sub> concentrations and the correspondingly smaller changes in annual mean NO<sub>2</sub>.

## 2 Emission projections

The emissions projections used here are the same as used in the projections presented by Stedman *et al*, 1998 and included in the review of the NAQS (DETR *et al*, 1999a, DETR *et al*, 1999b). It is reasonable to assume that road traffic emissions of NO<sub>x</sub> will determine the additional NO<sub>x</sub> or NO<sub>2</sub> measured at the roadside in comparison to that measured at nearby urban background locations (the roadside enhancement of NO<sub>x</sub> or NO<sub>2</sub>). Similarly, it was assumed that traffic emissions contribute 90 % of the total low level NO<sub>x</sub> emissions in areas with the highest urban background concentrations. It was also assumed that this remaining 10% of emissions will remain unchanged between 1996 and 2005.

The National Atmospheric Emissions Inventory (NAEI) road transport model was been used to calculate estimates of urban road traffic NO<sub>x</sub> emissions for 1996 and other years (Salway *et al*, 1999, Murrells, *pers comm*). The emissions in future years have been calculated for a 'business as usual' scenario, designed to represent the impacts of current national and international policies. The emissions estimates listed in Table 1 show that urban road traffic emissions of NO<sub>x</sub> are expected to fall to about 44% of 1996 levels by 2005. If the remaining 10% of emissions contributing to background concentrations of NO<sub>x</sub> remain at 1996 levels in 2005, then this leads to a reduction in 'background' emissions to 50% of 1996 levels.

**Table 1 UK urban road traffic emissions of NO<sub>x</sub> (kTonnes per year, based on 1996 NAEI)**

year	1990	1991	1992	1993	1994	1995	1996	1997
emissions	458	466	447	431	419	393	370	341
percentage of 1996	124%	126%	121%	117%	113%	106%	100%	92%
year	1998	1999	2000	2001	2002	2003	2004	2005
emissions	312	286	260	237	214	194	176	162

percentage of 1996	84%	77%	70%	64%	58%	52%	48%	44%
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The emissions projections used both here and in the work completed for the review of the NAQS were based on the NAEI road transport model as used to compile estimates of UK emissions for 1996 and subsequently published by Salway *et al* (1999). Emissions projections have recently been calculated using a revised model, which will be used to calculate an NAEI inventory for 1997. Table 2 shows a comparison of these new estimates with those that have been used in previous work. The revised estimates have lower emissions for 1996 and higher emissions for 2005. As a result urban road traffic emissions of NO<sub>x</sub> are expected to fall to about 51% of 1996 levels by 2005, as opposed to the estimate of 44% of 1996 levels used in earlier work. The site specific projections of NO<sub>x</sub> and NO<sub>2</sub> concentrations presented in this report have been based on the same emissions estimates as used previously for consistency with the review of the NAQS. This enables us to assess the reliability of the prediction methods that have been used in the review of the NAQS. Projections based on the revised emission estimates have been included in section 6 as a sensitivity analysis. Site specific and map based projections of future concentrations carried out for subsequent reviews of the NAQS will be based on the most up to date emission estimates available at the time. It will also be appropriate to revisit the assumption of 10% of 1996 emissions from non-road traffic sources at this stage and to compare historical trends in non-traffic emissions with monitoring results.

**Table 2 Comparison of UK urban road traffic emissions of NO<sub>x</sub> based on 1996 and 1997 NAEI model calculations (kTonnes per year)**

	1996	2005	2005 as a percentage of 1996
1996 NAEI inventory	370	162	44%
1997 NAEI inventory	361	184	51%

### 3 Predicting NO<sub>2</sub> from NO<sub>x</sub>

Nitrogen dioxide is often described as a secondary pollutant because the majority of ambient NO<sub>2</sub> is formed by oxidation of NO that has been emitted into the atmosphere. An understanding of the oxidation processes that lead to the current measured concentrations of NO<sub>2</sub> and how these processes are likely to be influenced by changes in future emissions are therefore essential for predicting future concentrations of NO<sub>2</sub>.

One of the main mechanisms by which NO is oxidised to NO<sub>2</sub> is by reaction with O<sub>3</sub>. If NO<sub>x</sub> emissions are reduced and O<sub>3</sub> concentrations remain approximately unchanged then this will increase the proportion of NO<sub>x</sub> emissions that will be rapidly converted to NO<sub>2</sub>. This means that annual mean concentrations are unlikely to respond in direct proportion to reductions in NO<sub>x</sub> emissions.

The trimolecular reaction of NO with O<sub>2</sub> is thought to be one of the dominant oxidation routes on rare winter smog episode days (Bower *et al*, 1994). The rate of this reaction depends on the square of the NO concentration and a reduction in NO<sub>x</sub> emissions to about half of the current values on these types of days will therefore tend to significantly reduce the efficiency of this pathway. Winter smog episode concentrations of NO<sub>2</sub> are therefore expected to be much reduced by 2005 and this is the reason why predictions in this report are focused on annual means.

Methods for predicting future NO<sub>2</sub> concentrations from predictions of NO<sub>x</sub> have been and are currently the subject of considerable research, because of the importance of these predictions within the review of the NAQS. The methods that were used to predict background concentrations of NO<sub>2</sub> in the report prepared for the review of the NAQS (Stedman *et al*, 1998) were not fully consistent with the methods that were applied at the roadside. Site specific projections of background NO<sub>2</sub> for 2005 were derived from 1996 NO<sub>2</sub> measurements. The results of the modelling work of Derwent (1999) that a 50% reduction in NO<sub>x</sub> emissions would be expected to lead to a 30% reduction in annual mean urban NO<sub>2</sub> concentrations were applied. Projections of roadside NO<sub>2</sub> concentrations for 2005 were derived from projections of roadside NO<sub>x</sub> concentrations using an empirically derived non-linear relationship between measured annual mean NO<sub>x</sub> and NO<sub>2</sub> concentrations.

Figure 1 shows a comparison of measured annual mean concentrations of NO<sub>x</sub> and NO<sub>2</sub> for both background and roadside monitoring locations in 1997. The solid line on Figure 1 shows a non-linear function that has been fitted to the measurement data for background sites. The advantage of using this type of non-linear curve is that future NO<sub>2</sub> concentrations can be directly predicted from NO<sub>x</sub> predictions by assuming that the curve will remain the same in future years.

Figure 1 also shows that roadside NO<sub>2</sub> concentration are generally lower than background concentrations for the same measured NO<sub>x</sub> concentration. This is because of the limited time that is available for NO to be oxidised to NO<sub>2</sub> at the roadside and the limited amount of ozone that may be available in the roadside environment. The dashed line shows the non-linear function that has been fitted to current roadside measurement data.

# 4 Projections for urban background sites

## 4.1 THE GRAPHS

Site specific projections for urban background sites are shown in Figures 2 to 22. Projections based on both 1996 and 1997 annual mean data have been calculated for sites for which at least five years of monitoring data are available for the period from 1990 to 1998. Sites classified as Urban Background, Urban Centre and Suburban (Broughton, *et al*, 1998) have been included in this analysis. The graphs show the following information:

- projected annual mean NO<sub>2</sub> based on 1996 NO<sub>x</sub> measurements, calculated from NO<sub>x</sub> projections using the non-linear relationship between NO<sub>x</sub> and NO<sub>2</sub> derived from 1997 UK national monitoring data at background sites.
- measured annual mean NO<sub>2</sub>.
- projected annual mean NO<sub>x</sub> based on 1996 NO<sub>x</sub> measurements.
- measured annual mean NO<sub>x</sub>.
- projected annual mean NO<sub>2</sub> based on 1997 NO<sub>x</sub> measurements, calculated from NO<sub>x</sub> projections using the non-linear relationship between NO<sub>x</sub> and NO<sub>2</sub> derived from 1997 UK national monitoring data at background sites.
- projected annual mean NO<sub>x</sub> based on 1997 NO<sub>x</sub> measurements.
- NAQS linear projected NO<sub>2</sub>, based on 1996 NO<sub>2</sub> measurements, these are the values included in Table A9 of the review of the NAQS report. A linear NO<sub>2</sub> based method was used for consistency with the mapped projections of background NO<sub>2</sub> in the same report.
- NAQS linear projected NO<sub>2</sub>, based on 1996 NO<sub>2</sub> measurements, these are the values included in Table A9 of the review of the NAQS report.
- 21 ppb line, the NAQS objective for the end of 2005.

## 4.2 GENERAL CONCLUSIONS

The non-linear NO<sub>x</sub> based projections of NO<sub>2</sub> are generally equal to or lower than the NO<sub>2</sub> based linear projections included in the review of the NAQS (DETR *et al*, 1999a, DETR *et al*, 1999b). The non-linear projections are lower at sites where the current measured NO<sub>2</sub>/NO<sub>x</sub> ratio is unusually high. The predictions of current NO<sub>2</sub> (derived from the current NO<sub>x</sub> measurements) are lower than the measured values for these sites.

Overall, the backwards projections show good agreement with the measurements, which gives confidence in the conclusions reached in the review of the NAQS. These conclusions were that inner London background sites are likely to be at risk of exceeding 21 ppb in 2005 but all other background sites should have concentrations lower than 21 ppb. There are however indications that one site in Scotland (Glasgow City Chambers, see below) has not seen the expected decline in measured annual mean NO<sub>2</sub> concentrations in recent years.

Measured annual mean concentrations of both  $\text{NO}_x$  and  $\text{NO}_2$  were generally higher in 1997 than in 1996, particularly set against the expected decline in emissions, presumably due to unusually poor dispersion of primary pollutants due to meteorological conditions during 1997. Backwards projections based on these two years therefore provide reasonable estimates of the range of mean values in earlier years. Measured concentration in 1998 were generally lower than would be expected due to the falling trend in emissions, again presumably due to the prevailing meteorological conditions which lead to unusually efficient dispersion of pollutants.

### 4.3 INDIVIDUAL SITES

Projected  $\text{NO}_2$  concentrations at London Bridge Place and London Bloomsbury are not consistent with the measurements. The projections consistently under predict  $\text{NO}_2$  concentrations at London Bridge Place due to the unusually high  $\text{NO}_2/\text{NO}_x$  ratio at this site. Preliminary results from a detailed examination of measured annual mean  $\text{NO}_x$  and  $\text{NO}_2$  concentrations at a range of monitoring sites over the period from 1990 to 1998 indicate that a linear  $\text{NO}_2$  to  $\text{NO}_x$  may provide a better fit with changes in measured concentrations over time. This linear relationship is very similar to the non-linear relationship adopted here at all sites except those with the highest measured  $\text{NO}_x$  concentrations, such as London Bridge Place. Both  $\text{NO}_x$  and  $\text{NO}_2$  concentrations show little reduction in concentrations at London Bloomsbury compared with that seen at other sites. These two inner London sites are therefore expected to be at risk of exceeding the annual mean  $\text{NO}_2$  objective in 2005.

Measured concentrations at Glasgow City Chambers show no reduction in  $\text{NO}_2$  as  $\text{NO}_x$  levels decline. Preliminary results from a detailed examination of measured  $\text{NO}_x$  and  $\text{NO}_2$  concentrations at this monitoring site over the period from 1990 to 1998 show that this unusual behaviour is confined to the winter months, with summer concentrations declining as expected with reductions in measured  $\text{NO}_x$ . The lack of response to changes in  $\text{NO}_x$  during the winter is sufficient to cause the annual mean concentrations to remain approximately constant from year to year. This unusual behaviour of winter mean  $\text{NO}_2$  concentrations is also seen at Edinburgh Centre but the effect is smaller at this site and annual mean concentrations are seen to decline with reductions in measured  $\text{NO}_x$  as expected between 1993 and 1998.

Sites such as Sheffield Tinsley, Walsall Alunwell and Southampton Centre show projected  $\text{NO}_2$  higher than measurements, due to the greater influence of nearby roads on  $\text{NO}_x$  than on  $\text{NO}_2$ .

There is excellent agreement between projections and measurements at West London, Cardiff Centre, Belfast Centre, Leeds Centre, Liverpool Centre, Birmingham East, Hull Centre, Leicester Centre and London Bexley.

# 5 Projections for roadside sites

## 5.1 THE GRAPHS

Site specific projections for roadside sites are shown in Figures 23 to 37. Projections have been calculated for all sites with at least one year of measurement data and are based on 1997 monitoring data for most of the sites. Sites classified as both Kerbside (within 1 m of the kerb) and Roadside (generally 1 - 5 m from the kerb) have been included. Projections for Cromwell Road are based on 1995 data, the most recent year with reasonably complete data capture. The projections for Norwich Roadside, London Marylebone Road and Hove Roadside are the same as those included in the review of the NAQS, which were based on provisional measurements covering most of 1998. The roadside enhancement of  $\text{NO}_x$  concentrations at each site was calculated by subtracting mapped estimates of background concentrations (from Stedman, 1998) for the 1 km grid square including the location of the roadside site from the roadside annual mean measurement. The roadside enhancement of  $\text{NO}_x$  was then projected forwards and backwards on the basis of the estimates of UK urban road traffic emissions listed in Table 1. The background component was projected on the same basis as the site specific projections for urban background sites in section 4, with 10% of 1996 emissions remaining constant throughout. The graphs show the following information:

- projected annual mean  $\text{NO}_2$  based on 1997  $\text{NO}_x$  measurements, calculated from  $\text{NO}_x$  projections using a non-linear relationship between  $\text{NO}_x$  and  $\text{NO}_2$  derived from 1996 UK national monitoring data at roadside sites. These are the values included in Table A10 of the review of the NAQS report.
- measured annual mean  $\text{NO}_2$ .
- projected annual mean  $\text{NO}_x$ , based on 1997  $\text{NO}_x$  measurements.
- measured annual mean  $\text{NO}_x$ .
- 21 ppb line, the NAQS objective for the end of 2005.

## 5.2 GENERAL CONCLUSIONS

The backwards projections generally show reasonable agreement with the limited amount of monitoring data that is available. This confirms the conclusions in the review of the NAQS report that many of these roadside sites are likely to be at risk of exceeding the 21 ppb objective in 2005.

## 6 Sensitivity analyses

### 6.1 SITE SPECIFIC PROJECTIONS USING REVISED EMISSIONS PROJECTIONS

Figures 38 to 41 show projections calculated using the emissions projections based on the 1997 NAEI road transport model, as listed in Table 2. Projections have been calculated for West London, Manchester Town Hall, Cromwell Road and Haringey Roadside. As expected projected concentrations in the early 1990s are lower than those based on the 1996 NAEI and projected concentration in 2005 are higher. The backwards projections based on the 1996 NAEI seem to show at least as good agreement with the measured  $\text{NO}_x$  concentrations between 1990 and 1998 as the projections based on the revised emissions estimates.

### 6.2 THE IMPACT OF INCREASES IN NORTHERN HEMISPHERE BACKGROUND OZONE CONCENTRATIONS

There are indications of a small upward trend in annual mean ozone concentrations over the last 10 years at many rural monitoring sites in the UK (PORG, 1997), presumably as a result of an increase in northern hemisphere background concentrations. While there appears to be a consensus among the ozone modelling community that background concentrations will continue to increase, there is less agreement on the likely magnitude of this increase. Some analyses of the likely impact of such changes in background ozone concentrations on future urban  $\text{NO}_2$  concentrations have been carried out by Derwent (1999). A 10 ppb increase in ozone concentrations for all of the hours in a year was found to increase the annual mean  $\text{NO}_2$  concentrations by about 7 ppb for a transect across London. The changes in background ozone concentrations are unlikely to be of this magnitude over the next ten years or so and any changes would also be likely to vary from hour to hour and across the country. Significant increases in background ozone concentrations would clearly lead to annual mean  $\text{NO}_2$  concentrations higher than those projected using the  $\text{NO}_x$  to  $\text{NO}_2$  relationship based on 1997 measurement data presented here.

## 7 Conclusions

The site specific projections presented in this report clearly illustrate the impact of emissions reductions on ambient  $\text{NO}_x$  concentrations and the correspondingly smaller reduction in annual mean  $\text{NO}_2$ .

Overall, the backwards projections of concentrations at urban background sites show good agreement with the measurements, which gives confidence in the conclusions reached in the

review of the NAQS. These conclusions were that inner London background sites are likely to be at risk of exceeding 21 ppb in 2005 but all other background sites should have concentrations lower than 21 ppb. There are however indications that one site in Scotland has not seen the expected decline in measured annual mean NO<sub>2</sub> concentrations in recent years.

Measured annual mean concentrations of both NO<sub>x</sub> and NO<sub>2</sub> were generally higher in 1997 than in 1996, particularly set against the expected decline in emissions, presumably due to unusually poor dispersion of primary pollutants due to meteorological conditions during 1997. Backwards projections based on these two years therefore provide reasonable estimates of the range of mean values in earlier years. Measured concentration in 1998 were generally lower than would be expected due to the falling trend in emissions, again presumably due to the prevailing meteorological conditions which lead to unusually efficient dispersion of pollutants.

The backwards projections at roadside sites generally show reasonable agreement with the limited amount of monitoring data that is available. This confirms the conclusions in the review of the NAQS that many of the roadside sites included in the analysis are likely to be at risk of exceeding the 21 ppb objective in 2005.

## 8 Acknowledgements

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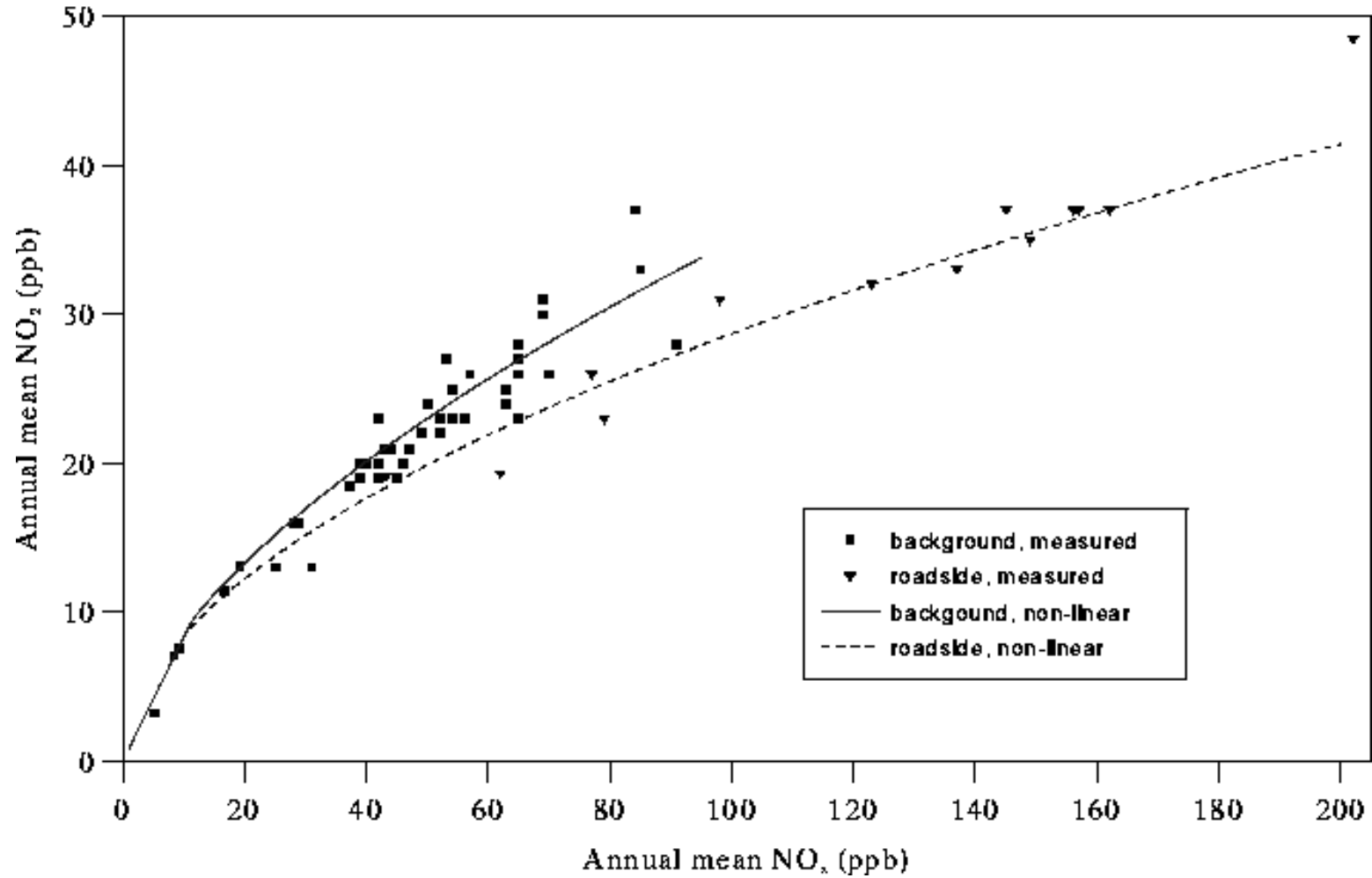
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Figure 1. The relationships between  $\text{NO}_x$  and  $\text{NO}_2$



# Appendices

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