Report

Comparison of health based metrics and modelling methods for ozone in the UK

The Department for Environment, Food and Rural Affairs, Welsh Assembly Government, The Scottish Executive and the Department of the Environment for Northern Ireland

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Executive Summary

This report details research undertaken as part of the 2006 Air Quality Strategy (AQS) review into the use of different ozone metrics to inform UK policy decisions. The health based metrics under examination are the annual mean of the daily maximum running 8-hour ozone concentrations with 0 ppb, 35 ppb and 50 ppb cut-offs.

The report includes:

- Identification and analysis of trends in each of the metrics at 7 long term monitoring sites in Defra's Automatic Urban and Rural Monitoring Network.
- Analysis of the relationships between the metrics using monitored data at each site type.
- Description and application of modelling techniques and comparison of model outputs and population-weighted means.

Time series charts indicated that background ozone levels in urban areas, which were declining until the 1990s due to increasing road traffic and NO_x scavenging, are increasing as a result of reduced NO_x titration associated with improving vehicle technology and more stringent engine emission controls. Trends in ozone concentrations at the rural sites were less readily identifiable – the 35 ppb cut-off metric showing a high degree of year to year variability. The 0 ppb metric has remained stable indicating little change in the influence of NO_x titration at rural locations and the 50 ppb cut-off metric has declined which shows a reduction in the impact of rural photochemistry of peak ozone levels.

Examination of the scatter plots of one metric to another indicated that the 35 ppb cut-off metric and the 50 ppb cut-off metric are more closely related to one another than the 35 and 0 ppb cut-off metrics, as both are sensitive to photochemical ozone episodes. The 0 ppb and 50 ppb cut-off metrics display no notable relationship as the 0 ppb cut-off metric best represents the influence of titration on background ozone levels and the 50 ppb cut-off metric best represents the influence of photochemical activity on peak ozone levels.

Two alternative modelling methodologies are described, an empirical method and **netcen**'s Ozone Source Receptor Model (OSRM), and have been implemented for each metric to provide map outputs for 2003 for comparison with each other. The outputs from these models have been verified against monitored data from the Automatic Urban and Rural Network (AURN). The empirical map outputs have a resolution of 1 x 1 km^2 and the OSRM maps have a resolution of 10 x 10 km^2.

The maximum modelled values from the empirical methodology were higher than the corresponding OSRM outputs. The empirical population-weighted mean results for the whole of the UK were 3% lower than the OSRM for the 0 ppb cut-off metric, 1% lower than the 35 ppb cut-off and 17% higher for the 50 ppb cut-off metric. At the 7 AURN monitoring sites for which the long term trends were analysed, empirical model values more closely resembled the monitored values for 2003 and the OSRM values less closely. This is as expected since the monitoring data has been used to calibrate the empirical models.

Both models appeared to perform better with the 0 ppb cut-off and not as well with the 35 and 50 ppb cut-offs. The 0 ppb cut-off metric is largely illustrative of background

hemispheric trends and local NO_x titration, while the other two metrics are more indicative of photochemical ozone generation.

Contents

1 Introduction

1.1 AQS REVIEW 2006

The UK Government and the Devolved Administrations are currently undertaking a review of the Air Quality Strategy. (Defra et al, 2006a, 2006b) This review will assess progress towards the achievement of the AQS objectives and assess the costs and benefits of possible additional measures to improve air quality in the UK. The focus of this review of possible measures will be on the impact of measures on concentrations of particles, nitrogen dioxide and ozone, the pollutants for which the achievement of the objectives is likely to be the most challenging. The AQS review requires the modelling of ozone concentrations for a number of possible additional measures scenarios and calculations of the associated health benefits. Ozone modelling has been carried out within the Tropospheric Ozone Modelling Contract (see Hayman et al, 2006) and the health benefit calculations are undertaken with in the Pollution Climate Mapping contract (see Defra, et al 2006c).

The cost-benefit analysis undertaken for the Clean Air for Europe programme (CAFE) is based on a new health-based ozone metric**:** effects of daily ozone on acute mortality will be quantified only at concentrations greater than 35ppb (maximum 8-hour mean). This requires the calculation of the annual mean of the daily maximum of running 8-hour mean concentration.

The quantification of the health effects of ozone within previous reviews of the AQS has followed the advice of COMEAP and ozone metrics with cut-off concentrations of 0 ppb and 50 ppb, but not 35 ppb, have been used. We do not currently have a very good understanding of the behaviour of these metrics in terms of historical measurements in rural and urban areas and OSRM model performance.

1.2 THIS REPORT

This work aims to provide Defra and DAs with an understanding of the behaviour of three different health effects related ozone metrics: the annual mean of the daily maximum of running 8-hour mean concentration, calculated with cut-off concentrations of 0, 35 and 50 ppb.

This report compares various the ozone metrics in 2003 as measured by Automatic Urban and Rural Network (AURN) monitoring sites; as modelled by the Ozone Source Receptor Model (OSRM) and by empirical modelling methods. The aims and objectives of the report are described below.
The study includes the following elements:

1) Subtask 1: An investigation of the temporal trends in these three ozone metrics in rural and, in particular, urban areas in the UK from the AURN monitoring data and a comparison of the behaviour of the metrics.
2) Subtask 2: The development of empirical mapping method to provide Defra and the DAs with maps of ozone concentrations in urban and rural areas for the metrics with cut-off concentrations of 35 and 50 ppb. Maps were calculated for 2003. An empirical method to map the metric with a 0

ppb cut-off is already available and has been used to calculate a map for 2003.

3) Subtask 3: A comparison of the automatic monitoring data results for these metrics with the empirical maps and available OSRM model results.

The metrics examined in this report are complex. The 0 ppb cut-off metric is determined by calculating the 8-hour running mean ozone concentrations and determining the maximum for each day of the year. The annual average of all these daily maximum values is then calculated. The 35 ppb and 50 ppb cut-off metrics are determined in the same manner as the 0 ppb cut-off metric but 35 or 50 ppb is subtracted from each daily maximum value. Any negative daily values resulting from this operation are converted to zeros and included in the calculation of the annual average of these values. (N.B. 1 ppb = 2 $\mu g\ m^{-3}$ ozone).

These studies are intended to provide Defra and the DAs with valuable background information to provide a context for the health benefit calculations to be carried out for ozone within the 2006 AQS review.

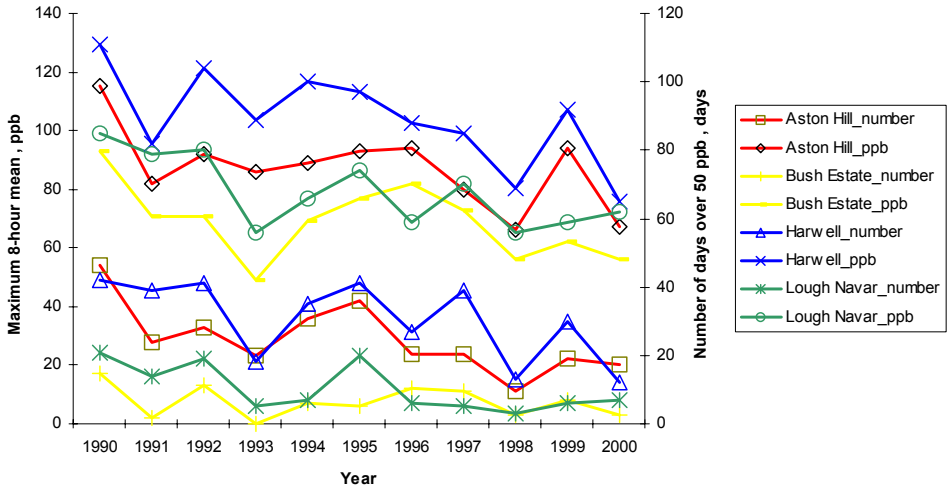
1.3 BACKGROUND TO OZONE

Ozone concentrations are determined by a complex series of inter-related factors including photochemistry, titration associated with NO_x and hemispheric background levels. Episodic peak ozone levels associated with photochemical action have declined over recent decades. The scavenging effect of NO_x titration has declined since about 1990 as improving technology has reduced road traffic emissions, resulting in increasing ozone levels in urban areas. Increasing hemispheric background levels of ozone have a more subtle impact on levels in the UK.

CAFE have chosen to express an ozone-related health indicator described as SOM 035. In contrast to the 35 ppb cut-off metric used here this indicator has units of $\mu g\ m^{-3}$ days (or ppb days) and is the sum of measured daily maximum 8-hour mean ozone over a year. This indicator is equivalent to the 35 ppb cut-off used here, but differs by a factor of 365. The CAFE indicator has the disadvantages that as a sum it needs to be corrected for data capture and the units are harder to interpret.

Prof. Derwent (pers. comm.) has presented data showing a downward trend in the maximum annual 8-hour running mean ozone concentration and the number of days when an 8-hour running mean ozone concentration exceeded 100 $\mu g\ m^{-3}$ at four UK ozone monitoring sites (see Figure 1.1). This decline has been attributed to reduction in the regional emissions of NO_x and VOCs.

Figure 1.1 The trend in the maximum annual 8-hour running mean and the number of days when an 8-hour running mean exceeded 100 μg m^{-3} at four UK ozone monitoring sites

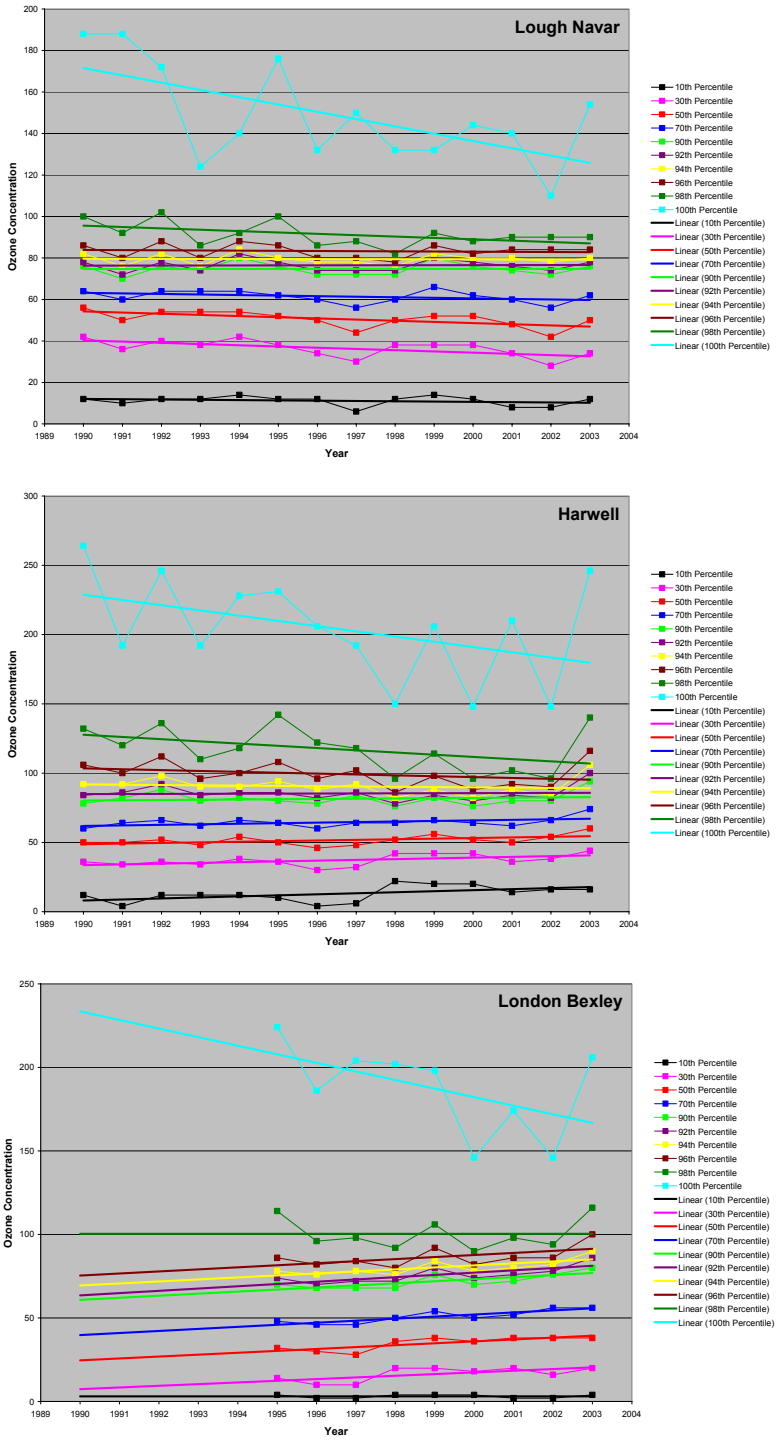


The temporal behaviour of different ozone concentration percentiles is shown in figure 1.2 for a number of UK ozone monitoring sites. In the figure, the change in the following percentiles - 10th, 30th, 50th, 70th, 90th, 92nd, 94th, 96th, 98th and maximum – are plotted for the years 1990 to 2003. At all sites, the peak hourly ozone concentration shows a downward trend. The figure also shows that downward trends are apparent down to the:

- 98th percentile: London Bexley
- 90th percentile: Harwell
- 30th percentile: Lough Navar

Stable or downward trends are seen in the lower percentiles at the rural sites in Figure 1.2 while the highest percentile is declining more sharply. However, while the downward trend in the maximum percentile is declining at London Bexley, the percentiles below the 98th percentile shows a much stronger upward trend. This illustrates the influence on ozone concentrations of NO_x controls in urban areas. The statistical significance of the trends has not been determined.

Figure 1.2 The trend in ozone concentration percentiles between 1990 and 2003 at 3 UK ozone monitoring sites



2 Analysis of monitoring data

2.1 INTRODUCTION

This section of the report aims to provide an investigation into the temporal trends in the three ozone metrics in rural and, in particular, in urban areas in the UK from the AURN monitoring data and a comparison of the behaviour of the metrics.

To assist in the understanding of the three previously described metrics, an investigation has been undertaken into the temporal trends in these three ozone metrics in rural and urban areas in the UK. Data for this comparison was available from Defra's Automatic Urban and Rural Network (AURN) sites and the following table (table 2.1) details the long running sites chosen. Data from long running AURN sites has been used to construct time series plots and scatter plots to investigate the temporal trends at the sites.

Table 2.1 Long running sites used to investigate temporal trends in the ozone metrics

Site Name	Site type	Date started
Belfast Centre	Urban Centre	08/03/1992
Birmingham Centre	Urban Centre	18/03/1992
Eskdalemuir	Rural	23/04/1986
Harwell	Rural	22/06/1976
London	Urban Background	(see below)
Lough Navar	Remote	02/04/1987
Sibton	Rural	01/07/1973

The London data set was compiled using data from 3 different but successive stations running in close proximity since 1973. This compilation produced a single continuous data set, shown in table 2.2.

Table 2.2 Sites used to compile the London data set

Site name	Operational period used in London data set
Central London	1973-1989
London Bridge Place	1990-1999
London Westminster	2001-2003

A data capture threshold of 75% was applied to the data prior to analysis. Table 2.2 indicates that the Central London data set is not continuous as it is comprised of data from 3 different monitoring stations (Central London, London Bridge Place and London Westminster) in close proximity to one another. These sites were operational over successive years and have been used in conjunction with one another to produce a data set that is almost complete between 1973 and 2003. The year 2000 is the only year for which no monitoring station was located in central London. The omission of an annual mean for 2001 was the result of low data capture (below 50%) in that year. By combining hourly automatic data sets from Central London and London Bridge Place, a 97.7% complete data set was produced with which to provide figures for 1990. The London time series chart

was produced using a data capture threshold of 50% to further reduce the number of gaps in the data set.

The following time series charts (figures 2.1 to 2.7) show the temporal trends in the three metrics at the various sites described in Table 2.1. Figures 2.8 to 2.13 are scatter plots illustrating the relationship between the metrics. Scatter plots are provided for the whole of the AURN data set from 1972 to 2003 and for the single year of 2003. Analysis of the long term trends and relationships between metrics is discussed in section 2.2, below.

2.2 TIME SERIES ANALYSIS

The influence of slight rises in hemispheric background ozone are not apparent in the 50 ppb cut-off metric at any of the sites for which long-term monitoring data are available. Trends in this metric are stable or show a slight decline. The decline is most noticeable at the sites with the longest runs of data. The highly publicised and widespread European heat wave and associated ozone episode of summer 2003 can be observed in each of the metrics (most prominently in the 35 ppb cut-off metric) at all sites.

Urban Centre sites
As Figures 2.1 and 2.2 show, trends in the 0 and 35 ppb cut-off metrics indicate a rise at urban centre locations. This is likely the result of declining titration associated with the reduction in ozone scavenging NO_x emissions from petrol-engined traffic due to the introduction of three-way exhaust gas catalysts and tighter regulations of diesel engine emissions. The 35 ppb cut-off at both Birmingham Centre and Belfast Centre appears to be rising but this metric is particularly variable rendering any meaningful trend very difficult to discern. Tighter controls on VOC emissions over the period, initiated to curb ozone levels, appear to have had very little influence in urban centre environments - the 50 ppb cut-off metric, which would most notably illustrate VOC related photochemical episodes, does not display a marked variation. This metric is most representative of trends in photochemical episodes. The stability of this metric, demonstrated by the time series, suggests that increasing controls of VOC emissions have been exceeded by the influence of improved technology on NO_x emissions in urban environments over the period since about 1990.

Central London sites
The time series for Central London using a 50% data capture threshold (Figure 2.5) is particularly erratic for each of the metrics. With the exception of 1985, the 35 and 50 ppb cut-off metrics show a decline in concentrations, particularly up to the 1990s, after which concentrations varied less. This might be attributed to controls on VOC emissions and the impact of their reduction on photochemical episodes – an effect that is most likely to be captured in the 50 ppb cut-off metric. However, no clear trend in this metric was observed at the other long term urban centre sites used in this analysis. This may be because the most significant declines in the more reactive VOC series took place prior to 1990. The 0 ppb cut-off trend appears to decline through the 1970s and 1980s before rising again through the 1990s until the present day. This corresponds with trends in NO_x concentrations and the policies used to control them. NO_x concentrations, associated with rising road traffic emissions, increased steadily until the late 1980s and would have reacted with and reduced ozone, particularly in heavily trafficked large urban areas such as London. Following the initiation of NO_x controls thereafter, this titration effect would have been notably reduced, resulting in increasing ozone concentrations. It is likely that this effect over the 1990s and beyond would have more than off set the decreases in ozone due to

VOC controls. This influence on titration would have an effect on all concentrations rather than just the highest episodic concentrations, hence it is notable in the 0 ppb cut-off metric rather than the 35 and 50 ppb cut-off metrics, which remove lower concentrations from the calculation.

Rural and Remote sites

Across the rural sites (Eskdalemuir, Harwell and Sibton), there is no obvious change in the 0 ppb cut-off metric, which remains relatively stable between 60 and 80 μg m^{-3}. The 35 ppb cut-off metric shows considerable year to year variation, particularly at the Eskdalemuir site. With the exception of 2003, during which there was a heat wave affecting much of Europe, the 35 ppb cut-off metric appears to be declining slightly at Harwell and Sibton. At all of these rural sites there is a slight decline in the 50 ppb cut-off metric, consistent with declining peak ozone concentrations across rural areas of Europe. The remote site, Lough Navar, exhibits a very slight downward trend in the 50 ppb and 0 ppb cut-off metrics. The 35 ppb cut-off metric, whilst much more variable, shows a more rapid decline.

Figure 2.1 Belfast Centre long term time series, 1992-2003, μg m^{-3}

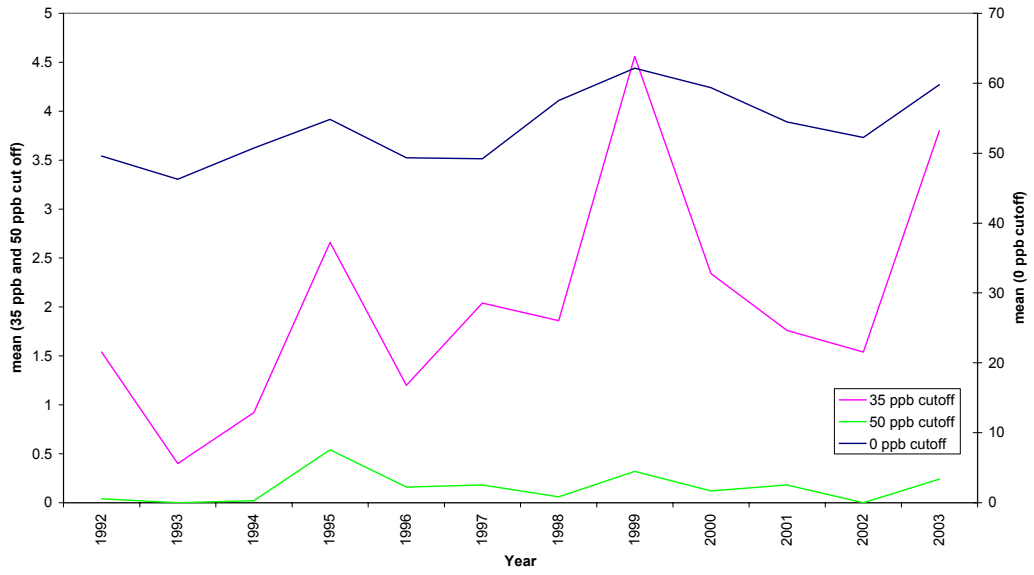


Figure 2.2 Birmingham Centre long term time series, 1993-2003, μg m^{-3}

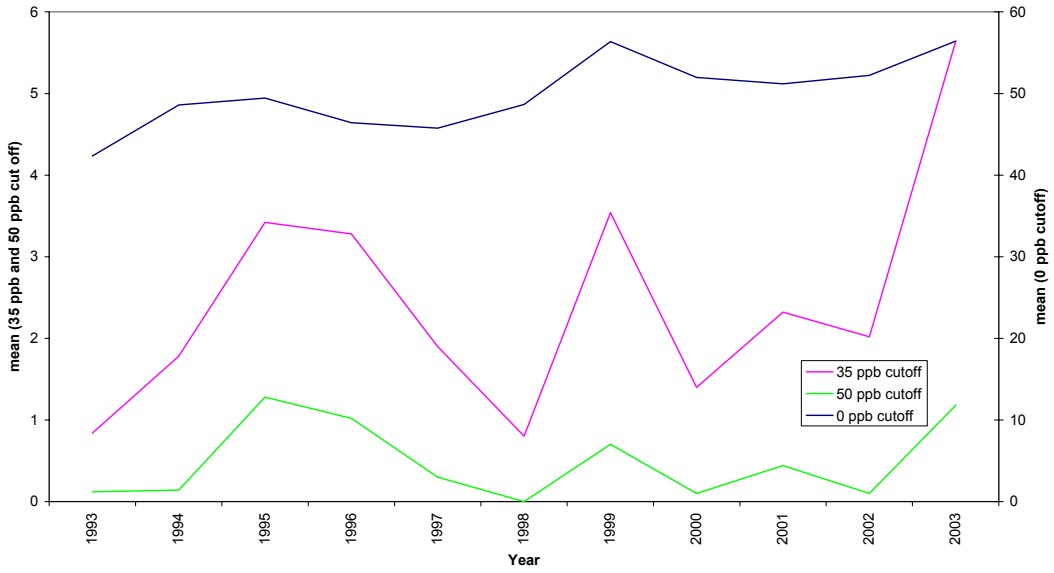


Figure 2.3 Eskdalemuir long term time series, 1987-2003, μg m^{-3}

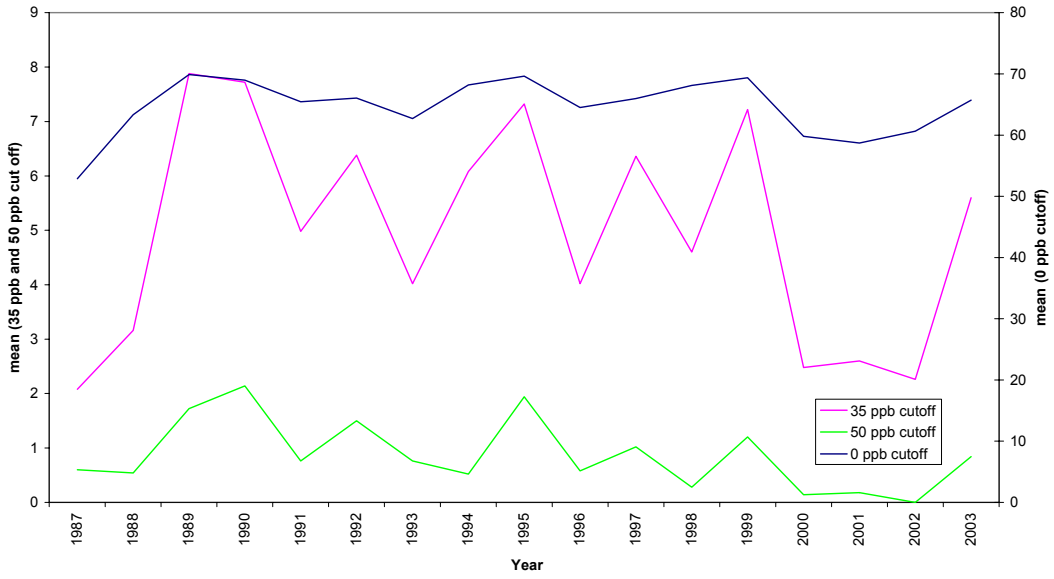


Figure 2.4 Harwell long term time series, 1984-2003, μg m^{-3}

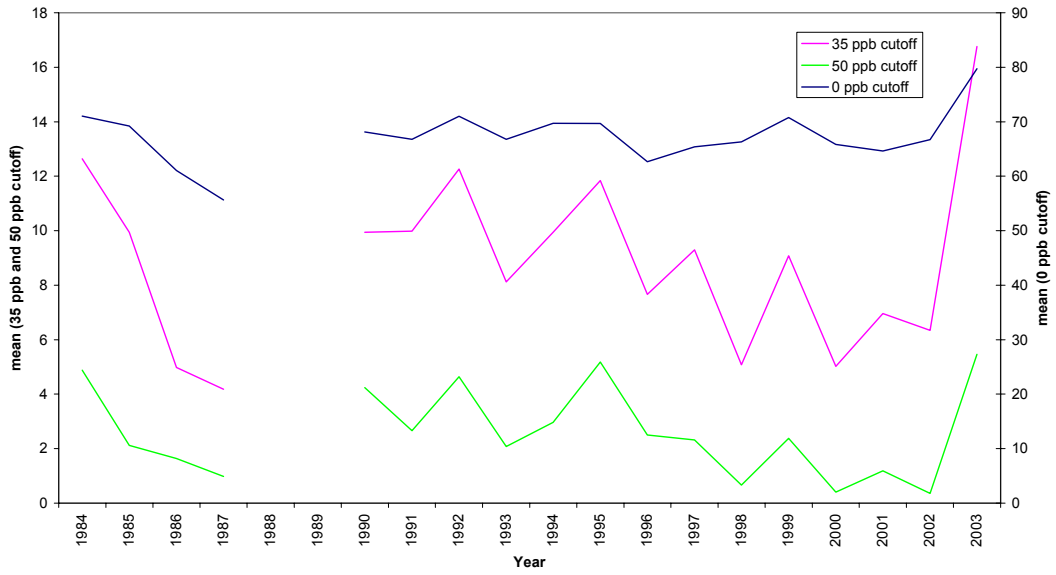


Figure 2.5 Central London long term time series (comprising Central London, London Bridge Place and London Westminster), 1972-2003, μg m^{-3} (using 50% data capture threshold)

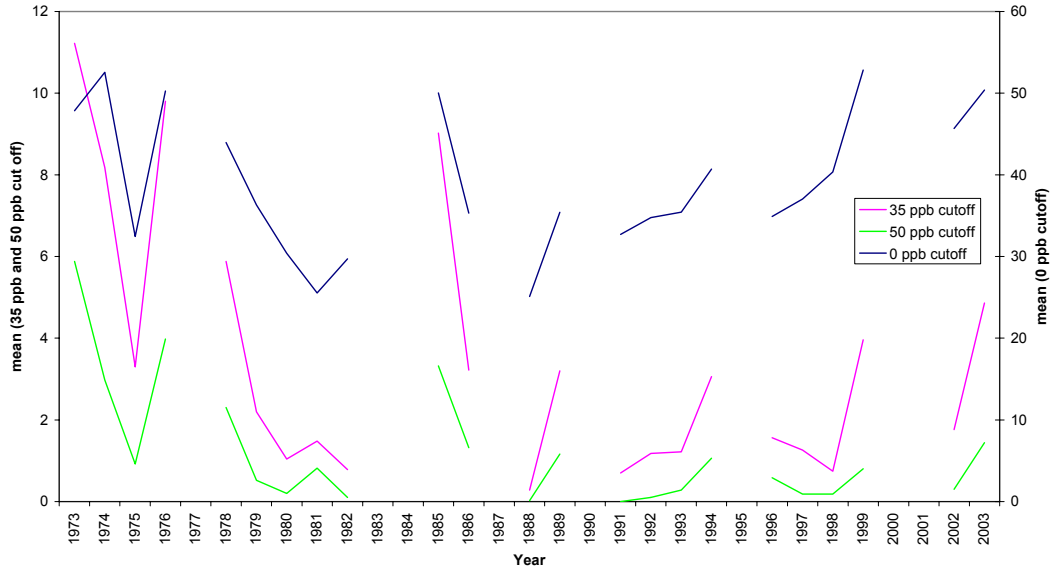


Figure 2.6 Lough Navar long term time series, 1988-2003, μg m^{-3}

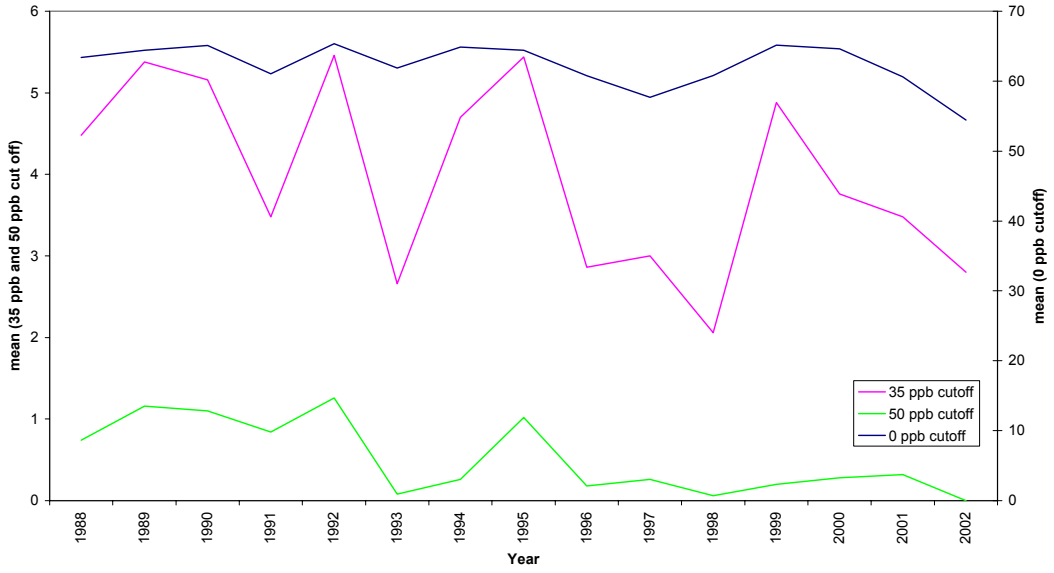
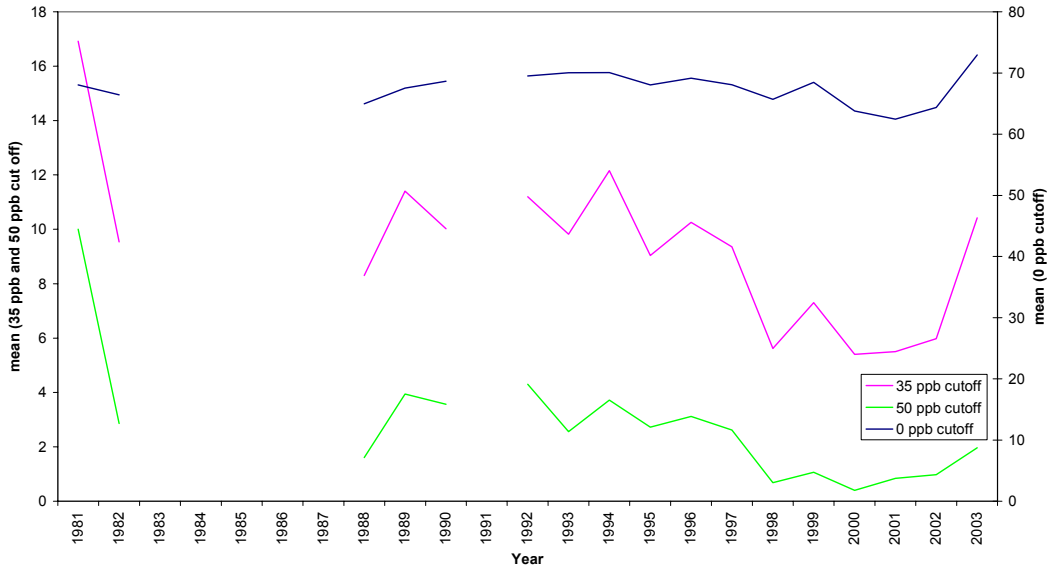


Figure 2.7 Sibton long term time series, 1981-2003, μg m^{-3}



2.3 SCATTER PLOTS

In Figure 2.8, the kerbside, roadside and urban centre observations range from 20-60 $\mu g\ m^{-3}$ for the 0 ppb cut-off metric and from 0-4 $\mu g\ m^{-3}$ for the 35 ppb cut-off metric, illustrating that this region is dominated by the effect of titration. The relationship between the two metrics for rural observations is steeper, showing the important influence of photochemical ozone at levels above 35 ppb. Figure 2.9 shows the same metrics as Figure 2.8 but only for the year 2003. The pattern in this chart is very similar to Figure 2.8, indicating that the trend seems to be spatial, with little significant temporal variation.

Figures 2.10 and 2.11 illustrate that there is little, if any, relationship between the 0 ppb cut-off and 50 ppb cut-off metrics. This is reasonable because the 0 ppb cut-off metric is driven by the influence of titration and the 50 ppb cut-off metric is driven by the influence of photochemistry. Given the influence of these different mechanisms on each metric it is likely that the response of these metrics to different emission reduction strategies would be quite different. The remote sites are clustered at high levels for the 0 ppb metric (60-90 $\mu g\ m^{-3}$) with very little variation in the 50 ppb cut-off metric (0-3 $\mu g\ m^{-3}$). The roadside observations are located at the lower range of both metrics. No clear relationship between these two metrics can be ascertained for the urban centre sites but the rural sites exhibit more of a relationship, indicating the influence of photochemistry on rural ozone levels. Figure 2.11 shows the impact that the 2003 photochemical even associated with a record heat wave had on the 50 ppb cut-off metric – levels of this metric were higher than many of the points in Figure 2.10, particularly at the urban centre sites.

Figure 2.12 shows that the 35 ppb and 50 ppb cut-off metrics are more closely related than the 0 ppb and 35 ppb cut-off metrics and that the nature of this relationship is closer to linear. The remote sites in Figure 2.12 show that there is little influence of photochemical ozone and little influence of NO_x titration. Therefore the trend may be the result slightly rising hemispheric background ozone levels. Compared with the remote sites, the rural sites in Figure 2.12 show a trend in which the 50 ppb cut-off metric rises at a greater rate with the 35 ppb cut-off metric. This shows the influence of more photochemical action and more titration at these sites whereas the remote sites are typically located further from NO_x sources and tend to be further from areas of high photochemical action. The trend at urban centre sites looks similar to the trend at rural sites but shows a steeper curve, indicating that the 50 ppb cut-off metric is higher at urban centre locations than rural locations at the same 35 ppb cut-off metric level. This is the result of more titration at these locations, restricting the 35 ppb cut-off metric to lower levels, and the result of more photochemical activity than at rural sites which causes higher levels of the 50 ppb cut-off metric. Figure 2.13 shows the linearity of these trends for 2003 only.

Figure 2.8 0 ppb cut-off against 35 ppb cut-off (μg m^{-3}) at all monitoring sites, categorised by site type, 1972-2003

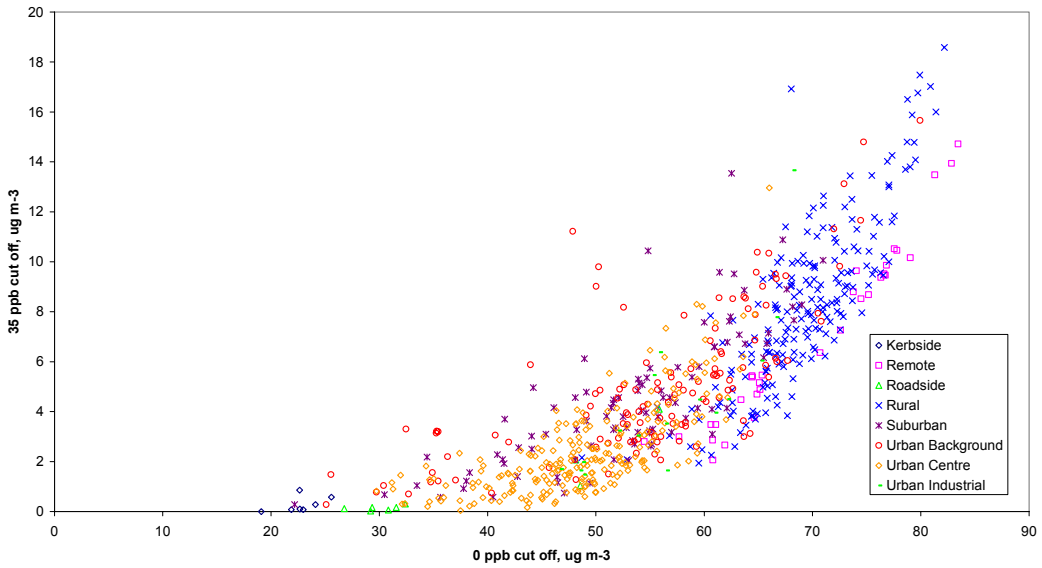


Figure 2.9 0 ppb cut-off against 35 ppb cut-off (μg m^{-3}) at all monitoring sites, categorised by site type, 2003

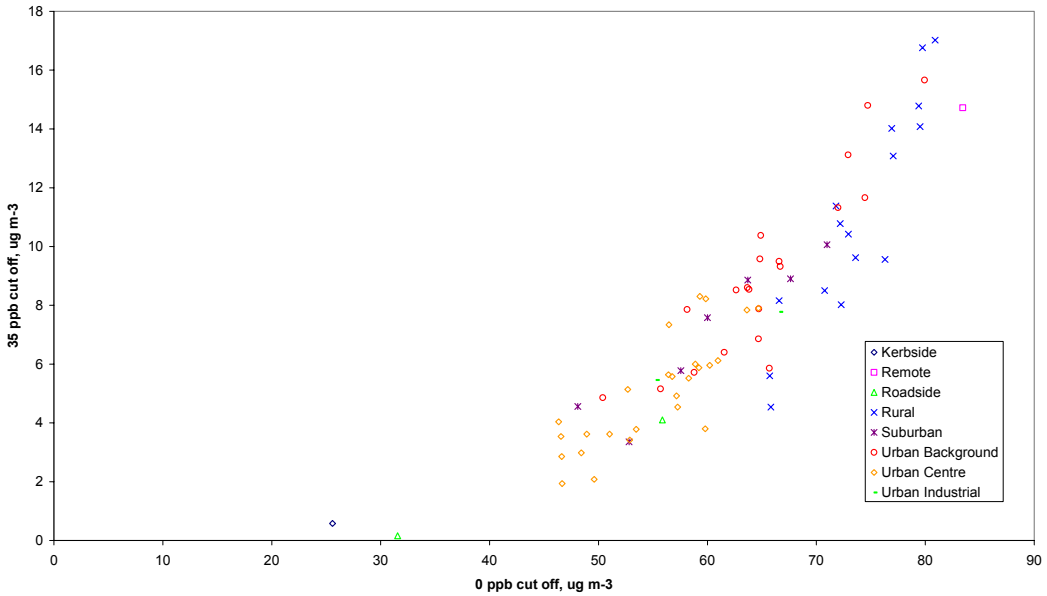


Figure 2.10 0 ppb cut-off against 50 ppb cut-off (μg m^{-3}) at all monitoring sites, categorised by site type, 1972-2003

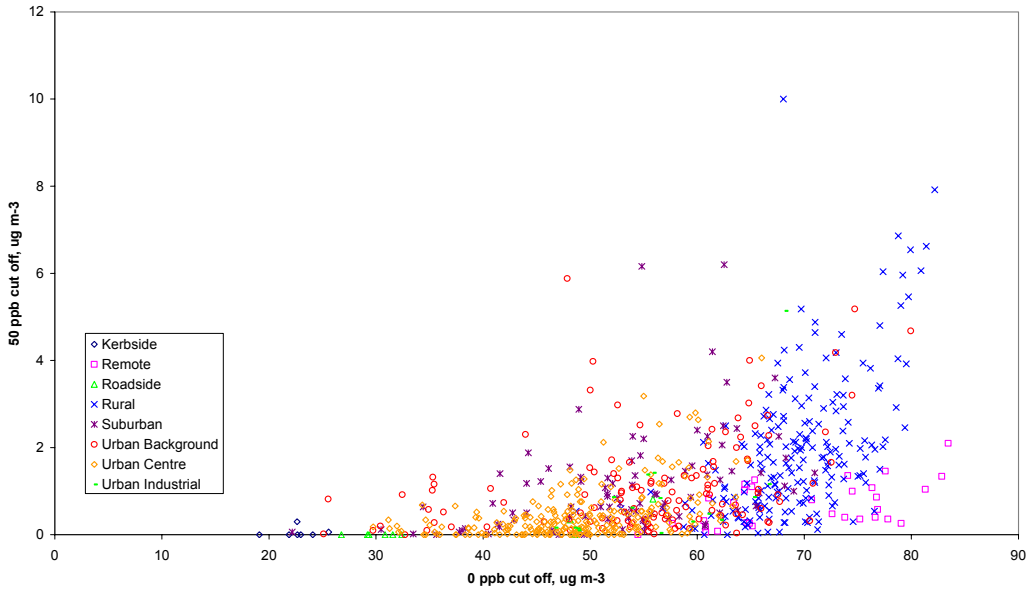


Figure 2.11 0 ppb cut-off against 50 ppb cut-off (μg m^{-3}) at all monitoring sites, categorised by site type, 2003

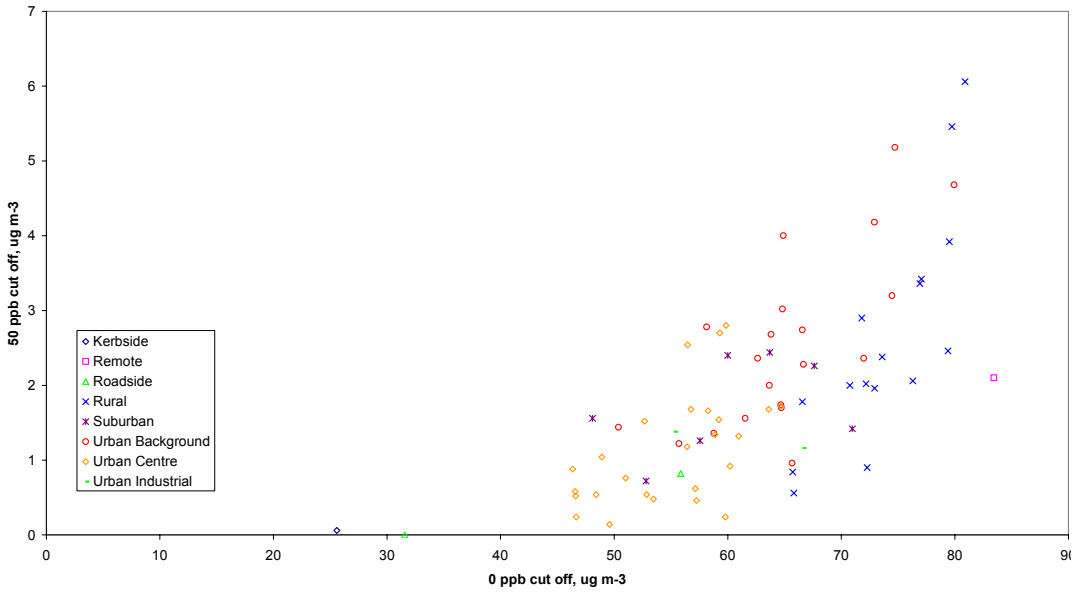


Figure 2.12 35 ppb cut-off against 50 ppb cut-off (μg m^{-3}) at all monitoring sites, categorised by site type, 1972-2003

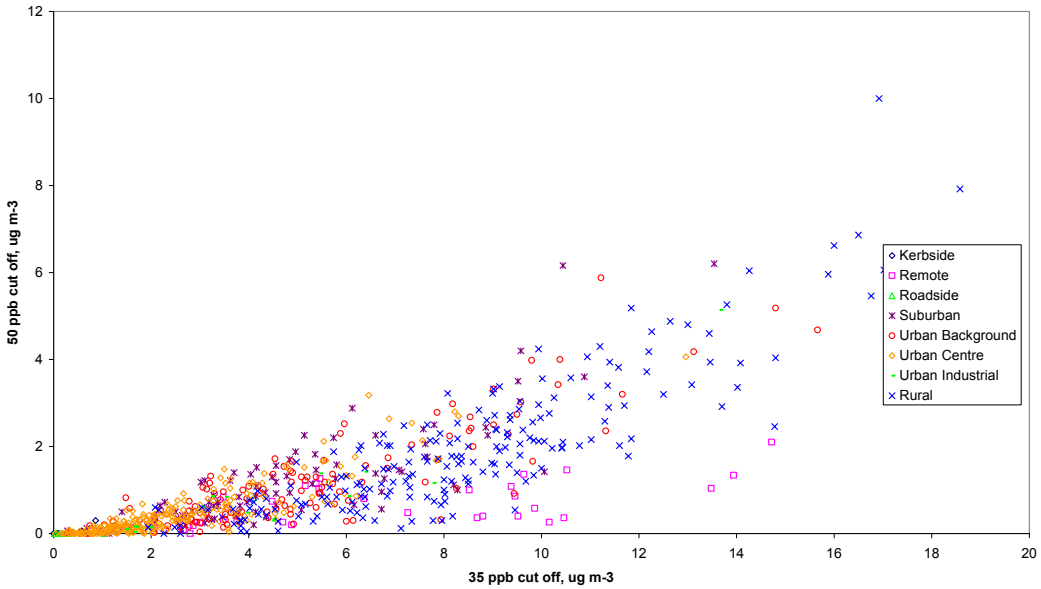
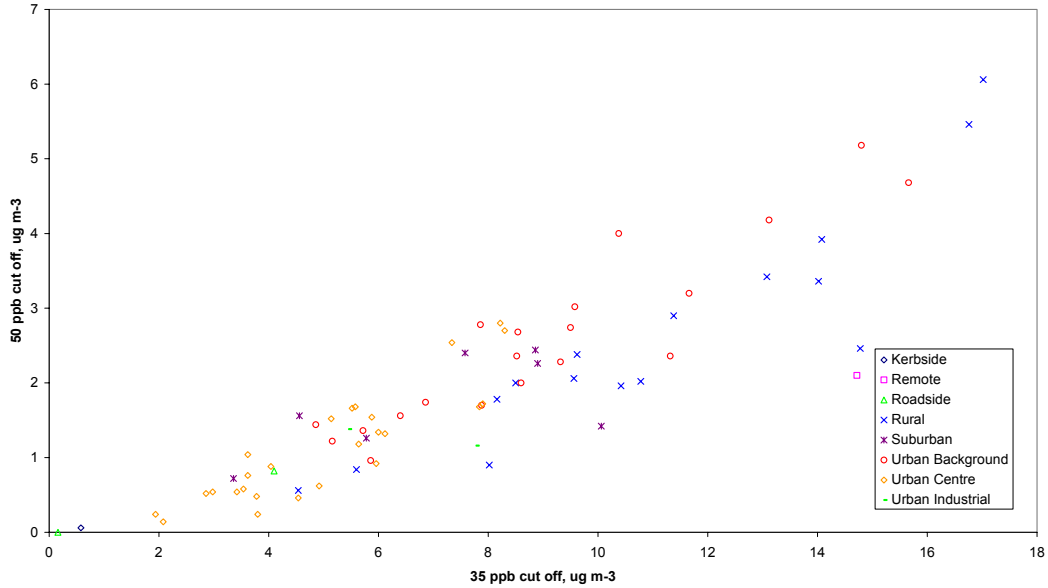


Figure 2.13 35 ppb cut-off against 50 ppb cut-off (μg m^{-3}) at all monitoring sites, categorised by site type, 2003



3 Mapping Method

3.1 INTRODUCTION

The empirical methods for modelling the 35 and 50 ppb cut-off metrics are similar and are described in section 3.2. Calculation of the 0 ppb cut-off statistic follows a very different methodology and is explained in section 3.3. The Ozone Source Receptor model used to map these metrics is introduced in section 3.4.

3.2 EMPIRICAL MAPPING METHODOLOGY FOR 35 PPB AND 50 PPB CUT-OFF METRIC MAPS

This section of the report explains the methodology used to generate maps of ozone in both the rural and urban areas for the metrics with cut-offs at 35 and 50 ppb respectively. Maps have been calculated for 2003.

Data from the national monitoring networks has been used to compile the annual mean of the daily maximum running 8-hour mean ozone concentration with 0, 35 and 50 ppb cut-off for 2003 for all sites with a data capture greater than 75%.

This data set contained ozone data for 72 sites across the UK. Using only the data from the rural sites, a bilinear interpolation method was used to generate 2 5 km resolution maps of ozone concentrations across all of the UK – one for the 35 ppb metric and one for the 50 ppb metric.

The corresponding interpolated ozone measurement at each of the 72 ozone sites was then found using the new map created for each of the metrics.

This map interpolated from the rural sites does not take into account the effects of urban areas and so it cannot be used as a representative map for the whole of the UK. To produce the final empirical map, the urban influence must be taken into account.

The urban influence term (UI) is calculated using the following equation (PORG 1997):

UI =
(interpolated rural measurement – measured) / interpolated rural measurement

This was calculated for each of the sites and a corresponding NO_x concentration for 2003 was also found from a map of NO_x concentrations calculated for 2003 (Stedman *et al* 2005). These 2 datasets were used to generate a graph of UI against NO_x concentration for both the 35 and 50 ppb metrics and are shown on figures 3.1 and 3.2.

Figure 3.1 NO_x vs UI 35 ppb cut-off metric

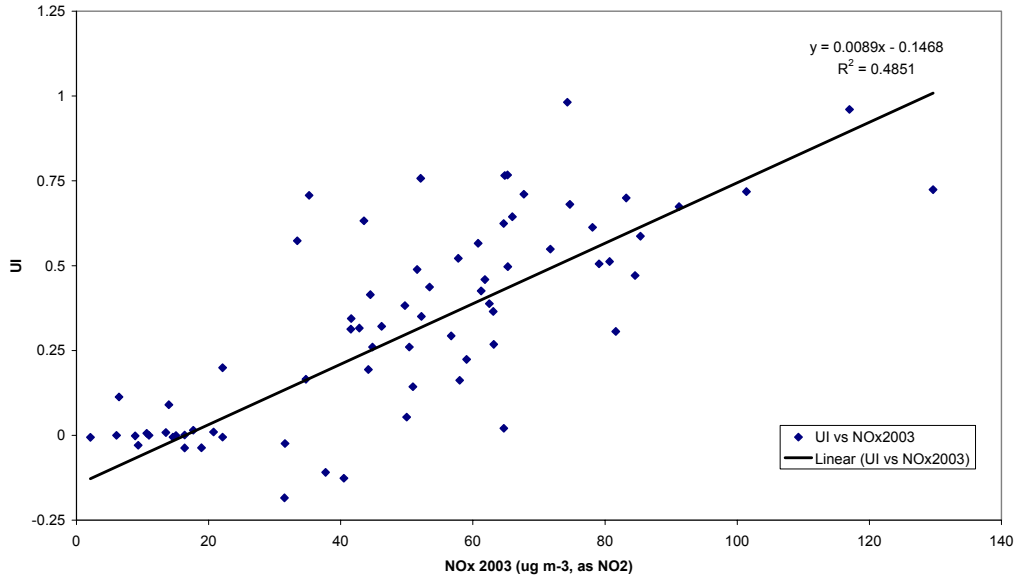
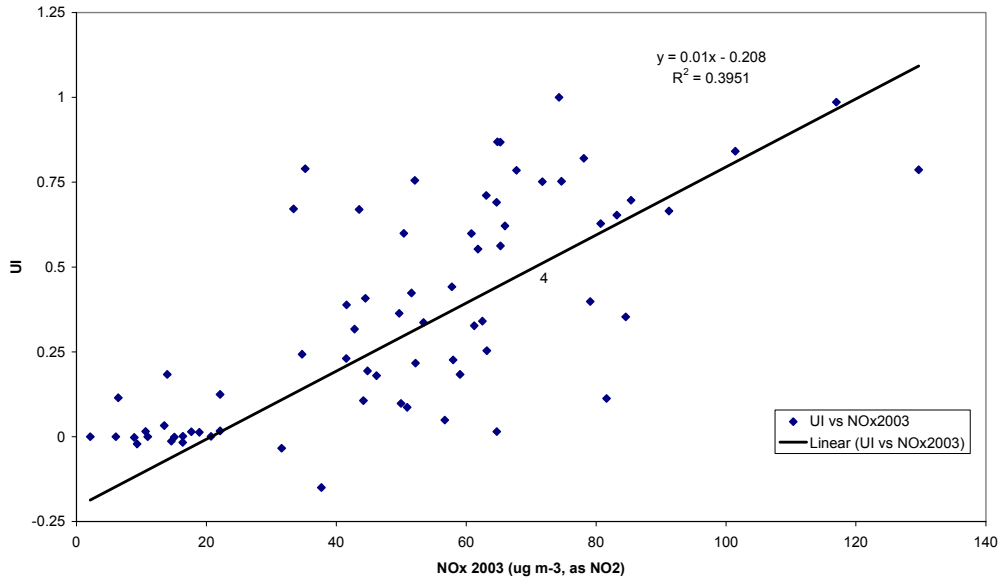


Figure 3.2 NO_x vs UI 50 ppb cut-off metric



The gradient from the graphs can be used in further calculations to determine the Urban Influence (UI). The intercept from the graph represents the NO_x concentration at which there is effectively no further UI, i.e. for figure 3.1, when the NO_x is 16.5 $\mu g\ m^{-3}$, the UI is 0. This value is subtracted from all the NO_x concentrations to create a corrected NO_x concentration and used in the following UI calculation.

UI = corrected NO_x * (gradient from graph)

Finally, this value is used to create the empirical maps for the metrics using the original interpolated rural map:

Resultant empirical map = interpolated rural map * (1 – UI)

This resultant map has been corrected for the influence of urban areas and as such is more reliable than simply interpolating rural measurements to obtain a UK wide map. The maps are shown and discussed in the next section.

3.3 EMPIRICAL MAPPING METHODOLOGY FOR 0 PPB CUT-OFF METRIC MAPS

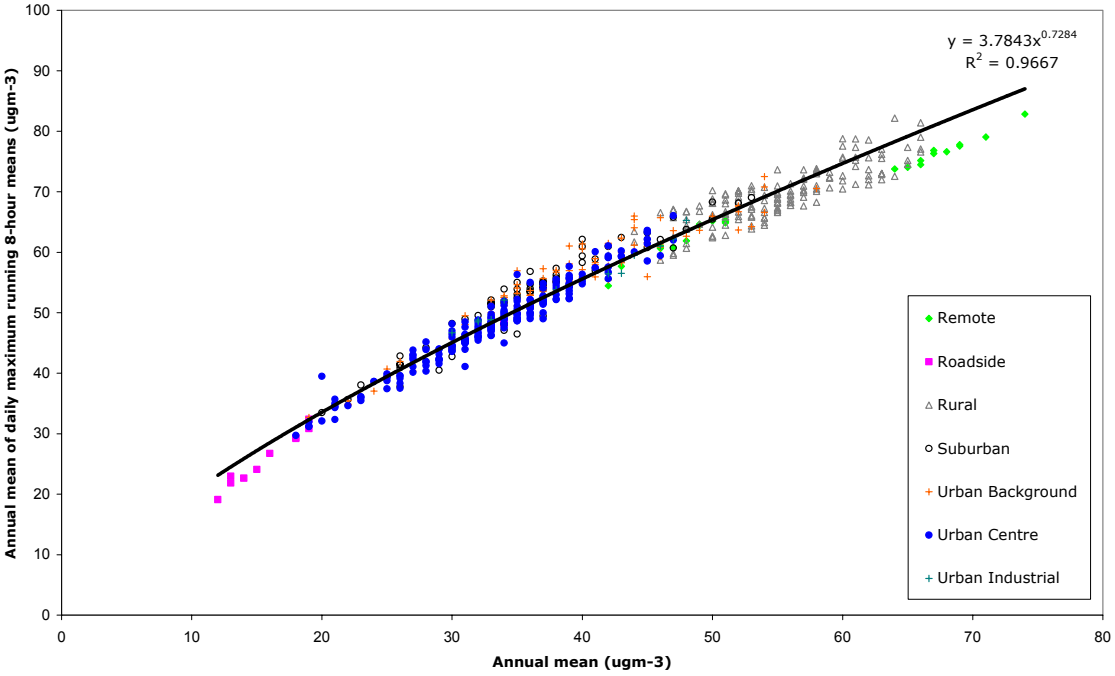
The maps of the annual mean of maximum daily 8-hour running means with a 35 or 50 ppb cut-off have been calculated using an urban influence term (UI) to describe the effect of NO_x upon the these metrics. For the annual mean of maximum daily 8-hour running means with 0 ppb cut-off we have the advantage of applying our understanding of ozone chemistry to estimate mean ozone in urban areas. Subsequently, the annual mean of maximum daily 8-hour running means with 0 ppb cut-off can be calculated using the empirical relationship between the annual mean of maximum daily 8-hour running mean metric and the annual mean (see Figure 3.3 below).

Previous work has used the oxidant partitioning model to predict NO_2 as a fraction of total oxidant using a function of NO_2 (Jenkins, 2004 and Stedman *et al*, 2005). By manipulating the terms within the oxidant partitioning model it can also be used to predict ozone as a fraction of total oxidant. In this case total oxidant is described as regional oxidant derived from rural mean ozone and interpolated NO_2 measurements, and a local oxidant component derived as a function of modelled NO_x concentrations (Stedman *et al*, 2005). Annual mean ozone is subsequently calculated as the product of total oxidant and manipulated oxidant partitioning model.

The rural ozone field described above has been calculated by interpolating the annual mean ozone concentrations for the well mixed period of the day (1200 to 1800 hours). Subsequently, a correction to account for the diurnal variation in ozone and thereby converting the mapped annual mean "well mixed" to a daily annual mean is applied (Coyle *et al*, 2002). The correction presented below, $\Delta O_{3,}$ is subtracted from the well mixed data and incorporates an altitude term (m):

$$\Delta O3 = 1.107 + 5.6 \times e^{(altitude \times -4.45 \times 10^{-3})} \text{ [ppb]}$$

Figure 3.3 The relationship between measured annual mean of maximum daily 8-hour running means and annual mean ozone concentrations 1992- 2002



3.4 OZONE SOURCE RECEPTOR MODEL (OSRM)

The Ozone Source Receptor Model (OSRM) has also been used to generate UK scale maps for the 3 ozone metrics. A brief description of the OSRM is provided here and the corresponding OSRM maps are presented alongside the empirically modelled maps in section 3.3. The information presented serves as an introduction to the model. More detailed description of the model can be found in Hayman *et al.*, 2002; 2004; 2005.

The OSRM is a recently developed model to describe photochemical ozone production in the UK [Hayman *et al.*, 2002, 2004; 2005]. The OSRM covers the EMEP model domain and uses global meteorological datasets provided by the Met Office to derive 96-hour back trajectories to specified receptor sites (UK/EMEP monitoring sites or a 10km x 10km grid covering the UK). The chemical scheme is based on that used in the STOCHEM model [Collins *et al.,* 1997; 2000; Steveson *et al.,* 1997]. The mechanism has ~70 chemical species involved in ~180 thermal and photochemical reactions. The mechanism represents ozone formation using 10 VOCs, which provides an appropriate description of ozone formation on the regional scale. The emission inventories are taken from EMEP for Europe with the option to use NAEI emission inventories for the UK, which have been aggregated to 10 km x 10 km and into 8 key sectors.

The OSRM is similar in concept to the UK Photochemical Trajectory Model (UK PTM) [Derwent *et al.*, 1998, 2004] in that it simulates the chemical development of species in an air parcel moving along a trajectory and to the ELMO source-

receptor model [Metcalfe *et al.*, 2002] in that calculations can be undertaken to a 10 km x 10 km grid covering the UK. The OSRM has a number of notable enhancements and advantages to these models:

- **Air Mass Trajectories**: Realistic air mass trajectories are derived from wind fields extracted from meteorological datasets. The UK PTM and ELMO model use linear trajectories. Meteorological datasets are available for use with the OSRM for the years 1995 to 2003;
- **Meteorology**: The boundary layer depth and other meteorological parameters characterising the boundary layer are interpolated in space and time from the input meteorological datasets;
- **Chemical Mechanisms**: The chemical mechanism used in the OSRM is an updated and extended version of chemical mechanism used in the ELMO or STOCHEM models. The chemical mechanism has been modified to include the formation of HONO and organic nitrates and a more extensive chemistry of NO_3
- **Photolysis Rates**: Photolysis rates have been calculated off line using a modified version of the PHOTOL code. The input database contains the dependence of photolysis rates for 17 species on zenith angle, cloud cover, land surface type and column ozone;
- **Emissions:** The model uses up-to-date emission inventories for nitrogen oxides, volatile organic compounds, carbon monoxide and sulphur dioxide taken from UK (National Atmospheric Emission Inventory) and European (EMEP) sources. The emissions of each pollutant have been divided into to 8 broad source categories (solvent usage, road transport, industrial processes, power generation, fossil fuel extraction and delivery, domestic combustion, natural and other). The assignment of the ~600 VOCs in the UK speciated VOC emission inventory to the 13 model VOCs was based on reactivity and structural considerations.
- **Temporal Emission Factors**: The OSRM converts the annual emission estimates to instantaneous emission rates using temporal profiles for the emissions of NO_x, VOCs, SO_2 and CO generated by Jenkin *et al*. [2000]. These profiles were derived either from real activity data or by using one of small set of default profiles.
- **Biogenic VOC Emissions:** An additional emission term is added to the emission rate of isoprene to represent the natural biogenic emissions from European forests and agricultural crops. The emission estimates can either be the same as those used in the UK PTM and taken from Simpson *et al.* [1995] or the new biogenic inventory produced using the PELCOM land cover dataset and the TNO tree species inventory. The later emission inventory gives emission potentials aggregated to the EMEP 50 km x 50 km grid for isoprene (from deciduous and evergreen trees: temperature and light[1] sensitive), monoterpenes (from deciduous and evergreen trees: temperature or temperature and light sensitive) and other VOCs (OVOCs, from deciduous and evergreen trees: temperature sensitive);
- **Dry Deposition**: Dry deposition processes are represented using a conventional resistance approach, in which the rate of dry deposition is characterised by a deposition velocity. Different deposition velocities are used over land and sea. The ozone deposition velocity over land has an imposed diurnal and seasonal cycle.

[1] This is the photosynthetically active radiation, typically about 45-50% of total global radiation, covering the wavelength range 400-700nm.

- **Initialisation**: The concentrations of O_3, CO, CH_4, C_2H_6, HNO_3 and PAN are initialised on each OSRM trajectory using output from the global tropospheric STOCHEM model. This improves the realism of the model as the seasonal cycle in ozone is now represented. It also allows for coupling between regional scale ozone production and the hemispheric circulation. A full set of daily concentration fields are currently available for 2 calendar years, one representing the climatology of the late 1990's (actually 1998) and the second a future atmosphere (IPCC SRES scenarios for 2030). This will allow model runs to be undertaken to assess the effect of climate change on regional ozone concentrations.

The OSRM describes the boundary layer by a single box and assumes that this is well mixed. While this is a reasonable assumption for rural areas, it is less valid in urban areas with high NO_x emissions. In such areas, there will inevitably be a gradient in the NO_x concentration profile, with higher concentrations at the surface. This will lead to lower ozone concentrations at the surface through reaction with nitric oxide.

An algorithm has been developed and implemented in the OSRM post-processor to convert the mid-boundary layer concentrations to surface concentrations on a hour-by-hour basis. This algorithm uses the local meteorological parameters characterising the boundary layer, surface roughness appropriate for the surface types considered, resistance parameters for O_3 and NO_2, the local NO_x emission rates and a simple NO-NO_2-O_3 photostationary state chemistry.

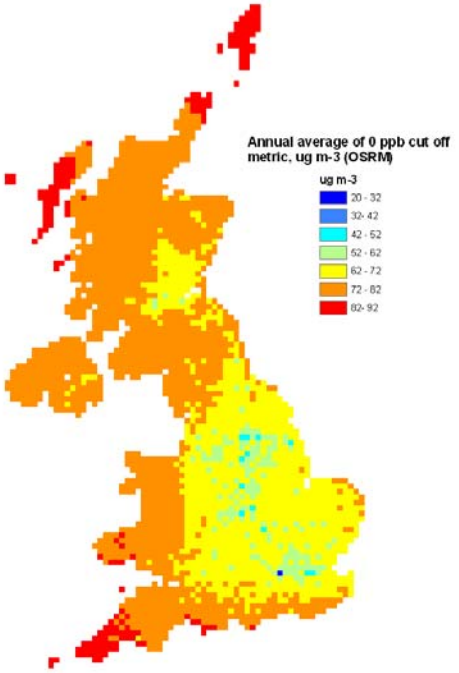
The use of this algorithm led to a significant improvement in the performance of the OSRM compared to measurements, particularly for the London and other urban sites. The surface-correction algorithm was used in the processing of all the subsequent OSRM output.

In this analysis, the OSRM has been run for the 0, 35 and 50 ppb cut-off metrics for 2003.

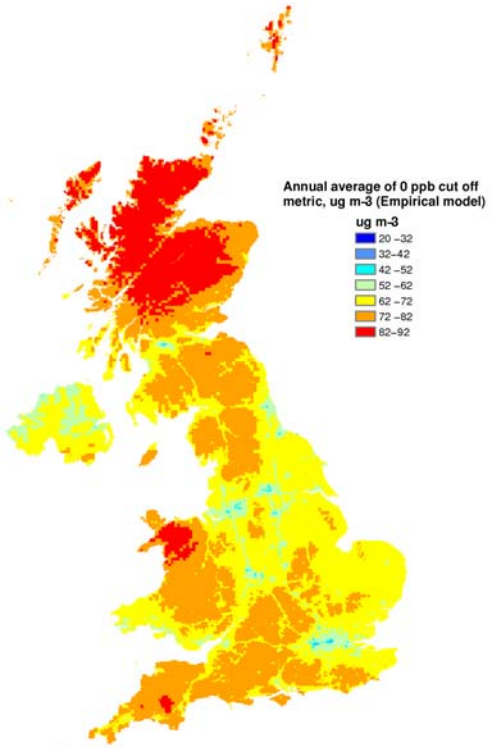
3.5 DISCUSSION

The following figures (Figures 3.4 to 3.6) show the empirical maps and OSRM maps created for the metrics with 0, 35 and 50 ppb cut-offs applied.

Figure 3.4 2003 Annual mean of daily maximum running 8-hour mean ozone with no cut-off applied (0 ppb), 2003

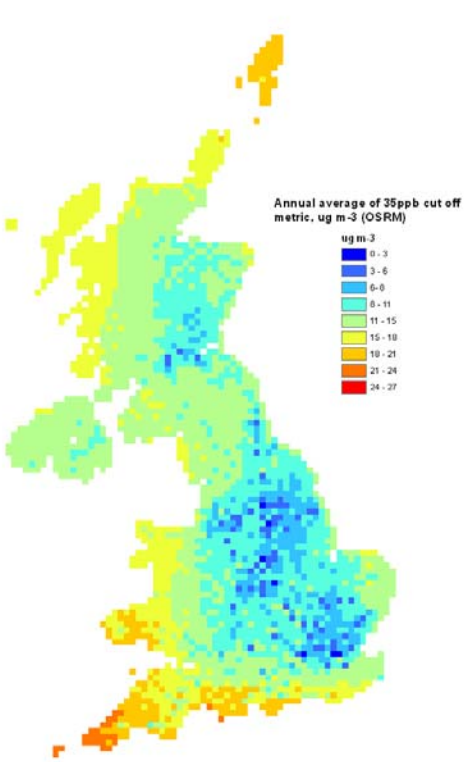


OSRM map

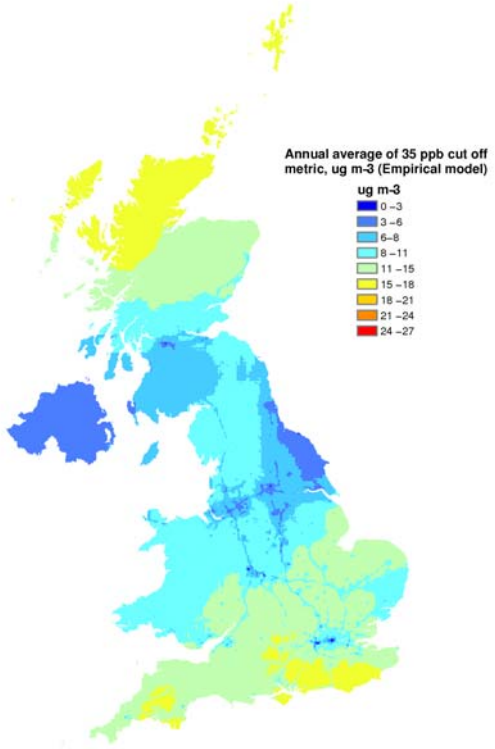


Empirical

Figure 3.5 Annual mean of daily maximum running 8-hour mean ozone with 35 ppb cut-off applied, 2003

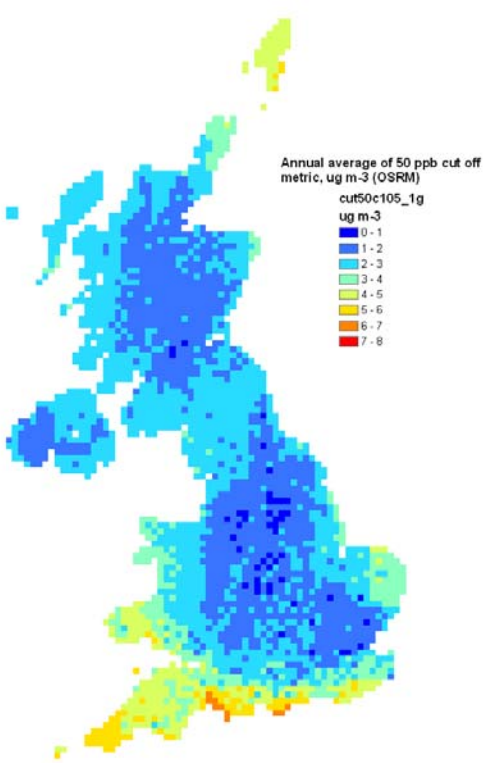


OSRM map

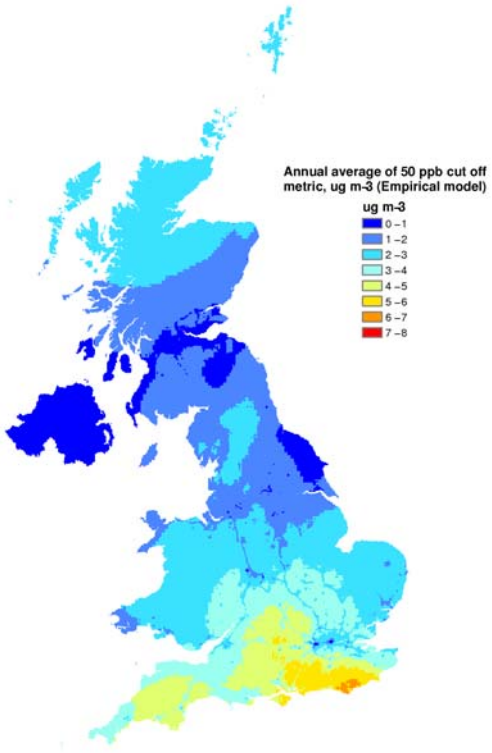


Empirical

Figure 3.6 Annual mean of daily maximum running 8-hour mean ozone with 50 ppb cut-off applied, 2003



OSRM map



Empirical map

In general, the empirical and OSRM maps show a similar distribution of ozone concentrations across the UK, although there are some differences.

For all three metrics, the empirical and the OSRM maps identify urban areas and major roads or motorways as ozone depleted areas. The OSRM map does not present ozone depletion in smaller roads or urban areas as well as the empirical maps due to the coarser spatial resolution of the OSRM maps (10 x 10 km^2 compared with 1 x 1 km^2 for the empirical maps).

Differences common to all three metrics include modelled concentrations in Northern Ireland where OSRM consistently predicts higher values than the empirical model. Additionally, for all three metrics, OSRM maps show generally higher concentrations in island areas (e.g. Outer Hebrides, Shetland, Isles of Scilly). These elevated values may be genuine, but may have been compounded by an artefact of the model in the way it addresses grid squares containing little land mass. If genuine, these high concentrations of ozone could be caused by the less significant ozone loss mechanisms to the sea compared with the land. The influence of very clean south westerly Atlantic air masses which, in the absence of primary NO_x pollution would best illustrate the effect of rising hemispheric background levels of ozone, could cause higher ozone concentrations in these locations.

The 0 ppb empirical map shows a slight east to west gradient with the highest modelled ozone concentrations occurring further west, particularly the rural areas of Dartmoor, North Wales and the Scottish Highlands. The OSRM map for this metric also shows a slight east to west gradient with highest concentrations in Cornwall and some of the Scottish islands. However, in contrast to the empirical map, the upland areas of Dartmoor, North Wales and the Scottish Highlands are not identified as having particularly high concentrations on the OSRM map. This may be because OSRM does not include an altitude correction.

The 0 ppb cut-off is the metric which for both models shows the clearest distinction between ozone in urban and rural areas. This is due to the nature of the metric – because there is no cut-off, the influence of NO_x titration on levels all year round is most apparent in this metric so there is a marked difference between high and low NO_x areas.

The 35 ppb cut-off metric empirical map shows most variability from north to south, with high concentrations in the northernmost parts of Scotland and in Southern England and lower concentrations in between. The OSRM map also has higher concentrations in northern Scotland and southern England, but additionally shows more pronounced east-west variability than the empirical map, with higher concentrations generally found in the western UK.

The empirical map for the 50 ppb cut-off metric exhibits a more pronounced north to south variability than the 35 ppb cut-off metric empirical map. The higher concentrations occur in southern and central southern regions and drop with increasing latitude until the Scottish Highlands where levels rise again slightly. This rise in Scotland is due to the rural Strath Vaich monitoring station which typically sees high ozone levels and from which data was used in the initial interpolation (see section 3.2). The OSRM map also identifies southern England as containing high concentrations of ozone for this metric. However, it differs from the empirical map in the extent of this south coast area of higher ozone, which does not extend as far inland as on the empirical map. It also identifies Pembrokeshire, and to a lesser extent Norfolk and the west coast of Wales as areas of high ozone, which are not identified on the empirical map.

The main influences on the 0 ppb cut-off map are from NO_x titration and background ozone levels. The most important influence on the 50 ppb cut-off map is photochemical

ozone episodes, which lead to the highest concentrations in the south. The 35 ppb cut-off map is influenced by all three. Thus, the empirical map for this metric shows highest concentrations in the south due to photochemical episodes and in the north of Scotland due to background ozone levels and little NO_x titration.

4 Model Comparisons

4.1 COMPARISON WITH MONITORED DATA

Results from both the empirical and OSRM modelling have been compared with monitored data at the 7 long-term sites in the UK's national AURN for which the time series analysis was performed in section 2. These are presented in Tables 4.1 to 4.3, which show the 2003 monitored and modelled values.

Table 4.1 Measured data, empirical and OSRM outputs at 7 AURN long-term monitoring sites for the 0 ppb cut-off metric (μg m^{-3})

Site	Monitored value	Empirical map value	OSRM map value
Belfast Centre	59.8	50.2	71.4
Birmingham Centre	56.4	50.5	60.9
Eskdalemuir	65.7	73.5	74.9
Harwell	79.7	73.5	66.0
London (Westminster)	50.4	47.8	58.6
Lough Navar	64.9	62.8	77.5
Sibton	72.9	69.1	69.5

Table 4.2 Measured data, empirical and OSRM outputs at 7 AURN long-term monitoring sites for the 35 ppb cut-off metric (μg m^{-3})

Site	Monitored value	Empirical map value	OSRM map value
Belfast Centre	3.8	3.6	10.2
Birmingham Centre	5.6	5.0	6.9
Eskdalemuir	5.6	6.9	12.7
Harwell	16.8	14.2	9.4
London (Westminster)	4.9	5.0	6.8
Lough Navar	4.3	4.7	13.1
Sibton	10.4	10.5	11.9

Table 4.3 Measured data, empirical and OSRM outputs at 7 AURN long-term monitoring sites for the 50 ppb cut-off metric (μg m^{-3})

Site	Monitored value	Empirical map value	OSRM map value
Belfast Centre	0.24	0.42	1.77

Birmingham Centre	1.18	1.27	1.30
Eskdalemuir	0.84	1.09	2.28
Harwell	5.46	4.71	1.80
London (Westminster)	1.44	1.27	1.47
Lough Navar	0.36	0.41	2.05
Sibton	1.96	2.07	3.03

Tables 4.1 to 4.3 shows the modelled data derived from the empirical method and the OSRM next to the corresponding measured value at each site. The difference between the modelled outputs and the measured value is shown as a percentage of the measured data in Table 4.4.

Table 4.4 Summary of the percentage differences between measured and modelled data at 7 AURN long-term monitoring sites

Site	Empirical			OSRM		
	0 ppb cut-off	**35 ppb cut-off**	**50 ppb cut-off**	**0 ppb cut-off**	**35 ppb cut-off**	**50 ppb cut-off**
Belfast Centre	-16.0	-5.6	75.8	19.3	168.7	637.5
Birmingham Centre	-10.5	-12.1	7.8	7.9	22.0	10.2
Eskdalemuir	11.9	22.9	29.2	14.0	127.3	171.4
Harwell	-7.8	-15.5	-13.7	-17.2	-44.0	-67.0
London (Westminster)	-5.2	2.8	-11.6	16.4	40.7	2.1
Lough Navar	-3.3	9.3	14.7	19.4	200.9	469.4
Sibton	-5.3	1.2	5.6	-4.7	13.9	54.6
Average	**-5.2**	**0.4**	**15.4**	**7.9**	**75.6**	**182.6**

Table 4.4 indicates that the empirical model compares quite favourably with the monitored data. Averaging the model's performance across all 7 sites, it under predicts the 0 ppb cut-off metric measured values by 5%, over predicts the 35 ppb cut-off metric by less than 1% and over predicts the 50 ppb cut-off metric by 15%. This is as expected since the monitoring data has been used to calibrate the empirical models. The equivalent OSRM performance across all 7 sites was an over prediction of 8% for the 0 ppb cut-off metric, an over prediction of 76% for the 35 ppb cut-off metric and an over prediction of 183% for the 50 ppb cut-off metric. The over prediction in the 50 ppb cut-off metric was dominated by several large percentage differences at Belfast Centre, Eskdalemuir and Lough Navar. It should also be noted that the smaller values in the 50 ppb metric give rise to larger percentage differences. Because there are only a couple of sites in each site type category among the 7 long-term monitoring sites analysed, it is hard to make a meaningful judgement on the performance of the models in different kinds of environment. The OSRM model appears to overestimate the values of all three metrics at Lough Navar and Belfast Centre.

4.2 POPULATION-WEIGHTED MEANS

Because the health implications of these ozone metrics are of interest, a population-weighted mean calculation of the various metrics has been used as this best represents exposure per person. Population-weighted mean concentrations have been calculated for each of the metrics as modelled empirically and by OSRM and are presented in Table 4.5. Table 4.6 presents the percentage difference between the empirical results and the OSRM results by region.

Table 4.5 Population-weighted means for empirical and OSRM methods (μg m^{-3})

Region	Empirical maps 2003			OSRM maps 2003		
	0 ppb cut-off	35 ppb cut-off	50 ppb cut-off	0 ppb cut-off	35 ppb cut-off	50 ppb cut-off
Scotland	66.3	8.1	1.0	69.8	9.6	1.7
Wales	66.5	9.2	2.3	73.2	13.4	2.7
Northern Ireland	60.7	4.7	0.5	74.0	11.5	2.0
Inner London	53.6	7.6	2.1	58.5	6.8	1.5
Outer London	57.8	9.5	2.7	59.1	6.9	1.5
Rest of England	64.0	9.4	2.5	64.7	9.1	2.0
UK	63.2	9.0	2.3	65.1	9.1	1.9

Table 4.6 Difference between empirical model and OSRM population-weighted means (as percentage of empirical data)

Region	0 ppb cut-off	35 ppb cut-off	50 ppb cut-off
Scotland	-5.3	-19.1	-63.8
Wales	-10.1	-44.9	-16.8
Northern Ireland	-21.8	-143.8	-285.5
Inner London	-9.1	10.7	27.1
Outer London	-2.3	26.9	43.2
Rest of England	-1.2	2.9	22.0
UK	**-3.0**	**-1.1**	**17.1**

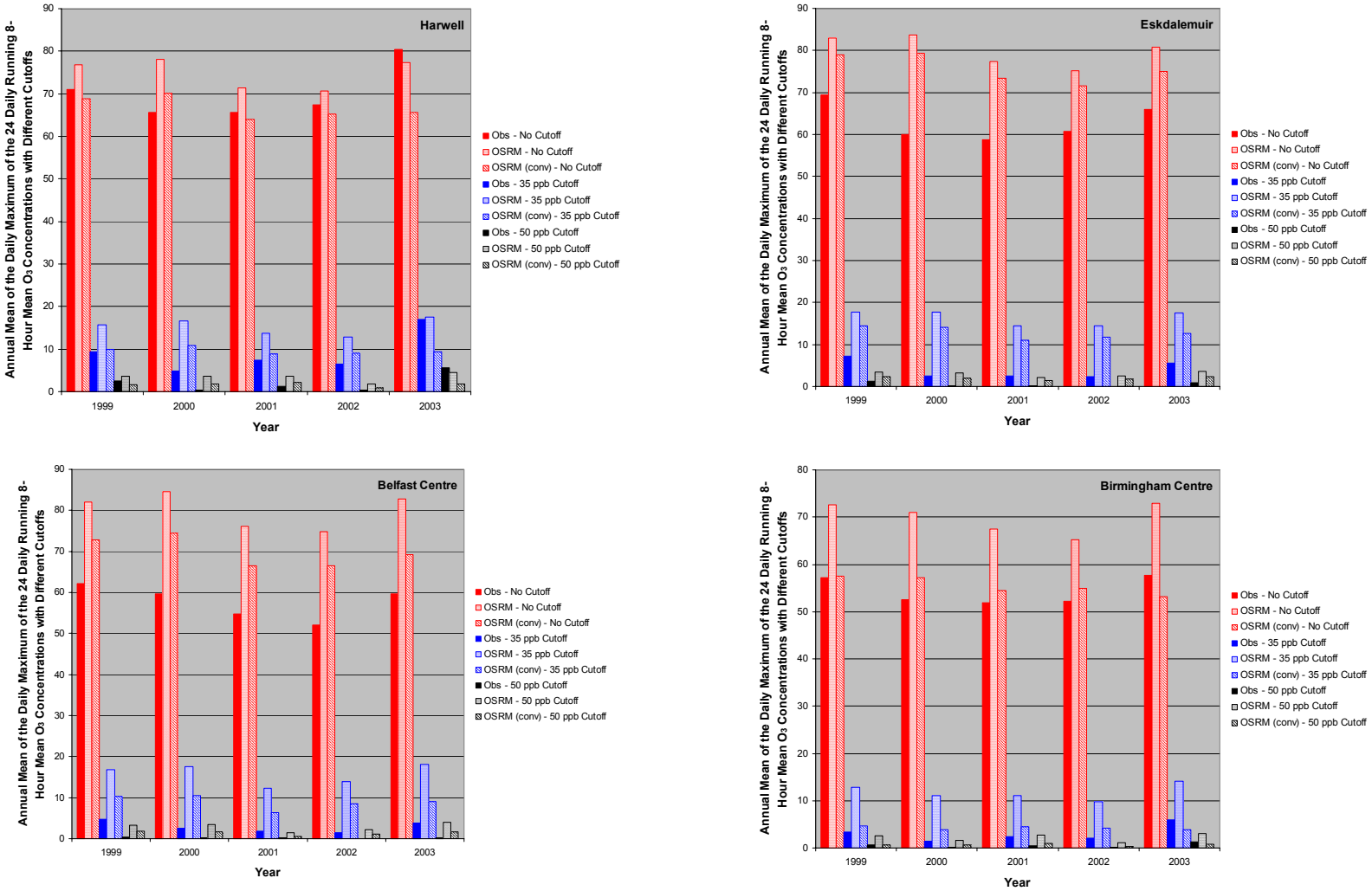
Table 4.5 shows the population-weighted mean concentrations by region for each metric as derived using the empirical method and OSRM. These differences are more concisely demonstrated in Table 4.6, which presents the differences as a percentage of the empirical output. The empirical model produced values for the 0 ppb cut-off metric that were lower than the corresponding OSRM output. For the 35 and 50 ppb cut-off metrics, the empirical model outputs were lower in Scotland, Wales and Northern Ireland, while in other UK regions, the empirically modelled population weighted mean concentrations were higher than for the OSRM. The most significant differences in population-weighted mean concentrations were in Northern Ireland and Wales for the 0 and 35 ppb cut-off metrics and in Northern Ireland and Scotland for the 50 ppb cut-off metric.

The analysis presented in this study is based on OSRM runs for a single year. In other work for the Department, OSRM model runs have been undertaken for a number of current years (1999-2003) using year-specific meteorology and emission inventories. Figure 4.1 compares the values of the three ozone metrics considered in this study, derived from ozone concentration measurements and from OSRM modelled concentrations for the following 4 sites: Harwell, Eskdalemuir, Belfast Centre and Birmingham Centre. Two OSRM modelled values are presented to show the effect of the surface conversion algorithm.

A number of points can be made:

1. The use of the surface conversion algorithm significantly improves the performance of the OSRM at all sites for the years shown, especially at the urban sites. There is some evidence that the surface conversion algorithm has overcorrected the mid-boundary ozone concentration;
2. As noted earlier, the OSRM tended to underestimate the metrics compared to observed values in 2003. For the earlier years (1999-2002), the model overestimated the metrics.

Figure 4.1 Comparison of the 3 ozone metrics derived from measurements with those calculated by the OSRM with and without the surface conversion algorithm activated (μg m^{-3})



4.3 MODEL VERIFICATION CHARTS

Figures 4.2 to 4.4 are verification plots showing the relationship between measured data and the equivalent modelled value at 72 sites in the AURN for which data capture exceeded 75%. Tables 4.7 and 4.8 provide summary comparison data such as the mean of measured and modelled values for each metric, the correlation coefficient (r^2), percentage of modelled values outside ±50% of the measured values and the total number of observations used.

Table 4.7 Summary of empirical model vs. measured data

Metric	Mean of measurements ($\mu g\ m^{-3}$)	Mean of empirical model estimates($\mu g\ m^{-3}$)	r^2	% outside data quality objectives	Number of sites
0 ppb cut-off	63.1	59.6	0.64	0.0	72
35 ppb cut-off	7.9	7.9	0.63	16.7	72
50 ppb cut-off	1.9	2.0	0.65	20.8	72

Figure 4.2 shows that the empirical model is reasonably accurate at predicting ozone concentrations. The accuracy is highest for the 0 ppb cut-off metric. Although the there appears to be greater variability between modelled and measured in the 35 ppb cut-off metric, it is actually the 50 ppb cut-off metric that exhibits more points outside the ±50% limits with 15 (20.8% of the total observations) outside this range compared with 12 (16.7% of the total observations) in the 35 ppb cut-off metric. There were no modelled values that fell outside the ±50% range in the 0 ppb cut-off metric.

Figure 4.2 Verification charts of empirical model results against monitored data

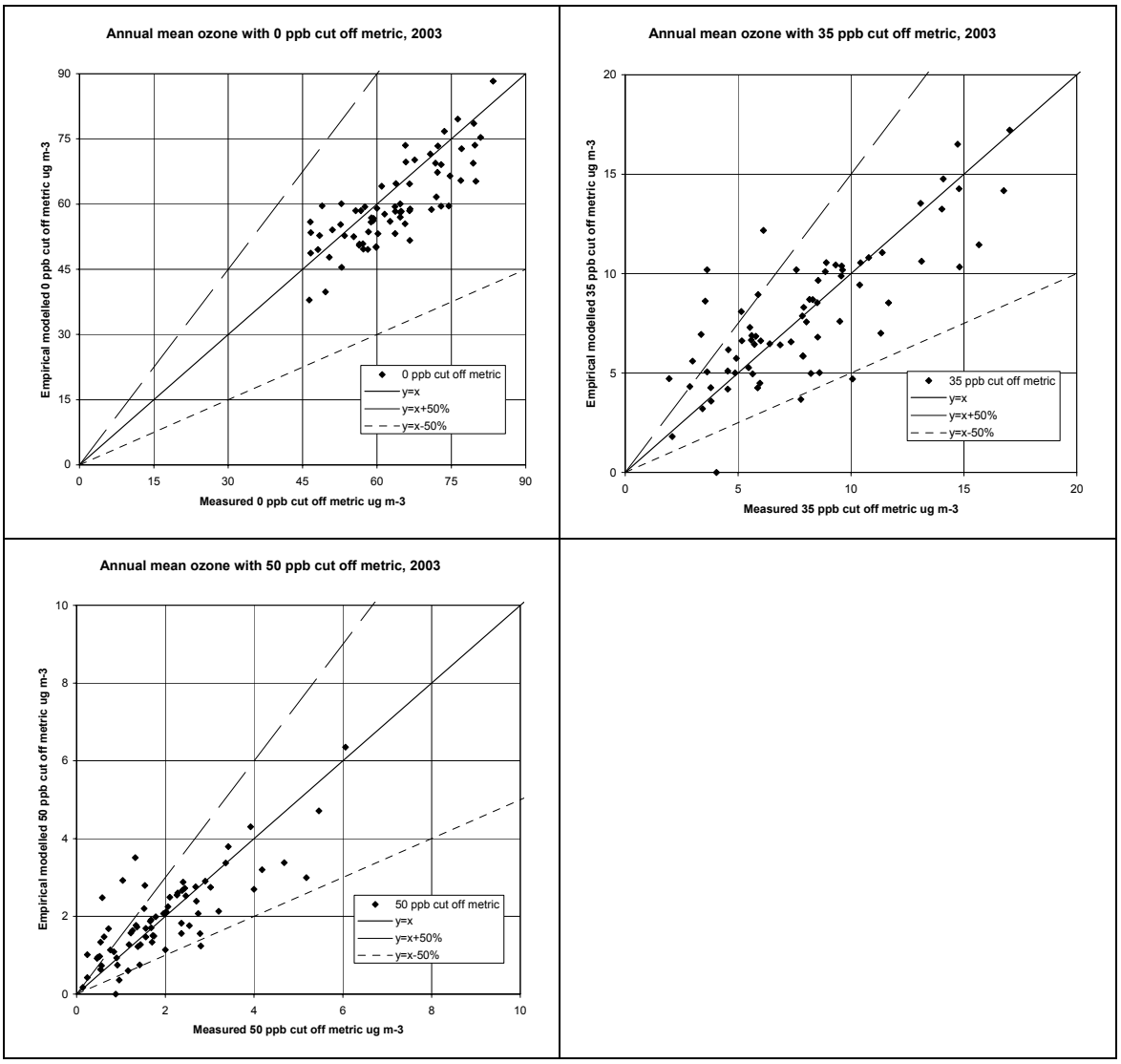
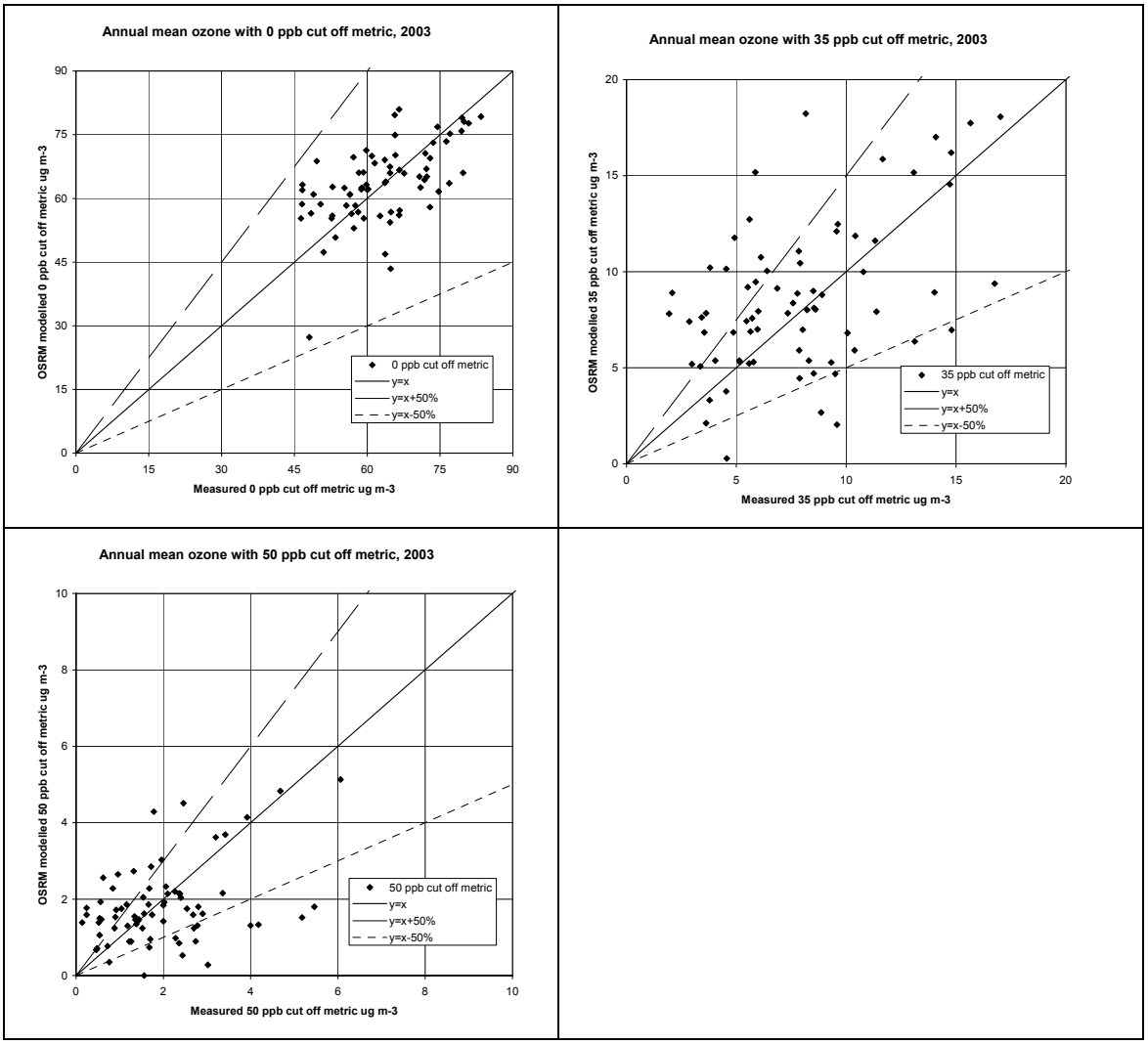


Table 4.8 Summary of OSRM vs. measured data

Metric	Mean of measurements ($\mu g\ m^{-3}$)	Mean of OSRM estimates ($\mu g\ m^{-3}$)	r^2	% outside data quality objectives	Number of sites
0 ppb cut-off	63.1	63.5	0.33	0.0	72
35 ppb cut-off	7.9	8.6	0.24	33.3	72
50 ppb cut-off	1.9	1.8	0.17	47.2	72

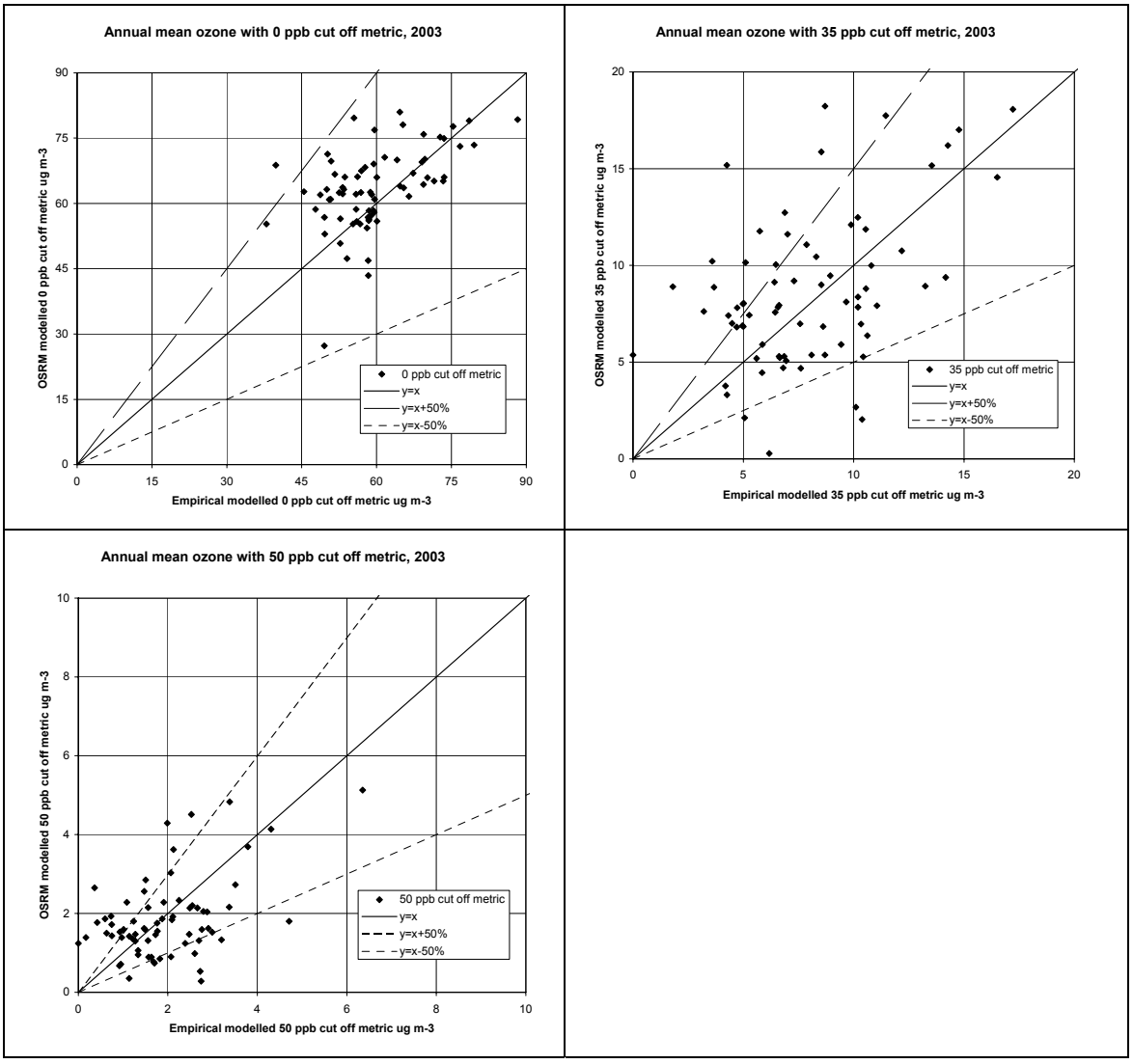
Results from the OSRM model are shown in Figure 4.3 and vary more than the corresponding empirical output when compared against monitored data. The 0 ppb cut-off metric shows that 3 points (0% of the total observations) are outside the ±50% range, 24 (33.3% of the total observations) are outside the range in the 35 ppb cut-off metric and 34 (47.2% of the total observations) are outside the range in the 50 ppb cut-off metric.

Figure 4.3 Verification charts of OSRM model results against monitored data



Direct comparison of the empirical model and the OSRM outputs illustrate that the OSRM results and empirical results predict broadly the same range of concentrations but, as might be expected, the empirical model results generally show less scatter when compared with measured concentrations. Both models generally show best agreement with the measurements for the 0 ppb cut-off and worst for the 50ppb cut-off metric.

Figure 4.4 Verification charts of OSRM model results against empirical model results



5 Conclusions

The different ozone metrics examined in this study respond differently over time and to trends in hemispheric background ozone levels, photochemical action and NO_x titration. As a result, the response of the metrics to various emission reduction scenarios is different, making the choice of metric an important factor in guiding policy decisions.

Empirical maps were more closely related to the measured data in 2003, largely because the model was calibrated using monitored data. The surface conversion algorithm in the OSRM improves the performance of the model. Both models appeared to perform better with the 0 ppb cut-off and not as well with the 35 and 50 ppb cut-offs. The 0 ppb cut-off metric is largely illustrative of background hemispheric trends and local NO_x titration, while the other two metrics are more indicative of photochemical ozone generation.

The comparison and verification analyses between the empirical and OSRM models presented here are for 2003. This was an unusual year and one for which OSRM did not perform as well as for some other years. Meteorology has a strong influence on ozone air quality and the year-to-year variation in the weather mean that general trends in ozone concentrations over time can only be readily identified over relatively long periods.

6 References

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