

Assessment of recent trends in NO₂ using CUSUM analysis methods

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

March 2005

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1 Summary

The purpose of this analysis is to investigate the nature and timing of trends in nitrogen dioxide concentrations as recorded by UK roadside monitoring sites between 2002 and 2004.

The hypothesis is that the large increase in NO₂ concentrations recorded at Marylebone Road in London is due in part to specific changes in the local emissions profile related to traffic management schemes such as the introduction of a bus lane, Congestion Charging etc. The analysis tests this hypothesis and is then extended to 150 further UK monitoring sites.

The CUSUM technique is used to identify sustained step changes in concentrations obscured from other analysis methods by measurement noise such as meteorological variation. CUSUM (Cumulative Sum) is the sum, over time, of deviations in the observed value of a variable from a reference value. If mean concentrations undergo a change, the CUSUM develops a linear drift centred on a line whose slope is approximately equal to the shift in the mean. Upper and lower control limits are set to identify when a process goes 'out of control' and whether this shift is sustained over time.

CUSUM analysis has identified sustained step changes in pollution concentrations at the Marylebone Road monitoring site. Towards the end of 2001, mean NO and NO₂ concentrations decrease, which may be related to the demarcation of the lane nearest to the monitoring site as a bus lane. In late 2002 or early 2003 there is an increase in mean NO₂ concentration, but a decrease in PM_{2.5} and a possible further decrease in NO. These changes may be related to introduction of regenerating particle traps on Transport for London's bus fleet. These step changes are independent of trends caused by gradually introduced measures such as fleet renewal and increased market share of diesel vehicles.

Screening using the Bland-Altman test of repeatability identifies six UK monitoring sites where abnormally high increases in NO₂ were recorded in 2004 over the previous two years. However, CUSUM analysis of these sites showed that in all but two cases such increases were episodic and not due to sustained step changes in concentrations. Sustained step changes were identified at two other UK monitoring sites besides Marylebone Road (Oxford Roadside and Chiswick High Road, London) although signals were less defined than that of Marylebone Road.

Improved sophistication of the CUSUM method is being undertaken to remove seasonal effects and improve method's sensitivity and ability to assess the 'timing' of changes. The method can be applied to any time series, be it air pollution, traffic, meteorology etc so has many potential applications.

More detailed analyses into the effects of regenerating particle traps on direct emissions of NO₂, including NO/NO₂/PM emissions testing, are required to aid understanding of their effects on ambient NO₂ trends.

2 Introduction

- 2.1.1 The purpose of this analysis is to investigate the nature and timing of trends in nitrogen dioxide concentrations as recorded by UK roadside monitoring sites between 2002 and 2004. During this period NO₂ concentrations at the central London kerbside AURN site Marylebone Road have increased significantly (annual mean increase from 80µgm⁻³ in 2002 to 110µgm⁻³ in 2004). The hypothesis is that this increase is due in part to specific changes in the local emissions profile related to traffic management schemes such as the introduction of a bus lane, Congestion Charging etc. The analysis tests this hypothesis and is then extended to all other monitoring sites in the AURN and regional monitoring networks in south east England.
- 2.1.2 This report should be considered in conjunction with complimentary studies carried out by the Institute of Transport Studies (ITS) at Leeds University and NETCen. The ITS report (Carslaw, 2005) identifies a marked increase in NO₂/NO_x emission ratio from road transport since 1997, based on analysis of 36 roadside monitoring sites within the London Air Quality Network. This trend is not uniform across all sites and some locations show a more rapid increase than others, the most extreme example being that of Marylebone Road.
- 2.1.3 The CUSUM technique used is particularly sensitive to sustained step changes in measurements reflecting small-scale emissions profile alterations such as local traffic management schemes. Such schemes can cause an immediate change to emissions local to a monitoring site, but a corresponding change in ambient levels can be difficult to identify due to the overwhelming influence of meteorology on pollution dispersion. The technique has the advantage of using unadjusted daily mean measurements with no pre-processing and can therefore be applied rapidly to a large number of datasets.
- 2.1.4 The novel application of CUSUM analyses to air pollution time series data presented in this report is intended for visual screening rather than quantification. The precise application of this technique to identify the presence and timing of distinct sustained step changes in pollution levels is being developed as part of a three-year Health Effects Institute research project (ERG, 2004).

3 The CUSUM statistical method

3.1 Basic description of method

- 3.1.1 CUSUM (Cumulative Sum) is a statistical method often used by quality practitioners to detect small and sustained shifts in a process. The CUSUM is the sum, over time t , of deviations in the observed value of a variable (x_t) from a reference value:

$$\text{Cumulative sum } S_t = S_{t-1} + z_t \quad \text{where } z_t = x_t - \mu_0$$

- 3.1.2 This running total is represented in a time series control chart. While X_n is normally distributed, CUSUM follows a distribution centred on the horizontal axis. If the mean undergoes a change, the CUSUM develops a linear drift centred on a line whose slope is approximately equal to the shift in the mean. Upper and lower control limits may be set at multiples of the standard deviation to identify when a process goes 'out of control' and whether this shift is sustained over time. There is a time lag dependent on the magnitude of the change and size of control limits before a process is identified as out of control.

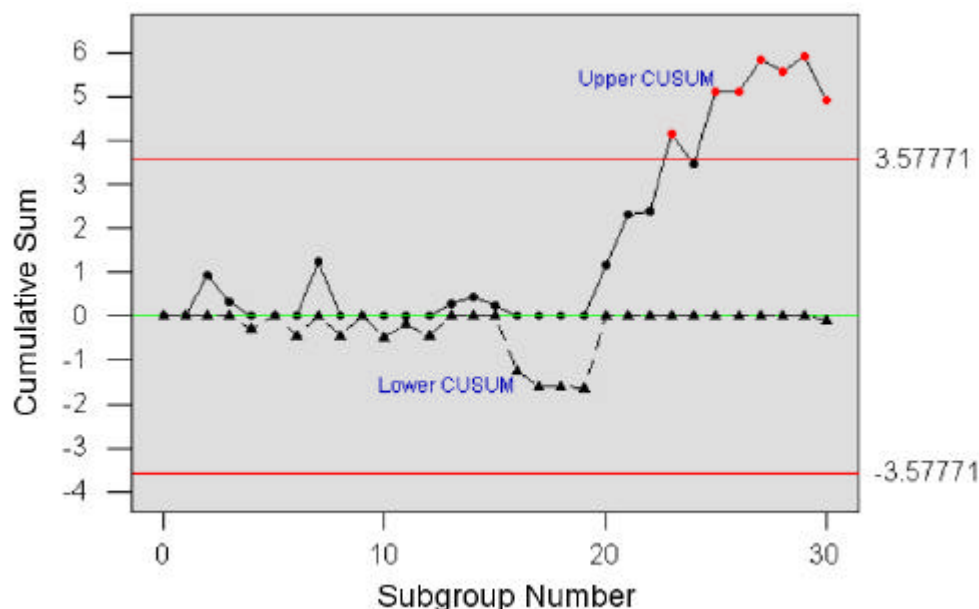


Figure 3-1: Sample CUSUM chart of manufacturing process (Bower, 2002).

- 3.1.3 The example in Figure 3-1 shows a CUSUM chart of a manufacturing process where a small shift in mean is introduced after sample number 20. By sample number 23 this shift is identified as being more than 4 standard deviations from the reference value. Note that there is a time lag of 3 samples following introduction of the change before the process is identified as being out of control.
- 3.1.4 In this analysis, the reference value will be the mean concentration of nitrogen dioxide at a given monitoring station for the calendar year 2002 (or 1st January 1999 in some cases). This reference value is subtracted from daily mean observations from the start date to 1st January 2005. The resulting (daily) deviations from this reference value are then added together sequentially to form a series; the first deviation forms the first CUSUM of the series, the sum of the first two deviations are added together to form the second CUSUM, the sum of the first three deviations are added together to form the third CUSUM and so on. A change in the mean concentration of the pollutant is proportional to the change in the slope of the CUSUM function.

- 3.1.5 Unlike long-term averaging charts, such as those showing running annual means, a step change in concentration is represented by a slope of constant gradient. This is because each subsequent value above the reference mean increases the CUSUM function by the difference between the reference mean and the new mean (as represented by the formula given in paragraph 3.1.1).
- 3.1.6 The sensitivity of the CUSUM procedure can be adjusted by the setting of k , a measure of the allowable 'slack' in the system. In the following analyses, k is set at 0.5, which means that any change in mean less than 0.5 of the standard deviation of the time series is treated as zero change (zero gradient in the CUSUM charts).
- 3.1.7 The CUSUM approach described is designed to identify whether or not a sustained step change in mean pollution levels has occurred and if so, when it occurred. The gradient of the CUSUM function can be used to estimate the magnitude of a step change, although other methods such as regression modelling may produce a more precise estimation.

3.2 *Limitations of use in this study*

- 3.2.1 There are a number of limitations to the straightforward application of the CUSUM technique as applied in this report, which limit the sensitivity of results. Firstly, the identification of an out of control process depends on readings being statistically independent and following a normal distribution. Furthermore, the true mean and standard deviation should be constant in a stable ('in control') process. Pollutant measurements can be expected to have some serial correlation, seasonality and underlying long-term trends. This has the effect that upper and lower control limits can be breached due to seasonal or short-term trends effects. To limit the number of such breaches, control limits have been set at 8 times the standard deviation ($h=8\sigma$) of the reference period (first 12 months of time series). Consequently, only strong signals can be clearly identified as being due to sustained step changes in long-term mean.
- 3.2.2 Techniques for de-trending and removing seasonality from pollutant results are currently being developed as part of a three year HEI-funded study into the Congestion Charging Scheme, but are not applied in this report.
- 3.2.3 It should also be noted that 2004 measurements at all sites were not fully ratified at the time of analysis.

4 Introductory analysis – Marylebone Road

4.1 Running annual mean trends

- 4.1.1 Figure 4-1 shows running annual mean trends in nitrogen dioxide at Marylebone Road kerbside site in Central London since 1999 (monthly increments). Mean concentrations are seen to remain level up until 2001 when they decrease up until the end of 2002. From the beginning of 2003 to the beginning of 2004 levels increase rapidly before stabilising. During this period rural NO₂ concentrations in south east England (as represented by the AURN site 'Teddington') have shown a relatively steady decrease ($-8.4 \mu\text{g m}^{-3}$ 1999-2004). Inner London background concentrations (as represented by the AURN site, 'North Kensington') have followed a similar trend to rural concentrations with an overall decrease of $6.3 \mu\text{g m}^{-3}$.

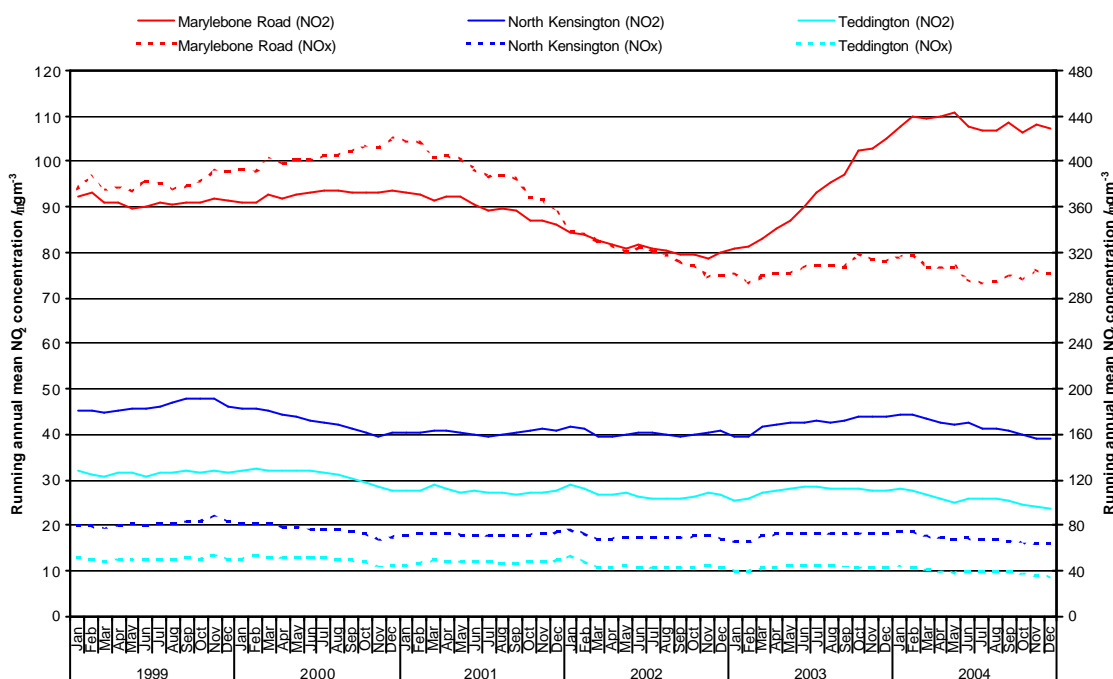


Figure 4-1: Running annual mean NO₂ concentrations 1999 to 2004 (monthly increments).

- 4.1.2 NO_x concentrations at the rural and background sites follow a very similar trend to NO₂, but the trends at Marylebone Road diverge rapidly during 2003. The changing NO₂:NO_x balance is more clearly illustrated in Figure 4-2, which shows running annual mean trends in NO₂/NO_x ratio. Ratios at the rural site show no long-term pattern until 2002, when a slight upward trend develops. At the North Kensington and Marylebone Road sites, ratios are steady up until 2002, then increase before becoming very stable during 2004. The rate of increase at Marylebone Road, however, is much more rapid (an increase of 36% at Marylebone Road, 9% at North Kensington).
- 4.1.3 As Figure 4-1 and Figure 4-2 use running annual means to illustrate trends, any step changes in recorded pollution levels will be slowly introduced over a period of 12 months. For example, if a step change of $5 \mu\text{g m}^{-3}$ were introduced into an otherwise stable situation, annual mean concentrations would slowly increase by $5/12$ per month until the concentrations stabilise at the new level after 12 months. While this has the advantage of reducing noise caused by meteorological seasonal variation, it can also obscure the timing of genuine step changes.

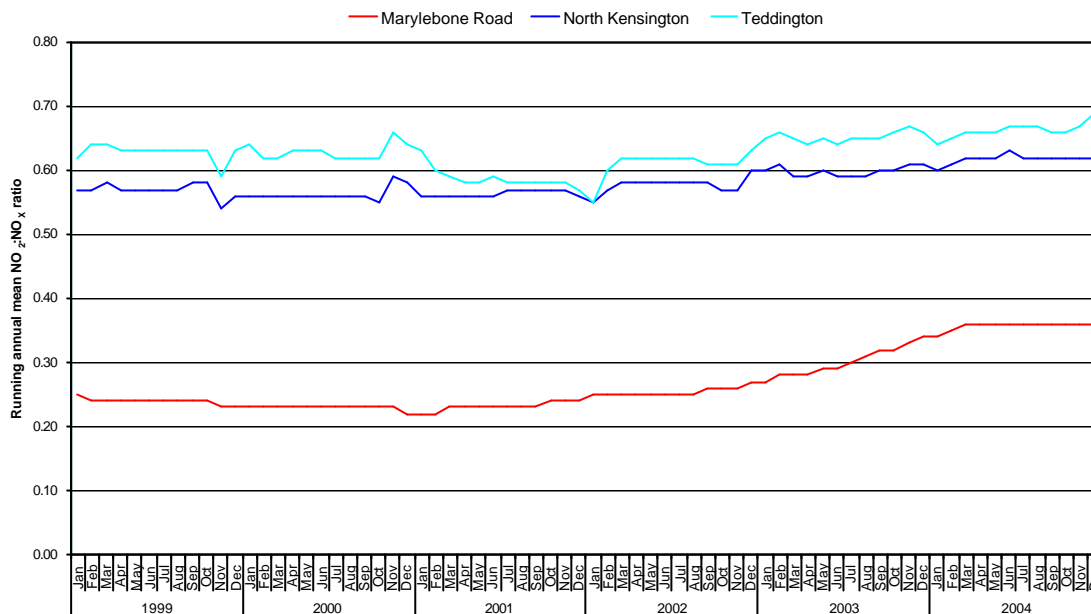


Figure 4-2: Running annual mean NO₂:NO_x concentrations 1999 to 2004 (monthly increments).

- 4.1.4 Two traffic management schemes have been introduced in recent years that have had a direct effect on vehicle flow or composition on Marylebone Road; a nearside bus lane was introduced in August 2001, followed by the start of the London Congestion Charging Scheme in February 2003. Introduction of the bus lane caused a 50% drop in vehicle flow in the nearside lane closest to the monitoring site inlet. The great majority of vehicles remaining on the nearside lane are diesel fuelled. While this measure produced a step change in local emissions, any effect on pollution levels would be seen as a gradual change on a running annual mean chart.

4.2 CUSUM analysis of nitrogen dioxide

- 4.2.1 While the running annual mean charts in Figures 4.1 and 4.2 are able to identify short-term trends in NO₂ concentrations, they do not show whether such trends are as a results of gradual change over a 12 month period or step change at certain date during that 12 month period. A chart using shorter averaging periods could help differentiate between gradual and step changes, but these can be difficult to discern because of measurement noise. Through the use of CUSUM charts the averaging period can be kept short (daily means) but step changes are more likely to be identified.

- 4.2.2 Figure 4-3 shows CUSUM analysis of nitrogen dioxide daily means 1st January 1999 to 2005. The reference value is 91µgm⁻³ (1999 annual mean concentration). There are four main patterns evident on this chart;

- January 1999 to December 2001 - No sustained deviation from the reference value. This represents steady daily mean values within the control limits (set at 8x standard deviation).
- December 2001 to October 2002 – Process out of control (lower limit) indicating negative shift in mean. A step change has occurred causing daily mean concentrations to be consistently lower than the reference value. The negative gradient indicates that this change is sustained throughout the period, *not that daily means become progressively lower*.
- October 2002 to May 2004 – Process out of control (upper limit) indicating a positive shift in mean. A second step change has occurred causing daily mean concentrations to be consistently higher than the reference value. The steep

positive gradient indicates that not only is this change sustained throughout the period, but also that the change is of greater magnitude than the previous step change causing an overall increase.

- d) May 2004 to July 2004 – A short period of level CUSUM indicates that daily means temporarily remain unchanged before stepping up again, possibly at an increased concentration (steeper gradient than in period c).

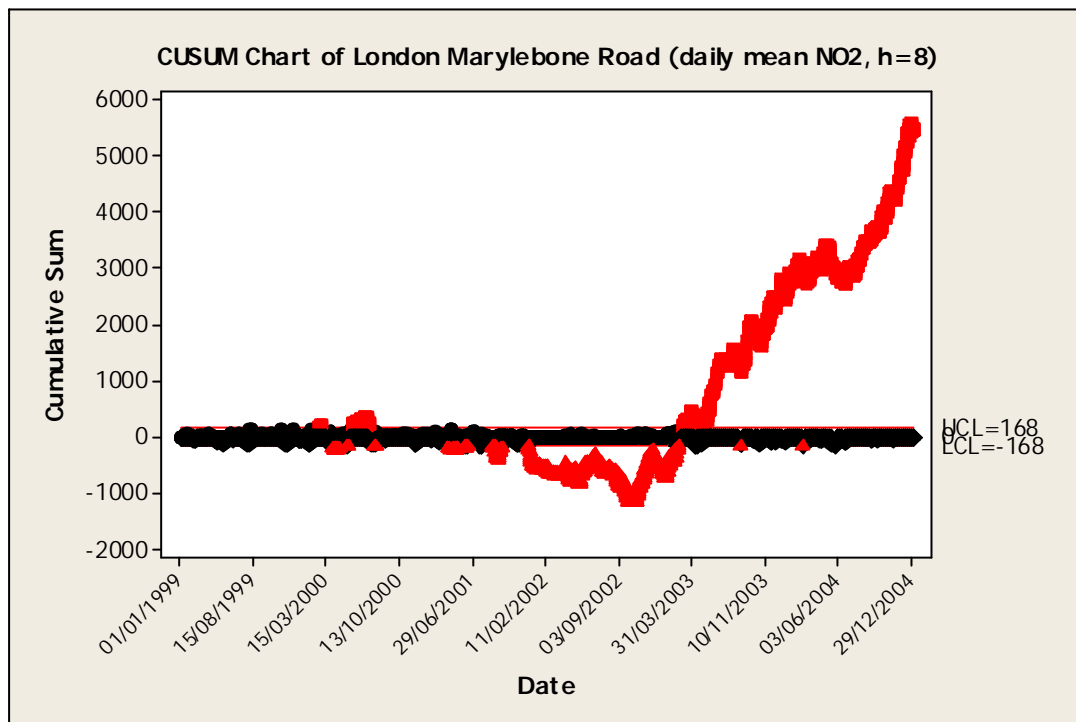


Figure 4-3: CUSUM chart of nitrogen dioxide at Marylebone Road using the 1999 annual mean as a reference value.

- 4.2.3 It is therefore likely that the bus lane introduced in August 2001 did cause a sustained decrease in NO₂ concentrations until a second measure introduced around October 2002 cancelled out this decrease and caused an overall increase. Since then, this increase has been sustained with some suggestion that a further increase may have occurred in 2004. This second measure does not coincide with the introduction of the CCS, but seasonal trends may have shifted the apparent change point, i.e., what appears to be a shift in mean due to a long-term traffic management measure, may *initially* have been a short-term shift due to meteorological conditions.

4.3 CUSUM analysis of other pollutants

- 4.3.1 The above analysis was repeated for NO, NO_x, PM₁₀ and PM_{2.5} measurements at Marylebone Road (Figures 4.4 to 4.7). Due to the strong influence of transboundary particulate matter, measurements from the rural AURN site 'Harwell' were subtracted from PM₁₀ and PM_{2.5} measurements. This has the effect of removing the background component of particulate matter, independent of local transport emissions. This is especially important during 2003 when a series of regional pollution episodes were caused by distant sources of particulate matter. Findings from this analysis are summarised below:

- 4.3.2 *Nitric Oxide* (Figure 4-4) – Process is strongly affected by seasonal variation with winter peaks and summer troughs making identification of step change timings difficult. However, the process goes out of control (below lower limit) in early 2002 with an increase in downward gradient during 2004. Outside of seasonal interference, there is no sustained positive (upward) gradient as seen with NO₂. Therefore, the second measure introduced

around October 2002 did not cause an increase in NO concentrations and may have caused a decrease.

- 4.3.3 *Oxides of Nitrogen* (Figure 4-5) – This chart represents the difference between Figures 3.3 and 3.4 ($\text{NO}_x = \text{NO}_2 - \text{NO}$). Although the process does go out of control for much of 2002–2004, overall, daily means return to the reference value by the end of 2004. Therefore decreases in NO concentrations are cancelled out by increases in NO₂ concentrations.
- 4.3.4 *PM₁₀ particulate* (Figure 4-6) – Aside from two short-lived episodes in October/November 1999 and November 2001 due in part to Bonfire Night celebrations, the process remains in control. This indicates that no upward or downward step changes have occurred during this period and traffic management measures have not had an identifiable effect on PM₁₀ concentrations.
- 4.3.5 *PM_{2.5} particulate* (Figure 4-7) – The chart for PM_{2.5} is significantly different to that of PM₁₀. There are two short-lived episodes at the beginning and end of 2001, but concentrations remain within control limits during 2002, indicating no identifiable change in mean concentrations. The process does, however, go out of control in early 2003 and is sustained until the end of 2004, i.e., a sustained decrease in mean concentration occurs. The pattern is similar to that of NO but without the initial negative step change in 2002 related to the introduction of the bus lane.
- 4.3.6 In summary, there appear to be three sustained step changes in pollution concentrations at the Marylebone Road monitoring site. After an initial period of stability, there is a negative step (i.e., a decrease in mean concentration) in NO and NO₂ towards the end of 2001, which may be related to the introduction of the bus lane. As the bus lane is on the traffic lane closest to the monitoring site, this would have the effect of moving all vehicles except buses and taxis further from the sampling point. Particulate concentrations are not affected by this change possibly because vehicles remaining within the nearside lane are almost entirely diesel fuelled.
- 4.3.7 The second step change occurs in late 2002 or early 2003 and causes an increase in mean NO₂ concentration, but a decrease in PM_{2.5} and a possible decrease in NO. This may be related to the increased frequency of buses on Marylebone Road and/or the introduction of regenerating particle traps on Transport for London's (TfL) bus fleet. These traps use an oxidising catalyst to convert NO to NO₂ within the exhaust system. The NO₂ is then used to help 'burn off' trapped particles, hence regenerating. The result is a greater proportion of NO_x emissions from vehicles fitted with such traps in the form of NO₂. Research is currently being carried out by ERG on behalf of TfL into the likely contribution of bus emissions to this observed increase in NO₂ concentrations at Marylebone Road and other roadside/kerbside sites in London. The potential effects of buses fitted with particle traps at Marylebone Road is also considered in more detail in the partner report by ITS.
- 4.3.8 The final step change occurs in mid 2004 when NO, NO₂ and PM_{2.5} concentrations increase. As this change occurs toward the end of the analysis period it is not clear whether it will be sustained or transient, i.e., due to meteorology or a change in local emissions.

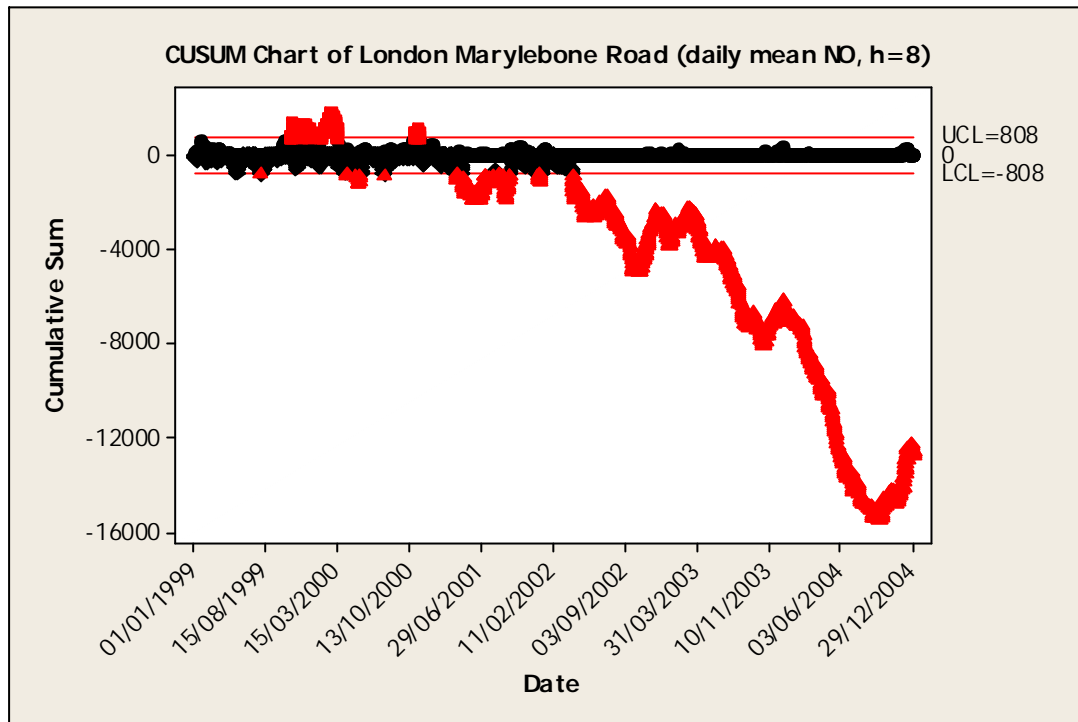


Figure 4-4: CUSUM chart of NO at Marylebone Road using the 1999 annual mean as a reference value.

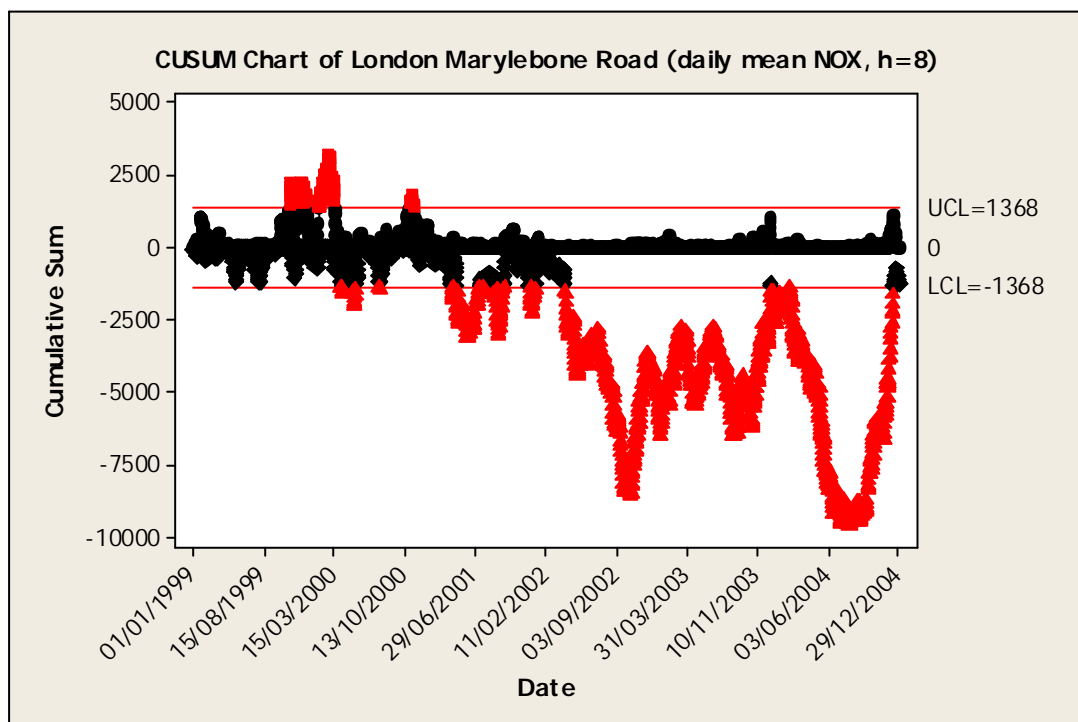


Figure 4-5: CUSUM chart of NO_x at Marylebone Road using the 1999 annual mean as a reference value.

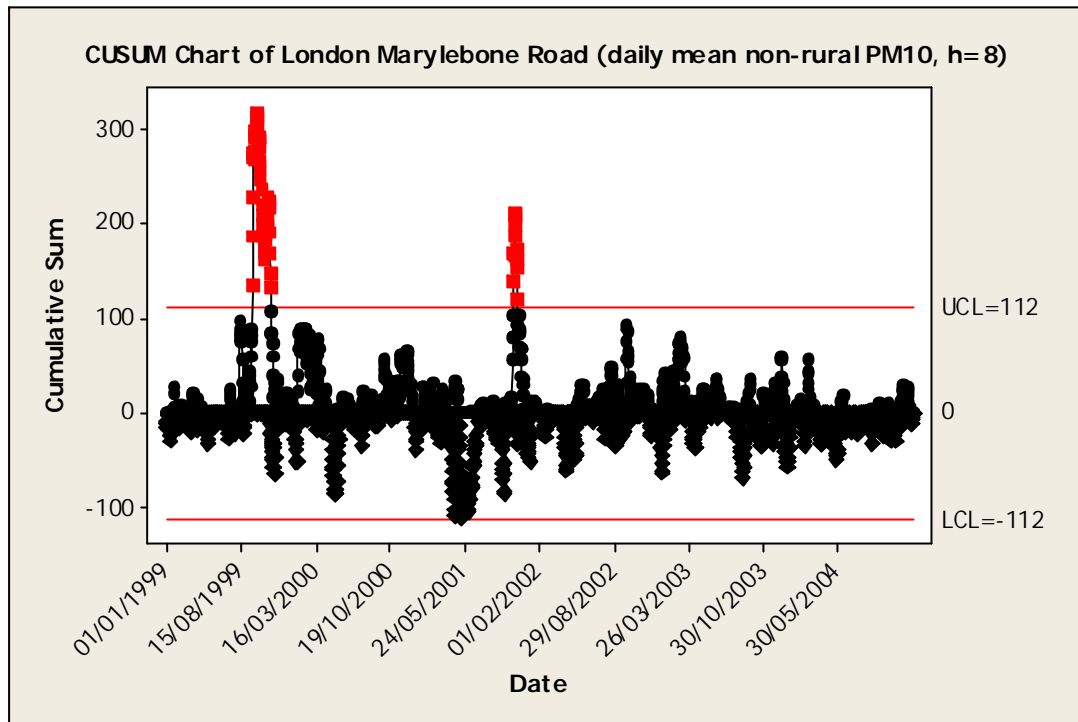


Figure 4-6: CUSUM chart of PM₁₀ at Marylebone Road using the 1999 annual mean as a reference value (rural component subtracted).

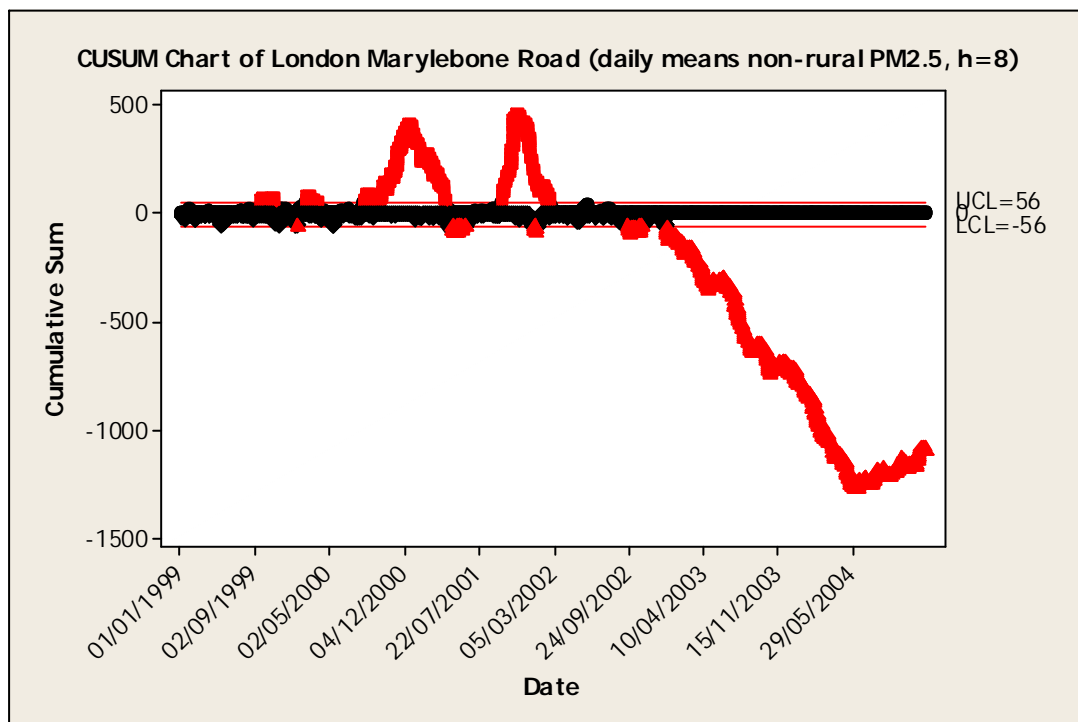
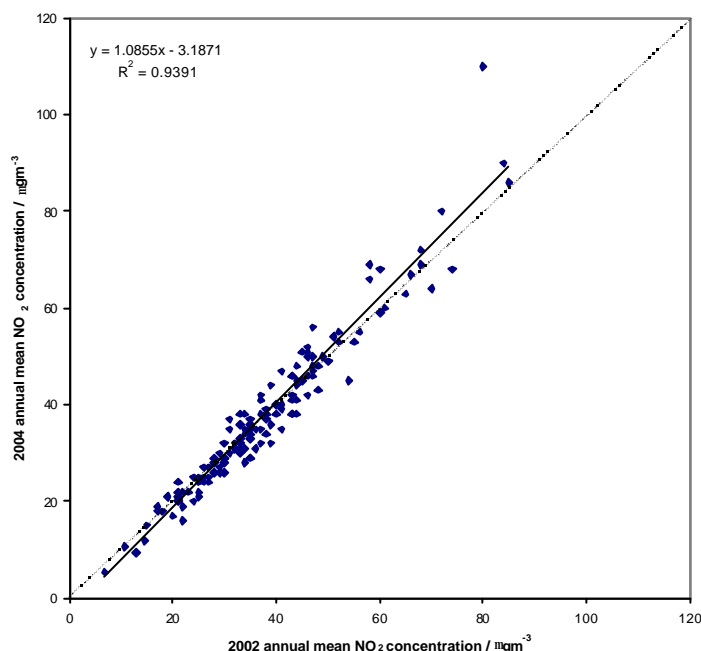


Figure 4-7: CUSUM chart of PM_{2.5} at Marylebone Road using the 1999 annual mean as a reference value (rural component subtracted).

5 National response

5.1 UK-wide nitrogen dioxide trends, 2002/2004

- 5.1.1 The initial analysis in Section 4 identified a step change increase in NO₂ concentrations at the central London kerbside site Marylebone Road. It is important to establish whether this response has been recorded elsewhere in the UK. Screening was carried out to identify locations where abnormally high increases in NO₂ were recorded in 2004 over the previous two years.
- 5.1.2 The Bland-Altman test of repeatability was applied to all AURN, LAQN and south east regional monitoring network sites with at least 75% data capture for 2002 and 2004. This approach tests the assumption that all sites respond to changes in overall meteorological conditions in a similar way over two defined periods. Those sites that deviate from this uniform response may have experienced local changes in emissions. Figure 5-1 compares annual mean nitrogen dioxide concentrations for 2002 and 2004 at all 150 sites. Even across such a wide geographical area, the linear relationship is statistically significant with an R²



value of 0.94.

Figure 5-1: Scatter plot comparing annual mean NO₂ concentrations in 2002 and 2004 across the UK.

- 5.1.3 While there are clear outliers from the regression line in Figure 5-1, the scatter plot does not quantify the statistical degree of deviation, i.e., how 'unusual' a particular site's change in annual mean levels is in comparison with the rest of the UK. The Bland-Altman test of repeatability (Bland and Altman, 1986) compares the mean of the two annual mean concentrations and the deviation from this mean. For example, Marylebone Road recorded annual mean concentrations of 80 μgm⁻³ and 110 μgm⁻³ in 2002 and 2004 respectively. The mean of these two values (95 μgm⁻³) is plotted against the difference in means (30 μgm⁻³). Limits are then added to highlight monitoring sites that exhibit a change in mean concentration greater than two standard deviations from the network mean change. If a normal distribution of data is assumed, these limits represent the 95% confidence interval. These sites can then be statistically defined as significant outliers to the analysis.

- 5.1.4 The results of the Bland-Altman test are shown in Figure 5-2. Six sites fall above the 95% confidence interval, i.e., recorded increases in annual mean concentrations more than 2σ above the UK network mean; five roadside sites in London and one in Oxford. One site, Trafalgar Road in Greenwich, fell below the 95% confidence interval. CUSUM analysis on daily mean data from 2002 to 2004 was then carried out on each of these sites to establish whether this increase was due to an identifiable sustained step change in NO₂ concentrations or due to short-term effects such as congestion caused by road works.

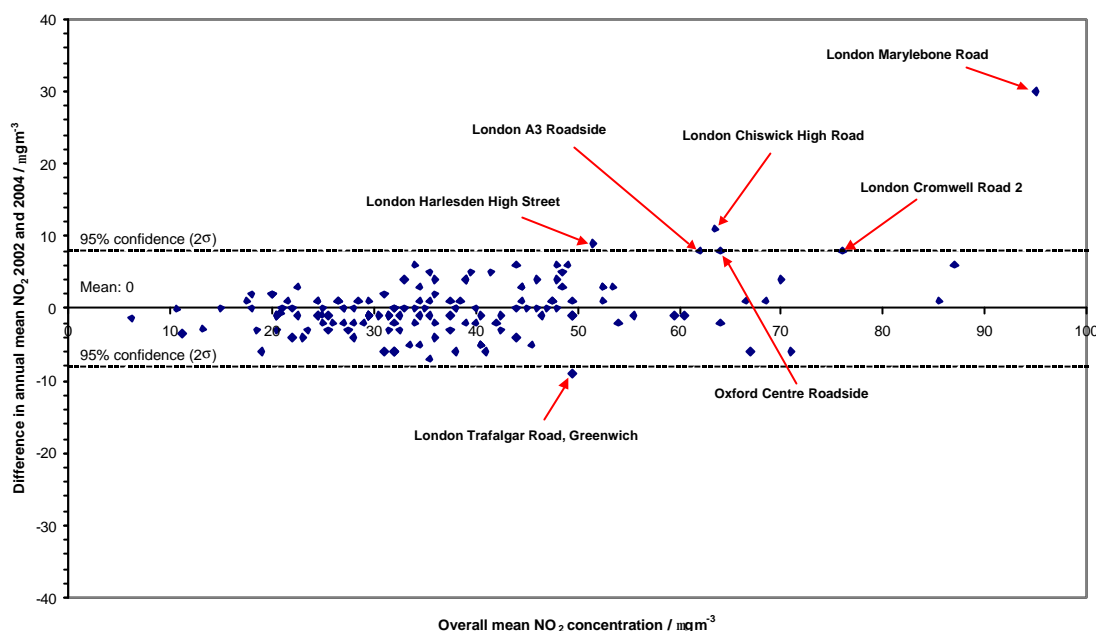


Figure 5-2: Bland -Altman plot for NO₂, annual mean concentration 2002 vs. 2004

5.2 Results of CUSUM analyses

- 5.2.1 CUSUM analysis results for the seven sites identified as outliers are presented in Figures 4.3 to 4.9.
- 5.2.2 Patterns at all sites show some degree of seasonal or episodic variation, i.e., periods where processes go out of control are not sustained. This indicates that step changes are unlikely to be due to permanent emissions changes or traffic management schemes. The exceptions are Oxford Roadside (Figure 5-7), London Chiswick High Road (Figure 5-8) and Marylebone Road (Figure 5-9).
- 5.2.3 The only site to show exceptional negative change (below the 95% confidence interval) was London Trafalgar Road in Greenwich. The CUSUM chart shows the process remains in control until mid-2004 when a steep negative gradient takes it out of control (lower limit). As this downward step occurs toward the end of the analysis time series, it is not possible to establish whether it is a sustained change. The upward shift at the end of 2004 is likely to be seasonal and is similar to patterns seen on other site's charts.
- 5.2.4 The chart for Oxford Roadside site shows the process going out of control (upper limit) in early 2003, coincidental with a widespread pollution episode and common with many other sites. Unlike other sites, however, the process does not return to an in control state for the remainder of the time series. It is possible, therefore, that there has been a sustained upward step in mean concentration sometime in 2003. As the signal is relatively small (compared to that at Marylebone Road), the analysis will have to be developed to remove seasonality before a more specific date can be estimated.

- 5.2.5 Chiswick High Road's chart also shows the process going out of control (upper limit) in early 2003, but the positive gradient continues until the beginning of 2004. As with Oxford Roadside, the irregularity of gradients means that a longer dataset and some refinement of the technique before more conclusive statements can be made, but an upward step change in concentrations may have occurred during 2003.

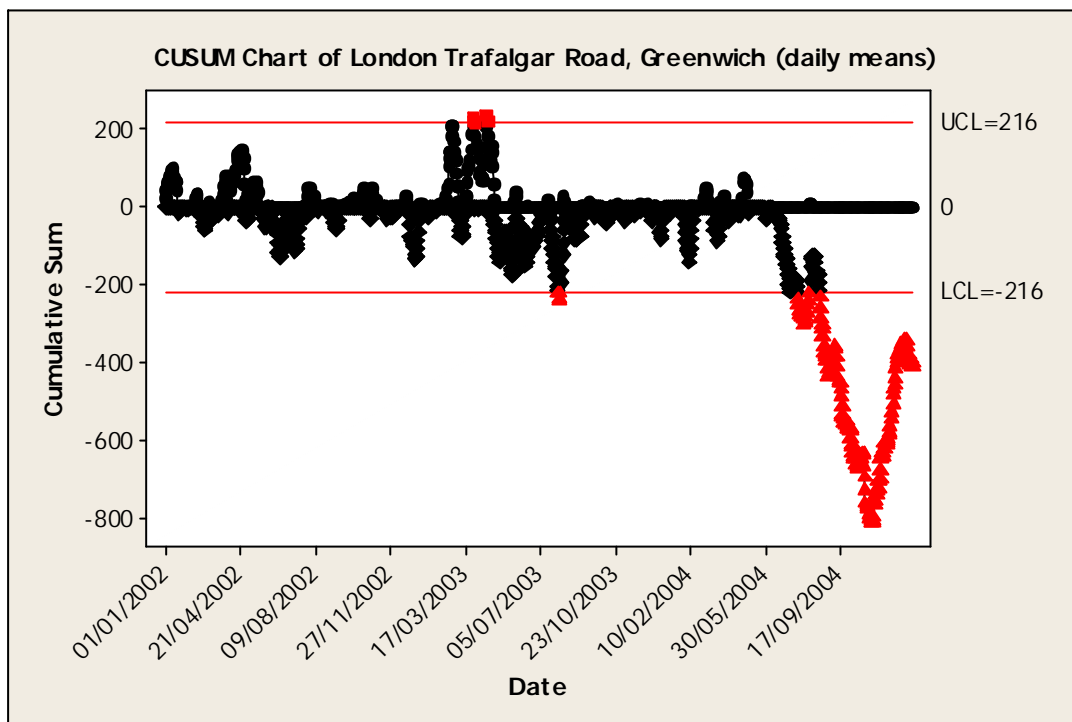


Figure 5-3: CUSUM chart of NO₂ at Trafalgar Road, Greenwich using the 2002 annual mean as a reference value.

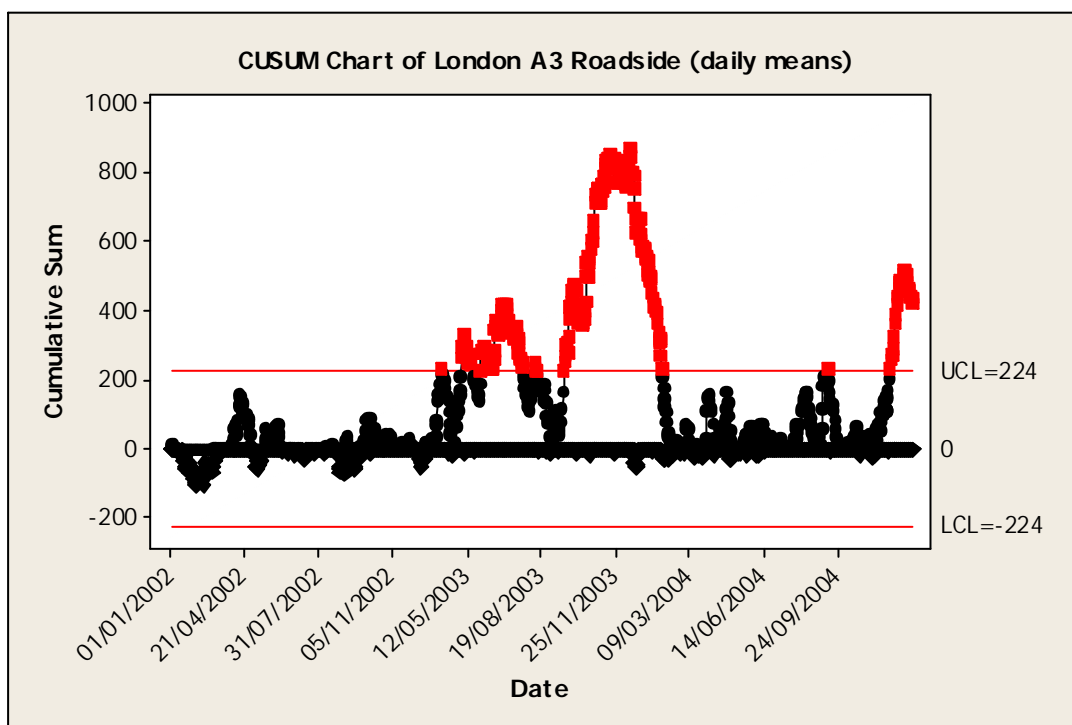


Figure 5-4: CUSUM chart of NO₂ at London A3 Roadside using the 2002 annual mean as a reference value.

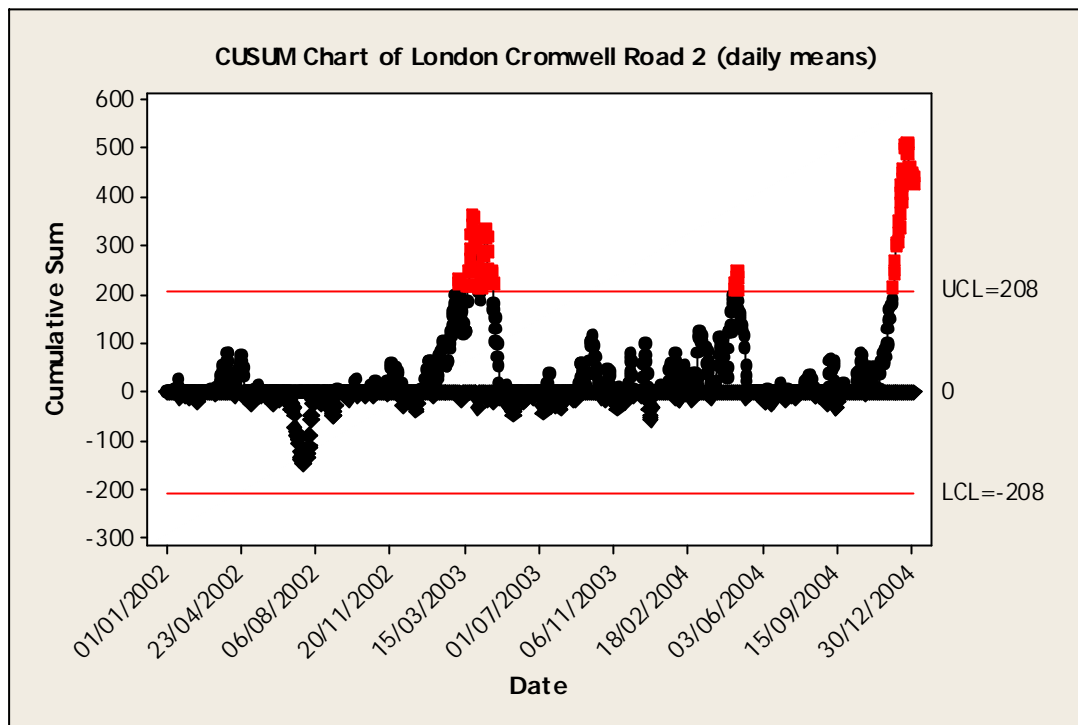


Figure 5-5: CUSUM chart of NO₂ at London Cromwell Road 2 using the 2002 annual mean as a reference value.

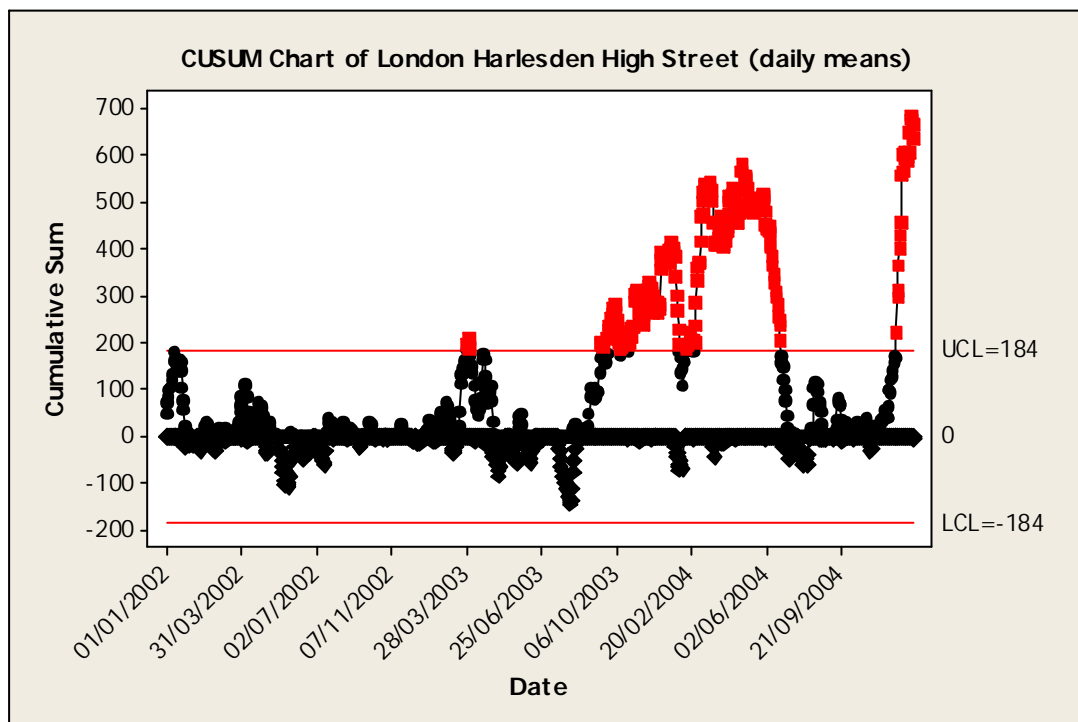


Figure 5-6: CUSUM chart of NO₂ at Harlesden High Street using the 2002 annual mean as a reference value.

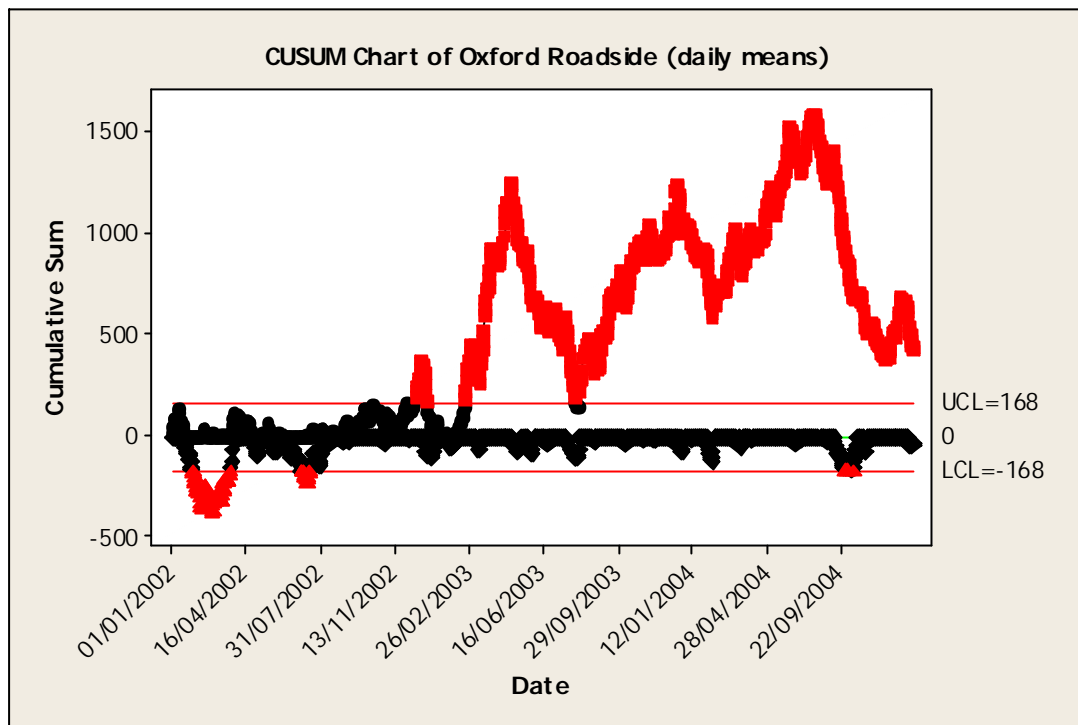


Figure 5-7: CUSUM chart of NO₂ at Oxford Roadside using the 2002 annual mean as a reference value.

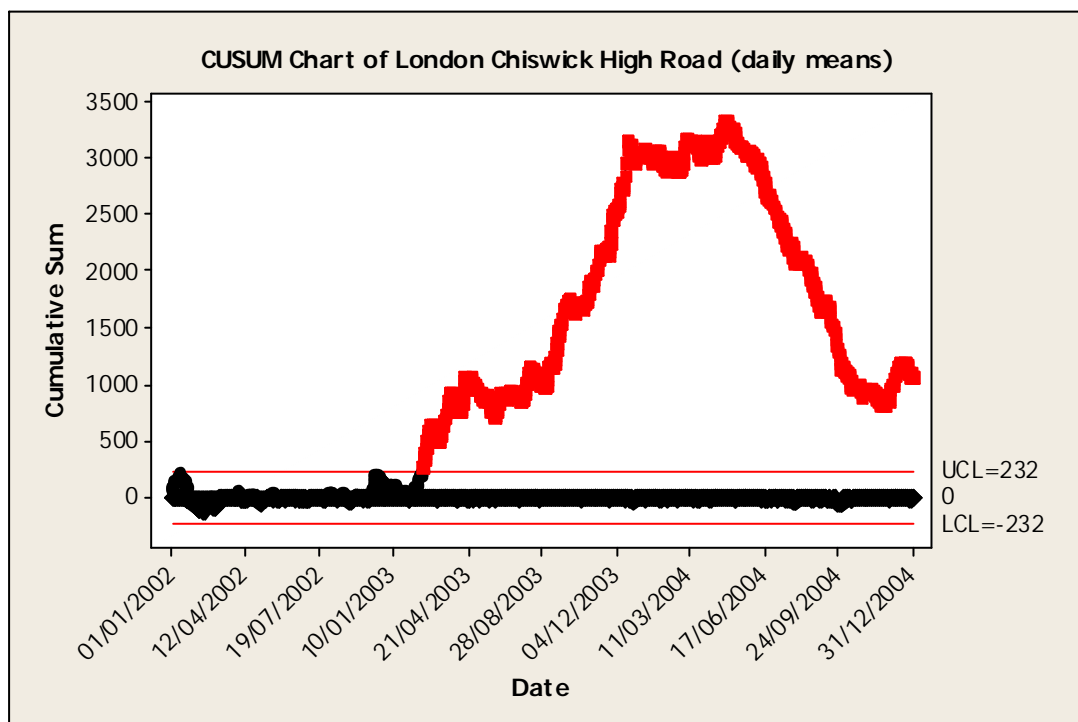


Figure 5-8: CUSUM chart of NO₂ at Chiswick High Road using the 2002 annual mean as a reference value.

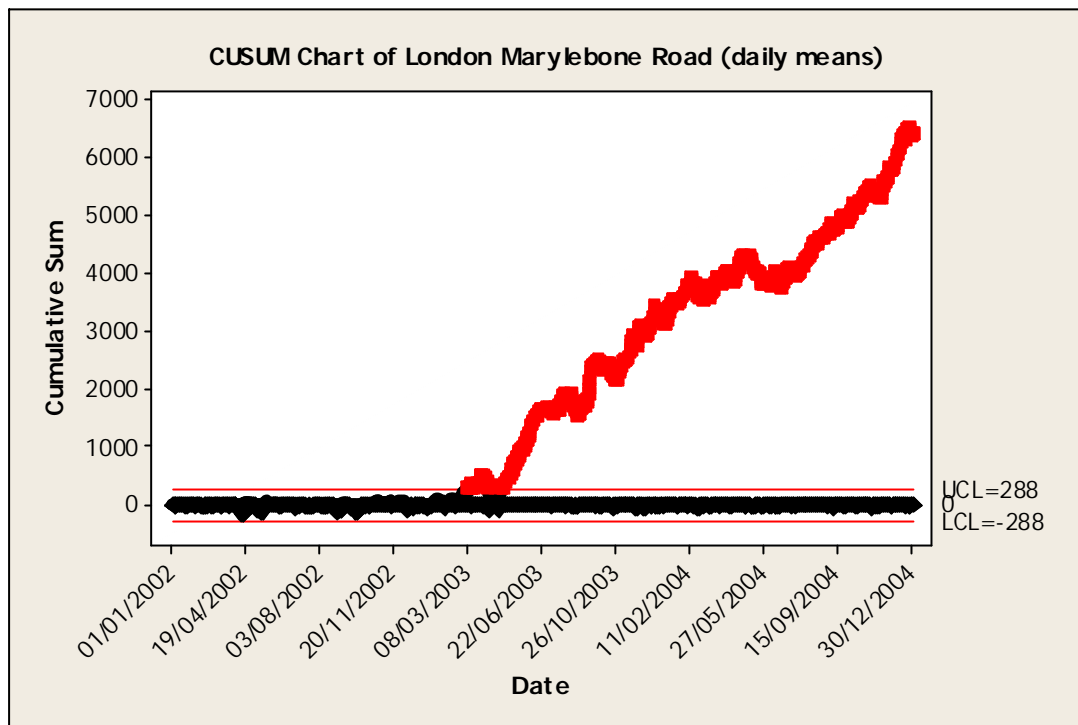


Figure 5-9: CUSUM chart of NO₂ at Marylebone Road using the 2002 annual mean as a reference value.

6 Conclusions and Recommendations

The CUSUM technique has proved useful at looking at the changes in time series air pollution concentrations. Where these can be related to other events, the analysis presents a much clearer picture of the most influential changes affecting concentrations. Improved sophistication of this method is being undertaken to remove seasonal effects and this will improve method's sensitivity and ability to assess the 'timing' of changes. The method can be applied to ANY time series, be it air pollution, traffic, meteorology etc so has many potential applications.

The straightforward application of CUSUM as presented in this report has identified the presence and approximate timing of step changes in mean pollution levels at those sites where there is a strong signal, most notably Marylebone Road. Investigation of step changes produced by the introduction of emissions or traffic management schemes should improve understanding of less distinct gradually introduced national measures. This should allow the translation of theoretical policy targets to identifiable changes in ambient pollution concentration.

It is clear from this analysis that traffic management schemes must consider each pollutant in isolation when assessment is made as to their affect on ambient concentrations. In the case of Marylebone Road, the introduction of the bus lane appears to have had the relatively intuitive effect of reducing NO_x and NO₂ concentrations, but not affecting PM₁₀ or PM_{2.5}. The second measure, which may be connected to regenerating particle traps on London Buses, is more unexpected; a large increase in NO₂ concentrations, little change in NO_x or PM₁₀ and a decrease in PM_{2.5}. These step changes, and the reasons for them, are independent of trends caused by gradually introduced measures such as fleet renewal and increased market share of diesel vehicles.

Sustained step changes in NO₂ have been identified at two other UK monitoring sites – Oxford Roadside and Chiswick High Road, London. At each site, the signal is less defined than that of Marylebone Road. Local conditions should be investigated at each site to establish reasons for these changes. Further research is currently being carried out by ERG at King's College London on behalf of Transport for London into the effects of bus particle traps and the effects of the Congestion Charging Scheme on NO₂ concentrations at London roadside sites. However, more detailed analysis of the effects of regenerating particle traps on direct emissions of NO₂ is required, including NO/NO₂/PM emissions testing.

7 References

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