Air pollution and emissions trends in London

Sean Beevers, David Carslaw, Emily Westmoreland and Hrishi Mittal

King's College London, Environmental Research Group
Leeds University, Institute for Transport studies

April 2009

Authors	Sean Beevers, David Carslaw, Emily Westmoreland and Hrishi Mittal
Approved by Date Version	David Dajnak April 2009 V1.0

Table of Contents

1 Summary key points....5
2 Recommended further work....7
3 Context....9
4 Aims of the research....9
5 Methods....9
5.1 Part 1 - Meteorological Normalisation of London's air pollution data....9
5.1.1 Accounting for meteorology in pollutant trends....9
5.1.2 Approach used for meteorological-averaging....10
5.2 Part 2 - The development of an hourly emissions inventory for road traffic in London....12
5.2.1 Introduction....12
5.2.2 Approach used for creating hourly traffic data....13
5.2.3 Creating hourly emissions data....14
5.2.4 Traffic trends between 2003 and 2007....17
6 Results and Discussion....19
6.1 Trends in meteorologically-averaged of PM_{10} measurements....19
6.2 Trends in PM_{10} hourly emissions....23
6.3 Trends in meteorologically-averaged for NO_X measurements....32
6.4 Trends in hourly NO_X emissions....35
7 Marylebone Road....39
8 Conclusions....45
9 Recommended further work....46
10 References....47

1 SUMMARY KEY POINTS

1. This report summarises a detailed and comprehensive assessment of trends in emissions and concentrations of NO_X and PM in London.
2. We have developed methods to create hourly road traffic and emissions estimates between 2003 and 2007. This has used a comprehensive set of traffic data in London combined with new vehicle data based upon Automatic Number Plate Recognition (ANPR) cameras. The use of both traffic and emissions data as hourly values has proved to be invaluable in helping interpret the air pollution measurement trends.
3. Statistical methods have been developed to account for meteorological variation for assessing trends in the concentrations of pollutants. Meteorological variation can falsely mask or emphasise trends. The focus of these assessments has been to develop a better understanding of the traffic-related component of trends by considering roadside concentrations with background values removed.
4. The statistical models have been used to calculate trends, weekday and diurnal variations in roadside increments of NO_X, PM_{10} and $PM_{2.5}$ under average meteorological conditions. This approach has the benefit that trends calculated in this way will more closely relate to emissions changes rather than changes due to meteorological effects. The focus is on determining trends in roadside increments above local background values.
5. The emissions modelling results have been summarised into monthly trends between 2003 and 2007 as well as diurnal profiles by day of the week to compare with the measurement data. Furthermore the emissions and traffic data can provide a breakdown of the most important vehicle types.
6. We find there is evidence that NO_X concentrations have not decreased over the period 2003-2008. In fact, in central/inner London the mean of 10 roadside sites suggests that concentrations have *increased* by around 7 % from 2003-2008 (Table 1). In outer London, concentrations of NO_X have decreased by about 8-9 %. The corresponding changes in emissions are reductions of 20 and 29 % respectively. These results therefore show there seems to be a considerable disparity between the expected change in emissions and the actual change in concentrations. The results also suggest that there is a difference in how central/inner London sites have responded compared with outer London sites.
7. For PM_{10} the average of 7 central/inner London sites shows that concentrations have decreased by 4-5 % compared with an estimated 25 % from detailed emission calculations (2003-2008) (Table 1). Similarly, in outer London the analysis of PM_{10} concentrations indicates a reduction of 13-14 % compared with a 25 % reduction estimated for the emissions. It should be noted that given the complexity of PM emissions/concentrations that these results will be less certain than those for NO_X. Nevertheless, there does seem to be a consistent difference in the way that central/inner London sites respond compared with outer London site, which is similar to the findings for NO_X.

Table 1. The percentage change in emissions between 2003 and 2007[1]

Zone	Pollutant	All vehicles	Light vehicles[2]	HGV[3]	Taxis	Cars	Bus	LGV	Rigid	Artic
Central	NO_X	-20	-32	-10	-25	-41	-17	-15	-4	3
	Measured NO_X	+7.3								
Outer	NO_X	-29	-35	-25	-17	-41	-20	-21	-32	-30
	Measured NO_X	-8.5								
Central	Total PM_{10}	-25	-25	-27	-43	-23	-50	-14	-17	0
	Measured PM_{10}	-4.6								
	Exhaust PM_{10}	-37	-36	-38	-47	-39	-73	-23	-25	-5
	Tyre brake PM_{10}	-3	-7	14	-18	-10	7	15	17	30
Outer	Total PM_{10}	-25	-20	-38	-35	-20	-48	-18	-38	-17
	Measured PM_{10}	-13.5								
	Exhaust PM_{10}	-37	-30	-49	-40	-35	-73	-24	-45	-22
	Tyre brake PM_{10}	-4	-5	0	-9	-8	7	8	-13	9

[1] Measurements from 2003-2008, [2] Car + Taxi + LGV, [3] Bus + rigid HGV + articulated HGV

8. While there does appear to be a clear difference in the trends in PM_{10} and NO_X in central/inner London and outer London, it remains difficult to identify a specific cause(s). However, one of the major differences between these two areas is the proportion of diesel vehicles. Diesel vehicles are known to be significant emitters of both NO_X and PM_{10}. The detailed analysis of traffic data suggests that flows of large diesel vehicles (particularly Bus, HGVs and LGVs) have increased in central/inner London between 2003 and 2007 and that there has been a more mixed response at outer sites. Interestingly, total traffic has reduced for both groups of sites between 2003 and 2007.

9. We have also shown that considering how emissions and concentrations vary by day of the week and hour of the day is an effective way in which to compare emissions variation with that for concentrations. In particular, removing the meteorological signal can in some cases significantly affect the diurnal profile of concentrations and provide a more robust means of comparing trends with emissions estimates.

10. An approach using multiple regression analysis on a combination of traffic data and meteorologically averaged measurements by day of the week has demonstrated that it can provide the relative emission rates for different vehicle categories. The example given showed that an average emissions ratio of 11:1 for HGV (bus + rigid + articulated) to Light vehicles (car + taxi + LGV) would result in closer agreement between the emissions and air pollution daily profiles of PM_{10}. By comparison the ratio using the emissions calculation is 3.9. Further refinement of this method to consider larger number of vehicle types and across a number of years would provide a vital step in identifying the vehicles which may be the cause of the disparity between emissions trends and measurements.

11. The overriding conclusion of this work is, however, the clear disparity between the estimated trends in emissions and the observed trends in concentrations. This disparity has important implications for the management of air pollution and raises question about the adequacy of emission factors and projected concentrations of NO_X (and NO_2), and PM.

12. A useful side-effect of the meteorological averaging is that step-changes in the concentration of pollutants can be revealed. For example, there is evidence from this work that at Marylebone Road a distinct reduction in NO_X occurred in 2001 coinciding with the introduction of the bus lane. Such information could provide useful information concerning the effect that traffic management has in London (and elsewhere) that would normally be masked by meteorological variation.

2 RECOMMENDED FURTHER WORK

The analysis described in this document is the beginning of an important process of evaluation of emissions inventories that will enable greater understanding of emissions trends, air pollution model performance and better policy making. However, having created a large and unique dataset as well as important methods of analysis a number of additional work packages are recommended:

1. While there are many roadside air pollution sites in London, there are very few sites that have coincident traffic measurements. A more concerted effort to link quality road traffic measurements with air pollution sites would yield considerably more insight into these issues.
2. Important understanding of the inadequacies of emissions inventories can be made through the development of the multiple regression approaches, described in #10, to include additional vehicle types over a number of years.
3. In this study use was made of the currently available UK emissions factors, i.e. those based on results from Barlow et al., 2001. These have since been superseded via a consultation process undertaken by DfT during 2009. Initial results show that differences between the two emission factor datasets can be substantial for specific vehicles types. It would therefore be worth considering undertaking a recalculation of emissions using the new factors.
4. A separate research effort is being undertaken by ERG in order to monitor the effect of the LEZ in London. Part of this research is to establish a chemical speciation PM_{10} model and will include identifying road traffic emissions of PM and estimating trends over time. This provides a unique opportunity to build on the analysis presented here.

This page has been left blank intentionally

3 CONTEXT

There is increasing evidence available to suggest that particulate matter (PM), and to some extent nitrogen oxides (NO_X) concentrations have not decreased with expectations at many locations where road traffic is important. Emissions inventory calculations suggest that concentrations of these pollutants should have decreased during the last decade, but long-term trends at traffic-influenced sites suggest that NO_X and PM concentrations have remained static or have increased. PM in particular is a multiscale problem and is influenced by long range transport of secondary PM requiring mesoscale model predictions to improve understanding. However, for PM_{10} there is growing evidence that local emissions are increasing (Fuller and Green, 2006) and for NO_X the majority is locally derived. Hence it is important to establish the magnitude of any difference between emissions and air pollution trends and to understand whether data analysis can identify a vehicle type that plays an important role in this problem.

4 AIMS OF THE RESEARCH

There are a number of aims to the research, which include:

- The analysis of air pollution trends, at roadside sites, for NO_X, PM_{10} and $PM_{2.5}$ using methods to remove the influence of meteorology.
- To develop a method for the prediction of hourly emissions from road traffic and to establish the trend in emissions over periods coincident with the air quality trends.
- To summarise the data by hour of day and day of week by location (outer and central London) to establish differences between zones as well as a differences between the air pollution and emissions results.
- Finally, identify the areas where the emissions inventory and air pollution data show poor agreement and to establish a possible link between these periods and individual vehicle types.

5 METHODS

5.1 Part 1 - Meteorological Normalisation of London's air pollution data

5.1.1 ACCOUNTING FOR METEOROLOGY IN POLLUTANT TRENDS

It can be very challenging to analyse air pollution data due to the often strong influence that meteorology has. If one is interested in trends or how concentrations vary by hour of the day, then often meteorology can strongly influence observations. This is a problem if the interest is in knowing whether concentrations have decreased (or increased) over time because trends can be falsely masked or emphasised depending on the prevailing meteorology. Furthermore, if there is interest in knowing whether changes in concentration are in keeping with emissions estimates, it can be challenging to reconcile the two. Often, it is said that a particular year was 'good' or 'bad' with respect to concentrations. However,

rarely are such assertions based on an analysis of the causes of high or low concentrations. For this reason, an approach that is sometimes used is *meteorological normalisation*. The idea here is to 'account' for meteorological variation by building explanatory models for pollutant concentrations. If good models are developed i.e. they explain most of the variation in concentrations, it is then possible to use the model to predict concentrations over a fixed assumptions for meteorology – concentrations are *normalised*. The term used here is **meteorologically-averaged** because the predictions most closely relate to mean meteorological conditions.

The principal reason for accounting for meteorology in the current work is to allow for a better comparison with estimated emissions. This comparison focuses on several aspects of the temporal variation in concentrations, including the trend, diurnal and day of week effect. These periods have been chosen because they are able to highlight various aspects of the variation in emissions and concentrations. The long-term trend is a key consideration because it is important to know whether observed trends in concentration reflect that expected from an assessment of emissions. By considering variations by day of the week and hour of the day, it is possible to relate these variations to a more specific analysis of road traffic emissions. This is because vehicle emissions can have a strong day of the week and hour of day effect. In particular, different classes of vehicles show different behaviour over these time periods. HGVs, for example, tend to have much lower flows on Saturdays and in particular Sundays. Similarly, the variation in HGV flows often differs from that of cars by hour of the day.

A wide range of approaches have been adopted over the years to account for meteorology in trends. These include the use of linear models, neural networks (e.g. Gardner and Dorling (2000)) and non-parametric smoothing (e.g. Libiseller et al. (2005), Carslaw et al., (2007)). More recently an approach based on regression trees has been used to analyse air pollution at a location of high source complexity (Carslaw and Taylor, 2009). The regression tree approach has several advantages when applied to meteorological normalisation.

5.1.2 APPROACH USED FOR METEOROLOGICAL-AVERAGING

The selection of sites was primarily based on the availability of data for both NO_X and PM_{10} concentration measurements; both in terms of the number of years a site has been operating and its data capture. The focus in this work has been to consider the increment in concentration above background. For this reason all sites analysed were roadside or kerbside. Background concentrations (used for subtraction) were used from the North Kensington (KC1) site for central and inner London sites and the Bexley 2 (BX2) site for outer London sites. Several tests were made of different background site choices and the selection of these two sites was based upon data capture, location and appropriateness. In many situations there could be a legitimate choice of several sites. Tests were undertaken during model development to choose the site leading to the best model predictions. This was achieved by leaving a random selection of 50 % of the data out for model testing, where statistics such as the R^2 and residual sum of squares (RSS) could be calculated. In general it was found that KC1 and BX2 produced the best estimates. BX2 also has the advantage of having measurements of $PM_{2.5}$ – used in the analysis of Marylebone Road.

For modelling purposes hourly mean concentrations were used throughout; although the results were often averaged in some way to improve interpretation.

Meteorological data were used from the Met Office Heathrow site. A wide range of variables were assessed for modelling purposes including, for example: wind speed, wind direction, temperature, relative humidity and cloud amount. We also assessed many other variables such as precipitation amount and many measures of cloud type and amount. In general we found very little model improvement with the addition of variables beyond the 5 noted above. Our preference, therefore, was to develop the simplest good model rather than using many with little effect on model performance.

For model development a range of other variables were used including a trend term, seasonal and diurnal variation.

The modelling was conducted as follows. Data were processed to provide an hourly data set containing all variables of interest. Hourly background concentrations were subtracted from the corresponding roadside concentrations, yielding a data set of 'roadside increments'. The models developed related the roadside increment to a wide range of meteorological and other variable types in the form:

$$NO_{X\,increment} = \bar{u} + \phi + T + t_{trend} + t_{weekday} + t_{hour} + t_{JD} + \ldots$$

where the increment in NO_X is related to mean wind speed (u bar), wind direction (phi), temperature (T), and temporal terms.

The models were then used to predict mean concentrations assuming a fixed set of conditions for meteorology. Generally, mean or median (in the case of wind direction) values were used to derive a new time series with constant meteorology. Similar methods were used to show meteorologically-normalised diurnal and weekday effects.

To illustrate the principles of meteorological normalisation it is worth considering the diurnal variation in the concentration of a pollutant dominated by diffuse sources and strongly affected by the change in atmospheric mixing throughout each day. A good example of such a pollutant is ethane, whose emissions are dominated by natural gas leakage. Because of the nature of this source, the emissions tend not to vary (as far as we know) by hour of the day, except from a contribution due to vehicular and possibly some other minor sources. This means that the diurnal variation of ethane concentrations tend to relate strongly to atmospheric mixing: concentrations are highest when the boundary layer height is lowest (just before sunrise) and lowest when the boundary layer height, and the strength of atmospheric mixing, is highest i.e. typically mid-afternoon. This behaviour is shown very clearly in Figure 1.

Figure 1 also shows what the meteorologically-adjusted variation in ethane concentration, which differs in several ways, compared with the 'raw' data. First, there is a shift in the peaks (4am to 8-9am and midnight to 7-8pm). In this particular case it can be shown that

much of the remaining variation is due to variations in the boundary layer height, which are not captured using these techniques.[1] The other characteristic to note is that the amplitude of the variation decreases. Again, this is expected because much of the peak to trough variation is driven by meteorology. The focus of the current work is accounting for *local* meteorological variation using increments in roadside concentrations and thus `bulk' properties such as boundary layer height become much less important and local mixing and thermal turbulence more so.

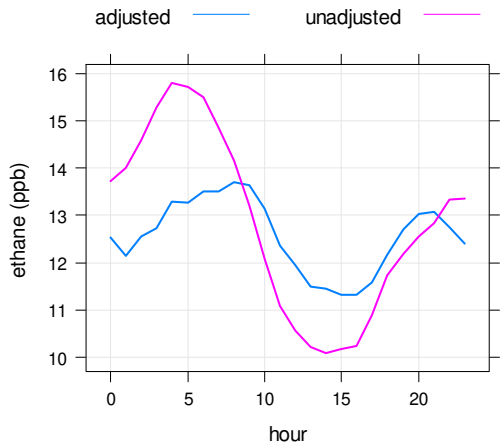


Figure 1. Diurnal variation in ethane concentrations at Marylebone Road. The 'raw' (unadjusted) data is shown by the magenta line and the meteorologically adjusted data by the cyan line.

The results shown for ethane also demonstrate the value when applied to other, more vehicle-related pollutants. The diurnal variation in NO_X, for example, will be affected by the same processes as that shown for ethane. If we want to know whether the diurnal variation is most affected by a particular class of vehicle (with a known variation) it is not easy to compare directly the concentration measurements with the emissions. If, however, the effect of meteorology is first accounted for, then the comparisons become more meaningful, which is the essence of this work.

5.2 Part 2 - The development of an hourly emissions inventory for road traffic in London

5.2.1 Introduction

To date emissions inventories in the UK have provided annual emissions using vehicle flow data which is based upon an typical day during each year, an estimate of vehicle stock, which is based upon UK vehicle licensing results at the year end and average vehicle speed by location. Enhancements to the models in London have included calculations of emissions for each hour of an average day using speed that also varies by hour, the use of 'on-road' vehicle stock based upon Automatic Number Plate Recognition (ANPR) cameras and specific fleet information for certain London fleets. Whilst emissions models have played an important role in understanding and developing policy to tackle air pollution this has been

[1] More comprehensive meteorological measurements would be required to capture these effects in a model.

via the use of dispersion models, which are themselves highly uncertain. Despite this they have remained the link between emissions and air pollution concentrations, with only a small number of researchers having compared air pollution measurement data directly with emissions calculations (Parrish, 2006).

Therefore, to make emissions estimates and meteorologically-averaged air quality data compatible an important next step is to increase the time resolution to hourly results. This is limited due to the lack of traffic count data associated with air pollution measurement sites and as a consequence we have developed methods to create modelled hourly traffic flows using surrogate datasets.

5.2.2 APPROACH USED FOR CREATING HOURLY TRAFFIC DATA

The approach to develop an hourly emissions estimate for road traffic used a combination of data, represented in Figure 2. The basis of the calculation was a 'London averaged' hourly traffic file based upon an average of Automatic Traffic Count (ATC) sites in central London, running between March 2003 and the end of 2007. The London average ATC data was assessed using a Generalised Additive Modelling (GAM) technique to estimate whether a long term trend existed in the data. GAMs are an extension of linear regression models and are useful when the relationships between variables are non-linear - as is the case here. It has been used extensively and in Carslaw et al., (2007) was used to assess trends in traffic related emissions. The GAM modelling established that total hourly traffic counts could be described using smooth functions of hour of day, day of week, season and trend and that on average these factors could account for R^2 values ~ 0.9. Furthermore, there was no significant long-term trend, thus avoiding any problems associated with introducing an artificial trend into the data between 2003 and 2007.

To calculate total traffic flows along individual roads the 'London averaged' data was scaled using manual classified count (MCC) data taken during weekday periods (7am to 7pm). Unlike the ATC data, manual counts are widespread and cover all of the major roads in London. This means that where a MCC count exists a specific hourly traffic file can be calculated. Since the manual count data is taken infrequently a number of tests were undertaken to compare MCC and ATC data taken over the same 12 hour period as well as for longer periods of the year. Furthermore, since manual count measurements may be highly variable due to specific local events the time series of these data was smoothed using a local regression (LOESS) smoothing function or where few measurements were taken an average of the data was used. For a detailed description of LOESS see Jacoby (2000). Finally, when rescaling the ATC data care was taken to maintain daytime and night time differences in vehicle flow as well as weekend totals.

The MCC data are classified into 11 vehicle types and these were used to split the total vehicle count for each hour of the day. Less data were available during weekday overnight periods, Saturdays and Sundays. Here a combination of datasets were used, including a set of MCC counts taken over a complete 24 hour period and Automatic Number Plate Recognition (ANPR) camera data. The former provided average proportions by vehicle type

during the overnight minimum traffic periods and a combination of ANPR and weekday MCC data was used to apportion the weekend periods.

The results of the total vehicle counts was tested against a separate data set of ATC data recorded by DfT during 2006 and 2007 and not used in the model development. These tests were made for a combination of 16 site years and are presented as a predicted and measured profile, averaged over all sites and by day of the week (Figure 3). Alongside these results a summary of bias and the RMS error has also been presented (Table 2). Comparison of the predicted total (red), measured ATC total (blue) and the residual, (predicted-measured in light green) across an average of all sites show that the methods for creating vehicle totals are robust and provide good results across all days of the week.

The results in Table 2 show that over a 24 hour period very little bias exists in the predictions and in all cases is below 7%. Lack of data during weekends is apparent and as a consequence the method has the poorest performance during Saturdays. Because manual count data is specific to each site the 12 hour weekday periods should have the lowest uncertainty and for these separate results are presented. Here too modest bias estimates are apparent with average values of the order of -5%. Given that these periods are also associated with vehicle proportions by hour means that there is some confidence in using these data in comparison with the meteorologically-averaged measurements. Furthermore, Sunday, is well predicted for total vehicles and is widely understood to have small proportions of HGVs so is also a period where the traffic and emissions data is relatively robust. Overall the predicted average data has a RMS error of ~ ±10%.

Finally, vehicle speed is applied by hour of the day based upon average hourly results over a number of Moving Car Observer surveys during weekdays as well as more limited datasets over night. Saturday speeds are assumed to be similar to weekdays because of similar traffic flows. For Sundays inter-peak daily speeds were used during 7am – 7pm along with the same overnight speeds as the rest of the week.

5.2.3 CREATING HOURLY EMISSIONS DATA

The same method as that described in the manual of the London Atmospheric Emissions Inventory (LAEI) was used in the creation of the hourly emissions data. The reader is referred to Mattai and Hutchinson, 2006 where the detailed model assumptions are described. The application of vehicle stock, which would normally be applied as a single estimate each year, was instead applied monthly by interpolating the annual figures.

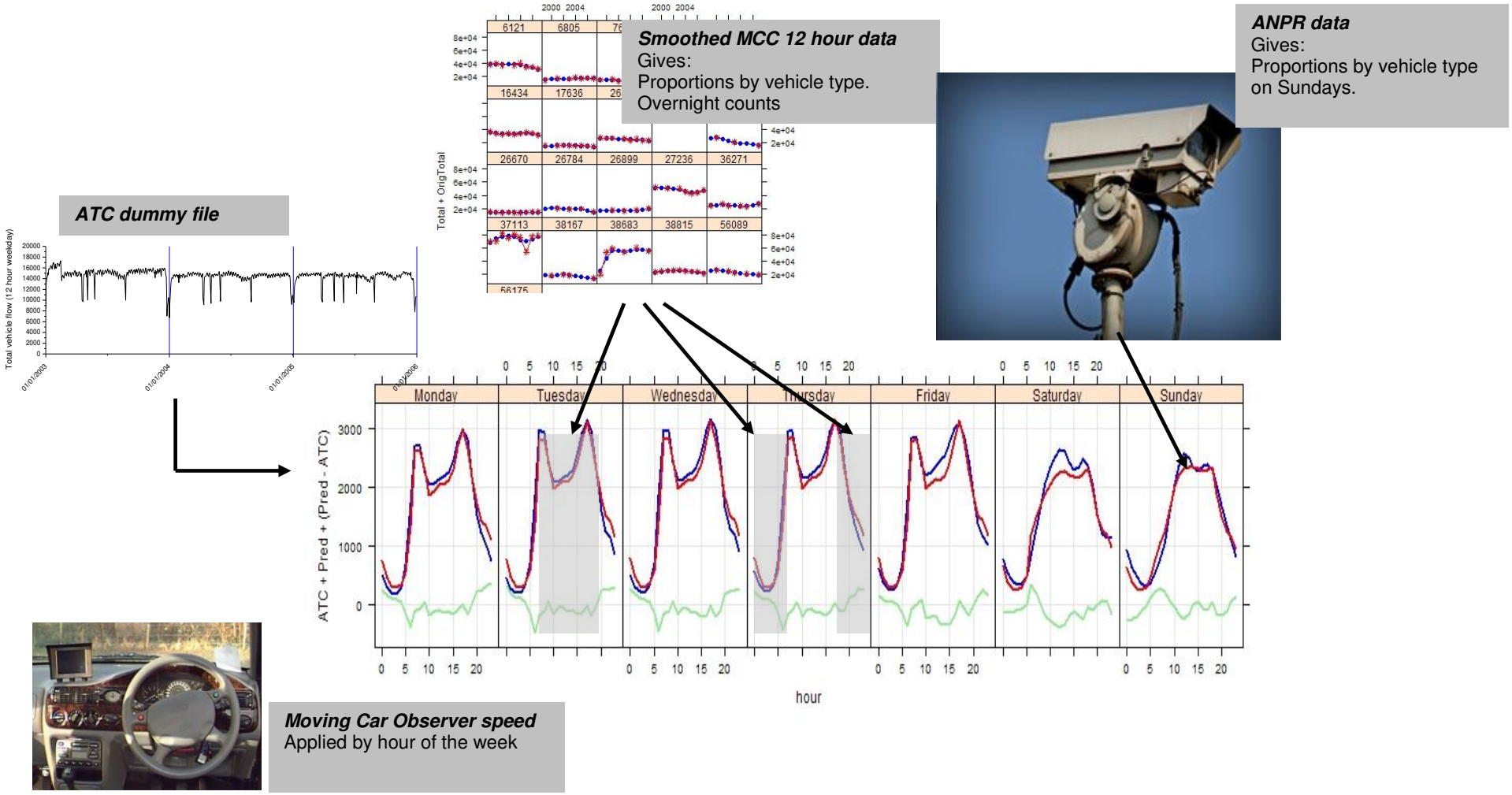


Figure 2. Application of the data in developing the 5 year hourly traffic file

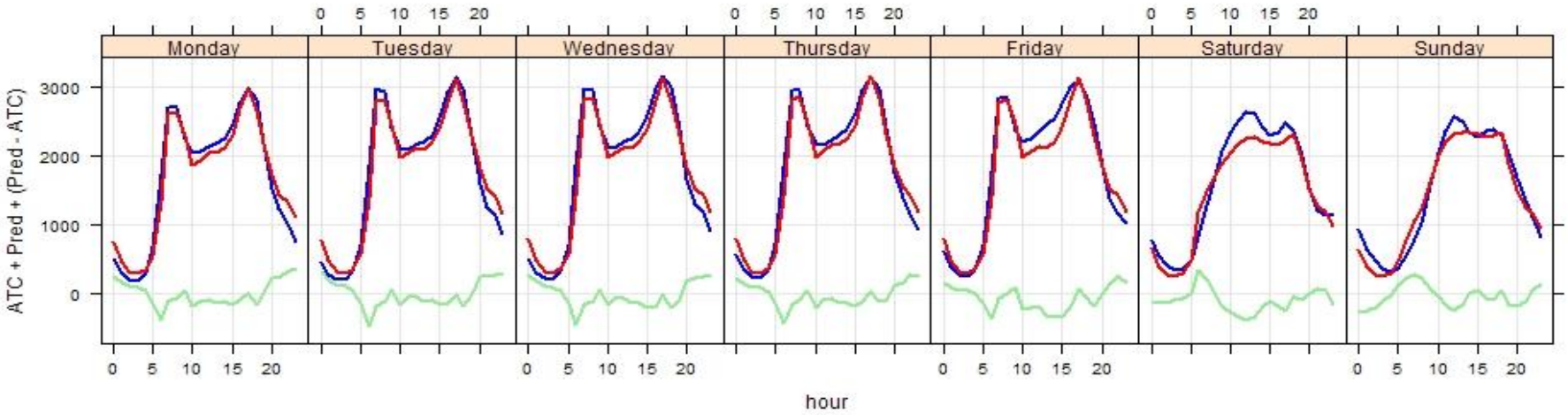


Figure 3. Comparison of predicted (red), measured (blue) and predicted-measured (light green) hourly total traffic flows. 16 site and year combinations for 2006 and 2007.

Table 2. Summary statistics from Figure 3

	Monday	Tuesday	Wednesday	Thursday	Friday	Saturday	Sunday	All days (RMS error)
Predicted	1659	1741	1753	1770	1753	1466	1419	
Measured	1657	1740	1773	1792	1820	1569	1461	**10.7%**
Bias (%) (pred-measured/meas)	**0.1%**	**0.1%**	**-1.1%**	**-1.2%**	**-3.6%**	**-6.6%**	**-2.9%**	(% of mean (measured))
Predicted (12 hrs)	2339	2455	2472	2495	2471	2066	2023	
Measured (12 hrs)	2438	2553	2588	2597	2625	2243	2036	**6.7%**
Bias (%)(pred-measured/meas)	**-4.1%**	**-3.9%**	**-4.5%**	**-3.9%**	**-5.8%**	**-7.9%**	**-0.6%**	(% of mean (measured))

5.2.4 TRAFFIC TRENDS BETWEEN 2003 AND 2007

A brief summary of road traffic changes between 2003 and 2007, for the sites used in the PM_{10} analysis described in chapter 6 show that total vehicles reduce by -8% at the central sites (Table 3) and –6% at the outer sites (Table 4). Furthermore, there is growth in the larger diesel vehicles at the central sites; bus (+8%), LGV (+20%), Rigid HGV (+20%) and Articulated HGV (+30%) (Table 3 and Figure 4) and in contrast a mixture of changes at the outer sites; bus (+18%), LGV (+12%), Rigid HGV (-17%) and Articulated HGV (+5%) (Table 4 and Figure 4).

Table 3 Traffic changes at the Central PM_{10} sites

Year	Cars	Bus	LGV	Rigid HGV	Articulated HGV	Total
2003	32072	1146	5580	1280	163	40241
2004	31062	1221	5466	1364	166	39279
2005	30006	1258	5648	1429	176	38517
2006	28709	1263	6055	1489	192	37708
2007	27246	1235	6682	1533	212	36908
Total change 2003 - 2007	-4826	89	1102	253	49	-3333
Percentage change 2003 - 2007	-15	8	20	20	30	-8

Table 4 Traffic changes at the Outer PM_{10} sites

Year	Cars	Bus	LGV	Rigid HGV	Articulated HGV	Total
2003	21850	750	4511	1074	331	28516
2004	22061	795	4635	1005	341	28837
2005	21851	831	4776	948	346	28752
2006	20908	867	4918	916	348	27957
2007	19625	888	5061	891	346	26811
Total change 2003 - 2007	-2225	138	550	-183	15	-1705
Percentage change 2003 - 2007	-10	18	12	-17	5	-6

Trends in traffic counts are also summarised in Figure 4, for both NO_X and PM_{10} sites separately and although there are small differences in HGV trends, overall both groups of sites show similar trends over the period.

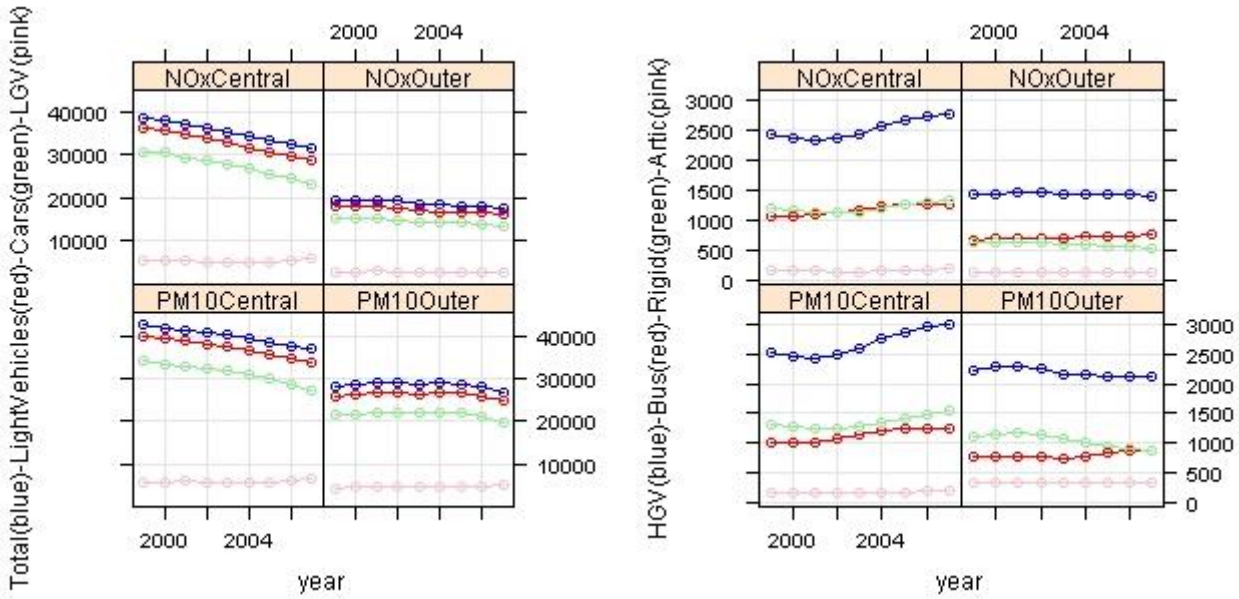


Figure 4. Vehicle flow trends (1999 – 2007) for all central/inner and outer sites. Note the number of sites used in the analysis of PM_{10} and NO_X is different. Total = HGV + LV, LV = Cars + LGV, HGV= Bus + Rigid + Articulated HGV. Note taxis are included in the car category.

6 RESULTS AND DISCUSSION

6.1 Trends in meteorologically-averaged of PM_{10} measurements

The sites analysed have been split into two broad categories: central/inner London and outer London. This area is approximately defined as being separated by the North/South Circular. This split was made in order to understand concentration observations behave differently. Central/inner London has a higher proportion of diesel vehicles and slower vehicle speeds compared with outer London, for example.

Developing models for particle concentrations is challenging for several reasons. First, there are many more sources of particles in the atmosphere than road transport – even when considering roadside increments. Examples of other sources include road and building works and non-exhaust vehicle emissions. There are also difficulties due to knowing precisely what an instrument actually measures. In principle, sophisticated models could be developed for particle concentrations, if there was sufficient knowledge of the sources etc. However, for almost all the measurements processed for this report, such additional information is not available. Occasionally it is known, for example, that concentrations during a particular period were affected by a specific event, but more generally this information remains an unknown.

Models developed for particle concentrations will therefore be less robust than those for the primary inorganic gases. This incomplete knowledge of particle concentrations is generally manifest in the models in two ways: increased noise in the predictions and additional features in the trend term. The latter tends occurred where, for example, there was an extended period of 'anomalous' measurements i.e. a month of building works. Nevertheless, good models with R^2 values around 0.6 for *hourly* measurements of PM_{10} were developed[2].

The sites analysed are shown in Table 5. Compared with NO_X there were fewer sites available to analyse. However, there were a sufficiently large number to categorise them by area of London.

Table 5 Sites used for the analysis of PM_{10}.

LAQN Site code	Site Name	Area
MY1	Marylebone Rd	Central/inner
CD3	Camden (3)	
KC2	Kensington & Chelsea 2	
CD1	Camden 1	
IS2	Islington 2	
LB1	Lambeth 1	
HF1	Hammersmith and Fulham 1	

[2] Remembering that the models were assessed using data independent of that used to develop the models in the first place.

RI1	Richmond 1	
HS4	Hounslow 4	
EN2	Enfield 2	
CR4	Croydon 4	
HG1	Haringey 1	
A30	A3 roadside	
BY7	Bromley 7	Outer London
EA2	Ealing 2	
GR5	Greenwich 5	
GB6	Greenwich-Bexley 6	

For sites in central and inner London (Figure 5) there is some evidence that traffic related PM_{10} concentrations have *increased* since 1998. Compared with the NO_X results (see next section), there is less certainty in the trend for the reasons already mentioned. Nevertheless, it is also clear from Figure 5 that that concentrations of PM_{10} have not decreased over this period.

The situation in outer London is more mixed (Figure 6), but there is more evidence of a decrease since 1998. Even in outer London though, there has been no clear trend in PM_{10} concentrations since around 2000.

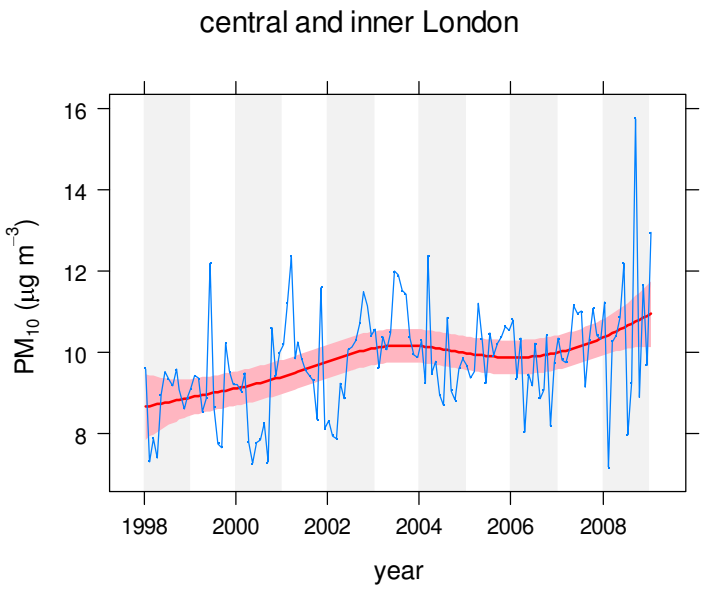


Figure 5. Meteorologically-averaged trend in the roadside PM_{10} increment, averaged for central and inner London. The red line shows a smooth fit to the data and the uncertainty (95 % confidence interval) is shown by the shading.

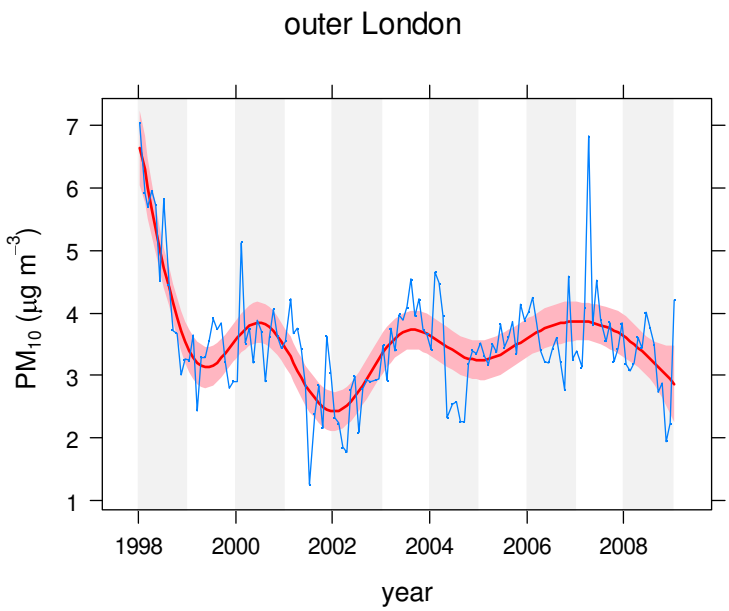


Figure 6. Meteorologically-averaged trend in the roadside PM_{10} increment, averaged for outer London. The red line shows a smooth fit to the data and the uncertainty (95 % confidence interval) is shown by the shading.

The diurnal variation in meteorologically-averaged PM_{10} reveals a different behaviour in central/inner London cf. outer London. In Figure 7 it is clear that PM_{10} concentrations tend to peak in late morning/midday in central/inner London, but later in the day in outer London (Figure 8). Another important difference is the timing of peaks on Saturdays and Sundays: in inner/central London PM_{10} concentrations tend to peak earlier in the day than in outer London. This contrast in the timing (and shape) of the diurnal/day of week PM_{10} concentration pattern between the two areas of London should provide clues as to the types of vehicle that dominate the concentrations.

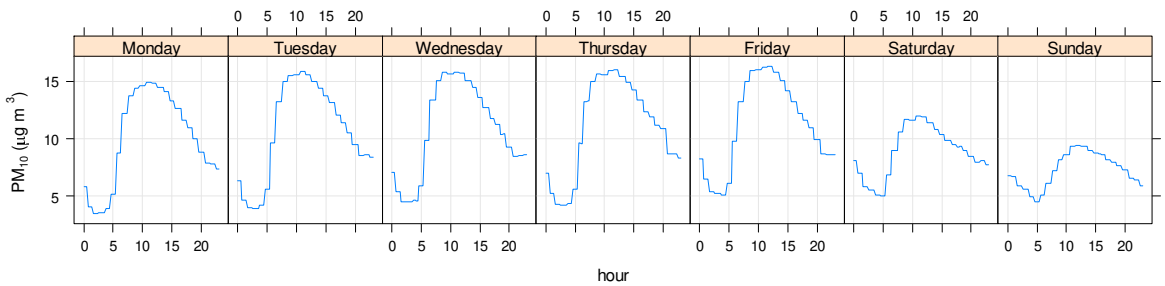


Figure 7. Diurnal and day of the week variation of meteorologically-averaged roadside increment PM_{10} concentrations averaged across the central/inner London sites.

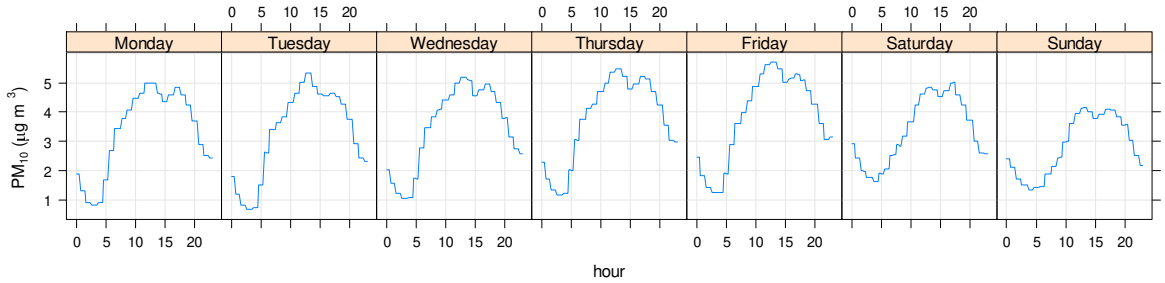


Figure 8. Diurnal and day of the week variation of meteorologically-averaged roadside increment PM_{10} concentrations averaged across the outer London sites.

The results for central/inner London suggest that PM_{10} concentrations may have increased in recent years and hence it is worth considering whether the trends differ by day of the week. Because there are far fewer HGVs on Sundays, for example, if there was an important effect by vehicle type it may be seen by plotting the trends in this way. Figure 9 shows the trends by day of the week for central/inner London sites. These results do not seem to suggest that the trends differ in overall shape by day of the week. It may tentatively be concluded that there does not seem to be a strong vehicle type effect that influences these trends. These results are discussed further in the section considering NO_X concentrations.

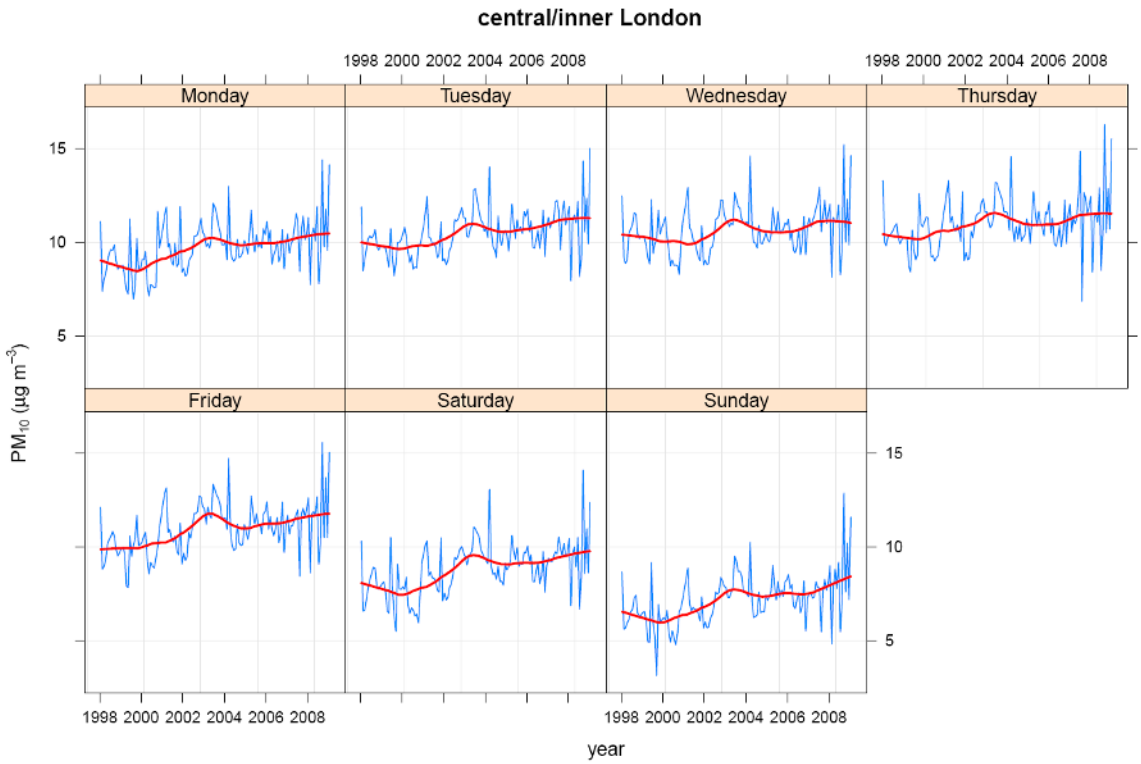


Figure 9. Trends in meteorologically-averaged roadside increment PM_{10} concentration averaged across the central/inner London sites by day of the week.

6.2 Trends in PM_{10} hourly emissions

Because of the limited availability of traffic data, hourly emissions estimates were calculated over a shorter period than for the air pollution measurements. Emissions were calculated for NO_X, Total PM_{10}, PM_{10} exhaust and PM_{10} tyre and brake wear, separately between early 2003 to the end of 2007 and for the collection of central and outer sites used in the measurement analysis[3]. The emissions were then summarised into monthly totals to provide trends over the period and into hourly average values summarised by day of the week and as diurnal profiles.

The average emissions across central and outer sites for 2003 and 2007 show that for total PM_{10} light vehicles dominate and at the central sites cars, taxis and LGV's make up 75% of emissions with HGV's making up ~ 25% (Table 6). At the outer sites taxis are much less important but light vehicles still represent 69% and 74% of the emissions in 2003 and 2007, respectively. Furthermore, by 2007 exhaust PM_{10} makes up approximately half (53%) of total PM_{10} (see Table 7).

Table 6. The percentage contribution to average emissions, by vehicle type, for 2003 and 2007

Zone	Pollutant	Year	Light vehicles	HGV	Taxis	Cars	Bus	LGV	Rigid	Artic
Central	Total PM_{10}*	2003	74	26	16	38	9	21	13	3
		2007	75	25	12	39	6	24	15	5
	Exhaust PM_{10}	2003	69	31	20	25	10	24	16	5
		2007	70	30	17	24	4	29	19	7
	Tyre brake PM_{10}	2003	83	17	7	63	8	14	7	1
		2007	81	19	6	58	9	17	9	2
Outer	Total PM_{10}	2003	69	31	4	39	12	26	12	7
		2007	74	26	4	42	8	28	10	7
	Exhaust PM_{10}	2003	63	37	6	26	13	31	16	9
		2007	70	30	6	27	5	38	14	11
	Tyre brake PM_{10}	2003	80	20	2	63	11	15	7	3
		2007	79	21	2	60	12	17	6	3

* Total PM_{10} = Exhaust PM_{10} + Tyre Brake PM_{10}

Table 7. The percentage of total PM_{10} emissions by source type

Year	Exhaust	Tyre and brake
2003	64	36
2004	61	39
2005	58	42
2006	56	44
2007	53	47

Comparisons between the trends in monthly emissions between 2003 and 2007 (Figure 10), and the meteorologically-averaged trends of Figure 5 and Figure 6, are in poor agreement and over comparable periods the emissions trends show a decline of 25%. The emissions of

[3] The emission factors used were based upon Barlow et al., 2001

total PM_{10} (exhaust + tyre and brake wear) have been split into separate categories; 'all vehicles', light vehicles (cars + taxis + LGVs) and HGVs (bus + rigid HGVs + articulated HGVs) and are also summarised in Figure 10. From this it can be seen that the emission trends are driven predominantly by the light vehicle category. Figure 11 and Figure 12 are also instructive as they break total PM_{10} into 2 components, exhaust and tyre and brake wear and this shows that the total PM_{10} trend is driven by the reduction in vehicle exhaust rather than tyre and brake wear, whose change is an order of magnitude smaller. For a complete breakdown of the emissions changes (and the equivalent measurement changes), by location and vehicle type, see Table 8.

Table 8. The percentage change in PM_{10} emissions and PM_{10} measurements between 2003 and 2007[1]

Zone	Pollutant	All vehicles	Light vehicles[2]	HGV[3]	Taxis	Cars	Bus	LGV	Rigid	Artic
Central	Total PM_{10}	-25	-25	-27	-43	-23	-50	-14	-17	0
	Measured PM_{10}	-4.6								
	Exhaust PM_{10}	-37	-36	-38	-47	-39	-73	-23	-25	-5
	Tyre brake PM_{10}	-3	-7	14	-18	-10	7	15	17	30
Outer	Total PM_{10}	-25	-20	-38	-35	-20	-48	-18	-38	-17
	Measured PM_{10}	-13.5								
	Exhaust PM_{10}	-37	-30	-49	-40	-35	-73	-24	-45	-22
	Tyre brake PM_{10}	-4	-5	0	-9	-8	7	8	-13	9

[1] Measurements from 2003-2008
[2] Car + Taxi + LGV
[3] Bus + rigid HGV + articulated HGV

The disparity between the emissions trends and those from the meteorologically-averaged measurements could not be clearer, with the emissions calculations showing a decrease of 25%, whereas the measured reductions are 5% at the central sites and 14% at the outer sites. As such the emissions inventory calculations present an optimistic view of change with the improved emissions performance of the vehicle fleet driving down emissions and the non-exhaust emissions sources such as tyre and brake wear remaining constant.

What is harder to establish from the trend data is whether the vehicle exhaust emissions are overly optimistic or whether other sources such as tyre and brake wear and/or resuspension are not stable, but increasing over the period. The component of emissions due to resuspension is not included in the emissions calculation due to lack of published evidence of a significant contribution to PM emissions from roads in London (Harrison et al., 2004 and Abbott, 2007). However, resuspension would almost certainly be reflected in changes to vehicle flow and within the modelling undertaken we have established the vehicle flow trends.

The trend in flows for all roads used in the emissions calculation is given in Figure 4. At some sites where the number of counts made during the study period was limited an average total vehicle flow was used, and a zero trend was assumed. The trend analysis shows that overall the manual count data does have a downward trend for light vehicles at central sites and a near zero trend at outer sites. At the central sites there is an increase in heavy

vehicles after 2002 and at outer sites again there is a near zero trend. In combination the vehicle km assumed in the emissions calculations would have fallen –6% to -8% (Table 3 and Table 4). Hence the fact that PM_{10} meteorologically-averaged concentrations are stable or increasing means that resuspension could only play an important role through a change in vehicle type, for example more LGV/heavy vehicles at the central sites. Alternatively, the influence of meteorological conditions could have played a role, however 2007 was exceptionally wet yet there is little evidence that PM_{10} measurements reduced during that year and in fact at the central sites they continued to increase. All of this points to their being a minor role for emissions associated with resuspension and although it cannot be ignored the focus of our initial analysis has remained elsewhere.

Comparison between the diurnal profiles of PM_{10} emissions summarised in Figure 13 with the meteorologically averaged equivalent graphs in Figure 7 and Figure 8 reveal very different results. What is most striking is the shape of the emissions curves. The emissions results rise to an early peak period (from 6am) and are then reasonably constant throughout the day. This is in contrast to Figure 7 and Figure 8, which is much more rounded and has no distinct morning or evening peak periods and a maximum at around 10am. There is also a difference between central and outer sites with the latter peaking approximately 1 hour later. The emissions results are also divided into contributions from light and heavy vehicles. Weekday heavy vehicle numbers peak at ~10am and fall steadily towards the evening. In addition, heavy vehicles make a small contribution to emissions on Sunday and so comparison of trends by day has the potential to establish the change in emissions by vehicle type.

By comparing the average PM_{10} concentrations for Sunday's and weekends in Figure 9 with their equivalent light and heavy vehicle flows it has been possible to establish the relative emission rates of the two vehicle categories, Light vehicles and HGVs. These data have been summarised into two simultaneous equations and solved to give relative emission rates for each vehicle category. This gives an emissions ratio of approximately 11:1 for HGVs:Light vehicles and does not agree well with the equivalent ratio from vehicle emissions inventories of ~ 4:1. Using the alternative 11:1 ratio to recreate the diurnal profile of emissions (Figure 14) begins to show closer agreement with the meteorologically-averaged PM_{10} concentrations in Figure 9. It also demonstrates that either the HGV emissions rates should be higher or that light vehicle emissions rates should be lower, or a combination of the two. This method has the potential to establish more clearly the limitations of emissions inventories for road transport, but only after further development of the technique to encompass different vehicle types and different years within the time series.

This page has been left blank intentionally

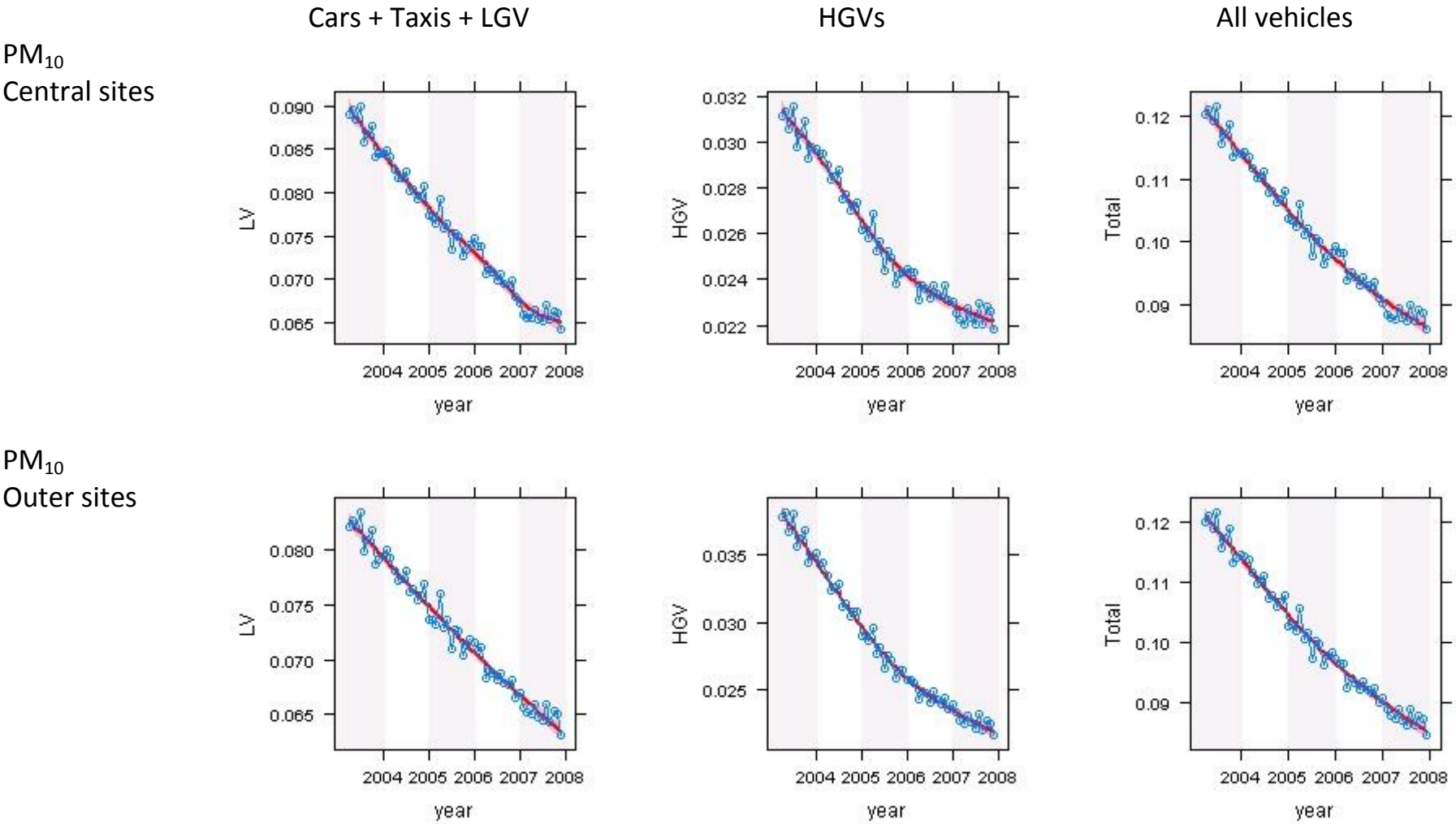
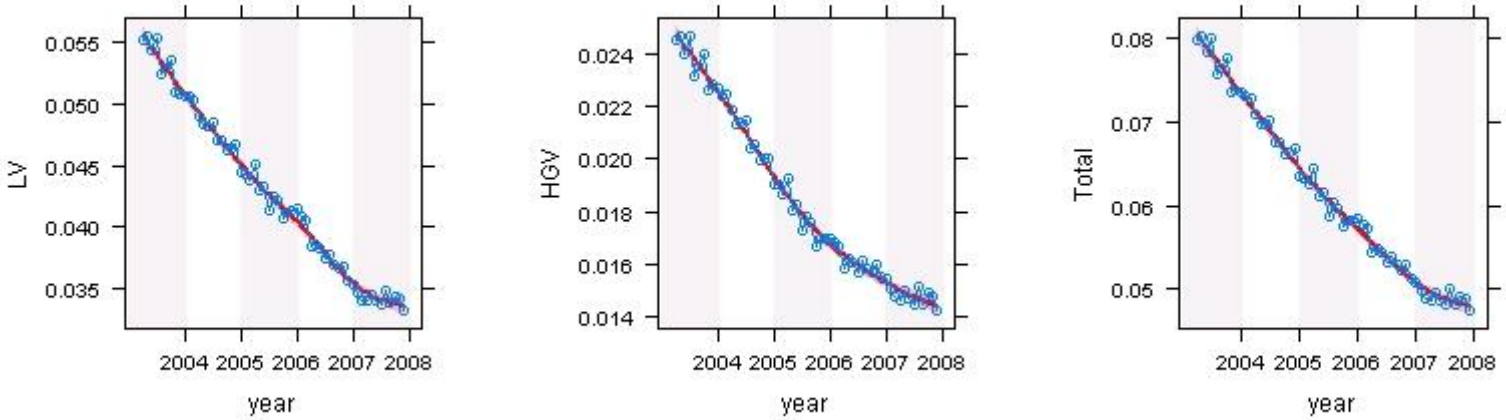


Figure 10. Monthly PM_{10} emission trends for central and outer roads (tonnes - exhaust + tyre and brake wear). The red line shows a smooth fit of the data and the uncertainty (95% confidence interval)

Cars + Taxis + LGV | HGVs | All vehicles

PM_{10} exhaust
Central sites



PM_{10} exhaust
Outer sites

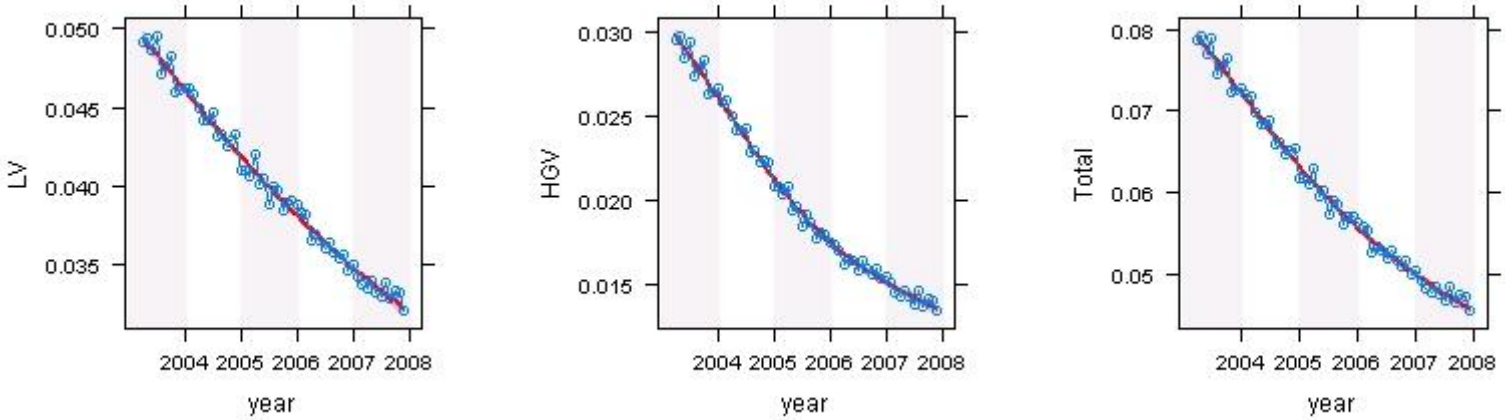


Figure 11. Monthly PM_{10} emission trends for central and outer roads (tonnes - exhaust). The red line shows a smooth fit of the data and the uncertainty (95% confidence interval)

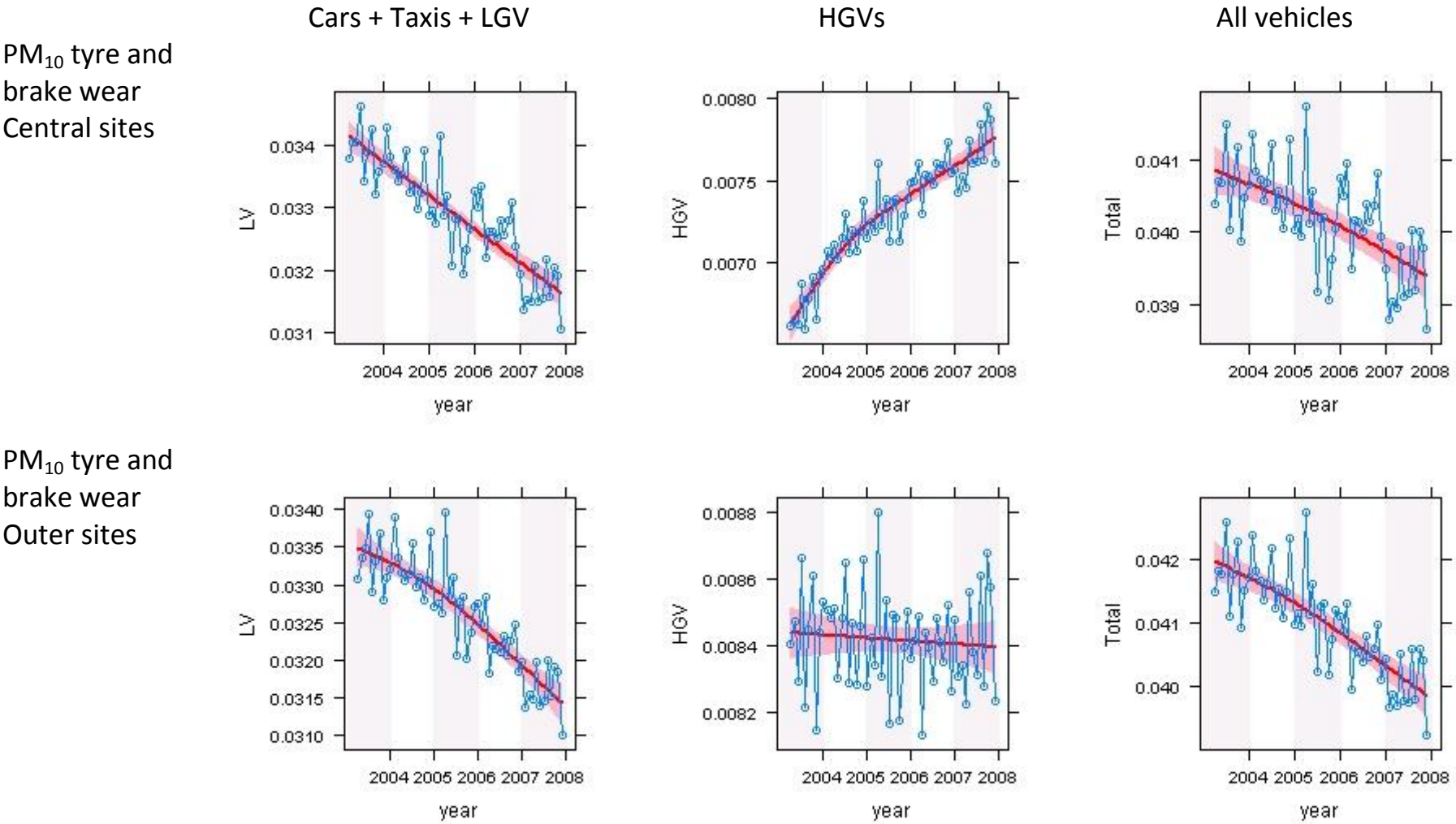
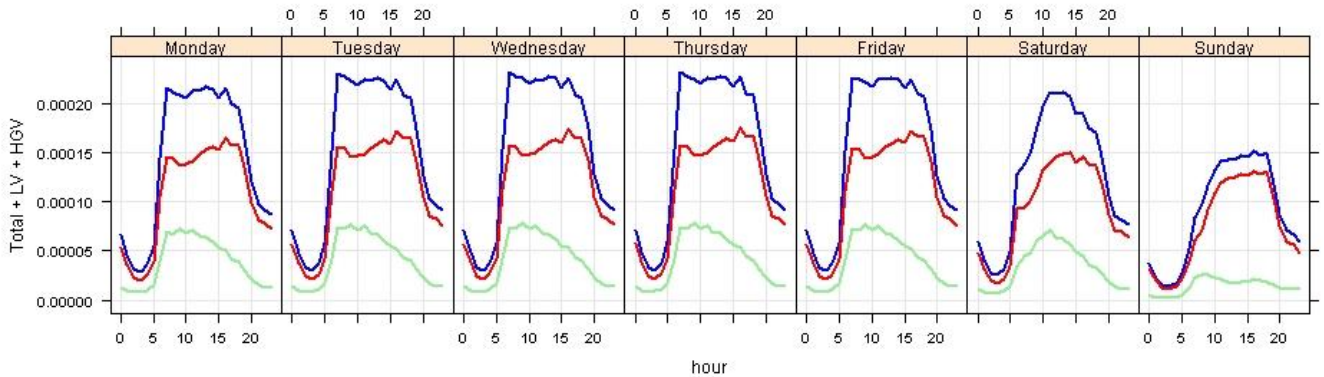


Figure 12. Monthly PM_{10} emission trends for central and outer roads (tonnes - tyre and brake wear). The red line shows a smooth fit of the data and the uncertainty (95% confidence interval)

PM_{10} central



PM_{10} Outer

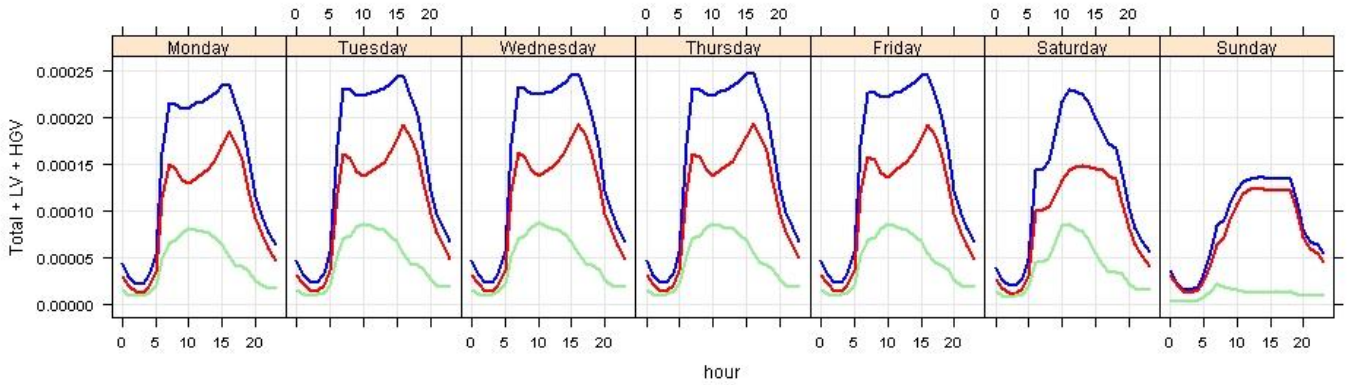


Figure 13. Diurnal PM_{10} emissions (exhaust + tyre and brake wear) for central and outer sites (tonnes/hr). The blue line shows the total PM_{10} emissions, the red line emissions from light vehicles (cars+taxis+LGVs) and the green line emissions from Heavy vehicles (HGVs + Buses).

PM_{10} central

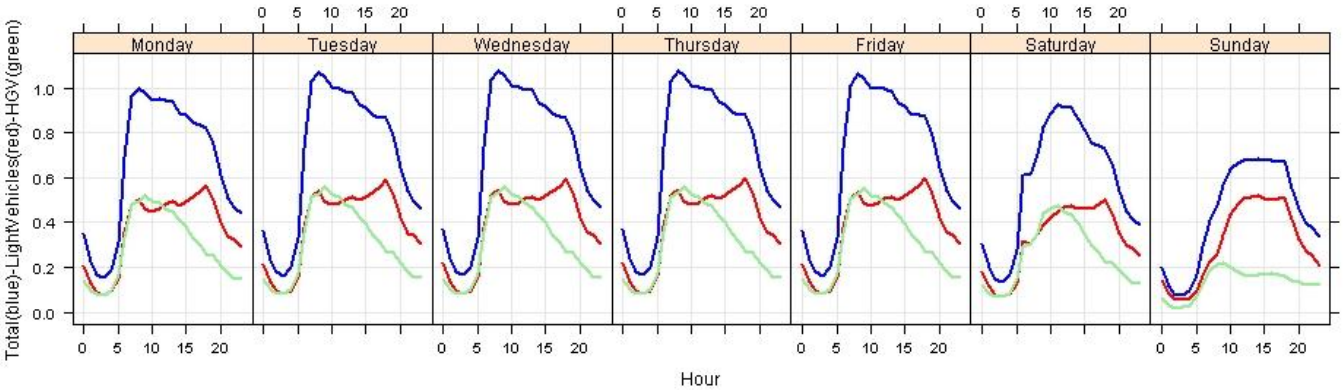


Figure 14. Diurnal PM_{10} emissions (exhaust + tyre and brake wear) for central/inner sites (tonnes/hr). The blue line shows the total PM_{10} emissions, the red line emissions from light vehicles (cars+taxis+LGVs) and the green line emissions from Heavy vehicles (HGVs + Buses).
The figure uses a HGV/LV PM emissions ratio of 11 calculated using a comparison between weekday and Sunday average meteorologically averaged PM_{10} concentrations and traffic counts. The resulting plot is in closer agreement with Figure 7. By comparison the actual ratio in the emissions inventory is 3.9.

6.3 Trends in meteorologically-averaged for NO_X measurements

The mean trend in NO_X calculated across all central/inner London sites is shown in Figure 15. It is clear that concentrations decreased markedly from 1998-2002. However, concentrations have changed little since 2002 and in fact since around 2006 there is some indication that they are *increasing*.

Table 9. Sites used for the analysis of NO_X.

LAQN Site code	Site Name	Area
MY1	Marylebone Rd	Central/inner
CD3	Camden (3)	
KC2	Kensington & Chelsea 2	
KC3	Kensington & Chelsea 3	
CD1	Camden 1	
KC4	Kensington & Chelsea 4	
IS2	Islington 2	
LB1	Lambeth 1	
HF1	Hammersmith and Fulham 1	
WA4	Wandsworth 4	
RB3	Redbridge 3	Outer London
RI1	Richmond 1	
HS4	Hounslow 4	
HV1	Havering 1	
EN2	Enfield 2	
CR2	Croydon 2	
CR5	Croydon 5	
HG1	Haringey 1	
A30	A3 roadside	
BY7	Bromley 7	
CR4	Croydon 4	
EA2	Ealing 2	
GR5	Greenwich 5	

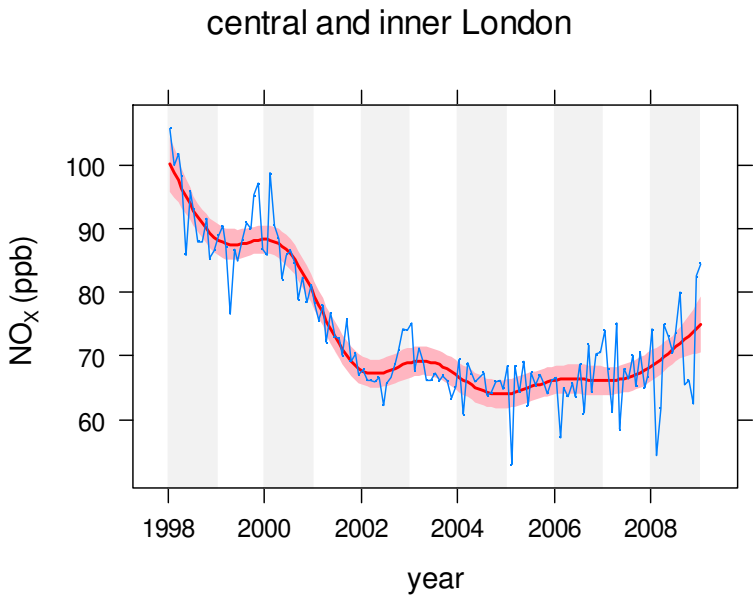


Figure 15. Mean normalised trend in the roadside NO_X increment averaged for central and inner London. The red line shows a smooth fit to the data and the uncertainty (95 % confidence interval) in this fit shown by the shading.

The situation in outer London differs in several important ways compared with central/inner London. First, the decrease in NO_X has been relatively smooth over time and is monotonic. Second, there is less evidence that concentrations have increased in recent years. Nevertheless, concentrations have remained approximately level over the past 5 years. It is clear comparing Figure 15 and Figure 16 that there do appear to be important differences in how concentrations of NO_X have changed in these two areas. Comparing the concentrations at the beginning of 1998 and the end of 2008, concentrations in outer London decreased by about 40 %. By contrast the decrease in central/inner London was around 30 %.

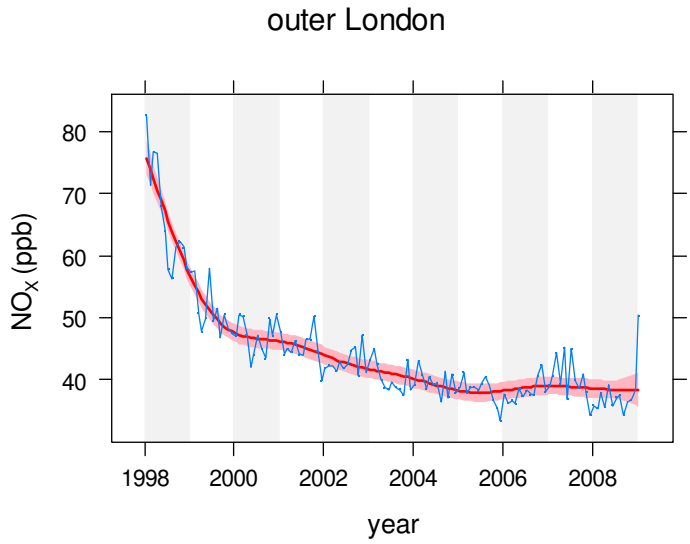


Figure 16. Mean normalised trend in the roadside NO_X increment averaged for outer London. The red line shows a smooth fit to the data and the uncertainty (95 % confidence interval) in this fit shown by the shading.

It is also useful to consider how the normalised concentrations vary by hour of the day and day of the week. For central/inner London (Figure 17) concentrations tend to increase with the morning rush hour, remain high before decreasing at the evening rush hour – for weekdays. This behaviour contrasts somewhat with the outer London sites (Figure 18) where there is evidence of a morning and evening rush hour peak. Both areas show a similar response at weekends: concentrations on a Saturday are lower than weekdays and Sundays are lower again. Furthermore, concentrations tend to peak later in the day on Sundays compared with other days.

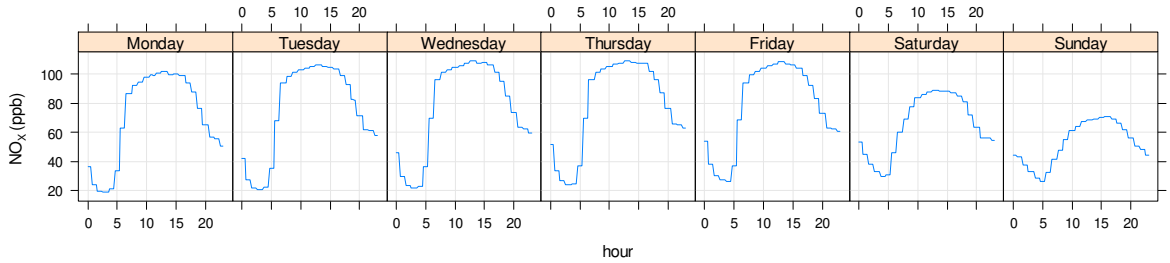


Figure 17. Diurnal and day of the week variation of meteorologically-averaged roadside increment NO_X concentrations averaged across the central/inner London sites.

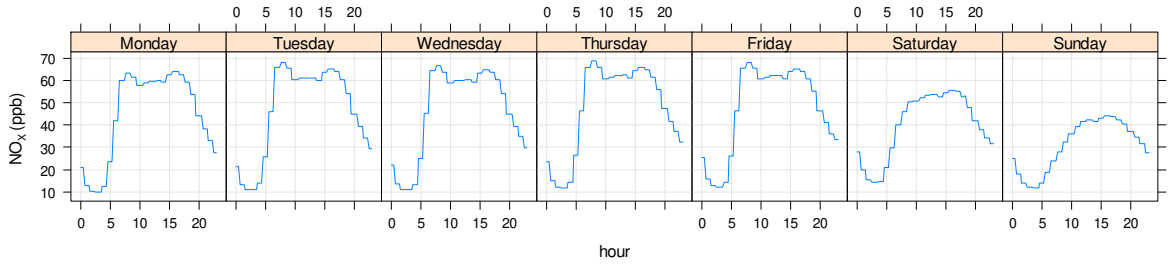


Figure 18. Diurnal and day of the week variation of meteorologically-averaged roadside increment NO_X concentrations averaged across the outer London sites.

Given that the results for central/inner London suggest that NO_X concentrations may have increased in recent years, it is worth considering whether the trends differ by day of the week. Because there are far fewer HGVs on Sundays, for example, if there was an important effect by vehicle it may be seen by plotting the trends in this way. Figure 19 shows the trends by day of the week for central/inner London sites. These results do not seem to suggest that the trends differ in overall shape by day of the week. It may tentatively be concluded that there does not seem to be a strong vehicle type effect that influences these trends. For example, if it were the case that HGVs were a major cause of the increase in NO_X concentrations, trends on Sundays would be expected to be different from other days of the week. Unfortunately it is difficult to draw firm conclusions from this analysis due to the large number of potential factors that can affect emissions.

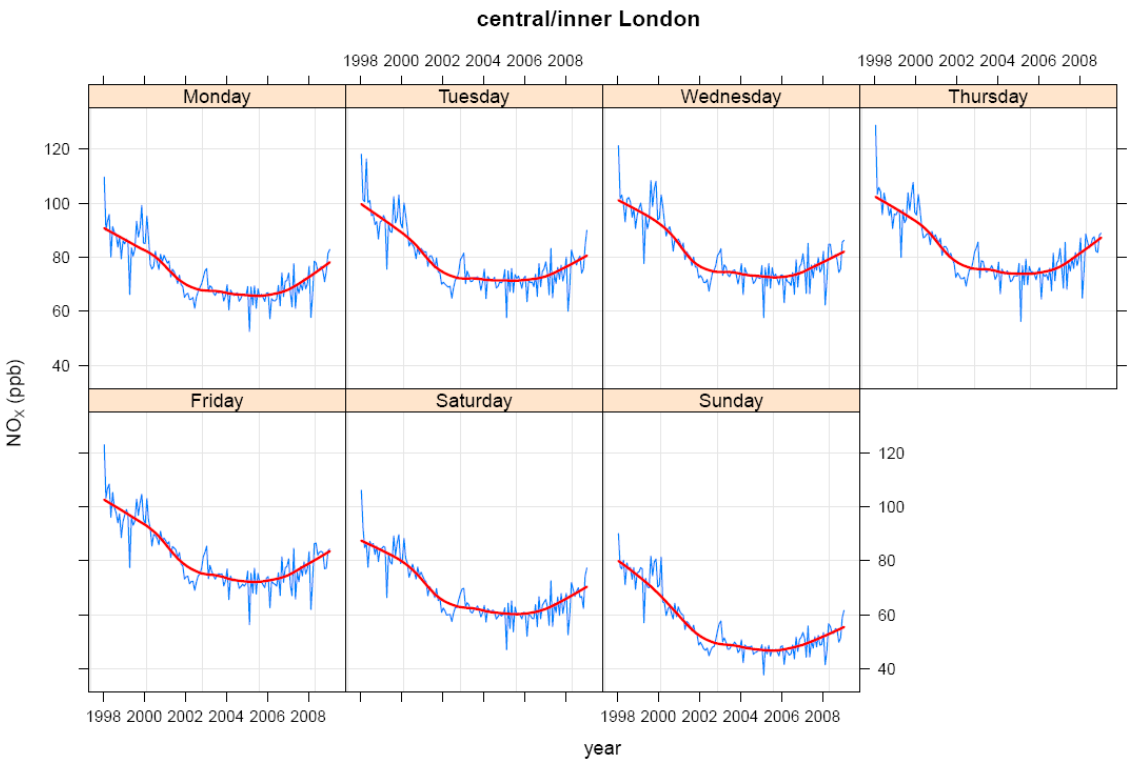


Figure 19. Trends in meteorologically-averaged roadside increment NO_X concentrations averaged across the central/inner London sites by day of the week.

6.4 Trends in hourly NO_X emissions

A brief summary of the contribution of emissions of NO_X by vehicle type shows that at central and outer sites and for 2003 and 2007, bus, car and rigid HGV emissions are dominant (Table 10). The split between the emissions contribution of light and heavy vehicles is 49% to 51% at the central sites in London in 2003 and 40% to 60% for outer sites in 2007.

Table 10. The percentage contribution to average NO_X emissions, by vehicle type, for 2003 and 2007

Zone	Pollutant	Year	Light vehicles	HGV	Taxis	Cars	Bus	LGV	Rigid	Artic
Central	NO_X	2003	49	51	9	28	25	11	21	6
		2007	42	58	9	21	26	12	25	7
Outer	NO_X	2003	43	57	2	30	29	11	19	9
		2007	40	60	2	25	33	12	18	9

The NO_X emissions trends between 2003 and 2007 and the equivalent trends in meteorologically-averaged NO_X concentrations are once again in poor agreement (Table 11). In addition, the meteorologically-averaged trend is different for central/inner sites and for outer sites, with the central sites showing an increase of 7% in NO_X concentrations and the outer sites a decrease of 9%. In contrast, the emissions calculations show a decline of ~25% across all sites, being dominated by light vehicles in central/inner London and shared between light and heavy vehicles at outer sites. As a result it is concluded that the emission

tends for NO_X are also optimistic and that this has important implications for policy development and meeting EU limit values for NO_2. Finally, comparison of the meteorologically-averaged diurnal profiles in Figure 17 and Figure 18 with those based upon the hourly emissions in Figure 21 show closer agreement between weekday and weekend profiles compared with the equivalent PM_{10} results, although further day of the week analysis of the kind described for PM_{10} is needed to assess the weaknesses that exist in the emissions calculations.

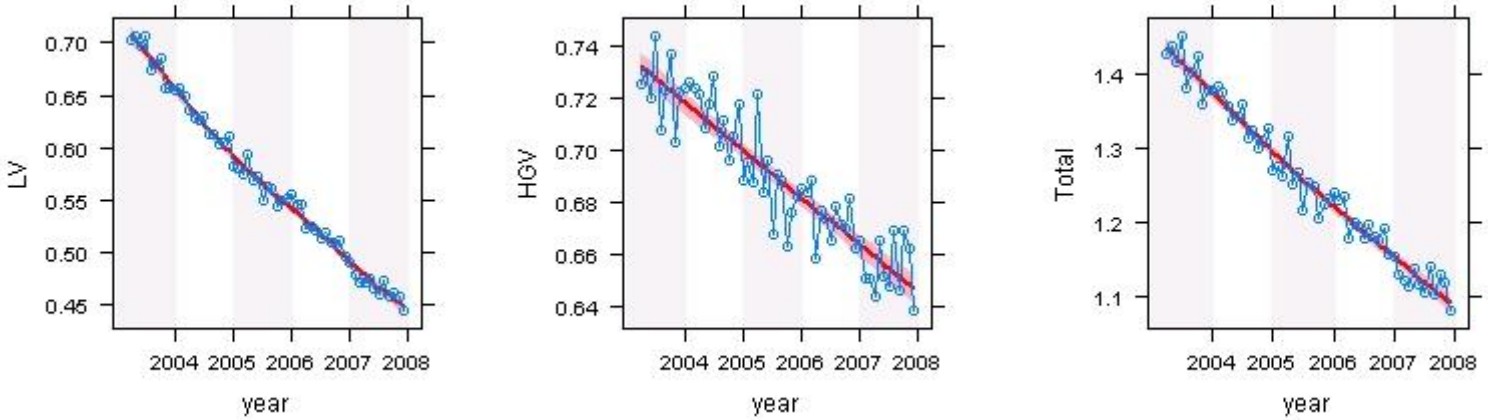
Table 11. The percentage change in NO_X emissions and NO_X measurements between 2003 and 2007[1]

Zone	Pollutant	All vehicles	Light vehicles[2]	HGV[3]	Taxis	Cars	Bus	LGV	Rigid	Artic
Central	NO_X	-20	-32	-10	-25	-41	-17	-15	-4	3
	Measured NO_X	+7.3								
Outer	NO_X	-29	-35	-25	-17	-41	-20	-21	-32	-30
	Measured NO_X	-8.5								

[1] Measurements from 2003-2008, [2] Car + Taxi + LGV, [3] Bus + rigid HGV + articulated HGV

Cars + Taxis + LGV | HGVs | All vehicles

NO_X emissions
Central sites



NO_X emissions
Outer sites

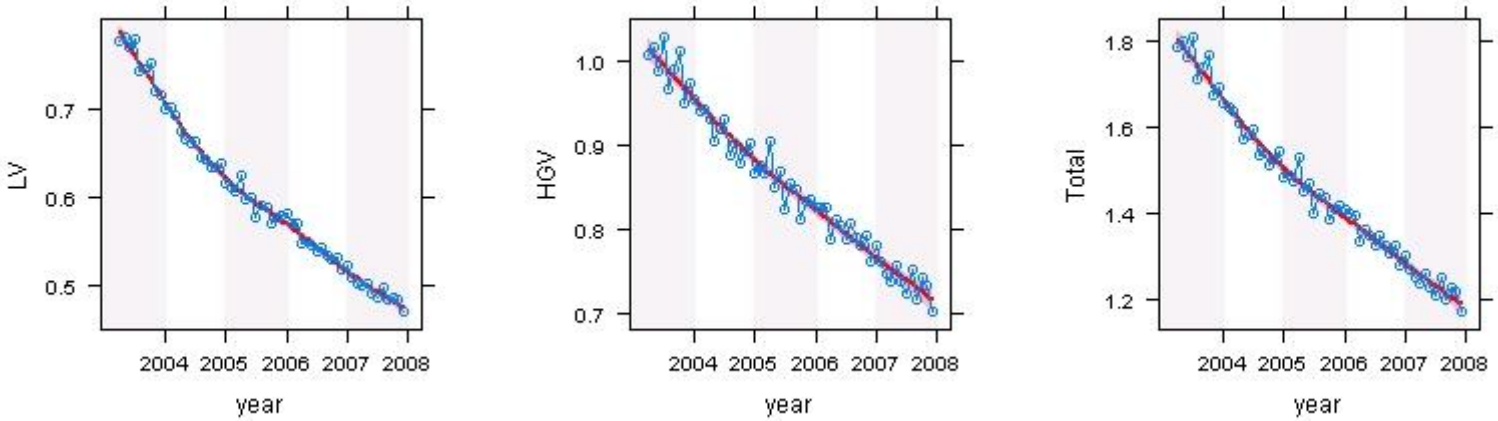
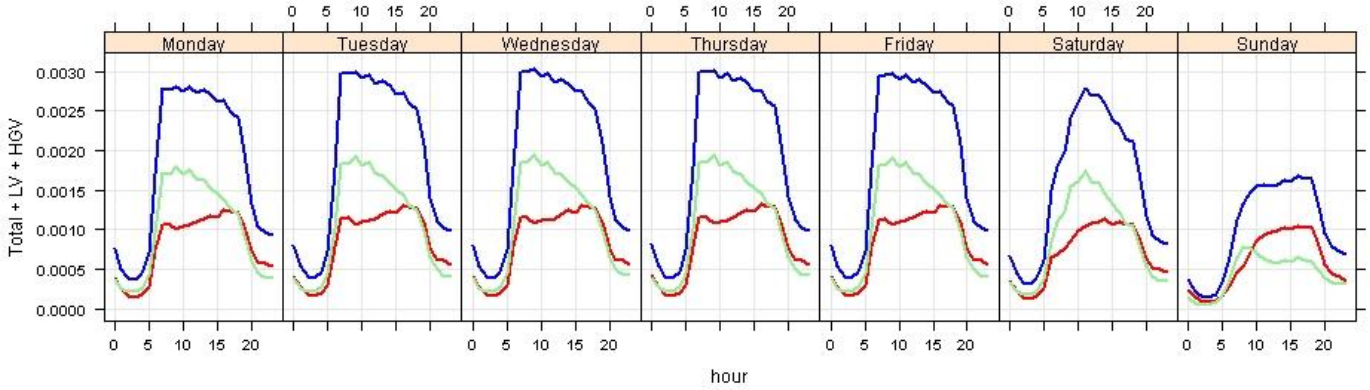


Figure 20. Monthly NO_X emission trends for central and outer roads. The red line shows a smooth fit of the data and the uncertainty (95% confidence interval)

NO_X central



NO_X Outer

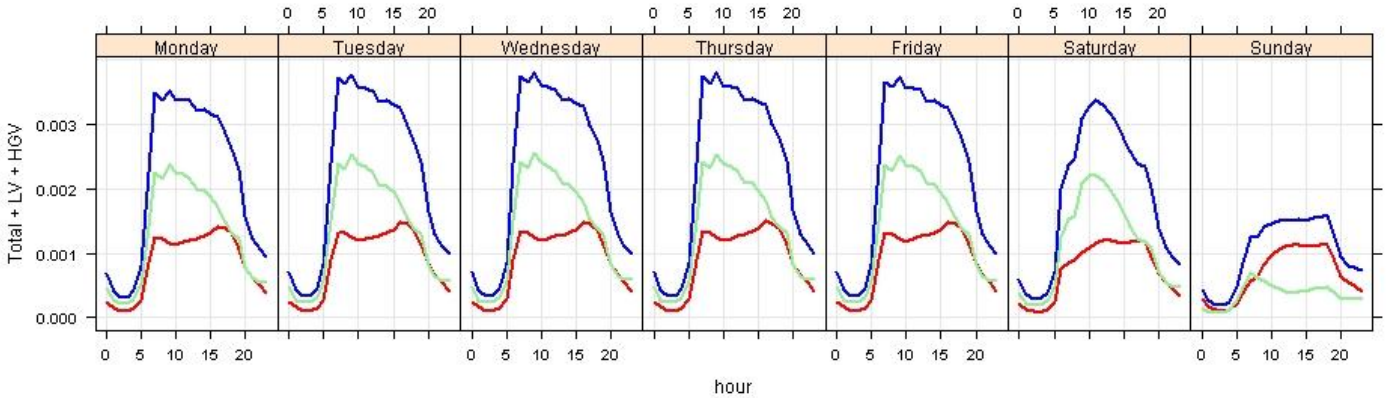


Figure 21. Diurnal NO_X emissions for central and outer sites (tonnes/hr). The blue line shows the total NO_X emissions, the red line emissions from light vehicles (cars+taxis+LGVs) and the green line emissions from Heavy vehicles (HGVs + Buses).

7 MARYLEBONE ROAD

The analysis in the previous sections focussed on providing an aggregated view of the likely change in emissions over time by central/inner London and outer London. The long time series of data available at Marylebone Road enables a more specific analysis to be undertaken. Figure 22 shows the meteorologically-averaged trend in PM_{10} concentrations at Marylebone Road. It is evident from this plot that there has been a tendency for the local component to increase over time since 1998; or at best, there is very little evidence to support the view that emissions have decreased. The large, short duration peaks in 1999 are known to be related to building works close to the site.

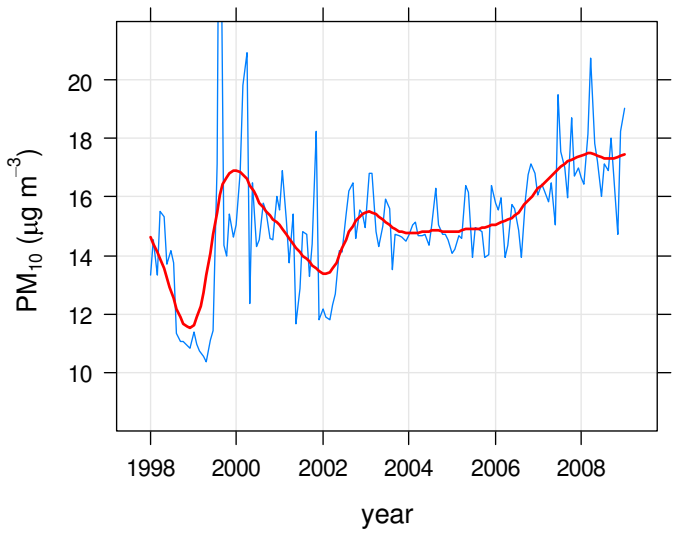


Figure 22. The trend in meteorologically-averaged roadside increment PM_{10} concentrations at Marylebone Road.

Because Marylebone Road has a long time series it is useful to consider in which ways PM_{10} concentrations may have changed over time. This has been considered by splitting the data set into an "early" period (1998-1999) and a "late" period (2007-2008). Figure 23 shows that the principal difference between the 1998-1999 and 2007-2008 is that concentrations have tended to increase during the early and late parts of the day. On average there has been little change or no change during the middle part of the day. Also apparent is that there has become less of a difference between Saturdays and Sundays cf. weekdays for 2007-2008 data. There could be several explanations for this behaviour, but one interpretation is that cars/LGVs have become relatively more important for PM_{10} emissions over time. This is because flows of these vehicles tend to be higher than HGVs early/late in the day and there is little difference in the flows of light vehicles by day of the week. However, this is only one possibility and some caution is needed in interpreting these data.

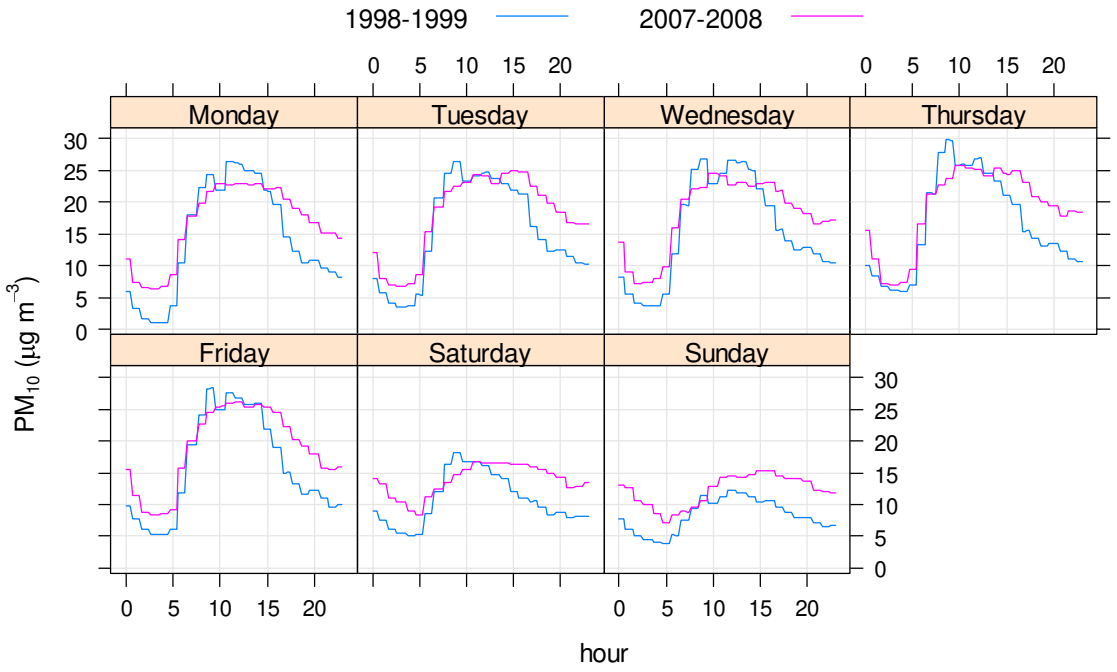


Figure 23. Meteorologically-averaged diurnal/day of week trends in roadside increment PM_{10} concentrations at Marylebone Road for two periods: (1998-1999) and (2007-2008).

The trend in NO_X at Marylebone Road is notably different to that for PM_{10}, as shown in Figure 24. One of the most striking features of Figure 24 is the sharp decrease in concentration towards the end of 2001. In this respect, sometime around 2001 looks to be a 'change-point' where concentrations shifted from one level to another. The only known change at this time was the introduction of the bus lane in August 2001 (Carslaw et al., 2006). Indeed, one of the benefits of accounting for meteorology in trends is that interventions such as this are more clearly shown.[4] The change points are better shown in Figure 25.

[4] We have not analysed the change in NO_X in detail, but clearly such an analysis could reveal the importance of an intervention such as a bus lane as well as providing information on the relative importance of vehicle emissions.

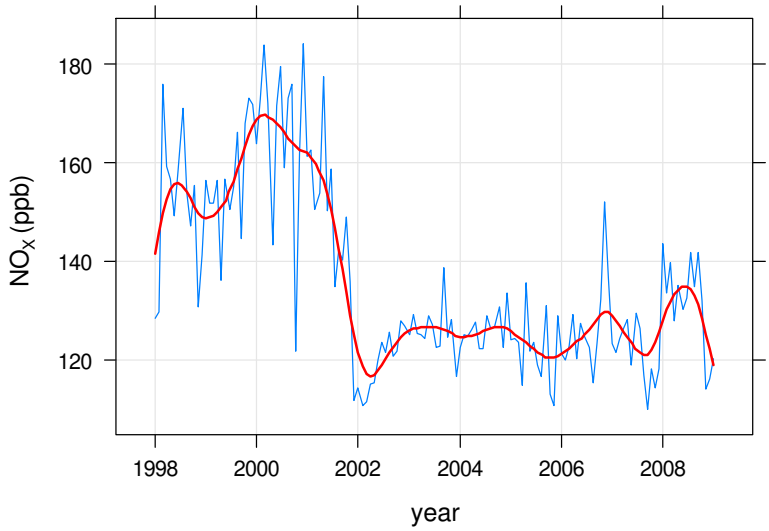


Figure 24. The trend in meteorologically-averaged roadside increment NO_X concentrations at Marylebone Road.

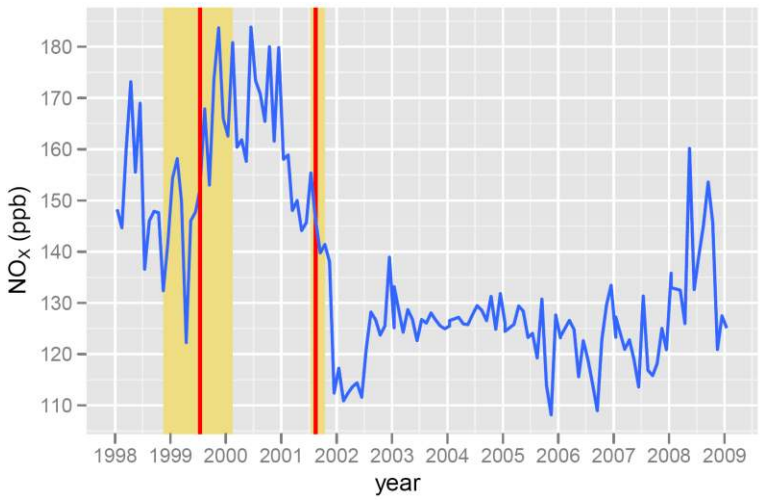


Figure 25. The trend in meteorologically-averaged roadside increment NO_X concentrations at Marylebone Road showing change-points. The shaded areas show the 95 % confidence intervals in the timing of the change according to the method of Carslaw et al. (2006)

Considering how the diurnal variation in concentrations of NO_X may have changed in a similar way to the PM_{10} analysis shows that reductions in NO_X since 1998/1999 to 2007/2008 have be proportionately similar across all hours of the day. This behaviour is again very different to that observed for PM_{10} concentrations.

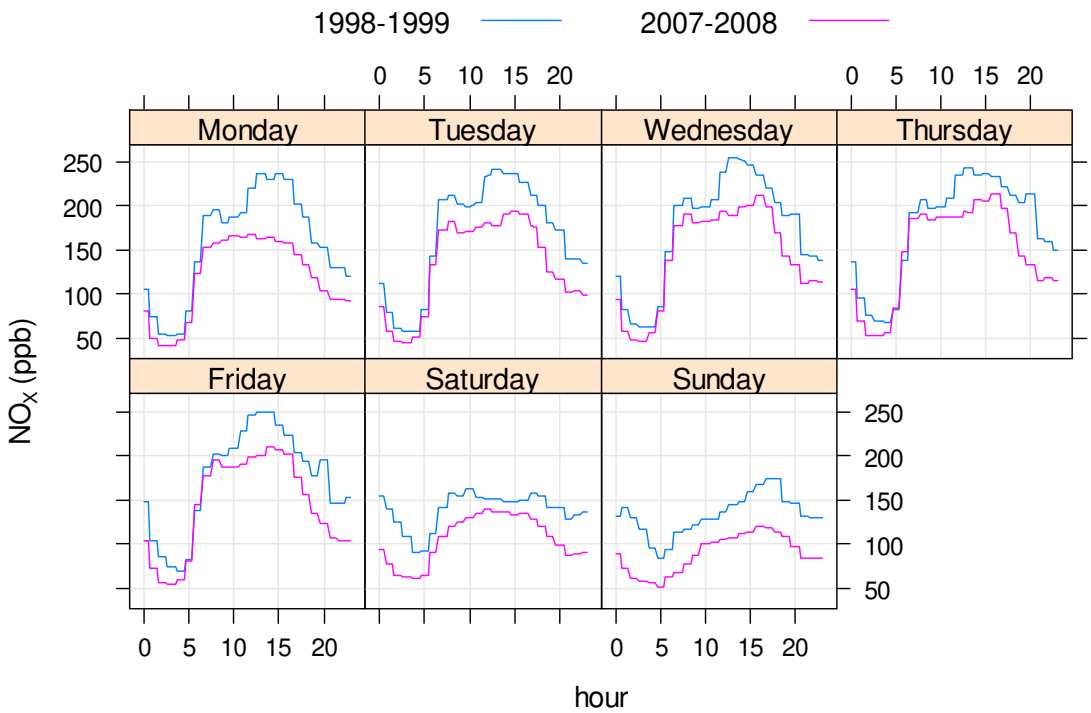


Figure 26. Meteorologically-averaged diurnal/day of week trends roadside increment NO_X concentrations at Marylebone Road for two periods: (1998-1999) and (2007-2008).

We have also considered $PM_{2.5}$ concentrations at Marylebone Road. For this time series the suburban BX2 sites was used for background subtraction; a decision based on data capture rate and a desire to have a representative urban background site. $PM_{2.5}$ TEOM measurements at Marylebone Road were not part of national QA/QC procedures prior to 2003 and some doubts concerning the validity of the measurements have been expressed for data before this date (AQEG, 2007). For this reason we only consider data from 2003-2008.

The meteorologically-averaged trend is shown in Figure 27 and indicates that the increment in $PM_{2.5}$ concentrations has increased since 2003. Less clear increases are observed if the London Bloomsbury site is used for background subtraction, shown Figure 28. However, the Bloomsbury site is itself in close proximity of busy roads with high flows of buses and may not be the most appropriate site for background subtraction; hence the general preference from North Kensington. Nevertheless, both sites show that the $PM_{2.5}$ increment has increased at Marylebone Road since 2003.

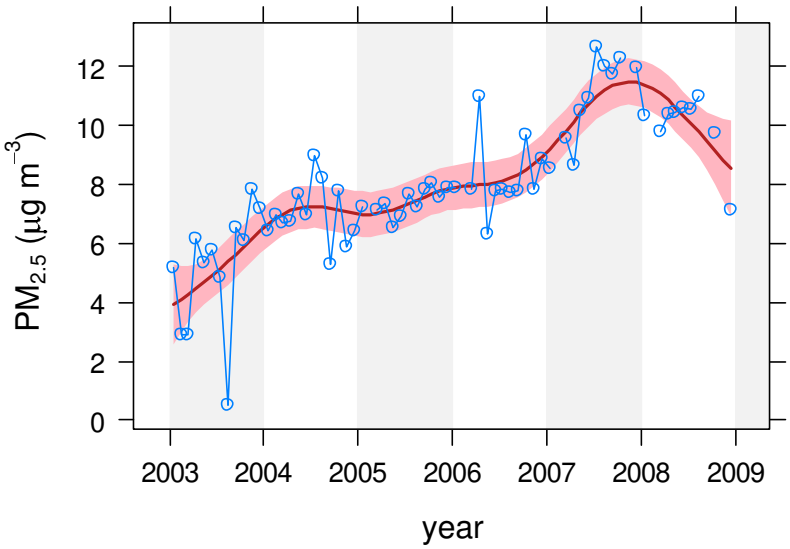


Figure 27. The trend in meteorologically-averaged roadside increment $PM_{2.5}$ concentrations at Marylebone Road using the BX2 site for background subtraction.

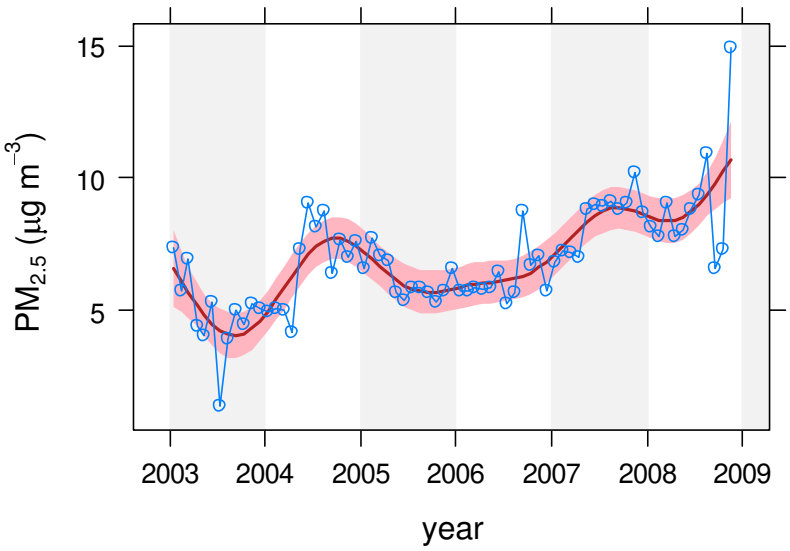


Figure 28. The trend in meteorologically-averaged roadside increment $PM_{2.5}$ concentrations at Marylebone Road using the London Bloomsbury site for background subtraction.

This page has been left blank intentionally

8 CONCLUSIONS

This report summarises a detailed and comprehensive assessment of trends in emissions and concentrations of NO_X and PM in London. We have developed methods to create *hourly* road traffic and emissions estimates between 2003 and 2007 using comprehensive traffic data in London combined with new vehicle data based upon Automatic Number Plate Recognition (ANPR) cameras. The use of both traffic and emissions data as hourly values has proved to be invaluable in helping interpret the air pollution measurement trends.

The statistical models were used to calculate trends, weekday and diurnal variations in roadside increments of NO_X, PM_{10} and $PM_{2.5}$ under *average* meteorological conditions. This approach has the benefit that trends calculated in this way will more closely relate to emissions changes rather than changes due to local meteorological effects; thus allowing for a more robust comparison between the two.

The overriding conclusion of this work is that there is a clear disparity between the estimated trends in emissions and the observed trends in concentrations. This disparity has important implications for the management of air pollution and raises question about the adequacy of emission factors and projected concentrations of NO_X (and NO_2), and PM.

From the analysis of the measurements, we find there is evidence that NO_X concentrations have not decreased over the period 2003-2008. In fact, in central/inner London the mean of 10 roadside sites suggests that concentrations have *increased* by around 7 % from 2003-2008. In outer London, concentrations of NO_X have decreased by about 8-9 %. The corresponding changes in emissions are reductions of 20 and 29 % respectively. These results therefore show there seems to be a considerable disparity between the expected change in emissions and the actual change in concentrations. The results also suggest that there is a difference in how central/inner London sites have responded compared with outer London sites.

For PM_{10} the average of 7 central/inner London sites shows that concentrations have decreased by 4-5 % compared with an estimated 25 % from detailed emission calculations (2003-2008). Similarly, in outer London the analysis of PM_{10} concentrations indicates a reduction of 13-14 % compared with a 25 % reduction estimated for the emissions. It should be noted that given the complexity of PM emissions/concentrations that these results will be less certain than those for NO_X. Nevertheless, there does seem to be a consistent difference in the way that central/inner London sites respond compared with outer London site, which is similar to the findings for NO_X.

While there does appear to be a clear difference in the trends in PM_{10} and NO_X in central/inner London and outer London, it remains difficult to identify a specific cause(s). However, one of the major differences between these two areas is the proportion of diesel vehicles. Diesel vehicles are known to be significant emitters of both NO_X and PM_{10}. The detailed analysis of traffic data suggests that flows of large diesel vehicles (particularly Bus, HGVs and LGVs) have increased in central/inner London between 2003 and 2007 and that there has been a more mixed response at outer sites. Interestingly, total traffic has reduced for both groups of sites between 2003 and 2007.

We have also shown that considering how emissions and concentrations vary by day of the week and hour of the day is an effective way in which to compare emissions variation with that for measured concentrations. In particular, removing the meteorological signal can in some cases significantly affect the diurnal profile of concentrations and provide a more robust means of comparing trends with emissions estimates.

Using multiple regression analysis on a combination of traffic data and meteorologically averaged measurements by day of the week has demonstrated that it can provide the relative emission rates for different vehicle categories. It is shown that an average emissions ratio of 11:1 for HGV (bus + rigid + articulated) to Light vehicles (car + taxi + LGV) would result in closer agreement between the emissions and air pollution daily profiles. By comparison the ratio using the emissions calculation is 3.9. Further refinement of this method to consider larger number of vehicle types and across a number of years would provide an important step in identifying the vehicles which may be the cause of the disparity between emissions trends and measurements.

A useful side-effect of the meteorological averaging is that step-changes in the concentration of pollutants can be revealed. For example, there is evidence from this work that at Marylebone Road a distinct reduction in NO_X occurred in 2001 coinciding with the introduction of the bus lane. Such information could provide useful information concerning the effect that traffic management has in London (and elsewhere) that would normally be masked by meteorological variation.

9 RECOMMENDED FURTHER WORK

The analysis described in this document is the beginning of an important process of evaluation of emissions inventories that will enable greater understanding of emissions trends, air pollution model performance and better policy making. However, having created a large and unique dataset as well as important methods of analysis a number of additional work packages are recommended:

While there are many roadside air pollution sites in London, there are very few sites that have coincident traffic measurements. A more concerted effort to link quality road traffic measurements with air pollution sites would yield considerably more insight into these issues.

Important understanding of the inadequacies of emissions inventories can be made through the development of the multiple regression approaches to include additional vehicle types over a number of years.

In this study use was made of the currently available UK emissions factors, i.e. those based on results from Barlow et al., 2001. These have since been superseded via a consultation process undertaken by DfT during 2009. Initial results show that differences between the two emission factor datasets can be substantial for specific vehicles types. It would therefore be worth considering undertaking a recalculation of emissions using the new factors.

A separate research effort is being undertaken by ERG in order to monitor the effect of the LEZ in London. Part of this research is to establish a chemical speciation PM_{10} model and will include identifying road traffic emissions of PM and estimating trends over time. This provides a unique opportunity to build on the analysis presented here.

10 REFERENCES

Abbott J, 2007. PM_{10} resuspension by road vehicles. AEA report produced for Department for the Environment, Food and Rural Affairs (DEFRA). Reference Number ED-48209/R2388.

Barlow T J, Hickman A J and Boulter P G. 2001. Exhaust Emission Factors 2001: Database and Emission Factors, Transport Research Laboratory, Crowthorne TRL report PR/SE/230/200.

Carslaw, D.C., Ropkins, K and M.C. Bell. (2006) Change-Point Detection of Gaseous and Particulate Traffic-Related Pollutants at a Roadside Location. Environmental Science and Technology. Vol. 40. Issue 22. 6912-6918.

Carslaw, D.C., Beevers, S.D. and J.E. Tate (2007). Modelling and assessing trends in traffic-related emissions using a generalized additive modelling approach. Atmospheric Environment. 41 pp. 5289-5299.

D.C. Carslaw and P.J. Taylor. Analysis of air pollution data at a mixed source location using boosted regression trees. *Atmospheric Environment*, In press, 2009.

Gardner, M.W., Dorling, S.R., 2000. Statistical surface ozone models: an improved methodology to account for non-linear behaviour. Atmospheric Environment 34 (1), 21–34.

Harrison RM, Jones AM, Lawrence RG. 2004. Major component composition of PM_{10} and PM2.5 from roadside and urban background sites. Atmospheric Environment 38, 4531 - 4538.

Jacoby, WG. 2000. Loess: a nonparametric, graphical tool for depicting relationships between variables. Electoral Studies 19 (2000) 577–613.

Libiseller, C., Grimvall, A., Walden, J., Saari, H., 2005. Meteorological normalisation and non-parametric smoothing for quality assessment and trend analysis of tropospheric ozone data. Environmental Monitoring and Assessment 100 (1–3), 33–52.

D.D. Parrish. 2006. Critical evaluation of US on-road vehicle emission inventories. Atmospheric Environment 40 (2006) 2288–2300.