

## PM<sub>10</sub> in London During 2003

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### Abstract

During 2003 London experienced a series of widespread PM<sub>10</sub> pollution episodes. Elevated particulate matter concentrations were measured at all background sites during February, March, April and August, with lesser incidents being experienced during September and November. Source apportionment analysis of PM<sub>10</sub> and PM<sub>2.5</sub> reveals that the first 5 episodes were predominately caused by secondary PM<sub>10</sub> from distant sources, with the summer episodes also being linked to photochemistry. The influx of secondary PM<sub>10</sub> during 2003 can be compared to the secondary episode experienced in London during spring 1996. During 2003 the secondary PM<sub>10</sub> was sufficient to cause the daily mean concentration of PM<sub>10</sub> to exceed 50 µg m<sup>-3</sup> on 20 days without additional PM<sub>10</sub> from primary sources. During 1996, 34 such days were experienced. The November 2003 episode was associated with Guy Fawkes Night fireworks. Roadside sites measured additional PM<sub>10</sub> from local traffic, which increased the roadside concentrations during PM<sub>10</sub> incidents and caused additional incidents not measured at background sites.

The incidents during 2003 reversed the established trend of declining PM<sub>10</sub> concentrations in London. Breaches of the 2005 EU Limit Value for PM<sub>10</sub> were largely confined to roadside sites in Central London and several roadside sites in Outer London. The difference between roadside and background PM<sub>10</sub> concentrations, with respect to the EU Limit Value, suggests that control of road traffic sources may be sufficient to manage any recurrence of these episodes. A required reduction of primary PM<sub>10</sub> emissions of 30% has been calculated for the typical Inner London roadside site Kensington & Chelsea 2. The practicality of implementing such road traffic measures is, however, an open question.

### Introduction

Air quality limit values for PM<sub>10</sub> were set at the European wide level by the first Daughter Directive (99/30/EC) under the Air Quality Framework Directive (96/62/EC). The Daughter Directive stipulates that by the start of 2005 the daily mean PM<sub>10</sub> concentration should not exceed 50 µg m<sup>-3</sup> on more than 35 occasions per year and the annual mean should not exceed 40 µg m<sup>-3</sup>. Additionally by 2010, the daily mean PM<sub>10</sub> concentration should not exceed 50 µg m<sup>-3</sup> on more than 7 occasions per year, and the annual mean should be less than 20 µg m<sup>-3</sup>. The first Daughter Directive stipulates that PM<sub>10</sub> should be measured gravimetrically for assessment against the EU Limit Value.

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The 2005 EU Limit Values for PM<sub>10</sub> have been incorporated into UK law in the Air Quality Regulations 2000. In London, the Greater London Assembly (GLA) has responsibilities for the management of air pollution in the capital in conjunction with the London local authorities. The Limit Values for PM<sub>10</sub> have been incorporated into the Mayor's Air Quality Strategy (GLA 2002).

This paper reports preliminary analysis of the PM<sub>10</sub> measured in London during 2003; apportioning PM<sub>10</sub> between different source categories to explain the cause of each episode, and these measurements were placed in the context of both the EU Limit Values and measurements since 1994.

## Method

### Measurements

Air quality in London is monitored by the London Air Quality Network (LAQN) and the UK Automatic Urban and Rural Network (AURN). During 2003 there were 99 monitoring sites in the LAQN area (including AURN sites); 71 sites measuring PM<sub>10</sub> with the vast majority (59) using the Tapered Element Oscillating Micro Balance (TEOM<sup>TM</sup>) method. Further information about these monitoring sites can be obtained from [www.erg.kcl.ac.uk](http://www.erg.kcl.ac.uk) and from [www.airquality.co.uk](http://www.airquality.co.uk).

This study used automatic measurements of PM<sub>10</sub> and PM<sub>2.5</sub> made using the TEOM method. Measurements of NO<sub>x</sub> used in this study were made using the chemiluminescent method with automatic equipment subject to fortnightly calibration traceable to National Metrological Standards. Automatic NO<sub>x</sub> and PM<sub>10</sub> instruments were subject to twice yearly independent audit by the National Physical Laboratory or AEA plc.

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The EU limit value requires gravimetric measurement of PM<sub>10</sub>. However, the vast majority of PM<sub>10</sub> measurements in (and around) London are made using automatic methods, mainly the TEOM (1400a and 1400ab). Allen *et al.* (1997), Smith *et al.* (1997), Green *et al.* (2001), Charron *et al.* (2004) and others have observed that the TEOM produces a lower measurement of PM<sub>10</sub> than that derived gravimetrically due to greater sampling losses of semi-volatile particulate and particle bound water from the TEOM. A 'correction' factor of 1.3 is recommended in the UK for comparison of TEOM PM<sub>10</sub> measurements with the EU Directive (DETR, 1999). This 'correction' factor was applied to all TEOM PM<sub>10</sub> measurements in this study. In the absence of a recommended 'correction' factor for TEOM PM<sub>2.5</sub> measurements, the 'correction' factor of 1.3 was also been applied to these measurements. It is, however, recognised that the 'correction' factor will depend on particle composition (Charron *et al.* 2004) and this is therefore likely to lead to inaccuracies when applied to PM<sub>2.5</sub>. In all cases TEOM instruments were operated using their default settings with the exception of Mass Concentration Averaging Time and Storage Interval, which were both set to 900 seconds.

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The analysis presented in this study is based on preliminary measurements of air pollution during 2003. More detailed analysis will be undertaken following the publication of a final measurement dataset later in 2004.

## Modelling method

The PM<sub>10</sub> modelling methodology is detailed in Fuller *et al.* 2002. The model apportions PM<sub>10</sub> by source through analysis of measurements of annual mean NO<sub>x</sub>, PM<sub>10</sub> and PM<sub>2.5</sub> across a network of monitoring sites. PM<sub>10</sub> is identified as arising from three source components; primary (associated with NO<sub>x</sub>), secondary (mainly the PM<sub>2.5</sub> not associated with NO<sub>x</sub>) and natural (the PM<sub>10</sub>-PM<sub>2.5</sub> component not associated with NO<sub>x</sub>). Daily mean secondary and natural components were then derived from long term monitoring sites across London. Total daily mean PM<sub>10</sub> concentrations at any location were then calculated by adding the secondary and natural PM<sub>10</sub> to primary PM<sub>10</sub> derived from NO<sub>x</sub> at the location.

**גמחק:** Modelling was undertaken for the two sites with the most fugitive events (Marylebone Road and Kensington & Chelsea 2 during the second half of 1999) and for three other sites during 2000 to demonstrate the range of fugitive source types.¶

The application of the modelling method in this preliminary study had three departures from that described in Fuller *et al.* 2002.

Firstly, the daily mean secondary and natural components were derived from a single background site in Bexley. This preliminary analysis does not contain sufficient PM<sub>10</sub> and PM<sub>2.5</sub> measurements to apportion PM<sub>10</sub> on each day of the year. More detailed source apportionment will be undertaken following the availability of a ratified dataset.

Secondly, allowance was made for the measurement offset of +3 µgm<sup>-3</sup> (raw TEOM) applied by the TEOM to all measured mass concentrations (Patashnick and Rupprecht 1991, Rupprecht and Patashnick Co. Inc. 1992, Rupprecht and Patashnick Co. Inc. 1996). Following the application of the 1.3 'correction' factor (DETR 1999) this offset has a value of 3.9 µgm<sup>-3</sup>. The model described in Fuller *et al.* 2002 apportioned this offset to secondary sources of PM<sub>10</sub>. Here the offset was introduced as a separate PM<sub>10</sub> source. Retention of the offset within the model ensured comparability between the source apportionment method and TEOM measurements, and also maintained the applicability of the 1.3 'correction' factor to the source apportioned PM<sub>10</sub>.

Thirdly, the factors for the calculation of primary PM<sub>10</sub> and PM<sub>2.5</sub> from measured NO<sub>x</sub> concentration were revised based on analysis of measurements during 2001. The following factors were used

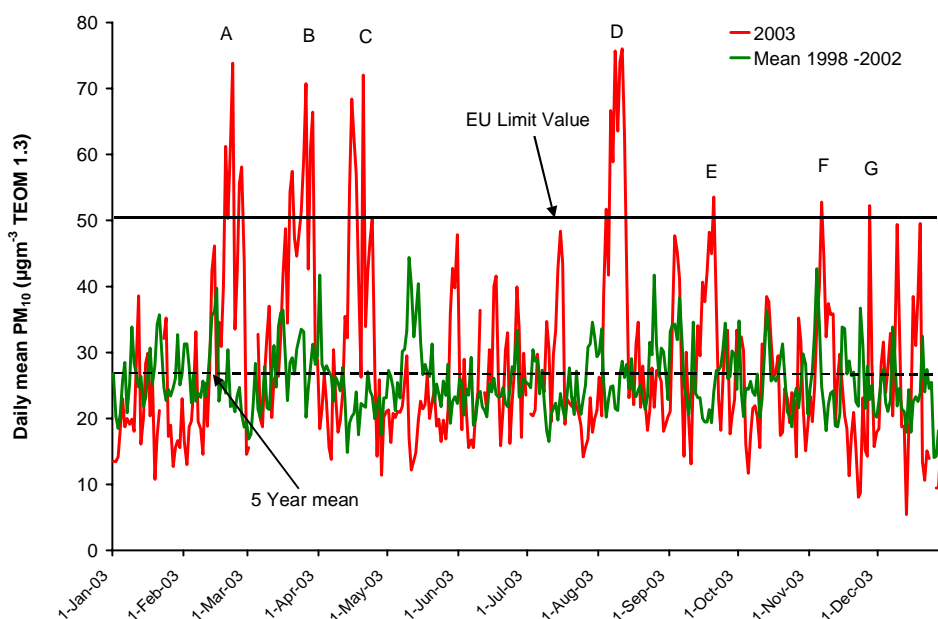
$$\text{Primary PM}_{10} (\mu\text{gm}^{-3}) = 0.105 (\mu\text{gm}^{-3} \text{ppb}^{-1}) * \text{NO}_x (\text{ppb})$$

$$\text{Primary PM}_{2.5} (\mu\text{gm}^{-3}) = 0.828 (\mu\text{gm}^{-3} \text{ppb}^{-1}) * \text{NO}_x (\text{ppb})$$

## Results

### Measurements

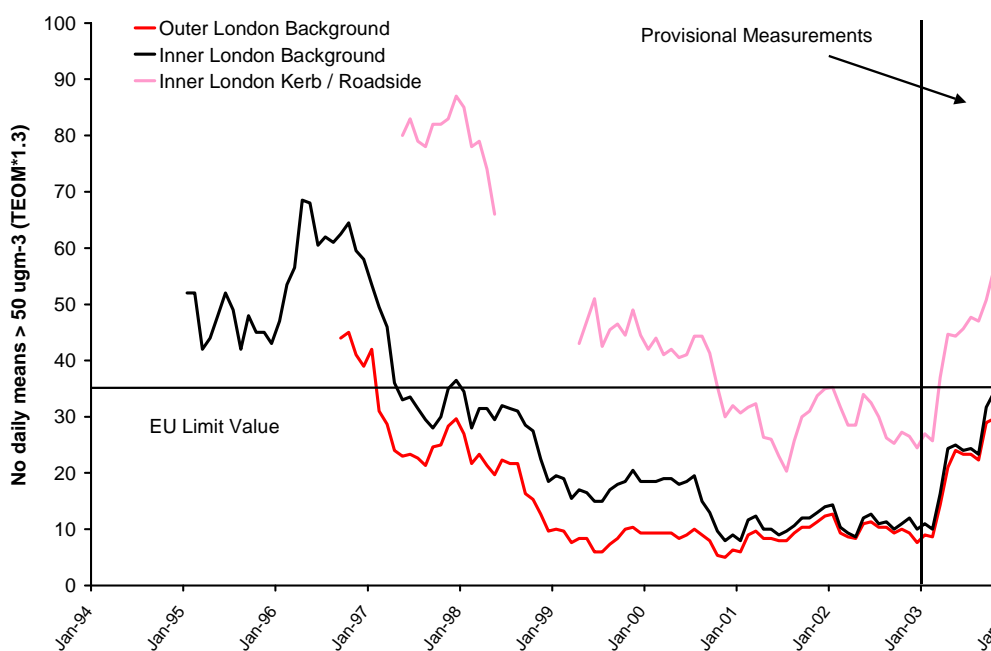
Figure 1 shows the daily mean PM<sub>10</sub> concentration measured at Kensington & Chelsea 1; a typical Inner London background site, and the mean PM<sub>10</sub> concentration on each day during the preceding five years. Figure 1 shows 7 distinct episodes during 2003 when the daily mean PM<sub>10</sub> concentration exceeded 50 µgm<sup>-3</sup>. These episodes are labelled A to G. Episodes A to D were obvious elevations above the 5 year mean.



**Figure 1 Daily mean PM<sub>10</sub> measured by TEOM (\*1.3) at Kensington & Chelsea 1. Measurements during 2003 and the mean concentration during 1998 – 2002 are shown.**

Figure 2 shows the number of daily mean PM<sub>10</sub> measurements above 50 µgm<sup>-3</sup>, as a running annual count, at three different types of location. The long-term measurements at Inner London background sites exhibit a downward trend from around 50 days above 50 µgm<sup>-3</sup> in 1995 to around 10 days in 2002. The similar downward trend of all site types reflects a reduction in secondary and primary PM<sub>10</sub> emissions, whilst the convergence in the number of daily means above 50 µgm<sup>-3</sup> reflects the reduction in traffic emissions of primary PM<sub>10</sub>.

During 1995 typical Inner London background sites exceeded the 2005 EU Limit Value, which implied a widespread breach of the Limit Value throughout London. The situation deteriorated during Spring 1996 due to the substantial secondary episode at this time (APEG 1999, Stedman 1997). As a consequence, 76 daily means above 50 µgm<sup>-3</sup> were measured in Inner London during the year ending April 1996; more than double the 2005 Limit Value of 35 days. A repetition of such an episode would clearly provide challenges for air quality management. The additional days above 50 µgm<sup>-3</sup> caused by the Spring 1996 episode left the running count in Spring 1997. Other events affecting the number of daily means above 50 µgm<sup>-3</sup> included a primary episode during Autumn 1997 and the unsettled weather in late 2000. Inner London background sites have consistently achieved the 2005 EU Limit Value since 1998. The number of daily means above 50 µgm<sup>-3</sup> measured at Outer London sites was only marginally below those measured in Inner London. A larger difference can be seen between the background and kerb/roadside sites in Inner London than between outer and Inner London background sites.



**Figure 2 Number of days when daily mean PM<sub>10</sub> exceeded 50 µg m<sup>-3</sup> (TEOM\*1.3) shown as a running annual count measured at three types of location in London.**

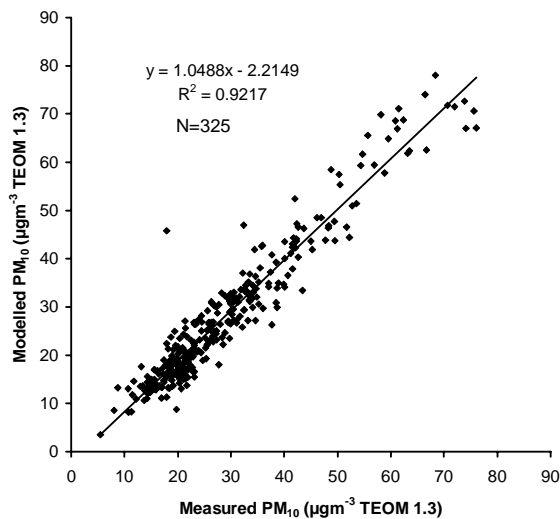
The number of daily means above 50 µg m<sup>-3</sup> at the kerb/roadside in Inner London follows a similar trend to background, albeit with additional days due to local traffic emissions. Inner London roadside sites have generally achieved the 2005 EU Limit Value since 2000. Measurements at Marylebone Road are not shown in Figure 2 but have been in the range 70–160 days per year and show variations in part due to local events such as building works.

The provisional measurements shown in Figure 2 suggest that the previously improving trend in PM<sub>10</sub> concentration was reversed in 2003; with 2003 PM<sub>10</sub> levels being comparable to those during 1998. Provisional measurements shown in Figure 2 reflect the impact of the PM<sub>10</sub> episodes in 2003. Compared to 2002, background sites measured around 20 additional daily means above 50 µg m<sup>-3</sup> during 2003, with kerb/roadside sites in Inner London measuring around 30 such additional days. The results presented in Figure 2 are means calculated from a sample of sites within each site type and therefore mask individual site variations. The majority of Inner London background TEOM sites did not exceed the Limit Value; the exception being Tower Hamlets 1 where building works may have caused additional local PM<sub>10</sub>. Regrettably, insufficient TEOM measurements of Central London background conditions were therefore available to determine if the EU Limit Value was exceeded in this area. An estimate of the PM<sub>10</sub> in Central London has been made using the available measurements from the Bloomsbury background site and measurements from Kensington & Chelsea 1 in Inner London. It is estimated that Central London background locations

measured at least 33 daily means above  $50 \mu\text{g m}^{-3}$ . There is hence a possibility that background locations in Central London exceeded the Limit Value. By the end of 2003, road and kerbside TEOM sites in Inner London had exceeded the 2005 EU Limit Value by a large margin. However the attainment of the Limit Value at background locations suggests that reduction of local primary  $\text{PM}_{10}$  at roadside and kerbside locations in Inner London may lead to the attainment of the Limit Value at these locations. The required reduction in local primary  $\text{PM}_{10}$  can be calculated following source apportionment of the measured  $\text{PM}_{10}$ .

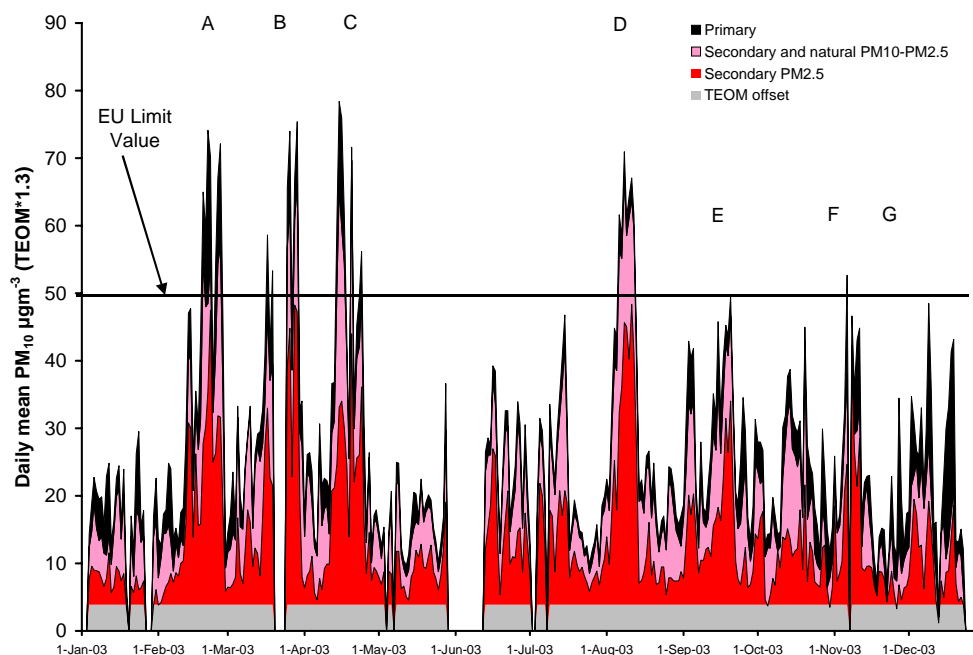
#### *Source Apportionment*

Figure 3 compares the measured  $\text{PM}_{10}$  and the total  $\text{PM}_{10}$  derived from the source apportionment model. Overall the modelled daily mean concentrations compare well with a co-relation coefficient ( $r^2$ ) of 0.92. The modelled annual mean ( $27.8 \mu\text{g m}^{-3}$ ) compares well with the measured annual mean ( $28.5 \mu\text{g m}^{-3}$ ).



**Figure 3 Regression analysis of daily mean measured  $\text{PM}_{10}$  and total  $\text{PM}_{10}$  derived from the source apportionment model for the Kensington & Chelsea 1 monitoring site.**

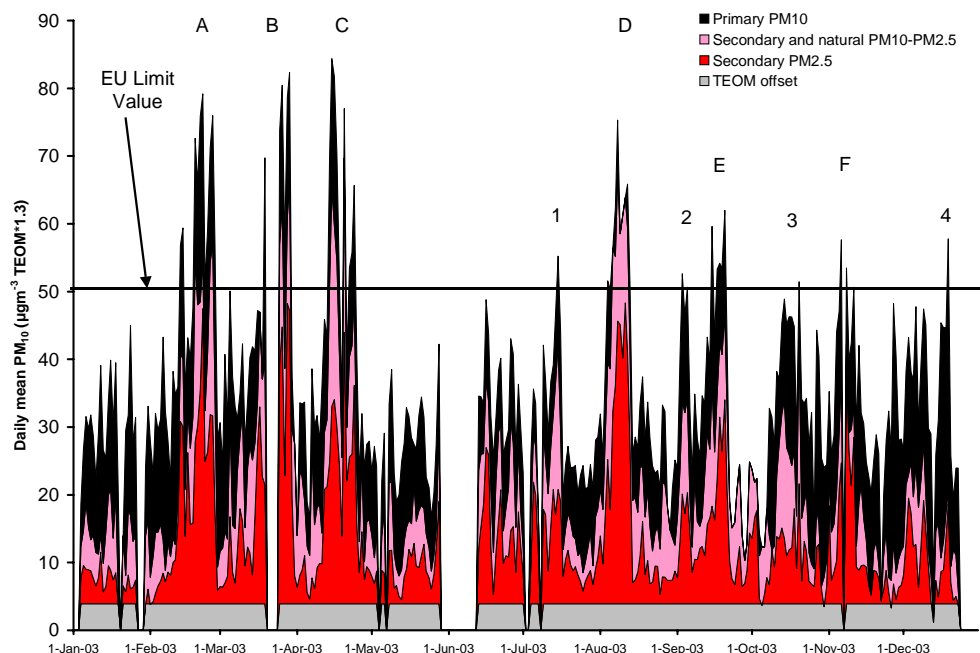
Figure 4 shows the source apportionment of  $\text{PM}_{10}$  measured at Kensington & Chelsea 1; a typical Inner London background site. Daily means above  $50 \mu\text{gm}^{-3}$  were due to distinct episodes; labelled A to F. The  $\text{PM}_{10}$  concentration during episode G and the modelled daily mean did not exceed  $50 \mu\text{gm}^{-3}$  at this time. During episodes A to E,  $\text{PM}_{10}$  was dominated by secondary particulate brought into London from continental sources. The imported secondary  $\text{PM}_{10}$  is present in the  $\text{PM}_{2.5}$  as expected and is also present in the coarse ( $\text{PM}_{10} - \text{PM}_{2.5}$ ) fraction which is indicated by elevated concentrations of the secondary and natural coarse particulate during the episodes A to E. The daily mean secondary and natural coarse particulate concentration reached an annual maximum of  $32 \mu\text{gm}^{-3}$  on the 15<sup>th</sup> April 2004 at the start of episode C which may be indicative of a Saharan dust episode similar to the episode that affected southern England during March 2000 (Ryall *et al.* 2002). Episodes B to E were associated with photo-chemical activity as indicated by elevated concentrations of ground level ozone. Episode D was dominated by secondary  $\text{PM}_{10}$  and coincided with record-breaking temperatures and the highest ground-level  $\text{O}_3$  concentrations measured in London since 1990. Episode F was caused by Guy Fawkes Night bonfires and fireworks. The source apportionment method is not accurate at this time due to large local sources of particulate that are not also sources of  $\text{NO}_x$ .



**Figure 4 Source apportioned daily mean  $\text{PM}_{10}$  at the Inner London background site Kensington & Chelsea 1 with episodes labelled**

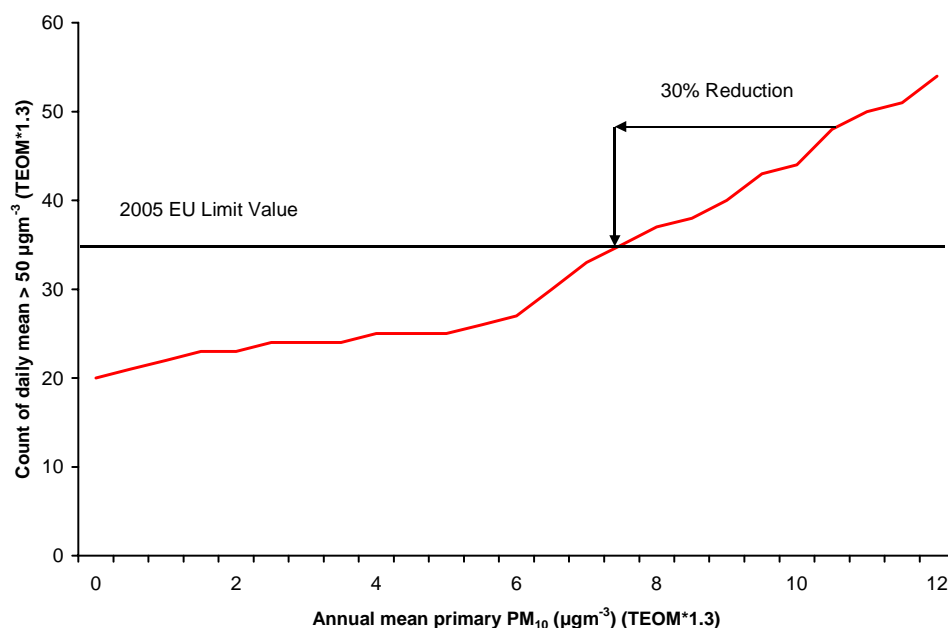
Figure 5 shows the source apportionment of  $\text{PM}_{10}$  measured at Kensington & Chelsea 2; a typical Inner London roadside site. Due to its proximity to a major road the site experienced more primary  $\text{PM}_{10}$  particulate than the background site. This additional primary  $\text{PM}_{10}$  increased the impact of episodes A to E, with additional daily means above  $50 \mu\text{gm}^{-3}$

(TEOM\*1.3). The additional primary  $\text{PM}_{10}$  also led to 4 further episodes not measured at the background site; these are labelled 1 to 4 in Figure 3. Episodes 1 to 3 were caused by a mixture of local primary and secondary particulate. Episode 4 was dominated by local primary sources and is a classic winter-time pollution episode occurring in poor dispersion conditions.



**Figure 5 Source apportioned daily mean  $\text{PM}_{10}$  at the Inner London roadside site Kensington & Chelsea 2 with episodes labelled.**

Air quality management initiatives on a local or even city-wide scale can only have impact on the primary  $\text{PM}_{10}$ . Given this it is possible to calculate the reduction in primary  $\text{PM}_{10}$  concentration necessary to achieve the 2005 Limit Value at the Kensington & Chelsea 2 site. Figure 6 shows the relationship between the annual mean primary  $\text{PM}_{10}$  at Kensington & Chelsea 2 and the annual number of days with mean  $\text{PM}_{10} > 50 \mu\text{gm}^{-3}$  based on the source apportionment of measurements during 2003. Changes in annual mean primary  $\text{PM}_{10}$  were assumed to apply equally to all primary  $\text{PM}_{10}$  at the site throughout the year. This analysis showed that the annual mean concentration of primary  $\text{PM}_{10}$  would have to be reduced by 30% to achieve the 2005 EU Limit Value for 2003 measurements at this location.



**Figure 6 Relationship between annual mean concentration of primary PM<sub>10</sub> at Kensington & Chelsea 2 and the annual number of days with mean PM<sub>10</sub> > 50 µgm<sup>-3</sup>**

Source apportionment of PM<sub>10</sub> has determined that the main episodes, during February, March, April and August were caused by increased PM<sub>10</sub> from secondary and natural sources. The secondary and natural PM<sub>10</sub> episodes are similar to those experienced during 1996. A comparison between the severity of the secondary PM<sub>10</sub> episodes during 2003 and those in 1996 is not straightforward and would need to account for the reduction in emissions of secondary PM<sub>10</sub> precursors. However, source apportionment allowed a simple comparison of the number of daily means above 50 µgm<sup>-3</sup> due solely to secondary and natural particulate, and showed that 34 such daily means were measured during 1996 compared to around 20 during 2003.

## Conclusions

During 2003 London experienced a series of PM<sub>10</sub> episodes. Provisional measurements show that the 2005 EU Limit Value was exceeded at roadside and kerbside sites in Inner London and at several such sites in Outer London. Source apportionment allowed the causes of the PM<sub>10</sub> episodes to be determined, and showed that the 2003 episodes were largely caused by regional sources; secondary and natural PM<sub>10</sub>. Nevertheless attainment of the 2005 EU Limit Value, during the circumstances experienced during 2003, can be achieved at roadside sites in Inner London by a reduction in the concentration (and therefore emissions) of local primary PM<sub>10</sub>. It is estimated that a local primary PM<sub>10</sub> reduction of 30% would be required to attain the 2005 EU Limit Value at the typical Inner London roadside site Kensington & Chelsea 2.

## Acknowledgements

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