Analysis of Partisol blank filter weights

Institute for Transport Studies
University of Leeds

David Carslaw

(*version 23rd May 2008*)

UNIVERSITY OF LEEDS

Summary key points

Blank filters

1. This report summarises the analysis of blank filter weights from Partisol measurements across the UK from 2000-2007. It focusses on characterising and understanding several artefacts in the time series of blank and exposed filter weights.

2. There is evidence of changes in the initial filter weight, most notably during 2005 with one set of filters weighing around 150 mg and the other 140 mg. It is shown that these two filter classes are associated with different blank filter weights with the heavier filters being associated with about 2.5 μg m^{-3} more equivalent blank filter concentration compared with the lower weight filters.

3. There is strong evidence of a seasonal cycle in the filter blank weights, with higher concentrations being observed during the summer than winter. The difference between summer and winter concentrations is ∼3 μg m^{-3}. The seasonal cycle accounts a considerable amount of the variation in blank filter weights (on average). However, there is significant random variation and evidence of a more systematic change during 2006.

4. Given the known important effect that water vapour has on quartz filters, the seasonal cycle of humidity may be important. However, the seasonal cycle more closely corresponds to water vapour *concentrations* rather than relative humidity, where the latter peaks during the winter months. Laboratory studies would be required to demonstrate the most relevant humidity metric affecting quartz filters.

5. A change-point analysis strongly suggests that there was a step-change in filter weights of ∼3 μg m^{-3} around April 2006 (95 % confidence interval from December 2005 – May 2006). No identifiable reason for this change has yet been found. However, it is likely that some currently unknown factor has influenced filter weights in a systematic way e.g. change in filter material.

6. There is no evidence of a statistically significant difference between filters used in PM_{10} and $PM_{2.5}$ instruments.

7. An important consideration is the date assumed in the analysis. Three dates were available for analysis: when the filter was initially weighed, when it was deployed and the final weight date. The most appropriate date to use is not clear.

Measured particle concentrations

8. Absolute gravimetric concentrations of PM_{10} and $PM_{2.5}$ have also been analysed in a similar way to the blank filter weights. The seasonal cycle of measured concentrations differs from the blank filters in that the maximum occurs during springtime. This finding shows that the artefacts affecting the blank filters do not dominate the exposed filters.

9. There is some evidence to suggest that the increased blank filter weights observed during 2006/7 is also manifest in exposed filters. For example, an analysis of $PM_{2.5}$ at Harwell indicates a similar change-point time to the blank filters (April 2006). However, there is greater uncertainty in the timing of the change compared with the blank filters – the 95 % confidence intervals spans February 2006 – April 2007. It is more difficult to detect and characterise such changes in exposed filters due to the dominance of the particles themselves.

Comparison with other data sets

10. Blank filter data were also available from AEA. The data set is more limited than the BV data but does span several sites and several years. The AEA filters were all Whatman QMA quartz filters. The analysis showed that there was neither evidence for a seasonal effect nor a shift in the mean concentration. In addition, the AEA data show much less scatter than the BV filters. It is important to note however, that most of the AEA blanks remained in the analysis laboratory and were not field blanks, and that field blanks were associated with higher weights than the laboratory blanks.

11. Emfab filter data available from King's College London and weighted by BV shows no indication of a step change during 2006, but does show a weak seasonal cycle with higher filter weights during summertime.

Laboratory effects

12. Laboratory conditions pre 2005 were frequently outside the relative humidity and temperature ranges set by EN12341. In addition, pre-2005 data show a strong seasonal effect for relative humidity (RH), peaking in August. This is consistent with outside air being brought in and maintained at a constant temperature. However, there does not appear to be a direct link between laboratory environmental conditions and blank filter weights based on the information available.

13. The RH post-2005 underwent additional systematic changes. From April/May 2006 there was a decrease in humidity until the end of 2006, then a levelling off during 2007. Only during 2007 has the humidity been within the required tolerances. Evidence of a change in laboratory humidity around April/May 2006 does coincide with the change in the mean of blank filter weights, although this does not mean they are causal but this may be an important factor.

Potential for data correction

14. An important issue is whether the filters can be corrected in some way. From the analysis of the data it would appear that correction at individual sites on a daily or monthly basis would not be possible in a robust way. In part this is due to the lack of frequent blank filters at individual sites, but also due to the large amount of scatter on a monthly basis.

15. Correction on an annual basis is also not without difficulties. For annual data, the systematic increase in blank filter weights in 2006/7 is the most important effect (which *may* also be detected in exposed filters). However, the lack of an underlying reason for this change is problematic. It is not known, for example, whether future exposed blanks will also show this change and that filters will continue to show a consistent artefact. It is important therefore that filters continue to be analysed in future and that Defra remain vigilant with respect to these issues.

Contents

1. Introduction

This report summarises the analysis of Partisol filters used on the UK network. Recent analysis by Bureau Veritas (BV) of blank filter weights showed that there was evidence of an increase in filter weight over the period 2000 – 2007 [3]. Further details of the background to these issues and a summary of sampling protocols etc. can be found in that report. The current report analyses these data in a variety of other ways with the aim of providing clues as to the cause of these changes and to characterise them. In particular, seasonal and change-point analysis techniques are used.

This report also considers exposed filters and analyses them in similar ways to the blank filters. The intention was to determine whether the types of changes observed in the blank filters are also manifest in exposed filters.

This report focuses on the empirical analysis of these data and does not consider issues to do with the metrology of particle measurements, which is treated in detail elsewhere.

2. General overview of data

2.1. Description of available data

A full description of the data is given in the BV report [3] and only a brief overview is given here. The analysis in this report focusses on quartz filters weighed by BV. Data were provided by BV in a spreadsheet containing the following information:

Filter weights Information was available on filter weights before and after deployment and their difference. These were converted into concentration equivalents, which have been used in this report.

Dates Dates were provided relating to when the filters were initially and finally weighed and their deployment date in the field.

Other information Information was available relating to the environmental conditions prevailing in the laboratory at the time the filters were weighed. This information includes the relative humidity (RH), temperature and person identified as weighing the filters. Information was also provide on the site at which the filters were deployed.[1]

The various dates are potentially important because of their link to any external (or laboratory) influence. If for example, external environmental effects were important a date somewhere between filter deployment and final weighing might be important. At the outset it is not known which date or dates are the most important. Therefore, most of the analysis in this report has been repeated using a range of dates. However, for clarity plots are analysed on the basis of the date the filters were placed in the sampler. The effect of choosing different dates is considered in detail in section (§4).

Note that blank filter weights have been expressed as a concentration equivalent in μg m^{-3} to allow a more direct comparison with exposed filter concentrations.

---

[1] Further detailed information on the laboratory conditions was also available e.g. maximum and 24 hour mean humidity.

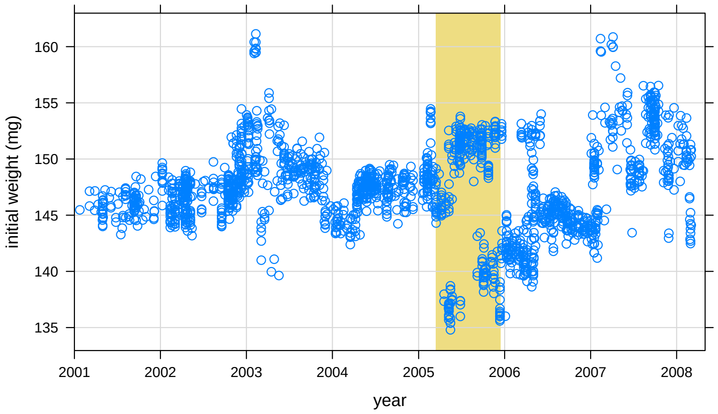


Figure 1: Time series of absolute filter weight based on initial weight measurements.

2.2. Absolute filter weights

This section considers the variation in initial *absolute* filter weight. Figure 1 shows the time series of weights, which are for the most part around 145 mg. However, it is also clear there is a disjoint in this series, shown by the shaded area during 2005. For this period it seems that two different filter types were used.

The two different initial filter types may also have some influence on the blank filter weights. To test this idea, a subset of data shown by the shaded area was analysed. This subset contains two discrete groups of filters, defined as having an initial weight above or below 145 mg. An important question is whether these two types of filter accumulate mass in different ways. Figure 2 shows how the initial filter weight varies with blank filter concentration. This plot shows there *is* a difference between the two types of filter: the filters with a higher initial weight result in higher blank filter concentrations. The filters with a higher initial weight are associated with blank filters that have concentrations of 2.8±0.4 µg m^{-3} (95 % confidence interval in the mean) and the filters with a lower initial weight with concentrations of 0.3±0.3 µg m^{-3}. Statistically, it can be shown that there is a clear difference in these two groups in their mean blank concentrations of 2.5 µg m^{-3}. This is a potentially important finding as it shows that nominally identical filters have different characteristics in terms of their blank filter concentrations. The difference of 2.5 µg m^{-3} is also a considerable fraction of absolute blank filter weights, as shown in the next section.

2.3. Summary of data

This section provides a brief overview of the data in order to develop an understanding of its characteristics. The data were first averaged by site and by month, as shown in Figure 3a. This plot highlights several features. First, blank filter weights are almost always positive (i.e. it is unusual for blank filters to lose mass). This behaviour might be expected due to the known issue of water absorption by quartz filters. Second, there is considerable variation on a monthly basis of filter weights, with some indication that weights tend to be higher during the summer months. Finally, there is evidence of a more systematic increase in filter weights during 2006/7. Figure 3b shows the individual points that make up Figure 3a, which highlights the variability in any one month of the blank filter weights.

It is also useful to consider at which sites the blank filters are placed and the variation in blank filter weight by individual site, which is shown in Figure 4. This plot also highlights

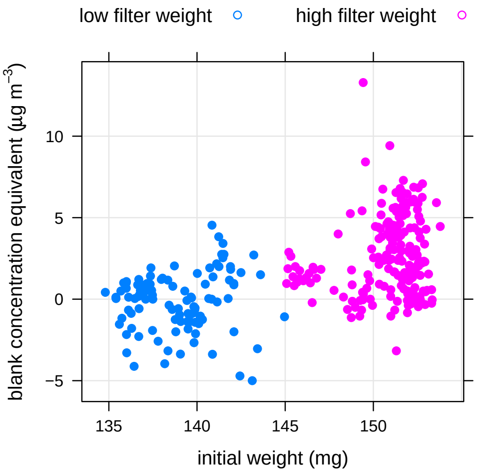
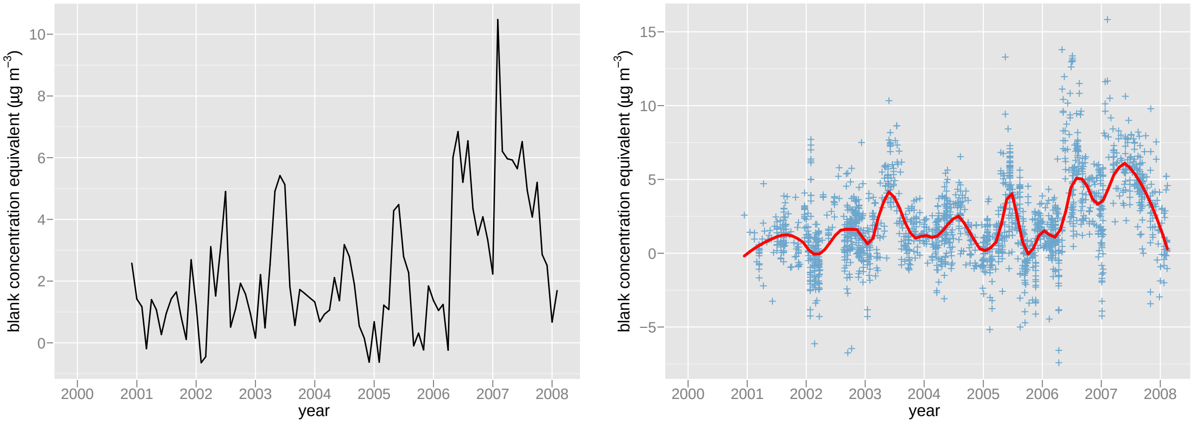


Figure 2: Relationship between initial filter weight and blank filter concentrations. The period covered is shown by the shaded area in Figure 1.



(a) **(b)**

Figure 3: a) Monthly time series of blank filter weights averaged across all sites, b) Time series of blank filter weights for all sites. Solid line shows locally-weighted regression smooth.

some important issues. At most sites, there is an irregular exposure of blank filters, making it difficult to assess specific sites for trends etc. However, at three of the sites (Auchencorth PM_{10} and $PM_{2.5}$) and Derby (non AURN) filters are exposed on a more frequent basis than other sites. These three sites appear to show similar characteristics, with a tendency towards increasing filter weights from the beginning of 2005 to the end of 2007, followed by a decrease. Unfortunately data are not available at these sites prior to 2005. It is difficult from these data alone to draw robust conclusions, but there does appear to be a consistent change across these sites.

No significant difference between PM_{10} and $PM_{2.5}$ blank filter weights was found using a t-test: they both undergo very similar changes (on average). This finding is better shown in Figure 5. Note that the disagreement in February 2007 is more likely due to the small sample size than any systematic difference (four PM_{10} filters and one $PM_{2.5}$ filter).

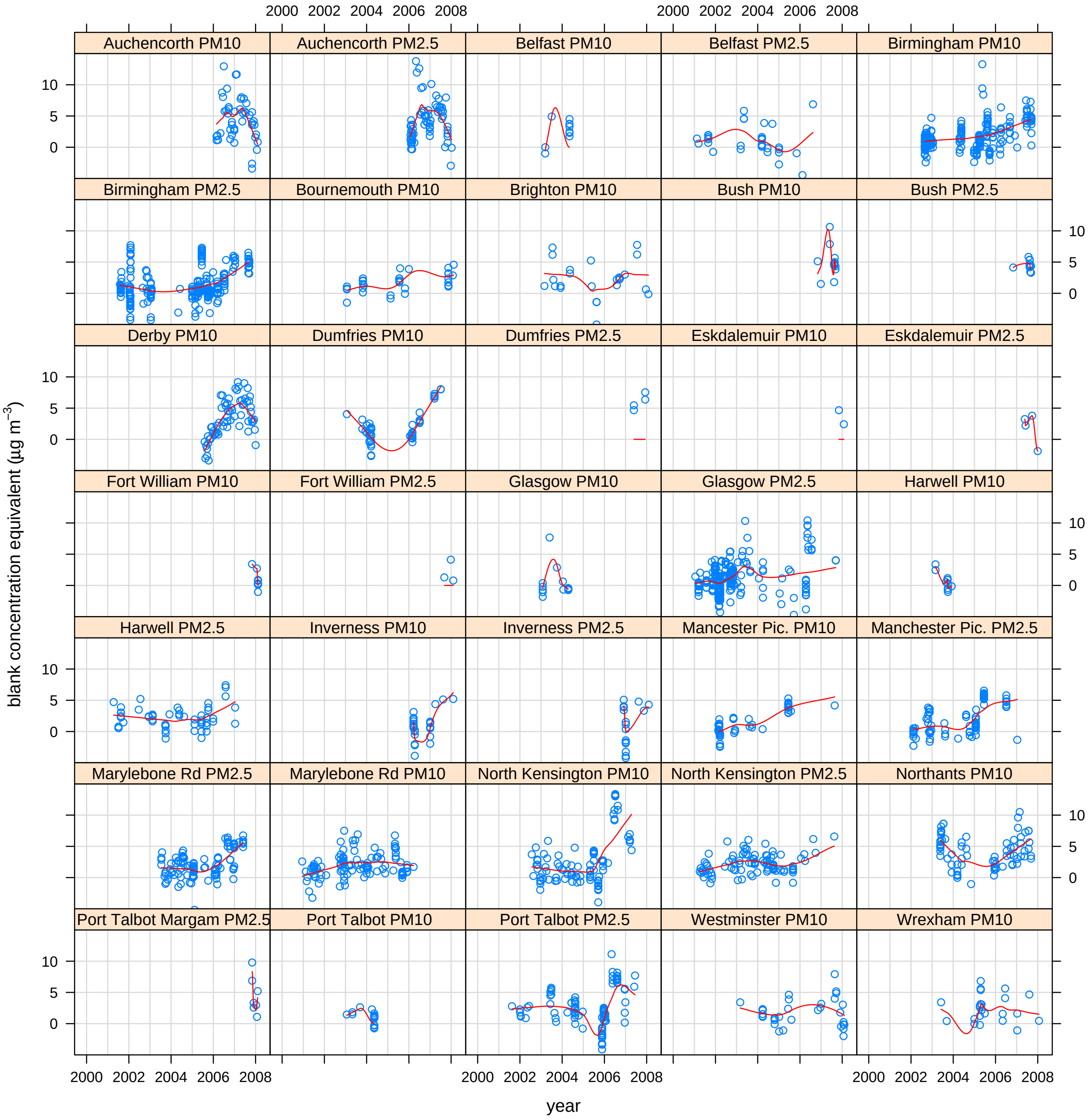


Figure 4: Individual blank filter weights by site.

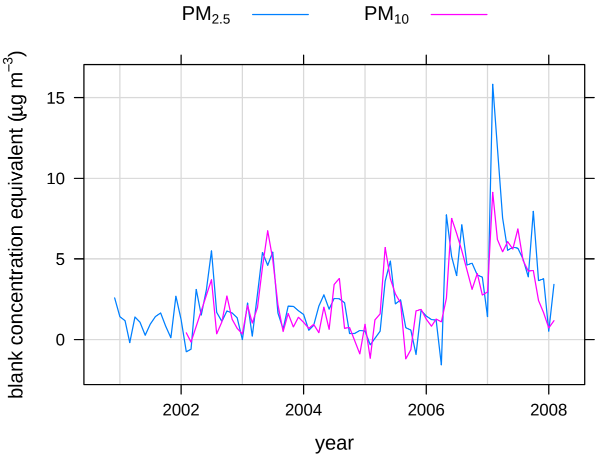


Figure 5: Monthly mean time series of PM$_{10}$ and PM$_{2.5}$ blank filter concentrations.

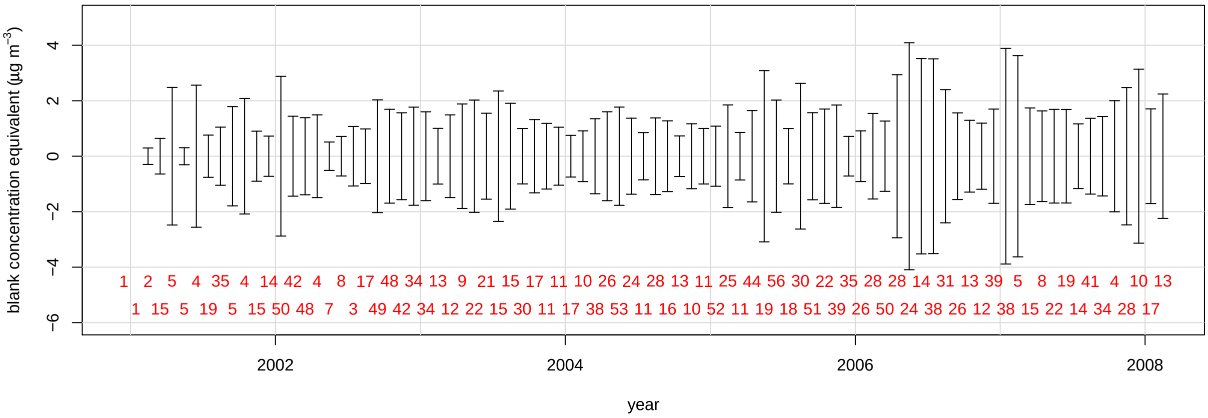


Figure 6: Monthly standard deviation ($\pm\sigma$) of blank filter concentration equivalents. The numbers adjacent to the x-axis show the sample size in each month.

2.4. Variability of blank filter weights

Figure 3b provides an indication of how variable the blank filter weights are by month. However, a better way to characterise the variability is to consider the standard deviation (σ) by month. A plot of the variability by month is shown in Figure 6. Overall the mean variation on this basis is ± 1.6 µg m^{-3} (range 0.3 – 4.1 µg m^{-3}). A summary of the mean, minimum, maximum and σ by month is given in Table 2 in Appendix A.

Figure 6 also shows periods where filter weights were highly variable. Two periods stand out: April – August 2006 (which coincides with a systematic change in filter weights as described in (§5), and January – February 2007, which coincides with the lab move date (and a month with only 5 filters).

Also calculated was the variation by site and by year. The intention here was to establish the extent to which changes were consistent across sites and explore whether there was evidence for systematic differences between sites. For each site and each year the 95 % confidence interval in the mean blank concentration was calculated; using sites that had at least 6 measurements during one year. Another motivation for this analysis was to help establish the robustness of any adjustment made to the data. As shown in Figure 7 there is some degree of consistency by site. Taking 2007 as an example, most points are clustered about 5 µg m^{-3}. There are also two outliers: Inverness and Bournemouth. It can be shown for these two sites that filters were placed in the sampler during the winter i.e. when blank filter weights tend to be low – see (§4). These results indicate that artefacts in blank filter weights are not site dependent, which may help if any correction were made.

The data for 2006 are more complex because these data straddle the change-point identified in April 2006. Indeed, if data from January – April 2006 are excluded the blank filter weights increase for 2006 compared with those shown in Figure 7.

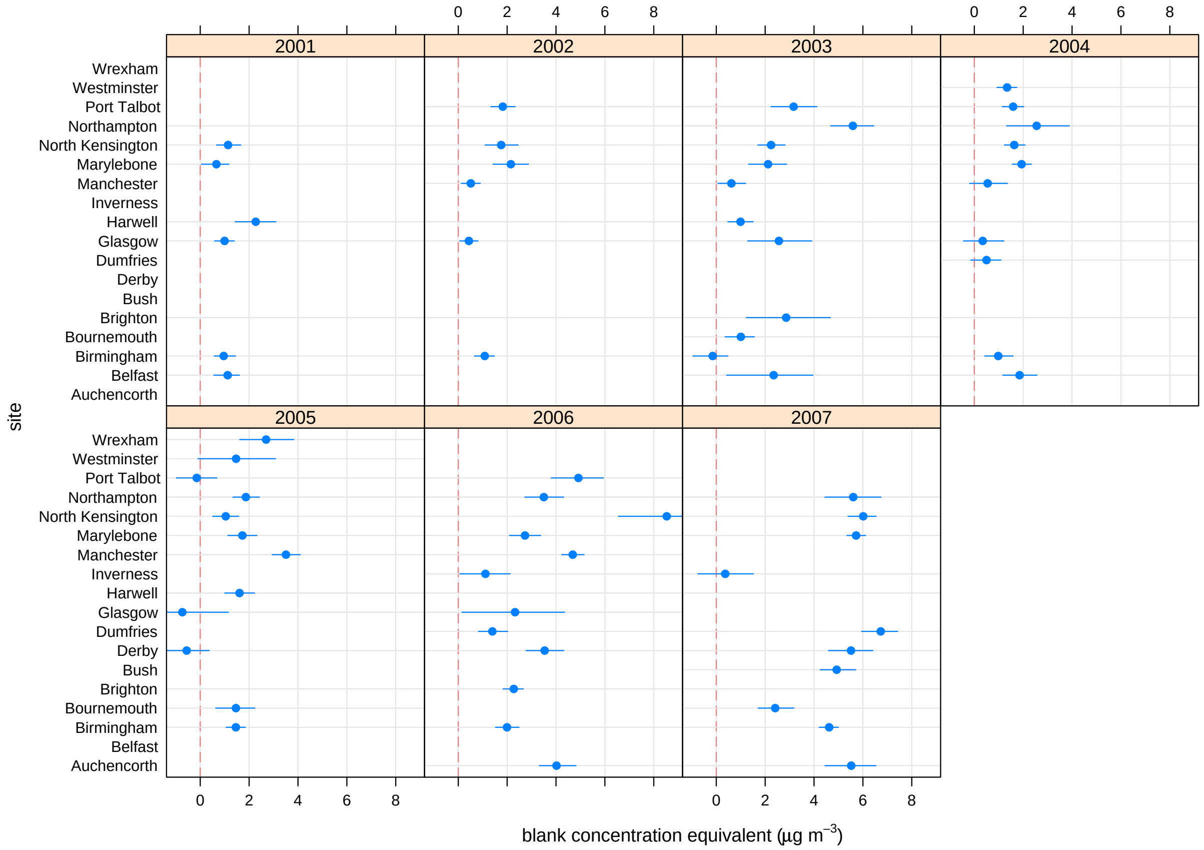


Figure 7: Means and 95 % confidence intervals in the mean of blank filter concentrations by year and by site.

3. Laboratory effects

The data set obtained from BV linked each filter with information from the laboratory used for filter weighing. Of particular interest is whether there are any obvious effects due to laboratory conditions e.g. humidity and temperature. These parameters (along with others) were recorded and compiled. Figure 8 shows the effects of temperature and humidity in the laboratory recorded at the initial and final weighing. Although there is considerable scatter in the plots, there is no obvious link between blank filter weights and RH or temperature. For relative humidity, the bulk of the points are close to 50 % humidity and temperatures are mostly between 20-21°C. However, there are significant departures from the tolerances allowed under EN12341 (RH 50±5 % and temperature 20 ±1°C). Most of the departures from these conditions occurred earlier in the time series. Note that these measurements are spot measurements and cannot provide information concerning the conditions throughout conditioning or weighing.

Also considered was whether there was any effect due to the analyst involved in weighting the filters – both before and after exposure. No effect was found. Furthermore, the length of time between the initial and final weighing was also considered, and again no significant effect was found. Overall therefore, there is no discernable effect on filter weights due to laboratory conditions on this basis.

There is evidence when considering the laboratory data overall of a seasonal cycle in humidity. Additionally, the time series of laboratory humidity shows several features that may be important, as shown in Figure 9. First, up until 2005 the laboratory conditions

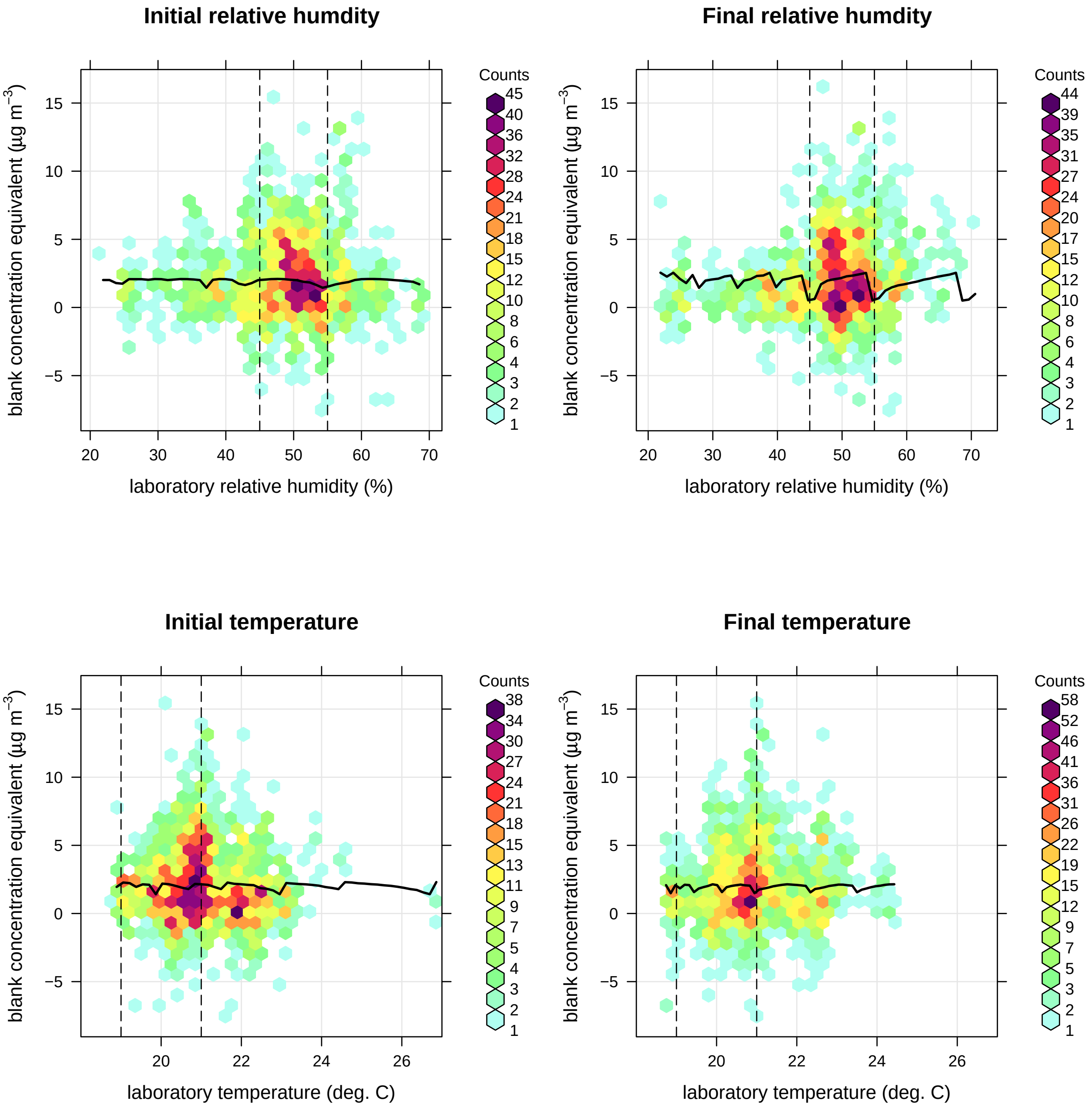


Figure 8: Relationship between blank filter weight and laboratory temperature and humidity. The plots show the number of counts in each 'bin' to highlight the overall distribution. Plots are split by initial and final conditions i.e. laboratory conditions when blank filters were first weighed and weighed after exposure. The solid line shows a smooth fit to the data. The dashed lines show the tolerances allowed under EN12341.

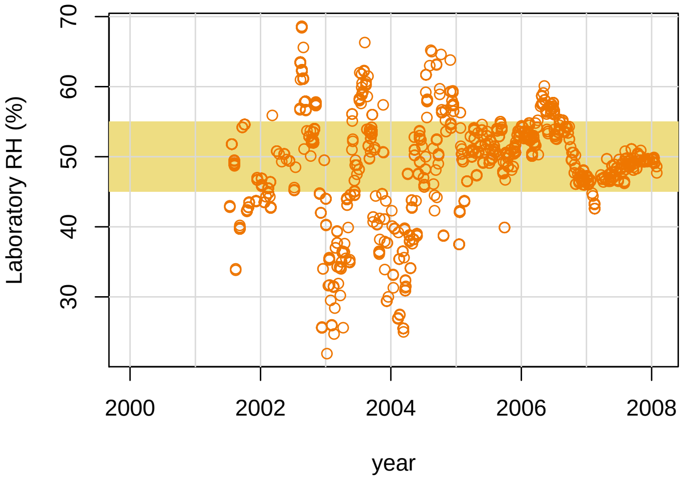


Figure 9: Time series of laboratory humidity at the time of initial filter weighing. The shaded area shows the tolerances allowed under EN12341.

were frequently (and significantly) outside the tolerances. Also, up until 2005 there was a strong seasonal effect. The seasonal cycle can be shown as peaking in August, consistent with the peak in *absolute* humidity shown in Figure 11 i.e. it does not follow the cycle of relative humidity. This might be expected when air is drawn from outside into a *fixed temperature* environment – in which case the RH will peak at the same time as the absolute humidity. Figure 9 also shows some anomalous behavior during 2006 in the spring/summer where there is a marked downward change in RH. The timing of this change (difficult to quantify using the methods used elsewhere in this report) does seem to be consistent with the changes that have been detected in blank and exposed filters i.e. around April/May 2006. However, how this type of change translates to a change in filter weight is more difficult to rationalise.

4. Seasonal effects

4.1. Analysis of blank filters

This section makes use of a seasonal trend decomposition technique based on locally weighted regression (STL) [2]. The idea here is to determine whether there is evidence of a seasonal cycle in the blank filter weights, which might help explain some of the variation and perhaps provide a way in which to correct the data.

Figure 10 shows the seasonal decomposition applied to the entire data set. The top panel shows the raw data and the subsequent panels show the extracted components (trend, seasonal and remainder). The orange bar adjacent to the y-axis in each plot covers the same range of concentrations in each plot and helps to show how much variation there is in each component. On this basis the remainder component is the most important – but this is affected by a few outliers, most notably in February 2007.[2]

---

[2] Decomposing the data into trend, seasonal and remainder components is actually useful for showing interesting outliers. In this case the February 2007 outlier is very clear in the remainder series. This date coincides with the laboratory move date, which may be a important factor for blank filter weights for that month. However, there are also few data avilable in this month (see Table 2. There is also perhaps some

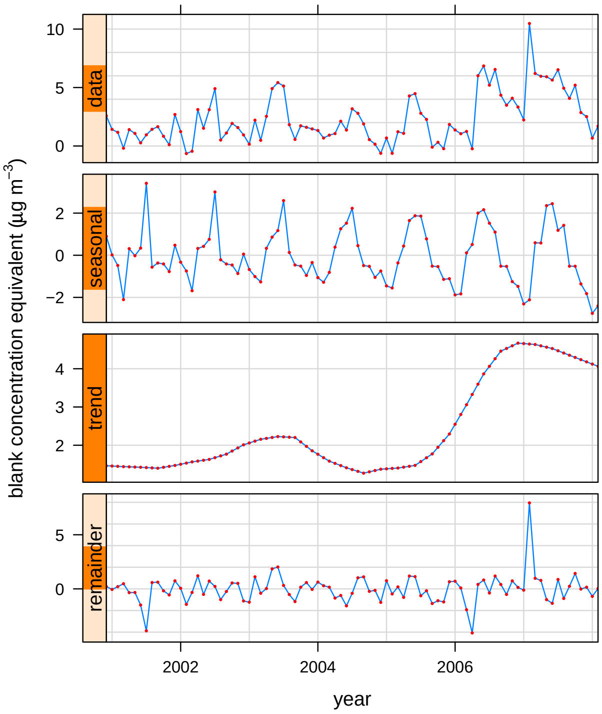


Figure 10: Monthly time series of blank filter weights decomposed into trend, seasonal and remainder components using the STL technique.

Figure 10 also shows that the filter weights tend to be higher during the summer months compared with winter months. Water vapour has been widely reported as having a strong influence of quartz filters. Most analysis of these effects have considered relative humidity (RH) as a measure of atmospheric water vapour content. RH values are lower during the summer months (typically around 65-70 %) and higher during the winter months (typically around 80-85 %). It might be expected therefore that the absorption of water would be more important during the winter months i.e. opposite to the findings here. However, RH might not be the most appropriate measure of atmospheric water vapour content. A more appropriate measure could be *absolute humidity*, which is a measure of the absolute water vapour content in air (typically measured in g per cubic metre). Based on an analysis of meteorological data from Heathrow Airport, it can be shown that the absolute humidity in June is almost a factor of two greater than that in January. The seasonal variation in temperature, relative and absolute humidity is shown in Figure 11.

4.2. Effect of chosen date on the seasonal cycle

As discussed previously there are several dates which can be used in the analysis of both seasonal influences and more systematic changes. The effect on the seasonal variation of the blank filter weights due to the date chosen is shown in Figure 12. The form of the seasonal cycle changes depending on which date is considered to be the most appropriate. If, for example it was thought that a date close to the final filter weigh date had most influence on filter weights, then the final plot in Figure 12, which peaks in July would be most relevant. However, if the analysis is based on the initial filter weighing date then the seasonal cycle would peak in May (first plot). Based on the average date between initial

---

evidence to suggest that the filter weights take time to ‘settle down’ after this time.

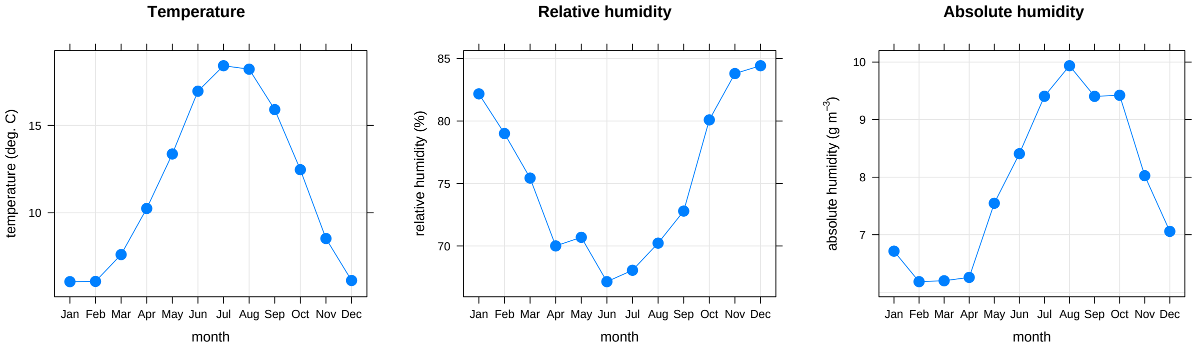


Figure 11: Variation in temperature, relative humidity and absolute humidity at the Met Office Heathrow site. The period analysed corresponds to November 2000 to December 2007.

and final weighings suggests a peak in July.

The plots shown in Figure 12 all suggest a strong summertime influence on blank filter weights. None of the plots convincingly match the seasonal cycles in temperature, relative or absolute humidity shown in Figure 11. Indeed, a regression analysis of monthly mean weights with different environmental variables shows the strongest effect with temperature rather than relative humidity.[3] The best correlation was obtained using the final filter weigh date, but even here the relationship is not strong ($R^2 = 0.30$). Clearly this analysis has had to make many assumptions. The actual environmental conditions affecting the filters while in the field are not known and if this information were available it might provide stronger clues as to the factors that affect the blank filters.

---

[3] The regression analysis was performed on monthly mean data by first subtracting 3 µg m^{-3} from May 2006 – February 2008 data i.e. removing the offset.

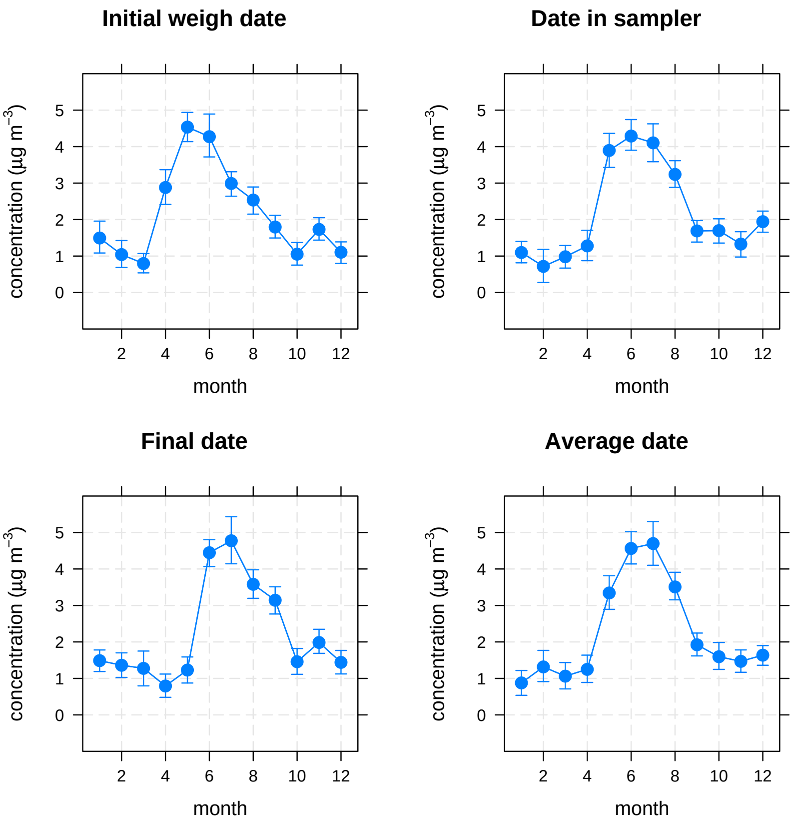


Figure 12: Seasonal variation in blank filter weights varying depending on the date chosen for the analysis. These plots were calculated using raw filter weight data and were not deseasonalised first.

5. Change-point analysis

5.1. Overview of method

It is clear from (§4) that seasonal effects do not account for all of the variation observed in the blank filter weights. In particular, there is evidence from Figure 3a that filter weights were much higher on average during 2006 and 2007. To help determine whether a particular "change-point" can be identified, when weights increased from one level to another, a change-point analysis has been applied.

The analysis is based on that discussed in [1]. Briefly, cusums (cumulative sums) are first calculated to identify candidate change-points – applied to data that have been rescaled such that the mean is zero. Thousands of cusums are then calculated by randomizing the data to determine whether the candidate change-point is statistically significant. The uncertainty in time of any change-points is then determined by applying bootstrap resampling.

The technique has been recently refined to account for correlated errors through the application of a block bootstrap [4]. Taking account of the correlation has the effect of increasing the uncertainty in the timing of the change-points; in this case by a small margin. In addition, the timing of the change-point is now based on the residual sum of squares (RSS), which is also applied to the randomised data. Testing showed that the cusum was in some cases affected by outliers close to the "true" date. In most cases there is however, good agreement between the cusum and RSS methods. The analysis here has only tried to seek

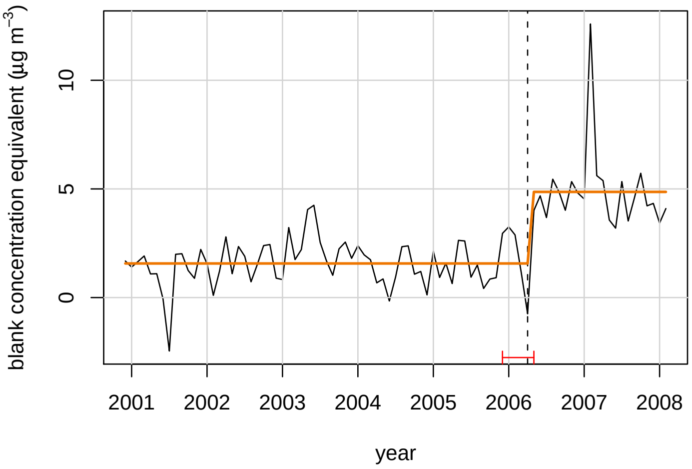


Figure 13: Change-point analysis of deseasonalised monthly mean blank filter weights. The black line shows the deseasonalised data, the thick line shows the mean level filter weights before and after the change-point, the dashed line shows the best estimate of the change-point timing and the error bars adjacent to the x-axis show the 95 % confidence intervals in the timing of the change-point.

evidence of a single change-point and not multiple changes. To improve the identification of the timing of any changes, the data were first deseasonalised as described in (§4).

5.2. Application to blank filters

Figure 13 shows the results of the change-point analysis. The black line shows the deseasonalised data, which correspond to the trend + remainder shown in Figure 10. The vertical dashed line shows the location of the best estimate of the detected (single) change-point in April 2006 (where May 2006 represents the first month after the change). The error bar adjacent to the x-axis show the 95 % confidence intervals in the timing of the change-point (December 2005 – May 2006). The thick line shows the mean level filter weights before and after the change-point. These results provide strong evidence that there was significant change in the mean blank filter weight around April/May 2006, where concentrations increased by about 3 µg m^{-3}.

The timing of the change-point may depend on the filter weigh date assumed. The analysis above was based on the date the filter was placed in the sampler. The analysis was repeated using the date the filter was placed in the sampler and the final weigh date. The results are shown in Table 1. The initial, mean and the final weigh dates give a consistent timing for the change around April 2006. The smallest uncertainty in the timing of a change-point is for the final weigh date and the date the filter was placed in the sampler.

Change-points of around April/May 2006 also coincide with increased variability in the blank filter weights (see (§2.4) and Figure 6). One explanation for such behaviour is that there is transition period from lower blank filter weights to higher ones – for example due to some change in filter material. Such a change would not occur instantaneously because of the different times batches of filters are placed in the field and differing exposure lengths etc. It would take time for such changes to "work through".

Apart from some short-term variability around the time of the lab move (January/February 2007), the variability of blank filter weights after the April/May 2006 change-point is similar

Table 1: Timing of change-points and variation with filter date assumed.

Date used	Change-point date	95 % uncertainty in change-point
Initial weigh date	April 2006	December 2005 – June 2006
Date in sampler	April 2006	December 2005 – May 2006
Final weigh date	April 2006	January 2006 – June 2006
Mean weigh date	April 2006	December 2005 – June 2006

to that prior to the change-point. Changes in filter weight post April/May 2006 are therefore mostly characterised by an increase in the mean and not an increase in variability.

6. Analysis of particle concentration measurements

6.1. Partisol measurements

The analysis so far has focussed on the weights of blank filters. An important issue is the extent to which similar patterns can be detected in the actual measurements of PM_{10} and $PM_{2.5}$. Daily data were downloaded for 34 time series of both PM_{10} and $PM_{2.5}$ and analysed. For simplicity, data were first averaged across all sites and monthly means calculated as shown in Figure 14. Note that only sites with > 1500 days of data were used in Figure 14 (17 sites) because many of the more recent sites are in Scotland and would result in a bias due to their lower concentrations. This plot shows that concentrations tended to increase from 2000-2007. Also shown are clear periods of elevated concentrations, with the most notable observed in spring 2003.

The monthly mean time series for these sites is shown in Figure 15.

6.2. Seasonal effects

The seasonal cycle has been analysed in much the same way as described in (§4). The measured data show a different seasonal cycle compared with the filter blanks. For the measured concentrations, the highest concentrations are observed during the spring, as shown in Figure 16. The peak in concentration in March is consistent with long-range transport of secondary aerosol. An exception appears to be 2006 where concentrations were not elevated during the spring. The trend component shown in Figure 16 confirms that at these sites concentrations increased from 2000-2007 by about 9 $\mu g\ m^{-3}$.

Averaging all months and calculating the 95 % confidence interval in the mean yields Figure 17. If the exposed filters (i.e. those used to sample) were affected in the same way as the blanks, the seasonal cycle would be different compared with that shown in Figure 17. In particular, measured concentrations during the summertime would be expected to be lower *if* they were for example affected by water absorption in the way described previously. These results do however show that different factors are likely to affect the blank and exposed filters.

6.3. Change-point analysis

A similar analysis to that described in (§5) was also applied to the site-averaged monthly measurements of PM_{10} and $PM_{2.5}$. These data were again deseasonalised to reduce the

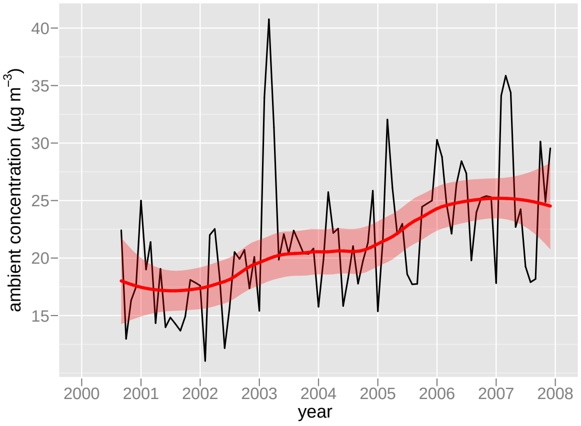


Figure 14: Monthly time series of PM_{10} and $PM_{2.5}$ concentrations averaged across all sites where there were at least 1500 valid days of measurements. The shading show the 95 % uncertainty intervals for the smooth fit.

variation first. The analysis showed that no significant change points could be detected. This finding is not too surprising because measured particle concentrations will be affected by a wide range of sources and "real" effects tend to dominate.

The analysis was therefore refined by specifically considering $PM_{2.5}$ concentrations at Harwell and at North Kensington. TEOM data were used to account for a large fraction of particle mass concentration by subtracting these concentrations from the gravimetric concentrations. The remainder would thus consist of particle mass not detected by TEOMs and any artefact due to filter issues. The Harwell site was chosen for several reasons – it is rural and not dominated by local particle sources and there is good data capture for both gravimetric and TEOM concentration measurements. For North Kensington there are no measurements of $PM_{2.5}$ made by TEOM. Therefore, $PM_{2.5}$ data were obtained from the Bexley 2 LAQN suburban site, which should provide reasonably representative urban background $PM_{2.5}$ concentrations.[4] However, it might be expected that the Harwell site would give a better indication of other filter effects because of its more rural location.

The results of the analysis are shown in Figure 18. A statistically significant change-point was detected in April/May 2006 for Harwell $PM_{2.5}$ concentrations, although the 95 % confidence interval in the timing of it is wide (February 2006 – April 2007). For North Kensington a different change-point was detected (December 2005, 95 % confidence interval from July 2005 to November 2006). For both sites the uncertainty interval for the change-point includes April/May 2006. The wider uncertainties are in part due to the noisier data compared with that for the blank filter weights. Taken together there is *some* evidence therefore that the *measured* particle concentrations reflect the patterns shown in the blank filter weights. This evidence is not definitive, but it appears likely that the problems that affect the blank filter weights in 2006/7 also affect the exposed filters.

Another outcome of this analysis is that the greatest difference in TEOM and gravimetric $PM_{2.5}$ concentrations at Harwell occurs during springtime conditions i.e. the time of year that appears to be consistently dominated by long-range transport.

---

[4] Six months data from the Bexley 2 site were missing during 2000/2001. These data were replaced by the mean values over the entire time series from 2000 – 2007.

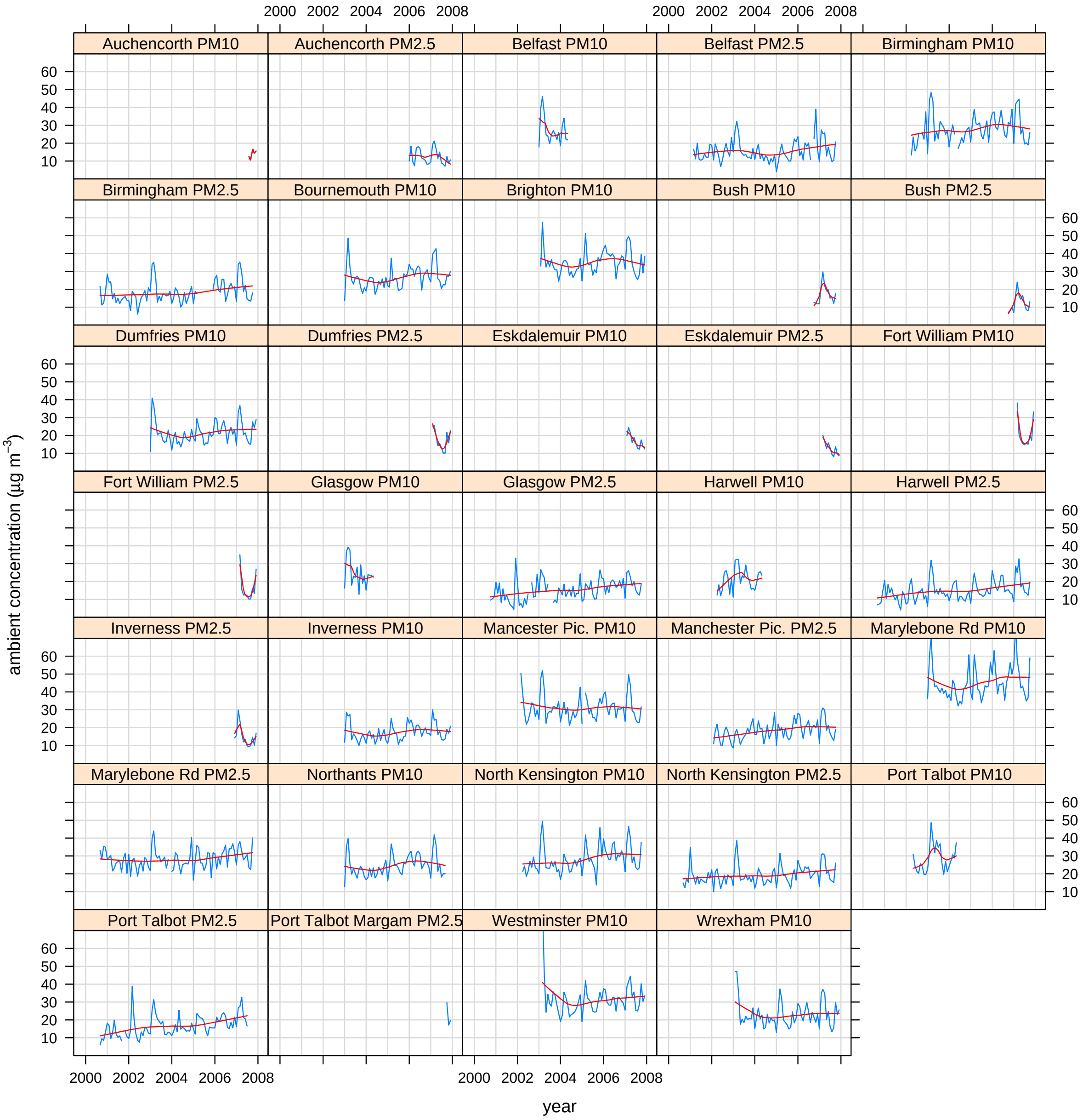


Figure 15: Monthly time series of PM_{10} and $PM_{2.5}$ concentrations by site.

6.4. FDMS measurements

It is useful to consider trends in other particle measurements to determine whether there is any evidence of increases throughout 2006/7. In particular measurements made by the Filter Dynamics Measurement System (FDMS), should provide a better indication of gravimetric particle mass compared with TEOM, due to unquantified volatile losses on the latter. Time series data for PM_{10} were provided by King's College London for the North Kensington site, the monthly means of which are shown in Figure 19. If anything, concentrations measured in this way *decreased* through 2006/7 as highlighted by the smooth fit line.

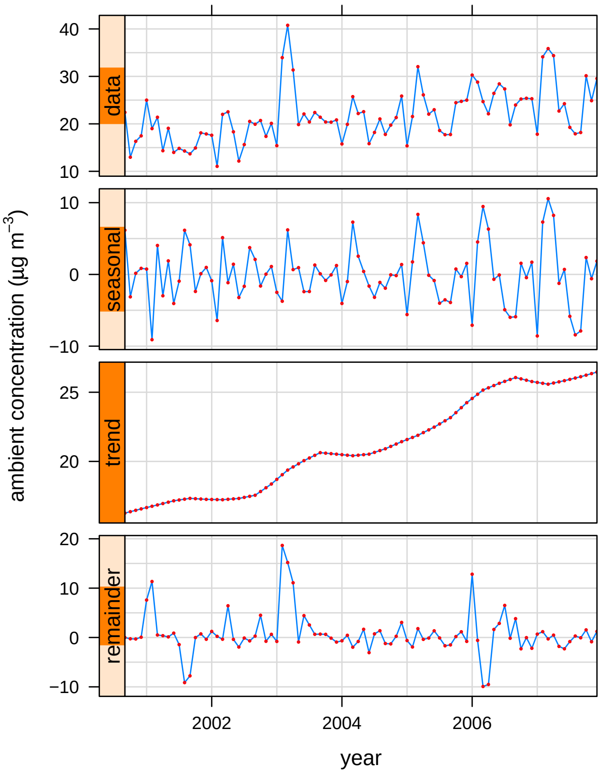


Figure 16: Monthly time series of mean PM_{10} and $PM_{2.5}$ concentrations decomposed into trend, seasonal and remainder components using the STL technique. Data are for the 17 sites where there were at least 1500 days of valid measurements.

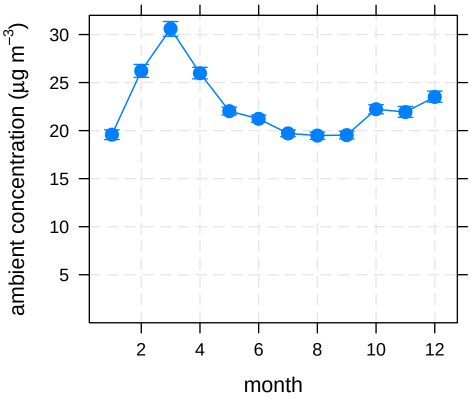


Figure 17: Variation in monthly mean concentrations of PM_{10} and $PM_{2.5}$. Error bars show the 95 % confidence intervals in the mean.

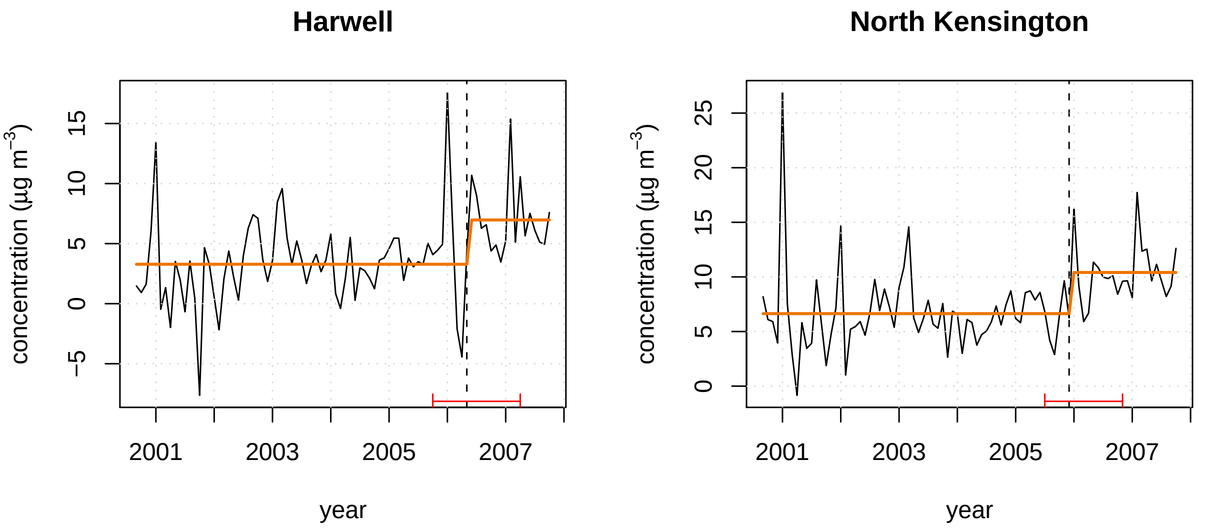


Figure 18: Change-point analysis of $PM_{2.5}$ concentrations at Harwell and North Kensington. The black line shows the deseasonalised data, the thick line shows the mean level filter weights before and after the change-point, the dashed line shows the best estimate of the change-point timing and the error bars adjacent to the x-axis show the 95 % confidence intervals in the timing of the change-point.

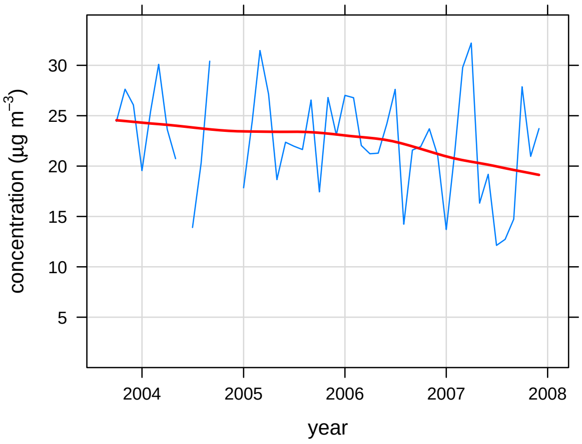


Figure 19: Monthly mean PM_{10} FDMS particle measurements at North Kensington. The red line shows locally-weighted regression smooth fit.

7. Analysis of other filter data

7.1. Analysis of filters from AEA

A more limited set of blank filter data was available from AEA as part of their work with specific local authorities. Data were available from one site in Liverpool and four sites in Scunthorpe and one close to Heathrow Airport spanning a range of dates. In addition, AEA data from pre-2003 from a research campaign were also available through BV and these were also included. The research filters comprised both field blanks (33 filters) and laboratory blanks (45 filters). It is interesting to note that the field blanks weight more than the laboratory blanks (1.7 vs. 0.1 µg m^{-3}). A total of 78 filters from the research trials from Marylebone Road, Glasgow, Belfast, Northants, Bournemouth, Inverness, Wrexham

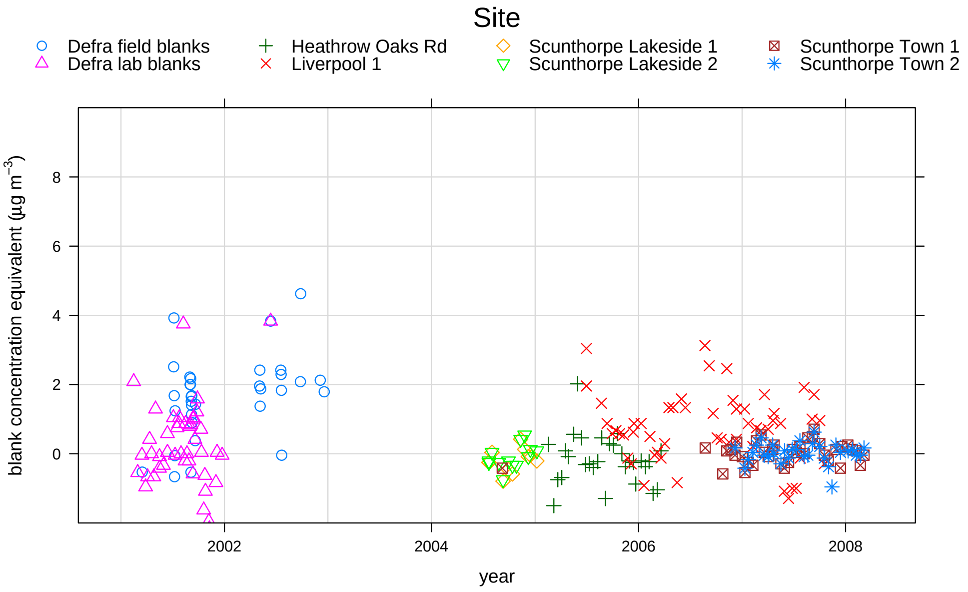


Figure 20: Time series of AEA blank filter concentrations at six sites using Whatman QMA quartz filers and results from Defra field trials. The y-axis scale is the same as Figure 3b for comparison.

and Dumfries. These data were processed to provide concentration-equivalent blank filter weights in the same as at the BV data. The dates used in this analysis relate to the initial filter weighing date. All filters tested were Whatman QMA quartz filters. In total there were 225 filters available. It is important to note however, that the AEA blanks remained in the analysis laboratory and were not field blanks.

The results from the analysis are shown in Figure 20, which shows individual blank filter concentrations by site. There are several points that can be made:

- There is no evidence of a systematic increase in blank filter weights during 2006/7 as is the case for BV filters.
- There is no evidence of a seasonal effect for these filters, although the sample size is limited compared with that for BV.
- There is more variability in the Liverpool data compared with other sites.
- Compared with the BV data, the scatter is considerably less.[5] The standard deviation (σ) of the AEA data is 1.1 $\mu g\ m^{-3}$ (254 samples) compared with 2.7 $\mu g\ m^{-3}$ (1943 samples) for the BV data.

Overall therefore, the AEA data seem to differ in two important respects compared with the BV data: there is no evidence of a seasonal cycle and no evidence of a shift in the mean during 2006/7.

Data were also available for other filter types and other sites, which have been included for completeness. These include:

- Neath Port Talbot (GN-4 Metricel membrane filters)

---

[5] Based on analysing the whole data set and not by month as before.

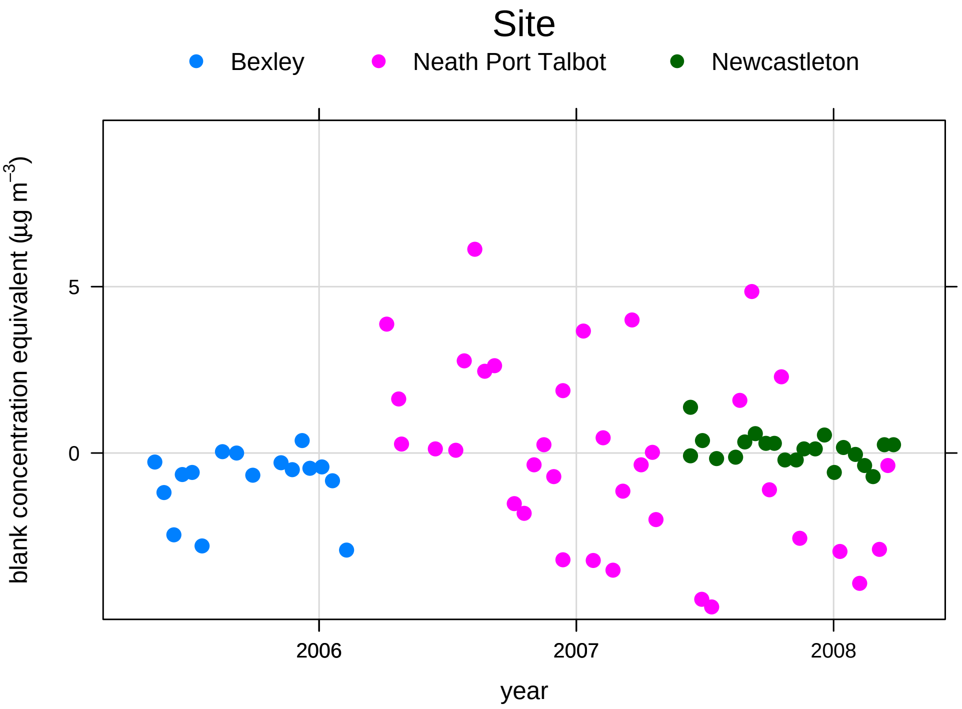


Figure 21: Time series of AEA blank filter concentrations at five sites and from research trials.

- Newcastleton (Emfab filters)
- Bexley (Millipore AQFA Quartz Fibre Filters)

The time series of blank concentrations is shown in Figure 21. While there is again scatter, there is no systematic evidence of either a seasonal influence or a step change.

7.2. Analysis of Emfab data from King's College London

Blank filter data were also available from King's College London from a Partisol 2025 PM_{10} instrument deployed at Earls Court roadside site. The individual blank data are shown on the left panel of Figure 22. The scatter in this time series is significantly less than that for the quartz filters. The quartz filters has a σ of 2.7 µg m^{-3} (1943 samples) while the Emfab data σ was 0.8 µg m^{-3} (311 samples). The analysis showed there was weak evidence of a seasonal cycle (right panel of Figure 22), with highest filter weights corresponding to summertime periods. However, compared with the quartz filters the seasonal cycle was much less important.

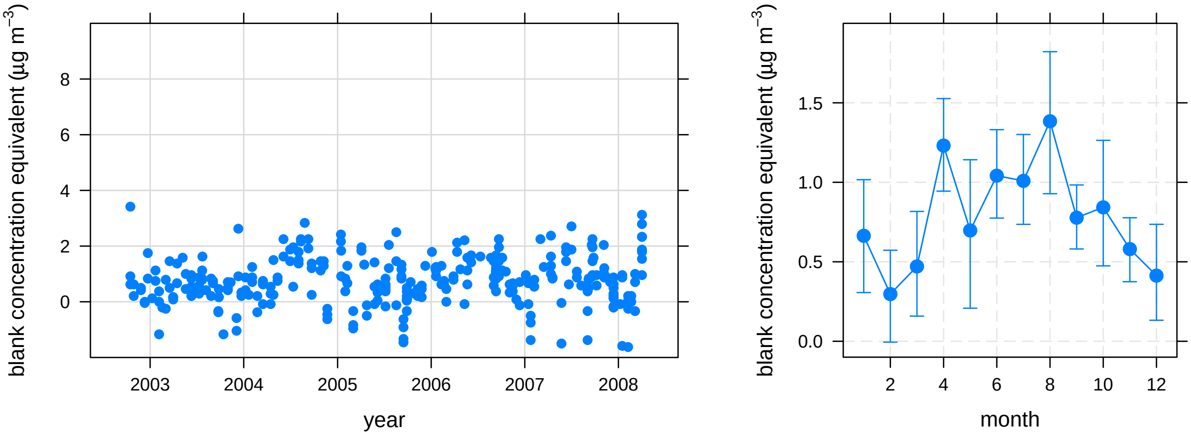


Figure 22: Left panel: Emfab blank filter weights for individual filters and, right panel monthly variation in Emfab filter weights with 95 % confidence intervals in the mean.

8. Data correction

An important consideration is whether there is an opportunity to correct the Partisol data on the UK network. To have confidence in any correction the underlying reasons for increased blank filter weights, and more importantly artefacts in exposed filters must first be established. This report identifies the presence of a seasonal cycle, which *could* be due to water absorption effects. It was suggested that absolute water vapour concentrations could be a more appropriate metric than relative humidity. However, laboratory analysis would be required to establish which metric is the most important.

An important issue is the large amount of scatter on a monthly basis in Figure 3b and 6. There are some months where the variation in blank filter weights is similar in magnitude to the measured particle concentrations. Correction on a monthly basis would therefore not be robust.

The correction of annual data is not without difficulty either. This is because of the presence of a more systematic increase in blank filter weights during 2006/7 is currently without explanation. It is also very difficult to establish to what extent any artefact may have affected the exposed filters. Although there is a similarity in the dates identified for the change-points in each case, the uncertainty intervals are wide in the case of the exposed filters. Furthermore, it cannot be assumed that any change affects blank and exposed filters in an identical way i.e. it is not known whether any artefacts are additive. Indeed it is reasonable to expect any material deposited on the surface of exposed filters will modify their physical and chemical characteristics e.g. their water absorption efficacy. Therefore, correction of annual mean data requires several assumptions to be made.

Acknowledgements

Thanks to Dr David Harrison and Dr Richard Maggs from Bureau Veritas for providing the data used in this analysis and for their input in clarifying data collection procedures etc. Thanks also to Dr Paul Quincey (NPL) for information on water absorption on quartz filters. Thanks to Gary Fuller, David Green and Ben Barratt for their helpful comments and the following data: $PM_{2.5}$ TEOM data for Bexley, FDMS data for North Kensington and Emfab (Earls Court) data. The provision of AEA filter blank data from Tony Clark is much appreciated.

References

[1] D.C. Carslaw, K. Ropkins, and M.C. Bell. Change-point detection of gaseous and particulate traffic-related pollutants at a roadside location. *Environ. Sci. Technol.*, 40(22):6912–6918, 2006.

[2] R. B. Cleveland, W. S. Cleveland, J.E. McRae, and I Terpenning. STL: A seasonal-trend decomposition procedure based on loess. *Journal of Official Statistics*, 6(1):3–73, 1990.

[3] D. Harrison and R. Maggs. Interim report in response to increasing PM concentrations. Technical report, Bureau Veritas, 2008.

[4] H. R. Kunsch. The jackknife and the bootstrap for general stationary observations. *Annals of Statistics*, 17(3):1217–1241, 1989.

A. Tables of results

Table 2: Summary blank data by month for both PM_{10} and $PM_{2.5}$ sites ($\mu g\ m^{-3}$). Dates are based on the date filters were placed in the sampler.

date	mean	minimum	maximum	σ	n
Dec-00	2.58	2.58	2.58		1
Jan-01	1.42	1.42	1.42		1
Feb-01	1.17	0.96	1.38	0.30	2
Mar-01	−0.19	−1.67	0.58	0.64	15
Apr-01	1.40	−2.21	4.71	2.48	5
May-01	1.08	0.75	1.54	0.31	5
Jun-01	0.27	−3.25	2.46	2.56	4
Jul-01	0.96	−0.38	2.21	0.76	19
Aug-01	1.43	−0.58	3.88	1.05	35
Sep-01	1.65	−0.96	3.83	1.79	5
Oct-01	0.83	−0.92	3.79	2.08	4
Nov-01	0.11	−0.92	2.08	0.90	15
Dec-01	2.69	1.04	4.08	0.72	14
Jan-02	1.23	−4.25	7.71	2.88	50
Feb-02	−0.65	−6.13	1.96	1.44	42
Mar-02	−0.45	−4.29	1.96	1.39	48
Apr-02	3.12	0.88	3.96	1.49	4
May-02	1.52	1.00	2.58	0.51	7
Jun-02	3.10	1.75	3.83	0.71	8
Jul-02	4.90	3.71	5.79	1.07	3
Aug-02	0.51	−1.21	2.50	0.98	17
Sep-02	1.11	−6.75	5.42	2.03	49
Oct-02	1.93	−6.46	5.75	1.69	48
Nov-02	1.58	−1.63	4.46	1.57	42
Dec-02	0.95	−1.96	7.50	1.77	34
Jan-03	0.15	−4.29	4.04	1.60	34
Feb-03	2.21	−0.21	3.79	1.00	13
Mar-03	0.48	−1.54	3.42	1.49	12
Apr-03	2.54	−0.29	5.50	1.88	9
May-03	4.91	−0.33	10.33	2.03	22
Jun-03	5.42	3.13	8.17	1.55	21
Jul-03	5.13	2.00	8.63	2.35	15
Aug-03	1.82	−0.79	6.17	1.91	15
Sep-03	0.56	−1.13	3.04	1.00	30
Oct-03	1.73	−0.79	3.54	1.32	17
Nov-03	1.60	−0.33	3.63	1.18	11
Dec-03	1.46	−0.13	3.71	1.04	11
Jan-04	1.33	−0.67	2.33	0.75	17
Feb-04	0.68	−0.46	2.54	0.91	10
Mar-04	0.93	−2.67	3.21	1.35	38
Apr-04	1.06	−1.96	3.88	1.60	26
May-04	2.11	−3.08	5.63	1.77	53
Jun-04	1.37	−1.04	3.50	1.37	24
Jul-04	3.18	1.00	4.33	0.85	11
Aug-04	2.80	−0.04	6.54	1.38	28
Sep-04	1.89	−0.79	4.21	1.28	16
Oct-04	0.55	−0.67	1.87	0.73	13
Nov-04	0.15	−1.04	1.92	1.17	10
Dec-04	−0.63	−2.37	1.92	1.00	11
Jan-05	0.68	−2.75	3.50	1.08	52
Feb-05	−0.63	−5.17	3.67	1.85	25
Mar-05	1.22	−1.08	1.96	0.86	11
Apr-05	1.08	−2.58	6.83	1.65	44
May-05	4.28	0.83	13.29	3.09	19
Jun-05	4.48	−1.04	8.42	2.02	56
Jul-05	2.80	1.21	4.33	1.00	18
Aug-05	2.27	−5.00	5.63	2.63	30
Sep-05	−0.10	−4.71	2.83	1.57	51
Oct-05	0.31	−3.38	4.54	1.70	22
Nov-05	−0.23	−4.12	2.83	1.85	39
Dec-05	1.84	0.33	3.38	0.71	35
Jan-06	1.37	0.33	3.88	0.91	26
Feb-06	1.05	−4.46	4.33	1.54	28
Mar-06	1.24	−1.58	3.75	1.27	50
Apr-06	−0.24	−7.42	6.38	2.94	28
May-06	6.01	0.08	13.79	4.09	24
Jun-06	6.84	2.13	12.62	3.52	14
Jul-06	5.20	0.46	13.37	3.51	38
Aug-06	6.55	1.29	11.50	2.40	31
Sep-06	4.35	1.21	6.54	1.56	26
Oct-06	3.49	1.29	5.08	1.29	13
Nov-06	4.08	1.96	6.04	1.19	12
Dec-06	3.33	−0.25	6.00	1.70	39
Jan-07	2.23	−4.25	11.62	3.89	38
Feb-07	10.48	6.50	15.83	3.63	5
Mar-07	6.20	2.13	9.17	1.74	15
Apr-07	5.96	3.88	8.29	1.63	8
May-07	5.92	3.25	10.63	1.69	22
Jun-07	5.64	2.21	9.00	1.69	19
Jul-07	6.52	4.54	8.04	1.17	14
Aug-07	4.94	1.25	8.21	1.37	41
Sep-07	4.08	0.00	6.88	1.43	34
Oct-07	5.20	3.25	7.96	2.00	4
Nov-07	2.86	−3.42	9.79	2.47	28
Dec-07	2.51	−2.96	7.54	3.14	10
Jan-08	0.67	−2.00	3.04	1.71	17
Feb-08	1.69	−1.04	5.21	2.24	13