Predicting black smoke levels from deposit gauge and SO_2 data to estimate long term exposure in the United Kingdom, 1956-61

Susan Hodgson[a], Fu-Meng Khaw[a,b], Mark S. Pearce[a], Tanja Pless-Mulloli[a]

[a] Institute of Health and Society, Newcastle University, Newcastle upon Tyne, UK

[b] North East Health Protection Unit, Health Protection Agency North East, Newcastle upon Tyne, UK

e mail addresses of the authors:

Susan Hodgson – susan.hodgson@ncl.ac.uk

Fu-Meng Khaw - f.m.khaw@ncl.ac.uk

Mark S. Pearce - m.s.pearce@ncl.ac.uk

Tanja Pless-Mulloli - tanja.pless-mulloli@ncl.ac.uk

Corresponding author:

Dr Susan Hodgson

Lecturer in Environmental Epidemiology

Institute of Health and Society

4th Floor William Leech Building

The Medical School

Newcastle University

Newcastle upon Tyne NE2 4HH

Tel: +44 (0) 191 222 3823

Fax: +44 (0) 191222 8211

e mail: susan.hodgson@ncl.ac.uk

Abstract

Background: In the UK air quality has been monitored systematically since 1914, providing valuable data for studies of the long-term trends in air pollution and potentially for studies of health effects of air pollutants. There are, however, challenges in interpreting these data due to changes over time in the number and location of monitored sites, and in monitoring techniques. Particulate matter was measured as deposited matter (DM) using deposit gauge monitors until the 1950s when black smoke (BS) filters were introduced. Estimating long-term exposure to particulates using data from both deposit gauge and BS monitors requires an understanding of the relationships between DM, SO_2 and BS.

Aims: To explore whether DM and/or SO_2, along with seasonal and location specific variables can be used to predict BS levels.

Methods: Air quality data were abstracted from hard copies of the monthly Atmospheric Pollution Bulletins for the period April 1956 - March 1961 for any sites with co-located DM, SO_2 and BS data for three or more consecutive years. The relationships between DM, SO_2, and BS were assessed using mixed models.

Results: There were 34 eligible sites giving 1521 triplets of data. There was a consistent correlation between SO_2 and BS at all sites, but the association between DM and BS was less clear and varied by location. Mixed modelling allowing for repeat measurements at each site revealed that SO_2, year, rainfall and season of measurement explained 72% of the variability in BS levels.

Conclusions: SO_2 can be used as a surrogate measure for BS in all monitoring locations. This surrogate can be improved upon by consideration of site specific characteristics, seasonal effects, rainfall and year of measurement. These findings will help in estimating historic, long-term exposure to particulates where BS or other measures are not available.

Keywords

Black smoke, Sulphur dioxide, deposit gauge, air pollution, exposure assessment

Introduction

Adverse health effects associated with exposure to air pollution are well documented. Short term increases in mortality and morbidity from respiratory and cardiovascular diseases have been consistently observed following air pollution episodes and in time series studies (Anderson et al. 2005; COMEAP 2006; Le Tertre et al. 2002). Furthermore, prospective studies have shown that prolonged exposure to air pollutants can lead to a reduction in life expectancy (Laden et al. 2006; Pope et al. 2002; Pope and Dockery 2006).

Studies of long term health effects as a result of cumulative exposure require good estimates of long-term personal exposure. As personal monitoring data cannot be collected retrospectively, such long term exposure estimates have to use data from nearby air pollution monitors as a starting point. When these air monitoring data are combined with residential histories and pollution source information, cumulative long-term exposures can be modelled and estimated for individuals.

In the UK, air quality has been monitored and reported systematically since 1914 (Department of Scientific and Industrial Research 1946), and as a result there is a potentially rich source of data available for studies of the long-term health effects of air pollutants. Data from the early air quality monitoring in the UK are also potentially valuable as levels experienced during that time in the UK are currently prevalent in many mega cities of developing countries (Bell et al. 2006; Bi et al. 2007; Braga et al. 2005; Karar et al. 2006; Wang et al. 1997). Although this wealth of historical air quality data is available there are difficulties in interpreting these data due to changes in monitoring techniques over time. Moreover, since the inception of the national monitoring network, the number of monitored locations has increased and the locations of monitoring stations have changed over time.

Up until the 1950s particulate matter was measured using deposit gauge (DG) monitors. This measurement involved exposing a standard deposit gauge for a period of one month before the solid (insoluble) and liquid (soluble) fractions were examined. These soluble and insoluble particles, when combined, gave a measure of 'deposited matter' (DM) expressed as tons of deposit per square mile (Department of Scientific and Industrial Research 1955). Over this period SO_2 levels were measured using the 'lead peroxide' method. For this measurement, a small cylinder, or 'candle', coated with lead peroxide was exposed to the air for one month. The lead peroxide oxidised ambient sulphur dioxide to sulphate, and the milligrams of sulphur trioxide per day per 100cm^2 of standard lead peroxide provided a comparative measure of SO_2 (SO_2(LP)) (Department of Scientific and Industrial Research 1955). The 1950s saw a radical change in the measurement of particulate matter, from DG monitors to black smoke (BS) filters. In this method, a known volume of air was drawn through a standard intake and through a white filter paper. The amount of smoke collected was estimated using the reflectance of particles (mainly elemental carbon)

trapped on the filter which could be converted into a measure of mass concentration expressed as milligrams of smoke per 100m^3 (Department of Scientific and Industrial Research 1955). A volumetric method was also utilised to measure acid-producing gases (excluding carbon dioxide) in the air, calculated and reported as SO_2 (net-acidity). In this method, a measured volume of air was passed through a solution of hydrogen peroxide over a period of 24 hours. The amount of acid collected was assessed via titration with alkali to provide a measure of sulphur dioxide expressed in milligrams per 100m^3 (Department of Scientific and Industrial Research 1955). By the late 1960s, many DG monitors were replaced by BS filters and whilst BS and SO_2 are still measured in the UK today by a network of 156 monitors (Loader et al. 2004), the use of DG monitors is now largely confined to the measurement of nuisance dust on an ad hoc basis. If estimates of long-term exposure to particulates rely on data from both DG and BS monitors, the association between the DM, SO_2 and BS measures requires exploration.

As part of an epidemiological study looking at air pollution and childhood respiratory infections in a cohort of 1142 babies born in Newcastle upon Tyne in 1947 we needed to model long term air pollution exposure at a high temporal resolution over the period 1947-1962. This period encompasses the change-over from DG to BS monitoring techniques for particulates, and from lead peroxide to net acidity testing for SO_2. Obviously any exposure model needs to take these changes into account. Ideally, we wanted to be able to satisfactorily predict BS levels at sites where this pollutant was not measured, and to be able to back-date BS levels for the late 1940s and throughout the 1950s when these data were not routinely collected.

In the present study we explored the relationships between DM, SO_2, and BS to ascertain whether DM and/or SO_2, along with other seasonal and location specific variables may be used to predict BS levels. To achieve this aim, data from co-located DG, BS and SO_2 monitors from across the UK over the period 1956-1961 were evaluated.

Methods

UK Air quality data were obtained from monthly Atmospheric Pollution Bulletins available as paper records for the period April 1956 - March 1961. These years were chosen as it was during this time that DG monitors were being replaced by BS filters and as a result monitors of DG, SO_2 and BS were most frequently co-located during this period; by 1961 BS and SO_2 (net-acidity) were routinely monitored on a national scale (UK Air Quality Archive a).

All sites with co-located DG monitors (for soluble/insoluble particles), SO_2 monitors (lead peroxide method) and BS monitors were identified. SO_2 levels measured by the lead peroxide (LP) rather than the net acidity method were of particular interest because the LP data is available for the period preceding the BS data. Monthly averages of SO_2 (LP) and BS, and monthly totals of

soluble/insoluble particles and rainfall levels were abstracted from sites where data was available for three or more consecutive years. Due to months with missing data for one or more of these parameters, the number of triplets of data available for analysis is less that the total number of months during which the co-located monitors were active. Parameters quoted as monthly totals (rainfall, insoluble/soluble particles) were computed as daily averages to enable comparison with the BS measure (presented as a monthly average based on the mean of daily average figures). The level of DM was calculated as the sum of insoluble and soluble particles.

The precise geographical locations of monitored sites were obtained from the UK National Air Quality Archive (AQA) (UK Air Quality Archive b). Monitor locations were classified into 15 categories (assigned by AQA) which characterised each site according to proximity to residential, industrial, urban and rural areas (Table 1). In addition, sites were classified as to whether or not they fell within a local authority 'smoke control zone'. Such control zones were established under the Clean Air Act 1956 to help the UK to meet air quality standards for SO_2 and particulates set by European law, and to avoid 'killer episodes' such as those experienced in London in the 1950s (Brimblecombe 1987a). Within these zones it is an offence to cause smoke to be emitted from a chimney of a building (UK Parliament 1956).

The relationships between DM, SO_2, and BS, with BS as the dependent variable, were assessed using mixed models to take into account the repeated observations made at each monitor site. The influences of other variables, namely year, season, rainfall, location in smoke control zone, and classification of monitor location were assessed for main and interactive effects.

To assess the validity of predicted BS levels at independent monitoring locations, BS, SO_2 and DM levels were abstracted for the period April 1956 - March 1961 for an additional 30 sites with less than three years of continuous data (511 triplets of data). To assess the validity of predicted BS levels for the period prior to April 1956, co-located data, where available, were abstracted for the original monitoring sites from April 1954 - March 1956 (354 triplets of data).

All statistical analyses were performed using the statistical software package SPSS 15.0 (SPSS Inc., Chicago).

Results

There were 34 co-located sites with data for three or more consecutive years. A total of 1521 triplets of co-located data were obtained. The sites were distributed throughout 11 English counties and 2 Scottish regions (based on 1991 boundaries), spanning an area of 520km (North-South) by 285km (East-West), see Figure 1.

There was a wide variation in the measured levels of DM, BS and SO_2 within and between sites as shown in figures 2 a, b and c. Seasonal variation was more pronounced in the levels of BS and SO_2 than DM, with the highest levels being recorded during the winter months (figures 2 d, e and f). The mean BS concentration over this period was 179µg/m^3 (range 10-1480µg/m^3), which is considerably higher than levels measured today (e.g. for 2007, mean BS levels from the 13 monitors in the UK were 9.9µg/m^3 (range 0-111µg/m^3) (UK Air Quality Archive c)).

Sites located in a smoke control zone had significantly higher levels of DM and SO_2 than sites not located in these zones (mean DM 0.55 versus 0.47 tons/sq mile/day, $p<0.01$; mean SO_2 2.1 versus 1.7 mg/100 cm^2/day, $p=<0.01$). However, this difference was not observed in levels of BS.

The AEA classifications of monitor location were associated with differing levels of pollution, with rural and medium-density residential areas having lower levels of all three pollutants than high-density residential, commercial and industrial areas, although variability was lost if the sub-categories were crudely pooled into these groupings. As a result, dummy variables were created for each subcategory for inclusion in the linear regression, with the 'unclassified sites, or mixed area' classification acting as the reference category.

Pearson correlation coefficients were used to explore the associations between levels of DM, SO_2 and BS. Using pooled data from all sites there was a significant positive correlation between DM and BS (Pearson correlation coefficient 0.29, $p<0.01$), and between SO_2 and BS (Pearson correlation coefficient 0.59, $p=<0.01$). When sites were assessed individually the correlation between SO_2 and BS persisted for each site (range of Pearson correlation coefficients 0.3-0.9, p values <0.05). The correlations between DM and BS varied between sites; at 12 out of the 34 sites a significant positive correlation was seen, at two sites a significant negative correlation was observed, and at the remaining sites non-significant positive correlations (11 sites) or negative correlations (nine sites) were observed (Figure 3). There was no obvious explanation for these different DM/BS correlations in terms of presence within a smoke control zone, AQA monitor location category, or sites with very high or low levels of pollution.

Mixed modelling revealed that SO_2, year, rainfall and season of measurement explained 72% of the variability in BS levels; there was a significant association between SO_2 and BS after adjustment for year, rainfall and season (B=56.84; 95% CI, 49.69-63.99) (Table 2). There was also a significant interactive effect between SO_2 and year, and SO_2 and season, although these additional parameters only explained an additional 1% of the variability in BS levels. Inclusion of the classification of monitor locations or location within a smoke control zone did not explain any additional variability in BS levels.

The validity of using the above variables to predict BS levels was assessed using additional co-located data abstracted from the Atmospheric Pollution Bulletins over the period April 1954 - March 1961. When SO_2, year, rainfall and season of measurement were used as predictors of BS levels, the correlation between the measured and predicted BS was 0.59 ($p<0.01$) and 0.57 ($p<0.01$) for the independent monitoring locations and earlier time period respectively, with these parameters explaining 35% and 33% of the variability in BS levels. The predicted BS levels tended to underestimate the measured BS levels, especially where higher concentrations were recorded (figure 4).

Discussion

The relationships between DM, SO_2, and BS in co-located sites have not been reported before. In this paper we assessed the relationships between these pollutants to ascertain whether DM and/or SO_2 may be used as a predictor of BS. We have shown that SO_2 is a better predictor of BS levels than DM, and that a simple model incorporating SO_2, year, rainfall and season of measurement can be used to predict BS levels at all monitoring locations and years. The relationships we have observed between BS, SO_2 and DM will enable us to reconstruct historic air pollution exposure at a high temporal resolution for a birth cohort established in 1947 and still ongoing (Lamont et al. 1998). This opens up the possibility of assessing the impact of early life and life-course exposures on health in infancy and later life in this and other historic UK cohorts, and as BS is still routinely monitored in the UK and elsewhere, findings will be able to be compared with more recently established cohorts. There are however several limitations to this study that need to be considered.

Although BS was correlated with SO_2 at all sites, just over two thirds of the variability in BS could be explained by SO_2 and the other variables in the model, suggesting that one or more influential parameters exist that we were not able to be take into account. The explanatory power of our relatively straightforward model (Pearson correlation co-efficient = 0.85, $p<0.01$, $R^2 = 0.72$), which utilises readily available data to fill gaps in the data series, is similar to other air pollution models. For example Abbey, 1995, extended an air pollution time series for California, USA, using total suspended particle data to estimate levels of PM10 from 1973 to 1987. The correlation between estimated PM10 and monitored PM10 was 0.93, with an R^2 of 0.86 (Abbey et al. 1995). Filleul, 2002, also extended a pollution time series using data from independent monitoring sites to predict monthly BS levels in Bordeaux, France. BS levels were estimated up to 1997, with correlation co-efficients between the measured and the predicted BS levels ranging from 0.43 to 0.88 at four sites (Filleul et al. 2002). Land-use regression models have also been used to characterize air pollution exposure, using variables such as road type, traffic count, elevation, and land cover rather than co-

located air pollution data. A recent review indicted that such models produce R^2 values of between 0.54 to 0.81 (Ryan and LeMasters 2007).

The usefulness of our model to predict BS levels outside of the period April 1954 – March 1961 is untested. As the model underestimates BS where higher levels of this pollutant are observed, the validity of the predicted BS levels are likely to be compromised at sites and/or times where BS levels are known to have been high. We acknowledge that the composition and quantity of particulate pollution has changed over time (as discussed below), and therefore advise caution in applying this model to years much beyond this period.

There were some monitoring sites where the observed DM and BS levels were negatively correlated, against expectation. We were unable to satisfactorily explain this finding in terms of known monitor characteristics, but it is likely that deposits from DG monitors often reflected material of a very local origin, and as such these monitors did not necessarily represent pollution over a wider area (Brimblecombe 1987b). Whether there were pertinent local conditions or pollution sources at these sites that may have led to high or low DM levels without a corresponding decrease or increase in BS levels is not known.

When BS measurement methods were introduced in the UK in the 1950s and 1960s, suspended particulate matter in the atmosphere was dominated by carbonaceous combustion products from domestic and industrial coal burning. Since then, fuel types and pollution sources have changed over time, affecting the size distribution, composition and toxicity of particulates (Anderson et al. 2001; Brunekreef and Holgate 2002). The formulae used to transform BS reflectance to mass concentrations were established based on a different composition of the air pollution mix, and as such BS figures could only really be interpreted as an indication of elemental carbon, not as mass concentrations (Roemer and van Wijnen 2001). As a result the reflectance method used for measuring BS has been superseded by gravimetric techniques that allow different size fractions of particulates to be assessed (Gotschi et al. 2002). The emphasis in monitoring has moved progressively from SO_2 and BS to pollutants such as ozone, nitrogen dioxide and particulate matter (PM) (UK Air Quality Archive a). Although measurements techniques have progressed, BS is likely to provide an appropriate proxy of particulate matter. The size of particles sampled by a black smoke filter has been reported to be below 5μm (Chow 1995) or 4.4μm (McFarland et al. 1982), and as such BS is likely to represent an important active component of both PM10 and PM2.5 (Anderson et al. 2001; Gotschi et al. 2002; Le Tertre et al. 2002) and may provide a better estimate of particles generated by traffic than PM (Roemer and van Wijnen 2001).

We have shown that it is possible to model long term exposure to BS using data on SO_2 levels, rainfall, year and season, but any resultant exposure measure will only be of use if BS is itself an

exposure of interest, or at least a good proxy of an exposure of concern. Recent evidence does suggest a correlation between BS and various adverse health outcomes (for example Brunekreef et al. 1997; Le Tertre et al. 2002; Schwartz et al. 2001; Timonen et al. 2002; van Vliet et al. 1997; Verhoeff et al. 1996).

Current evidence suggests that BS can provide a useful measure of ambient elemental carbon and of traffic related particulates in the UK, and in terms of health effects is likely to be an acceptable proxy for relevant particulate exposure. Although gravimetric techniques are now favoured for the determination of particulate levels, BS data remains a useful measure of particulate pollution for several reasons. Firstly, in the UK, BS data are available for a much longer period of time (over 30 years), and at more locations than other particulate measures (Solomon et al. 2003). Secondly, where health effects are primarily attributable to traffic exhaust, BS, which is dominated by diesel particles, may be a better proxy for particulate exposure than PM10 (Le Tertre et al. 2002). Thirdly, although gravimetric methods can separate out size fractions of PM they do not provide information on particle characteristics such as elementary composition, primary or secondary formation and emission sources; the importance of these characteristics in determining health effects is critical in formulating regulations that will best prevent ill health (Gotschi et al. 2002).

We have shown a clear relationship between SO_2 and BS, but the association between DM and BS is more complicated. We suggest that SO_2 may be used as a surrogate measure for BS in all monitoring locations, and that this surrogate can be improved upon by consideration of site specific characteristics, seasonal effects, rainfall and year. The findings from this study will help in estimating long-term exposure to particulates where BS or other measures of particulates are not available.

Acknowledgements

We are grateful to Nick Clement and Jane Tilbrook for abstracting the data from the hard copies of the monthly Atmospheric Pollution Bulletins.

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Figure 1: Map showing location of co-located monitoring sites with data for three or more consecutive years over the period 1956-1961

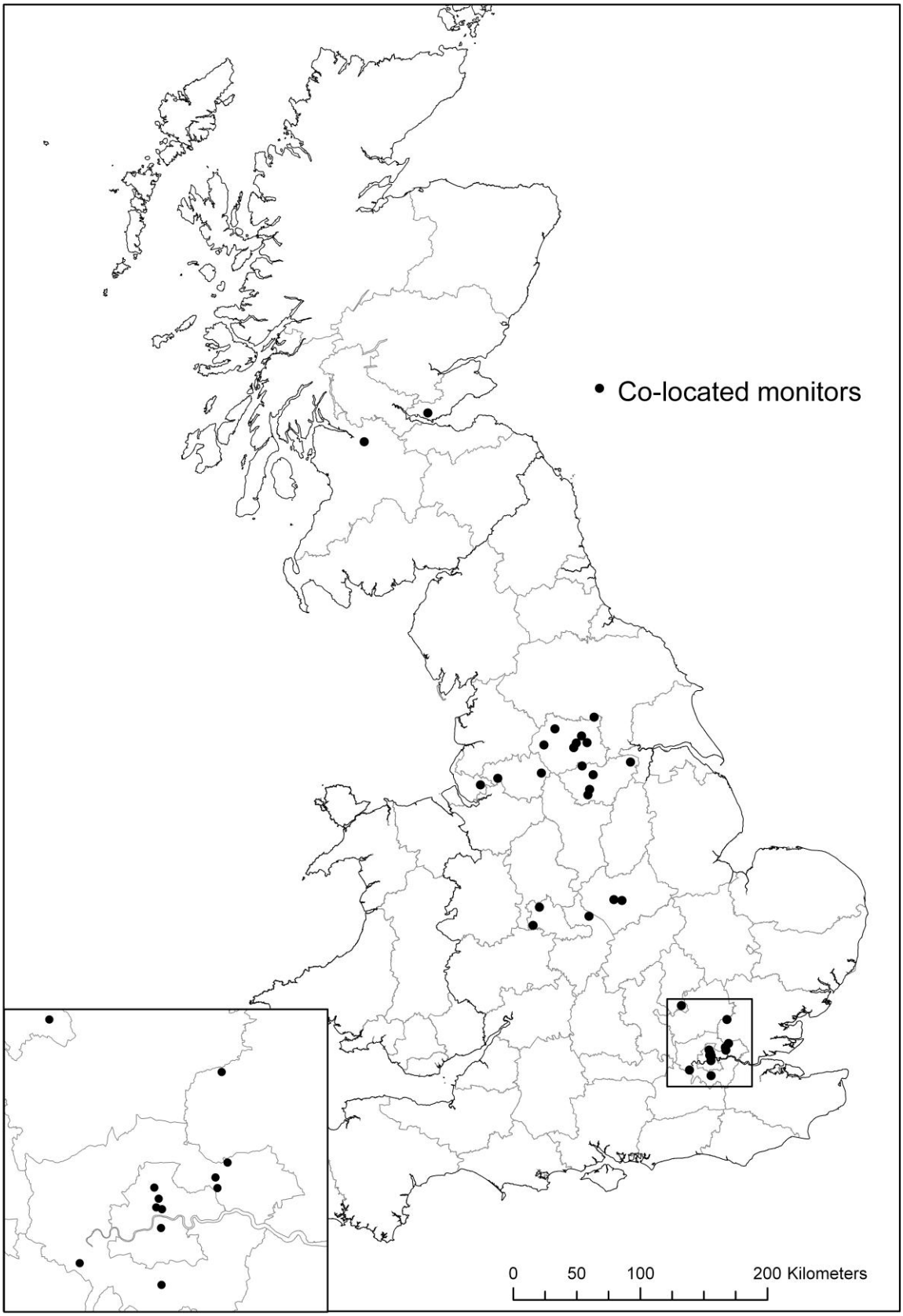


Figure 2: Box plots showing variation in measured levels of a) DM, b) BS and c) SO_2 within and between the monitoring sites. Seasonal variation in measured levels of d) DM, e) BS and f) SO_2 pooled across the sites.

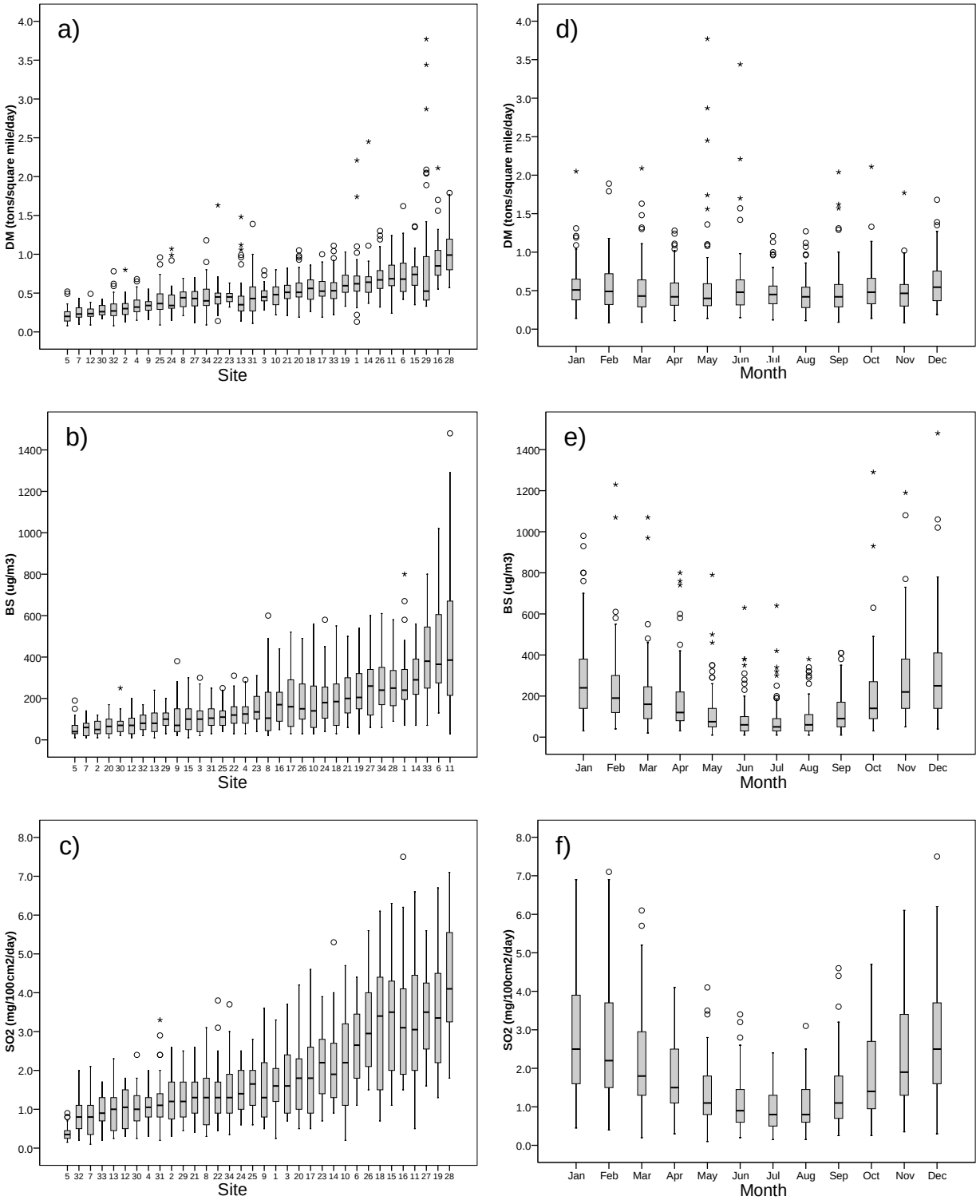
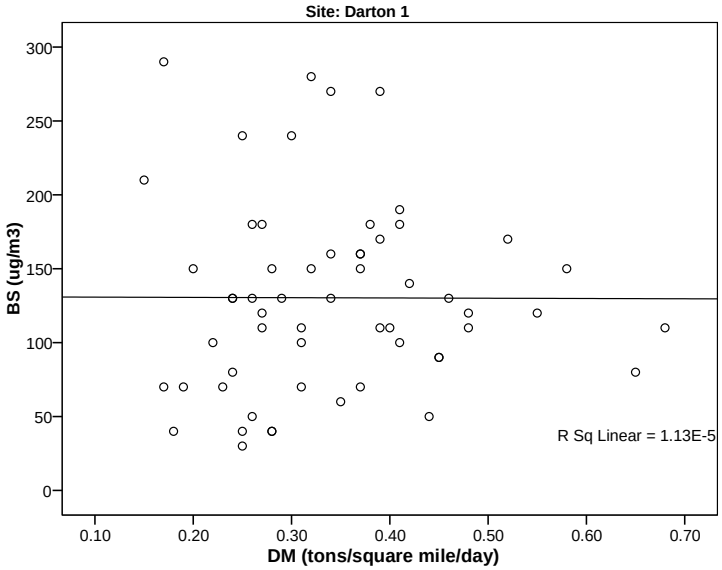
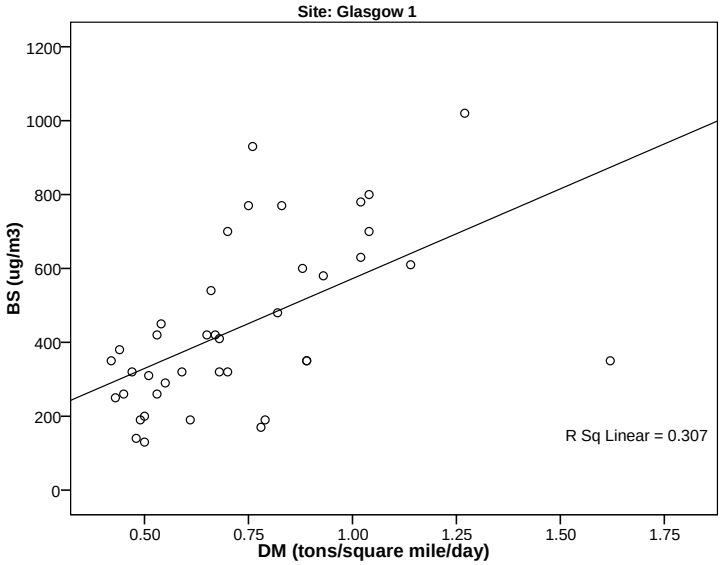


Figure 3: Scatter plots of DM and BS at three monitoring sites to show the variation in correlations observed between these two variables at individual monitoring sites.





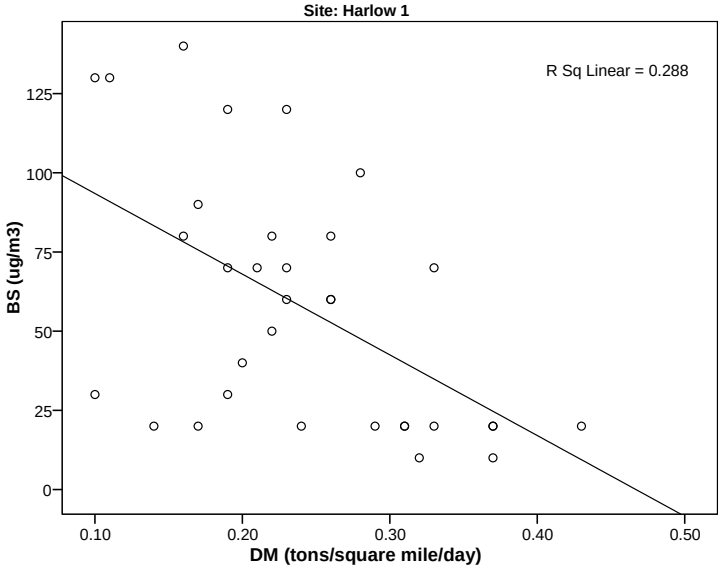


Figure 4: Measured BS versus predicted BS (predictors SO_2, rainfall, year and season) for a) independent monitoring locations, b) earlier time period (April 1954 to March 1956).